Light-matter interaction on the few- and sub-cycle timescale

Habilitationsschrift

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Kurzfassung

Licht-Materie-Wechselwirkung an der sub- und weniger-Zyklen Zeitskala

Eine wesentliche Herausforderung in der modernen Optik liegt darin, kürzere Zeitskalen und neue Frequenzbereiche zu erkunden, sowohl für höhere als auch für niedrigere Frequenzen. In den letzten Jahren wurden die verfügbare Zeitskalen in den Attosekundenbereich verschoben. Ein entscheidender Faktor hier ist die Möglichkeit, Pulse mit einer Dauer im Bereich von wenigen Zyklen oder sogar unterhalb des Einzelzyklus zu erzeugen. Zur gleichen Zeit wurden zudem die möglichen Frequenzbereiche für die Erzeugung von Pulsen mit wenigen Zyklen dramatisch erhöht.

In dieser Habilitationsschrift betrachtet der Autor Methoden zur Erzeugung von ultrakurzen Pulsen in sehr unterschiedlichen Frequenzbereichen, die vom Terahertz (THz) bis zum Vakuum-Ultraviolett (VUV) reichen. Es wurde aber auch Möglichkeiten entwickelt, diese Pulse zu verwenden, um die Dynamik der Licht-Materie-Interaktion zum Vorschein zu bringen. Im Falle von Ionisierung stellen diese neuen Frequenzen, so genannte Brunel-Harmonische, eine neuartige Quelle für ultrakurze Pulse im THz-Bereich dar, die es gleichzeitig erlauben, Prozesse innerhalb der Ionisationsereignisse zu untersuchen. In den Werken des Autors wird gezeigt, wie die elementaren Antworten aus der attosekundenlangen Ionisationsereignisse ein Interferenzmuster im Frequenzbereich bilden, welches das THz-Spektrum definiert.

Der Autor schlug auch vor, ultrakurze Pulse für neue ultraschnelle Bauelemente wie optische Schalter zu verwenden, die auf resonante Wechselwirkung basieren, obwohl resonante Effekte prinzipiell als “langsam” betrachtet werden.

Darüber hinaus demonstrierte der Autor einen neuen Mechanismus in Wellenleiter-Geometrien, der zu einen dramatischen Zusammenbruch eines Solitons führt, was ein sehr kohärentes Superkontinuum erlaubt, welches zu Unterzyklenpulsen komprimierbar ist. Der Autor studierte auch Superkontinuumsgeneration und verwandte Phänomene in Wellenleiterarrays und in einem freien Raum. Die Ergebnisse erlauben einen tieferen Einblick in fundamentale Fragestellungen der nichtlinearen Optik und Starkfeldphysik, und eröffnen neue Perspektiven für Zukunftstechnologien von extrem kurzen Pulsen.

Schlagwörter: Starke Licht-Materie Wichselwirkung, ultrakurze Pulse, Ionization, Solitonen.

Abstract

Light-matter interaction on the few-cycle and sub-cycle timescale

One of the main challenges in modern optics is to explore shorter times scales as well as to extend the frequency range to higher but also to lower frequencies. Last years, the time scales available have been shifted to the attosecond range — well below one femtosecond. The critical ingredient of this progress is the creation of strong few-, single- and even sub-cycle pulses. At the same time, the frequency range where few-cycle pulse generation is possible, quickly increases.

In this work the author considers methods for generation of ultrashort few- and sub-cycle pulses in very different frequency ranges, from terahertz (THz) to vacuum ultraviolet (VUV) and for the possibilities to use these generated pulses to test the dynamics of light-matter interaction. In the case of ionization, new frequencies, so called Brunel harmonics, appear, providing a novel tool both as a source of short pulse in THz range, but also as a meter allowing to investigate the processes taking place inside ionization events. Contribution in both of these directions has been provided by the author, who showed how the elementary responses from attosecond-long ionization events form an interference pattern in frequency defining the THz spectrum.

Furthermore, the author showed the possibility to use ultrashort pulses for novel ultrafast devices such as fast optical switches based on resonant interaction with atoms and molecules, despite the resonant effects often believed to be “slow”. In the waveguides geometries, the author demonstrated a new mechanism of a dramatic collapse of an optical soliton which gives birth to a very coherent supercontinuum, that is, a very broad spectrum, being compressible to sub-cycle pulses. The author also studied supercontinuum generation and related phenomena in the waveguide arrays and in a free space. The results allow a deeper insight into fundamental questions of nonlinear optics and strong field physics, at the same time opening up new perspectives for future applications of extremely short pulses.

Key words: strong light-matter interaction, ultrashort pulses, ionization, solitons.

Contents

List of Publications

This work is a compendium based on the following publications that are referred to in the text as [B1-B34]. For each of the works listed here, the author's participating in the work was important or deciding: he either issued the main idea, or supervised the project, or performed numerical calculations.

[B1] I. Babushkin, C. Brée, C. M. Dietrich, A. Demircan, U. Morgner, and A. Husakou, "Terahertz and higher-order Brunel harmonics: from tunnel to multiphoton ionization regime in tailored fields", J. Mod. Opt. **64**, 1078 (2017).

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Introduction

Since the advent of pulsed laser sources and short pulses, the main road of the progress in this field is the constant decrease of the pulse duration and increase of the peak power. Several decades ago optics has entered the intensity range where the external field is becoming comparable to or even higher than the intra-atomic one [1, 2]. In such strong-fields the dynamics of the outer electrons is determined not anymore only by the local electronic/atomic environment but also by the external field. In particular, when the electron is ionized and then returns to its core, very high-energy radiation in extreme ultraviolet appear (high harmonic generation, HHG).

But not only the electron return leads to photon emission. Also the electron ionization itself gives rise to a field-dependent change of the refractive index of the material due to ionization and thus to the nonlinear effects. Any nonlinearity, including ionization-induced one, leads to appearance of harmonics in the field. Thus, ionization and propagation of electrons in the field leads to generation of relatively low frequency harmonics up to tens of eV, which are referred to as Brunel harmonics [3, 4, B1]. Interestingly enough, if the field is asymmetric in certain sense (for instance it contains two frequencies ω and 2ω or contains only few cycles), the resulting radiation can be a “zero harmonics” with a frequency hundreds times less than the one of the pump, which can lead in terahertz (THz) frequency [5–8].

THz frequency range (often defined as the range of frequencies between 0.1 and 30 THz) is particularly important for optics since the rotational level of complex molecules belong to this range [9, 10]. That is, looking at complex molecules (for instance biological tissues) in THz range one can obtain a “fingerprint” identifying such molecules. On the other hand, until recently the THz frequency range was referred to as the THz gap because i) there were no compact tunable sources of THz light and ii) THz detectors were very insensitive. Although this situation gradually changes last years, it is still quite complicated to produce THz radiation with properties “on demand”.

The author considered both THz and higher order Brunel harmonics from ionization of noble gases such as argon in strong femtosecond single- and multicolor optical pulses The author made an impact into this field in the following ways: i) it was shown that sub-cycle attosecond-long ionization dynamics plays the key role in the formation of THz radiation, thus strengthening the ionization-based mechanism [B2–B6]. ii) the study of more-than-two color fields were initiated and preformed, showing that using, for instance, 3-color pumps allows to control the frequency and the waveshape of the THz radiation [B4]. iii) The mechanism of the THz energy scaling with the wavelength were studied [B5, B7]. iv) The ideal waveshape for most efficient THz generation were revealed, which appeared to be saw-like one [B8]. v) The mechanism of the spectrum formation for THz Brunel harmonics based on the "diffraction grating in frequency" was discovered [B4, B6]. vi) The study of higher-order Brunel harmonics in connection to the ionization dynamics on the sub-cycle scale were made [B1] vii) A new mechanism of THz radiation was found, based on the Freeman resonances [B9].

Another important ultrafast process related to sub-cycle light-matter interaction is the formation of supercontinuum (SC) [11–14]. SC is a pulse with very broad spectrum (typically at least one octave, that is, containing at least some frequency together with its second harmonics) which at the same time has a certain degree of coherence. SC can be routinely obtained from higher order solitons in waveguides and is very useful for stabilized laser sources and precision measurements. One of the fundamental problems of such SCs is that they are influenced strongly by quantum noise, and thus pulse-to-pulse stability is rather limited and degradates quickly with increasing the supercontinuum width. One of the key parts of SC generation is the breakup of the strong pulse into fundamental solitons, emitting dispersive radiation in the normal dispersion region. On the other hand very recently, with the advent of short strong mid-infrared laser sources soliton-like structures (so called light-bullets) were obtained not only in waveguides but also in a free space in the anomalous dispersion range of some materials [15], which also emit radiation in normal dispersion range. Up to recently, there were no clear understanding how this radiation was related to the Cherenkov radiation for solitons.

The author contributed into these problems in the following way: i) he has shown that if the higher-order soliton at the input of the waveguide is taken to be extremely short (only few-cycles long), the known dynamics of the SC formation becomes fundamentally different from what was known before [B10]. The new regime was called "soliton implosion" and is based on the formation of hundred-attosecond long shock front which is upon the further propagation transformed into the broad supercontinuum via Cherenkov radiation and dispersion and is completely insensitive to quantum noise and thus perfectly pulse-to-pulse coherent. ii) In the case of light bullets in a free space in anomalous-dispersion region it was shown that the emission into the normal dispersion region is indeed the Cherenkov radiation equivalent to the one of solitons in the waveguides, blue-shifted by the plasma [B11]. It was shown to play an important role to prevent the soliton collapse. iii) Generation of the supercontinuum in a weakly coupled array of waveguides was studied and shown that if the input is localized in only one waveguide of the array, the Cherenkov radiation tends to escape to the neighboring waveguides whereas the main solitonic part remains highly localized [B12]. iv) So called rogue waves (RW) were analyzed. A new possible physical mechanism of RW formation was found and the influence of the additionally injected dispersive wave on the RW statistics was revealed [B13, B14]. In addition, the of the pre-chirped dispersive wave to a single soliton in order to obtain soliton shortening was studied [B15] v) Short pulses generated in vacuum ultraviolet (VUV) were analyzed and the new regime of four wave mixing process (FWM) in the presence of phase- and group velocity mismatch was found [B16–B18]. The quasi-phase-matching of those FWM processes using ultrasound in gas-filled capillaries was studied [B19].

Finally, the author also addressed the resonant interaction of light and matter. It is typically believed that if the atom or molecule have a resonance at some frequency, the response close to this resonance is narrow and thus incompatible with short pulses. Recently it has been realized however that this is not necessary the case [16–18]. In particular, the Rabi oscillations can be arbitrary fast and a two-level system can experience the whole Rabi oscillation cycle on the few-cycles and even on a single cycles of the laser light. The author has contributed in this field by showing that: i) using Rabi oscillations one can create, erase and modify optical gratings on the few- and even sub-cycle time scale [B20–B22]. The grating period can be much smaller than the period of the pump pulse. ii) It is possible to engineer the excitation sequence of an oscillator chain to obtain the pulses with desired waveshape [B23]. iii) Even in the case of bare linear reflection from the metallic or dielectric nm-thick layer, a single-cycle input pulse can give a response in the form of a sub-cycle one [B24]. iv) The ultrashort pulses can be directly obtained from a two-section laser at the output with the help of so called coherent modelocking which uses self-induced transparency effects to stabilize the pulse in a cavity [B25–B27].

The work is organized as following: In the Sec. 1 the mechanism of THz and higher-order Brunel harmonics generation is discussed, in Sec. 2 the dynamics of solitons, supercontinua resonant radiation is addressed. In the Sec. 3 the coherent effects in few- and sub-cycle regime are investigated.

Chapter 1

Generation of THz and higher order Brunel harmonics

Nonlinearity created by fast tunnel ionization process in strong optical field produces new frequencies which are referred to as Brunel harmonics [3, 4, 19]. In contrast to the high-harmonic generation which appear due to return of electrons to the parent cores [20, 21], Brunel harmonics do not require such return. The change of refractive index due to free electron creation is already enough to create necessary nonlinearity. However, this macroscopic picture is not enough when the details of the spectrum are to be understood [8, 22]. The works of the author allowed to shed light to the role of the electron dynamics on the fast sub-femtosecond scale in this process as described below.

1.1 Ultrashort THz transients in gases in two- and multi-color fields

Many contemporary methods to obtain extremely high or extremely low frequencies make use of, in one or the other way, nonlinear processes in laser-induced plasma. In particular, it was demonstrated that using light-induced plasma dynamics, it is possible to produce frequencies in THz range, hundreds times smaller than the optical pump frequency [7, 8, 22–24].

Such a mechanism is especially useful to generate ultrashort intense pulses. Denotation “ultrashort” in this context assumes a pulse duration comprising of only few oscillations at the corresponding frequency, which translates into (sub-)picosecond pulse durations in the THz range. Compact THz lasers are still unable to provide short pulses. Table-top sources exploiting solid-state materials are based on down-conversion from the optical frequency range, either using semiconductor photoconductive switches [25], or via optical rectification using the second- or third-order nonlinearity in crystals [1, 26–31]. These methods provide high THz fields up to hundreds kV/cm and single-cycle pulse durations. However, it is difficult to scale them to higher energies, and the available spectral range is limited by material absorption bands. In addition, a perfect setup alignment is required. In contrast, THz generation in gases does not require complicated alignment and is not limited by any damage threshold. THz emission in plasma created by a single-frequency femtosecond pulse in a filament as well in a plasma spot were experimentally observed [32–34] and is attributed to a wake field generation [35]. This mechanism has rather low efficiency below 10^{-6} and produces radiation in the direction perpendicular to the optical axis. In contrast, the method based on the transition-Cherenkov radiation [36, 37] is much more collimated in the direction of propagation. However, its efficiency is still considerably low and the emission itself is cone-shaped around the propagation direction. Several other ideas to produce THz emission in gas are based on the interaction of pairs of filaments [38] and on the transition through the gas-plasma interface [39, 40].

In contrast to the previously described methods, THz generation from light-induced plasma produced by two-color laser pulses has higher potential in respect to higher pulse energies, tunability and spectral width. Besides, the setup is extremely simple: a linearly polarized pulse of the fundamental frequency and its second harmonic are focused into the spot where the photo-induced plasma is produced. If the plasma is generated, a low frequency signal with the electric field up to several hundreds kV/cm also appears on the detector [5, 41, 42].

According to the recent studies, ionization is necessary for such scheme to produce THz radiation [6–8, 22, 23, 42]. It is thus supposed to be produced by ionization in an asymmetric field and by the subsequent dynamics of plasma current [7, 8, 22, 23, 35, 43, 44]. In contrast to these findings, a FWM rectification mechanism was also suggested by other authors [5, 41, 44–46], corroborated by THz energy scaling arguments and by the energy dependence on the polarization of both beams [46]. The author of the present work contributed to this discussion about the leading mechanism by showing that the scaling of the spectral width with the pressure corresponds indeed to plasma-based explanation [B2]. This scaling was found in a good agreement with experiments and corroborated the importance of the plasma-based explanation.

The author also contributed by proposing a new method for pulse generation with a tunable frequency and carrier-envelope phase in a very broad frequency range from THz to MIR [B2, B4] based on plasma-based mechanism. It allows controlling the characteristics of the pulse such as its spectral shape, center frequency, and phase, using modification of the pump pulse waveform. The approach allows to establish an unambiguous correlation between the pump and the signal pulses. In particular, the framework was developed in [B4], which allowed to consider sub-femtosecond ionization events [47] as a kind of diffractive grating in frequency domain as well as to predict the spectral shape and energy in dependence on the pump pulses. In [B8], the best pump waveshape was calculated, based on the developed grating analogy, which appeared to be the saw-tooth waveshape or it finite-frequency approximation. In the plasma spot geometry, the important details of the spatial localization and the directionality of the THz generation were revealed [B6].

The potential of the THz emission using two-colored light fields was further explored in several directions: The asymmetrical field required for THz generation can be produced by using a single-color pulse with a near-single-cycle duration [6]. The asymmetry can also be created by strongly chirped pulses [48]. The electron dynamics can be influenced by variation of the light polarization [49, 50]. In addition, the frequency of the fundamental and second harmonics can be shifted separately, allowing to control the position of the maximum of the low-frequency part of the spectrum [51].

To develop this research direction further on, the author studied the details of THz light generation in different geometries, such as optical filament [B5] and hollow gas-filled waveguide [B3]. For the case of filament, using rigorous 3+1D simulation and comparisons to experiment, it was shown that the mechanism of THz formation after a critical intensity is indeed related to the plasma formation due to photo-induced ionization. More than one order of magnitude of increase in THz generation efficiency was predicted if mid-infrared instead of near-infrared pump pulses would be used. The scaling with the pump frequency for the filament and plasma spot geometries was improved and investigated in details in [B7]. It was shown that there is no well defined scaling, in contrast to other less precise investigations [52]. In the case of a hollow waveguide geometry, significant broadening of the spectrum was predicted [B3], leading to a supercontinuum spanning more than 5-octaves from THz to near-infrared, as well as significant blue-shift of the generated radiation upon the propagation. It was shown that the process of THz generation is self-influenced by the THz radiation generated at earlier propagation distances [B28].

1.2 Higher order Brunel harmonics

Dynamics of higher order Brunel harmonics has been studied by the author in [B1]. They do not need a two-color pump or other symmetry breaking mechanisms for their generation. In [B1] it was shown that the dynamics of Brunel harmonic generation is significantly dependent on the ionization regime and on the dynamics of the electrons leaving the atom. If the ionization rate is much smaller than the optical cycle (multiphoton ionization regime) the nonlinearity created by the ionization is too slow, so that the Brunel harmonics do not appear in spectrum. On the other hand, if the ionization is fast enough (tunnel ionization regime) the Brunel harmonics do show up. Nevertheless, even in the multiphoton ionization regime one can create Brunel harmonics by tuning the second frequency field from the double of the fundamental one by the detuning δ. In this case the beatings appear, which are slow enough for the ionization nonlinearity to respond. Brunel harmonics reappear at the frequencies detuning to the fundamental by the δ.

1.3 THz radiation from subcycle Freeman resonances

As it was already said, the dynamics in the multiphoton ionization regime is too slow to create new frequencies. Nevertheless, in the recent work of the author [B9] it was shown that this is not anymore true in the vicinity of so called Freeman resonances [53, 54]. Freeman resonances are the sharp peaks in the ionization rate arising at certain values of the pump field intensity. They appear because of the Stark shift of the bounded levels with the field intensity. Because of such shift, at certain values of the intensity a resonant transition of the electron to the highly living Rydberg state can takes place, after which the electron can go to the continuum via an absorption of a single photon. In that case, the population transfer in the atom is dominated by two major competing mechanisms: Freeman-resonance-enhanced ionization and population trapping in high- lying, laser-dressed and strongly distorted states. The latter can be viewed as the extension of the Kramers-Henneberger concept [55, 56] to the Rydberg manifold. [57–59]. Freeman resonances take place at very well defined values of intensity. If the input pulses have, for instance, Gaussian form, the intensity changes from zero to the maximum, and can pass through the Freeman resonances. As such crossing takes place, the ionization rate suddenly increases and quickly decreases again as the resonance has been passed. This dynamics takes place at the subcycle time scale, which leads to asymmetry in the ionization-created current. Modification of these currents in time lead to generation of low-frequency fields. Because the Freeman resonances take place at more than one place over the pulse duration, there is an interference between the fields generated at different events which creates beatings in time and thus a frequency comb structure.

1.4 Modeling issues

The author contributed to the modeling of THz generation. In particular, the author's works were the first where a model including both THz generation and propagation effects due to pump were treated in a consistent way on extended propagation distances. Namely, on the early stage of the research development, propagation effects were treated by many authors in a simplified way [8, 22, 23], which gave a qualitative picture of the process but many details of the complex spatio-temporal dynamics were disregarded. An alternative is a microscopic semiclassical description of the medium, which uses computationally demanding particle in cell (PIC) simulations [43, 44, 48]. Such simulations are limited to very small spatial box sizes of the order or few micrometers only. A separate class of models uses a direct quantum-mechanical simulation of the electron dynamics [46, 49, 60]. Although such modeling gives a possibility to check the details of microscopic process, it suffers from the impossibility to model "real world situations" where spatio-temporal propagation effects play significant role.

In contrast, in the works of the author the approach based on the the unidirectional pulse propagation equation (UPPE) was adopted [B2, B3]. It should be noted that these equations are well known [12, 61–63] and widely used for studying propagation of light in waveguides and filaments, in a good agreement with experiments [64]. However, it was first shown [B2, B3] how these equations can be used to model Brunel harmonic generation, also in THz range.

UPPE is the equation describing the dynamics of pulse propagation in a free space (a similar equation for waveguides is referred to as forward Maxwell equation (FME); here for simplicity the term UPPE is used for both cases since both FME and UPPE are based on very similar principles). UPPE does not use the slowly varying envelope approximation, the standard approximation of optics. On the other hand, UPPE is obtained under the condition of neglecting backward-propagating waves and is therefore much more efficient for propagation over relatively long distances than solving the full nonlinear wave equations. The description of ionization via the tunnel ionization formula [65] model was used. Finally, the author made an impact into the general theory of the UPPE by investigating the general structure of the fundamental solution of its linear part which appeared to be a projector onto some functional space [B29]. I also appeared to include the waves propagating in both directions, despite of the common belief (and the equation's name). It has been also shown that the casually principle is in general not respected by UPPE.

Chapter 2

Sub-cycle pulses and supercontinua from high-order soliton dynamics

2.1 Soliton implosion

The Heisenberg principle possesses a trade-off between the temporal pulse duration and its width in spectrum. That is, very short pulses correspond to broad spectra. This trade-off becomes especially dramatic as the pulse duration approaches the single cycle limit [66–70]. If the spectrum is “good enough” in the sense of coherence/spectral phase, then it is not necessarily to generate directly short pulses - instead it is enough to produce a broad spectrum and then shorten the resulting pulse using purely linear dispersion management. One useful method to obtain short pulses is a compression of not-so-short ones using dynamics of solitons [71–76]. Fundamental soliton is a stable structure which exists in the frequency region where the dispersion is anomalous [71, 76, 77]. If the energy of the input pulse is increased comparing to the one corresponding to a fundamental soliton, a higher order soliton appears [71, 76, 77], which, in contrast to the fundamental soliton, experiences a periodic sequence of compression/decompression cycles — the behavior which is unstable in realistic waveguides and leads to a breakup [12, 76, 78–80].

The dynamics of this breakup leads to supercontinuum (SC) generation [11–14, 80, 81] that is, to arising of a very broad spectrum at the output of the fiber. In SC, the temporal shape does not consists of a single pulse anymore. In course of the self-compression and breakup process so called dispersive (or Cherenkov) radiation is emitted at frequencies phase-matched with the solitonic one [82]. As a result, a complicated dynamics takes place, which includes many fundamental solitons interacting with each other as well as with their Cherenkov radiation [14, 81]. Generation of dispersive radiation leads also to a frequency shift of the soliton (different for every particular soliton). This dynamics takes place during relatively long propagation distance before the walk-off takes the solitons and the dispersive waves apart. It is important to note that during the pulse evolution an instability arising from the quantum noise takes place. It grows exponentially upon the propagation. The corresponding destabilization mechanism is either the modulation instability (MI) [13] which leads to formation of modulations on the shape of the pulse, or soliton fission [12], in which case the fundamental solitons escape the pulse one-by-one.

In the work [B10] an alternative mechanism of soliton breakup was discovered, which has been called soliton implosion. In this mechanism, the key role is played not by the noise but by the intensive Cherenkov radiation and the shock formation. The fundamental pre-requirement for this scenario to appear is the very short, few- or single- cycle input pulse of high enough intensity to form a soliton of higher order. In this case, at the initial state the high-order soliton compression takes place as usual. However, in contrast to known mechanisms, instead of the stage of noise amplification, a short sub-cycle shock is formed, of the duration of few hundreds attoseconds. This shock leads also to significant broadening of the spectrum. In addition, this process takes place on the small propagation scales of few tens of micrometers and is accomplished by very efficient and broadband generation of Cherenkov radiation. As it was shown previously by other authors [83, 84], for the very short input pulses the Cherenkov radiation is especially effective since it dependents critically on the spectral overlap between the soliton spectrum and the Cherenkov frequency. Besides, because of the fast interaction process, the Cherenkov radiation is spectrally very broad. In the case of soliton collapse described in [B10] the Cherenkov radiation itself forms a one-octave supercontinuum. Very importantly, the whole process is insensitive to noise and thus shows excellent pulse-to-pulse coherence. This allows to compress the output to sub-cycle, sub-femtosecond pulse durations and makes it an excellent candidate for comb generation.

Another way of pulse compression is based on an interaction of the dispersive wave with the fundamental soliton [85, 86]. In this scenario, reflection of even a weak dispersive wave from the soliton creates a back-action to the soliton itself, leading to its compression and frequency shift. The author contributed into studying of this mechanism by considering a chirped dispersive wave and its influence on the compression behavior [B15]. The chirp allows to create a tunable interaction along the propagation, enabling to increase the compression efficiency.

2.2 Rogue waves

The complex interaction of solitons in course of the supercontinuum formation attracted attention also in respect of so called rogue waves (RW), or killer waves [87–92]. Those are defined as solitons/waves with amplitudes at least two times larger than the "typical" ones. They appear on a statistical basis and are characterized by a statistical distribution with a heavy tail in the high peak range, much stronger than the non-interacting waves can typically have. Such optical RW posses certain analogy to the rogue waves in ocean, which attracted much attention recently [87].

In the work of the author [B13] a yet-another candidate for optical RW formation mechanism was presented, namely the fusion of two solitons at the advanced stage of the supercontinuum formation. Furthermore, in the work [B14] the author showed that by sending an additional low-amplitude dispersive wave allows to control the rogue waves. Namely, the whole statistics of RW formation in the supercontinuum generation can be modified. The control is achieved because the dispersive wave interact with every of the solitons but also, quite unsuspectingly, with the whole soliton fission dynamics on the early stage of supercontinuum.

2.3 Resonant radiation in filaments

Cherenkov radiation mentioned in the previous sections seems to be important also in the context of the filamentation in the anomalous dispersion regime. The fundamental concept of filamentation developed several decades ago is based on the picture of a filament as a relatively stable propagation structure where the Kerr-nonlinearity-induced self-focusing is balanced by the plasma defocusing [63, 93–96]. This picture recently was accepted to be far from complete for the short input pulse durations or/and long wavelengths. In particular, in [97], a super-high-power filamentation regime in air has been revealed, which is quantitatively and qualitatively different from a conventional filament. The main point is that leaking energy into higher harmonics plays an important role in arresting the collapse due to the Kerr nonlinearity. This fact mentions a significant paradigm shift in the whole nonlinear optical community. In addition to this, if we pump in the anomalous dispersion regime, the dynamics was also significantly different from the normal filamentation process [15]. Observations revealed generation of blue-shifted radiation [98] in such case. The author of the present work showed [B11] that these waves have the same nature as the Cherenkov radiation for solitons propagating in a waveguide. In addition, the corresponding Cherenkov frequency is significantly shifted by the generated plasma to the blue side. Moreover, it was also shown that the intense resonant radiation is an important mechanism preventing the collapse of the soliton.

2.4 Waveguide arrays

The dynamics of soliton propagation, supercontinuum generation and generation of dispersive waves has attracted attention last years also in context of array of elements coupled weakly to each other [99, 100, B30, 101–104]. In this situation, if we consider a linear pulse propagation, the light localized in one waveguide leaks to the neighboring ones, then to the neighbors of neighbors and so on, so that discrete diffraction takes place. This diffraction however happens on the scales much longer than the usual diffraction in a free space. In the case of a strong pulse launched into a free space, self-collapse due to Kerr effect takes place. In the same way, a strong enough pulse launched into several waveguides simultaneously will be collapsed into a single one [105] and then propagating in it. This happens however for very long pulses and moderate intensities. The author addressed the question what happens if the propagation in arrays of relatively short pulses is considered, which would, in the case of the single waveguide, form a supercontinuum [B12]. It was shown that indeed, a strong pulse breaks up into fundamental solitons and resonant radiation as it is usual for the SC formation. Nevertheless, after the breakup the discrete diffraction starts to play an important role. It can not overcome self-focusing for the fundamental solitons but is able to do so for the much weaker dispersive radiation. As a result, the solitons continue to propagate in the "central" waveguide whereas the dispersive waves quickly disappear.

2.5 Ultrashort pulses in waveguides using four wave mixing

In general, obtaining short pulses in waveguides does not necessary requires soliton-related dynamics. Short pulses at certain frequencies can be obtained from the short pulses at another frequency using nonlinear frequency conversion. In waveguides this implies most often a collinear for-wave mixing processes [27, 28, 30, 76, 106], for instance generation of 5th harmonic (using the fundamental frequency and its third harmonics via the process $5\omega = 3\omega + 3\omega - \omega$) [B16, 107, 108]. The phase matching for the process is achieved in the isotropic media using waveguide-induced modification of the refractive index [107–109]. This additional impact is anomalous [109] in a very broad range of frequencies and thus can help to achieve phase matching. In particular, such process was studied in [B16, B18] in a argon-filled waveguide where the phase matching for the 5th harmonics of the 800nm pump at ∼160 nm in a hollow-core capillary of ∼100 μm diameter can be achieved at low pressures of several mbar. This method is advantageous in comparison to other methods, for instance using crystals because due to the possibility of relatively high repetition rate and short pulse duration. In [B16, B18] the author used the model developed by him (see Sec. 2.6) to describe the propagation of short strong ionizing pulses in the waveguide taking into account also the transverse dynamics. In [B19], it was proposed to improve the efficiency of this process using quasi-phase-matching via pressure modulation which is created by a strong ultrasonic wave.

Besides, in [B17] it was shown that if the phase matching does not take place, there is a possibility to generate even shorter pulses on the cost of some decease of the pulse energy. Namely, when a phase and a group-velocity mismatch takes place at the same time, and ultra-short pulse of the fundamental frequency and a longer and weaker pulse of the third harmonics are sent to the waveguide, in the presence of the group-velocity mismatch the output pulse at the 5th harmonics contains two parts. One is a relatively long one propagating with its own group velocity (that is, with the velocity of 5th harmonics) whereas the another one propagates with the velocity of the pump (1th harmonics), that is, has the "incorrect" group velocity and at the same time $\sqrt{2}$ times smaller duration than the fundamental one. Interestingly, its amplitude quickly increases to some maximum and does not decreases anymore as it is usual by phase-mismatched processes. This strange and untypical behavior is explained by the fact, that this pulse, co-propagating with the pump frequency, does not really "propagates", but is constantly "depleted" and "regenerated" by the pump pulse at every moment of propagation. In all cases the dynamics of the pulse propagation was modeled with UPPE equation described in Sec. 1.4 and Sec. 2.6.

2.6 Modeling issues

The modeling of the equation was used with the UPPE approach described in Sec. 1.4. It allows to describe the propagation beyond the slowly-varying envelope approximation and thus are capable with arbitrary short pulses and broad spectra. Depending on the particular problem, two forms of these equation were used: one is for the fast electric field directly and the other is for the so called analytic signal [110, 111]. The latter is obtained from the fast real electric field by projecting the electric field to the positive frequency components. Despite of the projection, the analytic signal contains the same information as the electric field. The modified version of the equations for the analytic signal is also almost equivalent to the initial equation for the electric field, only the 3d harmonic generation is eliminated [110, 112].

Chapter 3

Few cycle dynamics in presence of resonant effects

Interaction of light with matter is the strongest if the optical frequency is in resonance with some transitions in the nonlinear medium. The dynamics of light-matter interaction near resonances is the oldest and one of the very successful topics of nonlinear optics [113–116]. Typically dynamics of light near resonances are used to create or control very long pulses, much longer than the single-cycle duration. This is because spectrally narrow resonances do assume long pulses. Nevertheless, recently the understanding started to develop, that this picture is far from being complete [16–18, 115, 117, 118]. In fact, Rabi oscillations can be arbitrary fast, even of the order or less than an optical cycle [1, 16, 117]. As an example of application, ultrafast resonant transitions were recently proposed theoretically and realized experimentally as an alternative mechanism to generate attosecond pulses [119]. Of course, as the pulse duration approaches the single-cycle, the influence of the other levels must be taken into account [18, B21, 118, 120].

In general, the period of Rabi oscillations in comparison to the dissipation/relaxation times determines also another important aspect of the resonant light-matter interaction. If the Rabi frequency is much less than the relaxation times of population and of the polarization of the medium (commonly denoted as T_1 and T_2 correspondingly) the light-matter interaction is called to be “coherent”. In this case, a phase memory in the medium plays an important role [113–116]. That is, the phase oscillations persist for a long enough time to significantly change the pulse dynamics.

The author proposed several devices based on the coherent interaction where the coherent effects play the central role [B27]. In particular, a two-section laser operating in the so called coherent mode locking regime (CML) which is capable to provide ultrashort pulses with down to few cycle duration and high repetition rate [B25–B27]. Furthermore, he proposed an ultrahigh-speed deflector of laser radiation that uses the possibility of inducing population gratings without overlapping pulses in a resonant coherent medium [B20, B22], and, finally a method to obtain unipolar pulses based on a coherent control of a response of Raman-active oscillator chain by means of a sequence of ultrashort excitation pulses [B23, B31].

3.1 Coherent (self-induced transparency) modelocking

Nowadays, passively mode-locked lasers have been actively used to generate short light pulses [121–124]. At present, the mechanism of the pulse generation in all types of two-section lasers operating in the passive mode-locking regime is based on the effects of absorption saturation in the absorber and saturation of the gain in the amplifier. Because the pulse interacts with the amplifying and absorbing media in the cavity incoherently, the spectral pulse width is limited by the gain line width $1/T_2$. Thus, the pulse duration is limited from below by T_2. In contrast, if the pulse interacts with the gaining and absorbing media in the cavity coherently, this restriction is not anymore valid, and the physical mechanism of the passive mode locking turns out to be the mechanism of stabilization of a self-induced transparency (SIT) pulse in the cavity, that is, the one with the area 2π. First theoretical investigations of the coherent mode locking (CML) regime (or self-induced transparency mode-locking as it is sometimes called) [117, 125, 126] were based on a homogeneous mixture of particles with different dipole moment and were not-self starting. In [B25, B32] the occurrence of a self-starting CML regime in a two-section laser was predicted, which also supports few-cycle pulses [B27]. In [B26], the analysis of the self-starting behavior and dynamics based on the area theorem was accomplished.

3.2 Subcycle population gratings

An another example of coherent-effects-based devices is an ultrafast deflector of laser radiation, which utilizes the diffraction of light on spatial gratings of population inversion in resonant media [127–130]. Traditionally, spatial gratings are created by monochromatic laser radiation, upon the interference of two or more light beams. Absorption of light in a resonant medium leads to changes in the population of the atomic levels, and, as a result, a population grating arise, repeating the shape of interference fringes of the laser beam [127]. If we would apply this traditional approach to very short pump pulses, the region of their overlap would be extremely small which would make it impossible to create more than just a few interference fringes.

However, if the duration of pulses is shorter than the relaxation times T_2 of the medium, a population grating can arise in the regime of coherent interaction of short light pulses with the resonant medium even without overlapping of the beams. This approach is referred to as echo-holography [131]. In [B20, B21] this approach was extended not only to create gratings but also to manipulate them, in particular by erasing and multiplying their spatial period. Interestingly, this period is can be made much smaller than the wavelength of the exciting pulse. This is because the polarization waves excited in the medium mediating the interaction between the pulses can be in fact arbitrary slow because the polarization waves do not obey the dispersion relation for light and, strictly speaking, even do not propagate, at least in the common sense.

In the case if the grating is excited by extremely short pulses, the dynamics of the levels other that the working ones can become important. In [B21, B27] this question was studied by simulating of a multilevel system of a typical alkali-metal atom. Surprisingly, the other levels plays rather small role in the dynamics even for nearly single-cycle exciting pulses.

3.3 Unipolar and quasi-unipolar pulses

The next possibility to use coherent effects proposed in the works of the author is the generator of “unipolar” pulses. Unipolar pulses (or video pulses) contain a considerable constant component of the electric field. At first glance, although the wave equation admits unipolar solutions, unipolar pulses seem to be unphysical, since the electric and magnetic components of the radiation field far from the emitter are proportional to the acceleration of charges [111]. In the case of periodic motion of bound charges the acceleration indeed cannot be unipolar. However, it was shown that, if a single initially bipolar pulse of a high power propagates in a nonlinear medium [117, 118, 132, 133] there is a possibility of generating unipolar or quasi-unipolar pulses.

In the works of the author [B31, B33, B34] quasi-unipolar pulses were shown to be possible if excitation of a Raman-active medium by a sequence of two or several extremely short pulses is utilized and the region of the interaction moves with a superluminal velocity. This can be realized, e.g., upon an oblique incidence of a small light spot on a thin layer. The light spot can be created, for instance, by a rotating mirror located at large distance from the layer. Such spot can have any velocity, also higher than the speed of light in vacuum.

Finally in [B24] the author has shown that, surprisingly, there is much easier way to generate quasi-unipolar pulses using a nm-thin layer of a metal or a dielectric, irradiated by a single-cycle pulse. In this case, the response of the medium can be a sub-cycle pulse which has the quasi-unipolar shape. Modification of the pulse shape by reflection is determined by the fact that in one-dimensional geometry the response is proportional to the velocity of oscillating charges/dipoles (and not to their acceleration), as it is prescribed by the Greens function of the one-dimensional D‘Alembert operator.

3.4 Modeling issues

Propagation of pulses in resonant media require methods beyond the slowly-varying envelope approximation for the field as well as beyond rotating wave approximation for the matter. In the most of the works of the author on this topic [B20, B21, B24, B27] the Bloch equations for the arbitrary fast field were used together with the direct modeling of the nonlinear wave equation. On the other hand, the direct comparison shows that slowly-varying-amplitude-based methods are working relatively well up to some limit in the few-cycle range. Therefore, in some of the works such slowly-varying approaches were indeed used [B25, B32].

In the works related to the study of the response of the oscillator chains, the response of the chain was calculated as a linear sum of all responses from every element of the chain [B31, B33, B34], the latter calculated using a suitable classical model.

Chapter 4

Conclusion and Outlook

4.1 Conclusions

The main goal of this work was to study the light-matter interaction with ultrashort pulses as well as new frequencies produced by such ultrashort pulses, in different situations, which arise in modern short-pulse experiments. Several setups were considered with various leading light-matter interaction mechanisms and different geometries, all of them beyond the standard slowly-varying envelope approximation in optics.

From the point of view of modeling, propagation in gases and solids with single- and multicolor fields, formation of supercontinuum in different frequency ranges from THz to VUV governed by Kerr nonlinearity, four-wave mixing processes and ionization- induced nonlinearity require unidirectional pulse propagation equation beyond the slowly-varying amplitude approximation. In the case of ionization this equation is was supplied with the equation for the free electron current. For the study of resonant light-matter interaction Bloch equations (without rotating wave approximation) as well as full nonlinear wave equation were solved.

Direct numerical modeling as well as analytical study allowed better fundamental understanding of the interaction processes as well as led to new proposals for ultrashort pulse generation at the frequencies on the current edge of optics such as THz and VUV. For instance, the diffractive grating analogy for the structure of the spectrum of Brunel harmonics in multicolor fields provided an unified view of Brunel harmonic generation process and made it possible to find new ways to control the generated radiation and to extract the information about ionization dynamics from it.

Furthermore, study of the propagation of few-cycle solitons in waveguides allowed to reveal a new mechanism of supercontinuum generation, based on a coherent soliton breakup process. Important role in this breakup is played by resonant radiation emitted by solitons. Such resonant radiation was also shown to play an important role in the arresting of collapse for light bullets in the anomalous dispersion range in a free space.

Finally, in the case of resonant interactions, the author used the less-known fact that “resonant” does not mean “slow” and demonstrated the possibility of generation of ultrashort, single- and even sub-cycle pulses in resonantly-interacting media. The possibility to generate population gratings with a period on the sub-cycle length scale with very short input pulses was also demonstrated.

4.2 Outlook

Ultrashort pulses at optical frequencies in novel spectral ranges such as THz and vacuum ultraviolet is one of the keys for future optical technologies and will be certainly important for many future discoveries in the solid-state physics, atomic physics, biophysics, etc..

As a possible future direction, it is very attractive to use Brunel radiation to study the details of dynamics of ionization in solids. Such works are deeply related to the newly developed field of ultrafast switches based on the transient ionization of dielectrics on femtosecond and sub-femtosecond scale [134–140]. Generation of new frequencies via resonant radiation in process of the soliton breakup is also a promising way to produce extremely broad spectra compressible to sub-femtosecond pulses as well as to produce optical combs covering the unprecedented frequency range at once, from the fine atomic transitions in the microwave to the low frequency nuclear transitions in VUV. The works on ultrashort pulse generation and control can pave the way to new highly efficient compact laser sources capable for few-cycle pulses in optical and even in THz range.

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