Summary of the PhD thesis

Phase and polarization changes of pulsed Gaussian beams during focusing and propagation

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1 Introduction

Light is an essential element for life, and not just by reason of being the primary energy source of Earth's flora, and through plants and unicellular organisms of its whole fauna, but also by serving as the most important intermediary agent between our planet's residents and their habitat. Visible light is, of course, just a small segment of the broad set of electromagnetic waves, and the other slices of the spectrum are at least as important as the optical regime, but light plays a fundamental role, as seeing is our dominant method for observation, not only by accident. Instead, it is evident, as the physical, chemical and biological mechanisms, which we encounter in our everyday life, are accompanied by the emission or absorption of light. As such, observing the interaction of light and matter gives us an evident way to study nature and its processes scientifically. Necessarily, to carry out these investigations effectively, we have to know the participants of this interplay well enough.

In 1960, just a few decades after the birth of quantum theory and a whole new bunch of questions, a new experimental device, the *laser* was created. This new appliance became one of the most important tools to further broaden our knowledge of light and matter, or the field ultimately connecting them, quantum science. The laser, the apparatus which emits light with extraordinary properties, has since been further developed to meet the demands of curious scientists who aim to gain insight on processes taking place in biological samples, chemical compounds or nanometer-size material structures.

One of the exceptional features of lasers, that scientists frequently utilize, is the ability to produce packets of light with extremely short duration. The importance of this capability lies in the fact that it opened the gate to electric field strengths and time scales which were unavailable before the laser came on scene. When the laser beams are *focused*, these extreme field strengths can even overwhelm the ones that the electrons feel in the vicinity of the atomic core. This physical feature, the unprecedented electric field strength, is the one that is usually utilized in the experiments of biology, physics, material science or medicine.

Now going back to the thought that emerged earlier, that is, to use light as a tool for our investigations we have to know its properties well enough, one influential question arises here: how does focusing affect pulsed beams? Does focusing distort the precisely set properties of the laser pulse?

2 Scientific background

Laser technology has substantially developed since the first demonstration in 1960. An aim of these advancements is to produce shorter and shorter pulses of light with as high power as possible. As a result of the technological progress, pulses of a few femtosecond ($1\,\mathrm{fs} = 10^{-15}\,\mathrm{s}$) duration can be produced by using laser oscillators or amplifiers nowadays. This means that the wave packet, which leaves the laser, contains only a few oscillations of the electric and magnetic fields. For this reason, these pulses are often termed as "few-cycle". These electromagnetic fields make it available to study nonlinear optical phenomena, or the explosively developing field of "attophysics". At the same time, they provide better pattern quality during material processing, or higher resolution in optical imaging techniques. Pulsed sources of other wavelength regimes (like terahertz waves or coherent radiation in the far ultraviolet) are usually also driven by laser sources. Furthermore, this laser technology made it possible to initiate the research topic of femtochemistry, found to be worth a Nobel prize in 1999.

The reason for the widespread application of lasers is that the light pulses they produce can be very well controlled and their parameters can be accurately set to the needs of the specific experiments. One of these relevant parameters is the so-called carrier-envelope phase (CEP). The value of the CEP is most relevant in the studies using few-cycle pulses, where the experimental outcome heavily depends on the exact temporal variation of the electric field. This was realized shortly after the appearance of the first few-cycle pulses, leading to many studies also recently.

Another property of electromagnetic pulsed waves, which is getting large attention nowadays as an important controlling parameter for experiments, is their polarization. While the pulses emitted by the oscillators or amplifiers are usually linearly polarized, wave packets with other polarization states (like circular), or even with a polarization that is time-dependent, can be produced. As mentioned earlier, the laser beams are focused in most of the applications. As a result, an important question here is whether the polarization state or the CEP of pulsed waves change upon focusing, and if yes, what kind of modifications they suffer. To answer these questions, firstly, a short review is given on the theoretical methods to describe ultrashort laser pulses and their properties. Then a brief overview is presented on the models of focusing and on the experimental methods that allow us to study few-cycle pulses.

2.1 Mathematical description and properties of ultrashort pulses

One of the most important steps for the mathematical description of ultrashort pulses is the Fourier transform. This mathematical transformation — which also enlightens the nature of pulsed waves — allows us to construct our temporally restricted wave packet from infinitely long, monochromatic components of different frequencies according to Fourier's law. This way Fourier transformation makes a connection between the temporal shape and the spectral representation of the electric field. The Fourier theory is used to study pulsed waves because monochromatic waves can be treated easier theoretically, description of their propagation properties are well-known, and this mathematical procedure is what makes it possible to solve the wave equation in many cases.

The solution of the propagation problem of wave packets can be obtained with the following three main steps. First, the pulse is decomposed into its monochromatic components using Fourier transform. Then the propagation of each monochromatic wave is calculated individually. Finally, the temporal shape of the electric field is reconstructed from the components in the point of interest by applying the inverse Fourier transform. The step which needs most attention is the second one describing wave propagation. The way this problem is treated depends on the approximations which can be used, the aspects which we are interested in, or the conditions involved. A short description of these methods are given later in this text, while the following few paragraphs focus on different parameters of electromagnetic pulses.

The carrier-envelope phase The expression describing the temporal variation of the electric field is often expressed as the product of a slowly-varying envelope and a rapidly changing carrier term. While the exact definition of the CEP can vary depending on the exact topic, it is generally true that it describes the relative phase of the carrier with respect to the envelope. Here a CEP definition is used which describes a single pulse, and not a series of pulses. According to this description, *the CEP of a single pulse is the phase of the wave packet at the instant of time when the envelope reaches its maximum.* This sentence is not just a definition, it also gives a numerical method to evaluate the CEP of any pulse shape, and it makes the possibility to generalize the meaning of CEP, in a physically meaningful way, even for pulses with arbitrary polarization state. When the light-matter interaction happens in a larger volume, or in planes not coinciding with the focal plane, it is important to know the CEP changes of pulses while they propagate from the focusing element to the interaction point.

Polarization Another relevant property of electromagnetic waves is their polarization. To represent the polarization state of ultrashort pulses, the expressions for the same characteristics of monochromatic waves can serve as a basis. It is important to note here that, in general, it needs special attention if someone wants to describe polarization of focused fields. It is due to the fact that only plane waves are purely transverse waves, and beams — or especially focused beams — can have longitudinal component of the electric field, that is, there is nonzero field component in the direction of propagation. Fortunately, in most of the cases, the paraxial approximation can be used, where this longitudinal field component can be omitted. As Gaussian beams are solutions of the paraxial wave equation, they can also be treated as transverse waves, like plane waves. Of course, focusing can result in situations when the paraxial approximation is not valid anymore. The benefit of paraxial approximation is that the polarization state of beams can be described the same way as that of plane waves, they can be given as two perpendicular, oscillating, scalar fields. One of the approaches to give the polarization state of transverse waves is the description by the polarization ellipse. As the perpendicular oscillations, in general, describe a trajectory with elliptical shape, the properties of this ellipse (orientation, ellipticity and semi-major axis size) can be used to depict the polarization state. An other characterization is the one using Stokes parameters, which expression form is often used in experiments. This is because they give their measurement method by their definition. Of course, the two descriptions — the one with the polarization ellipse and the other with the Stokes parameters — can be converted into each other.

Compared to the polarization state of monochromatic waves, the main difference for pulses is that their polarization can be time-dependent. There are more and more experiments where this feature is exploited. For example, it is a tool for the method called coherent control. It can be shown that both the description by the polarization ellipse and the one by Stokes parameters can be extended to represent time-dependent polarization. One has to be careful, however, as the "instantaneous" polarization ellipse has less meaning than the time-independent one. The instantaneous polarization ellipse represents the changes of the instantaneous electric field under phase transformations, and it does not give the trajectory which the electric field traverses as time passes. In the case of Stokes parameters, for instantaneous portrayal, the first Stokes parameter looses its meaning, as unpolarized field can not be interpreted. However, both descriptions give a unambiguous way to depict the instantaneous polarization state, so the changes during propagation or focusing can be studied using them.

Phase and group velocity The phase velocity is relevant in nonlinear optics, as — through phase matching — it affects the efficiency of several processes studied in this field of science. For plane waves, the phase velocity can be given by very simple formulas, while the same for beams needs more effort. Phase velocity gives how fast the points of equal phase travel. The shape of these surfaces, given by a constant phase value, differ from a plane in general. The spatial phase of beams vary not just in the direction of propagation, but also perpendicular to it, giving an important distinction from the case of plane waves. An interesting consequence of this is the velocity of the phase front of a focused beam, which can be bigger than the speed of light, due to Gouy's phase shift. This, however, does not contradict special relativity, as the velocity of the phase does not give the speed with which information travels. It means an issue in nonlinear optics, however, as in the vicinity of focus the previously mentioned phase matching can be destroyed.

The group velocity gives how fast the wave packet travels. For beams it can be calculated using a more complex expression than for plane waves, and it can have a superluminal (faster than light) value in the vicinity of focus, like the phase velocity. This still does not contradict causality, because the speed of energy propagation (related to the information carried) does not coincide necessarily with group velocity. The importance of the group velocity in this work lies in the fact that, together with phase velocity, it influences the CEP of the pulsed wave.

2.2 Theoretical description of beam propagation and focusing

The size of wave packets originating from lasers are restricted not just in time, but also in space. This way the description of their propagation is much more complicated than that of plane waves. In the field of optics, it is a huge advantage that, due to the short wavelength, simplifications can be introduced regarding the model used for the propagation of light. Still, focusing, for example, means a case that can not be treated properly this way. In the following, a few methods are briefly summarized which mean a way to do the second step — the one between the two Fourier transforms — of the three-part approach to describe pulsed-beam propagation, given before in this text.

Ray tracing A widely used and robust way to model light propagation is ray tracing, a method of geometrical optics. In this approach, light is modeled by rays, which travel along straight lines until they reach a spatial region of different refractive index. Along the boundary of such regions, the change in propagation direction can be calculated using Snell's law or the law of reflection. Due to the simple rules and few parameters, this way even complex optical systems can be modeled with relatively low computational resources. On the other hand, the wave nature of light is partially lost, so the diffraction or the interference of light can not be described using ray tracing. These wave-nature related phenomena occur in regions where ray tracing results in several intersecting rays. So, until the rays of the simulation are “far” from each other, ray tracing gives a good way to depict light propagation. In these cases it can even be used to calculate geometrical wave fronts. With the latter the wave nature is at least partially persistent.

ABCD formalism When cylindrical symmetry can be presumed along with the usual approximations of geometrical optics, the analytical theory of ABCD formalism (or ray transfer matrix analysis) can be used. ABCD formalism can be applied to describe the propagation of Gaussian beams, as they are solutions of the paraxial wave equation. While this method is not appropriate for the description of diffraction and interference either, it can properly give the focal shift of Gaussian beams when focusing is loose. So, the ABCD matrix analysis is a very efficient way to study pulsed beam propagation, especially if the Fourier transform can be treated analytically as well, because this way the whole theory can be analytical.

Scalar diffraction theory The tools of geometrical optics is inappropriate to treat problems related to the wave nature of light. It is widely known that along the boundary of light and shadow geometrical optics gives false description. This also contains a relevant part of the focal region. So, to describe focusing, a model is necessary which takes it into account that electromagnetic radiations are waves. A wave optical approach is scalar diffraction theory, which is based on the Huygens–Fresnel principle, but its tools can be derived also directly from Maxwell’s equations. The Kirchhoff diffraction integral, for example, was first formulated in the 19^{th} century, and since it has been shown several times that it gives theoretical results which nicely reproduce the experimental observations. This integral can be used to analyze focusing, so it gives a perfect way to calculate the amplitude and phase properties of the electric field in the vicinity focus.

2.3 Experimental methods to measure the changes of the carrier-envelope phase

Measuring the relative phase of the carrier and the envelope in the optical regime is a very laborious task. Most of the methods aiming to give information on the CEP are not able to measure its value, they only support data on its change from pulse to pulse. The CEP, often called “absolute phase” in this context, can be measured in a reliable manner only with one method. This technique is called stereo-ATI, referring to the fact that the strong electric-field dependence of above threshold ionization (ATI) is utilized, and that a double detector apparatus facing each other is used. Nevertheless, this solution needs an expensive experimental toolkit. As a result, finding an easily reproducible physical phenomena that can be turned into a simple measurement device for CEP detection is an actively studied research topic.

Fortunately, there is no need for complex physical processes if we only want to measure the changes of the CEP. In this case, linear optical methods can be utilized, like spectral interferometry. While the first application of spectral interferometry for such measurements aimed to detect CEP-drifts from pulse to pulse, the principles of this method makes it possible to use it for the detection of spatial phase changes. Spectral interferometry, true to its name, means the spectrally resolved measurement of the interference of two waves (the recording of an interferogram). With the different evaluation methods, each having its advantages and disadvantages, the spectral phase of the broadband radiation can be obtained from the interferograms. The way the CEP can deduced from these data depends on the exact conditions, as it is exemplified also in this work. It is important to note that the phase measured with spectral interferometry is a relative phase with respect to a reference wave. This is the reason why only phase change information can be deducted, and the “absolute” phase is not measurable this way.

3 Results

The main goal of this thesis is to study the phase and polarization-state changes of focused, pulsed Gaussian beams upon focusing and propagation. An important topic of the work is the phase and group velocity of focused beams. Related to these properties, special attention is paid to the carrier-envelope phase variations of focused, pulsed beams in the vicinity of focus. An other appointed goal of this study is to examine the influence of focusing, as a diffraction effect, along with that of beam distortions (truncation, chromatic aberration, monochromatic aberrations - spherical aberration, astigmatism, coma, curvature of field, distortion) on the phase properties in the focus. A relevant topic assigned for examination is the time-dependent polarization state of short pulses, specifically to investigate if it changes upon focusing and free-space propagation, and if yes, how. In short, the most important scientific results can be summarized in the following points:

I. I developed analytical formulas for the on-axis phase velocity and group velocity of focused Gaussian beams which take into account the wavelength-dependent properties of the focused beam. I specified the parameters which determine the changes of group velocity, and I established expressions for the calculation of these focused beam features using the characteristics of the source beam and the focusing system. Based on the previous attainments, I gave the specific conditions only under which the group velocity of a focused Gaussian beam is constant during propagation through the focal region. Using these results I highlighted that a chromatic aberration-like effect can occur not solely due to the focusing element, but also because of beam features.
I presented formulas for the calculation of the phase velocity and the group velocity of focused Gaussian beams when focusing is affected by chromatic and primary monochromatic aberrations. Specific attention is paid to the analytical evaluation of expressions to increase the precision and to widen the applicability of the numerical simulations. I used these results to analyze the effect of beam truncation, monochromatic aberrations and chromatic aberration on the on-axis phase-velocity and group-velocity variations of pulsed Gaussian beams. I made a comparison between recent and previous results on primary aberrations and their affect on phase properties in the vicinity of focus [T1].

II. I developed an accurate model of focusing with lenses and lens systems based on ray tracing calculations and scalar diffraction theory. I used this model to verify numerically that it is possible to focus few-cycle pulses with refractive optics without relevant distortion of the pulse envelope by simply compensating for the material dispersion of a dielectric slab corresponding to the phase-modifying properties of the focusing system on axis. I also confirmed the validity of an analytical model describing the on-axis CEP variation of focused, pulsed Gaussian beams. Based on the above two findings, I showed that focused, few-cycle pulses can propagate in the focal region of lenses with unmodified temporal shape at the electric field level [T2–T4].

III. I designed and built an experimental setup which is capable of measuring the spatial phase changes of pulsed beams with high spatial resolution. This setup was used to deduce experimental information on the carrier-envelope phase variations of few-cycle pulses in the focal region of an achromatic lens. I carried out wavelength-resolved measurement of properties of the source beam that was focused, and the phase changes of which were examined. With the measured beam properties I performed numerical simulations on the focused phase variations that correspond to the analyzed experimental case. With the comparison of the simulation and experimental results, I verified the main aspect of the theory, that is, I showed the effect of the wavelength dependence of the focused beam on the carrier-envelope phase changes, and that it results in variations which are different from Gouy phase shift [T5].

IV. I verified with numerical calculations the instantaneous polarization state changes of pulsed beams predicted by an analytical propagation theory. By choosing proper examples I also proved the simple polarization-change rule that the temporal variation of one polarization ellipse property (orientation or ellipticity) induces the change in the other property during propagation. With the help of the numerical study it was also highlighted that these propagation-induced changes are the consequences of beam diffraction, and as a result, they are general for all polarization-shaped pulsed beams. I also showed that the signatures of these local changes of instantaneous polarization state could be detected with simple experimental setups using spatially and temporally integrating detectors [T6, T7].

Publications

Publications related to the thesis published in peer-reviewed journals or conference proceedings

[T1] **B. Major**, Z. L. Horváth, and M. A. Porras. "Phase and group velocity of focused, pulsed Gaussian beams in the presence and absence of primary aberrations". *J. Opt.* 17.6 (2015), 065612. DOI: 10.1088/2040-8978/17/6/065612.

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