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*To my parents, wife, son, and daughter*

# Preface

The goal of this book is to show the most recent findings of newly emerged field of high-order harmonic generation (HHG) of laser radiation in the extended laser-produced plasma plumes. Ablation targets for HHG differ from the conventional gas targets used for harmonic generation in a number of ways. They are usually preformed plasmas with a density distribution that is determined by the ablation dynamics and subsequent evolution of plasma, which depend on the target composition and ablation pulse parameters in a complex way. In this connection, I would like to say a few words about different methods of generation of the coherent short-wavelength radiation using HHG approach.

The HHG in gases was for the first time observed in the second half of the 1980s using picosecond Nd:YAG lasers (as well as an excimer laser at 248 nm). The harmonics from different gases up to the 21st and 33rd orders of 1064 nm radiation were reported at an intensity of $3 \times 10^{13}$ W cm$^{-2}$, which led to an enormous growth of interest in this area of nonlinear optics. Those studies have demonstrated that the application of gases as nonlinear media can be used as an advanced method for generation of coherent extreme ultraviolet (XUV) radiation using picosecond driving pulses. Those early developments were further transformed in the field of gas HHG spectroscopy when new, predominantly femtosecond, lasers became involved in this field of study. Currently, harmonics up to the 5000th orders have been reported, although most recent studies are related with the development of the attosecond sources of laser radiation based on the gas HHG. Other applications include the analysis of the orientational features of some gaseous molecules through the study of variable harmonic spectra from these species, as well as the applications of gas harmonics for surface science, biology, medicine, and different branches of physics and chemistry. The attractiveness of this method is based on the availability of moderate-level femtosecond lasers in many laboratories worldwide and the simplicity of handling the gas-jet technique.

Another method, harmonic generation from the surfaces, which is based on completely different physical principles than HHG in gases, is less popular due to the sophisticated equipment required for its implementation. However, this method is well elaborated and used in advanced laboratories. Very high fluences and intensities (of the order of $10^{18}$ W cm$^{-2}$ or higher) and, most importantly, very high contrast ratios between the driving pulse and the prepulse already existing in any laser system are the main requirements for this technique. Not many laboratories can afford these conditions for surface HHG. Nevertheless, high cutoffs (up to the 2000th harmonics) and high conversion efficiencies for the lowest orders of harmonics were reported using this technique. Two distinct generation mechanisms have been identified to contribute to surface HHG: the coherent wake emission and the relativistic oscillating mirror process. Both mechanisms emit coherent XUV radiation in the reflected direction through nonlinear conversion processes at the plasma front surfaces.

One has to note that there are many good publications showing a whole picture of developments of the HHG in gases. The mechanisms of odd and even harmonic generation in the reflection of laser radiation from the surfaces are also frequently discussed in the literature. These HHG techniques are beyond the scope of this book. My aim is to familiarize the reader with the most recent approaches of harmonic generation in the XUV range with the use of the extended plasma plumes, which have never been used and are different from commonly used gas-jet sources.

Two monographs on the history of harmonic generation in the narrow plasmas and the achievements using new approaches that emerged between 2005 and 2013 have already been published (R.A. Ganeev, *High-Order Harmonic Generation in Laser Plasma Plumes*, Imperial College Press, 2012; R.A. Ganeev, *Plasma Harmonics*, Pan Stanford Publishing, 2014). Meanwhile, the developments in this field have shown new opportunities in further amendments of this technique. I will discuss some of these new findings appeared during HHG experiments in the narrow plasma plumes, while mostly concentrate on the studies using the extended plasmas.

What differs this book from my above-mentioned publications? The main difference is related to the application of the long plasma medium for HHG, instead of short one, to demonstrate the attractive properties of extended laser-produced plasmas from the point of view of the high-order nonlinear optical properties of various materials. Particularly, the quasi-phase matching in modulated plasmas demonstrated in a few recent studies has revealed the opportunity to tune the groups of enhanced harmonics along the XUV by different means. Another interesting finding is related with the opportunity to analyze the electron density of extended modulated plasmas. Other developments are related to the new advanced methods of harmonic generation emerged during recent time. Those methods include the application of double-pulse scheme for the analysis of the nonlinear optical properties of extended targets without the preliminary ablation by the picosecond heating pulses. The application of extended nonmetal targets (semiconductors, crystals, clustered carbon structures, etc.) allows the analysis of the relation between the excited ionic species emitting incoherent light and the harmonics coinciding with these ionic transitions. The studies of resonance enhancement in the nonmetal ablations reveal new opportunities to move farther toward the shortest wavelengths. A search of the enhanced response of carbon-contained clusters, study of the morphology of nanoparticle deposits from the ablated long targets, applications of the graphene-wrapped sheets for HHG, amendments of the two-color pump of plasmas caused by the positive dispersion of the extended medium allowing better overlap of the fundamental and second-harmonic orthogonal fields, etc., are among other distinctive moments differing this book from the two previous monographs.

All these amendments of plasma harmonics could not be realized without the collaboration between various research groups involved in the studies of the nonlinear optical properties of ablated species. Among numerous colleagues, I met and had the privilege to collaborate, I would like to thank H. Kuroda, M. Suzuki, S. Yoneya, M. Baba, and F. Mitani (Saitama Medical University, Japan); T. Ozaki, L. B. Elouga Bom, J. Abdul-Haji, and F. Vidal (Institut National de la Recherche Scientifique, Canada); P.D. Gupta, P.A. Naik, H. Singhal, J.A. Chakera, R.A. Khan, U. Chakravarty, M. Raghuramaiah, V. Arora, M. Kumar, and M. Tayyab (Raja Ramanna Centre for Advanced Technology, India); J.P. Marangos, J.W.G. Tisch, C. Hutchison, T. Witting, F. Frank, T. Siegel, A. Zaïr, Z. Abdelrahman, and D.Y. Lei (Imperial College, United Kingdom); M. Castillejo, M. Oujja, M. Sanz, I. López-Quintás, and M. Martín (Instituto de Química Física Rocasolano, Spain); H. Zacharias, J. Zheng, M. Wöstmann, and H. Witte (Westfälische Wilhelms-Universität, Germany); T. Usmanov, G.S. Boltaev, I.A. Kulagin, V.I. Redkorechev, V.V. Gorbushin, R.I. Tugushev, and N.K. Satlikov (Institute of Ion-Plasma and Laser Technologies, Uzbekistan); M.B. Danailov (Sincrotrone Trieste, Italy); B.A. Zon and M.V. Frolov (Voronezh State University, Russia); D.B. Milošević (University of Sarajevo, Bosnia and Herzegovina); M. Lein and M. Tudorovskaya (Leibniz Universität Hannover, Germany); E. Fiordilino (Università degli Studi Palermo, Italy); V. Toşa and K. Kovács (National Institute of R&D Isotropic and Molecular Technologies, Romania); V.V. Strelkov and M.A. Khokhlova (General Physics Institute, Russia); M.K. Kodirov and P.V. Redkin (Samarkand State University, Uzbekistan); A.V. Andreev, S.Y. Stremoukhov and O.A. Shoutova (Moscow State University, Russia) for their activity in the development of this relatively new field of nonlinear optics. Among them, I would like to underline the role of Prof. H. Kuroda who actively pursued the field of plasma harmonics starting from the very beginning of these studies in 2005. Professor Kuroda has supported my proposal to initiate the studies of the extended plasma media for the HHG. His invitation to carry out these studies in Saitama Medical University, Japan, played a crucial role in the developments of this field.

My wife Lidiya, son Timur, and daughter Dina are the main inspirations of all my activity. Now, becoming a grandfather, I would like to include Timur's wife Anya and our beloved grandson Timofey in the list of most important people, who help me to overcome various obstacles of the life of scientific tramp.

This book is organized as follows. Theoretical and experimental aspects of HHG are considered in the Introduction section. In Chap. 2, a review of most important results of the HHG in narrow plasmas is presented. Here, I also show recent studies of the small-sized plasma plumes as the emitters of high-order harmonics. In Chap. 3, various findings during application of extended plasmas for harmonic generation are analyzed. One of the most important applications of extended plasmas, the quasi-phase matching of generated harmonics, is demonstrated in Chap. 4. Here, I show various approaches in modification of perforated plasma plumes. Chapter 5 depicts the nonlinear optical features of the extended plasmas produced on the surfaces of different nonmetal materials. Chapter 6 is dedicated to the analysis of the new opportunities of extended plasma-induced HHG. The advantages of application of the long plasma plumes for HHG, such as resonance enhancement and double-pulse method, are discussed in Chap. 7. Finally, I summarize all these finding and discuss the perspectives of extended plasma formations for efficient HHG and nonlinear optical plasma spectroscopy.

Moroyama, Japan
January 2016

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# Contents

# Chapter 1
# Introduction. Theory and Experiment of High-Order Harmonic Generation in Narrow and Extended Media

High-order harmonic generation (HHG) is a technique for producing spatially and temporally coherent extreme ultraviolet (XUV) light. HHG in isotropic media occurs when an intense pulsed laser beam is focused into a gas jet or plasma. The intensity of the laser light is chosen such that its electric field amplitude is comparable to the electric field in atoms and ions. Such fields are able to detach electrons from above species by tunnel ionization, as opposed to photo-ionization by a weak field with high enough photon energy. The detached electron is accelerated in the field and under certain conditions has significant probability to hit the ion left behind upon return. The "collision" results in the emission of high energy photons. This description is called three-step model. The details of this process will be discussed below.

The use of extended media may offer new opportunities in application of XUV radiation for various needs. In Introduction, we discuss the use of extended gaseous media for HHG and show the advantages of this approach, which has prompted the use of long medium for the studies of the high-order nonlinear optical properties of various materials through the plasma harmonic approach.

## 1.1 High-Order Harmonic Generation in Isotropic Medium: Three-Step Model and Macroscopic Consideration of Frequency Conversion

When a high-intensity laser pulse passes through a gaseous or plasma medium, its atoms and ions emit odd harmonics. For a laser radiation wavelength $\lambda$, a superposition of the components $\lambda$, $\lambda/3$, $\lambda/5$, $\lambda/7$, etc. is observed at the output of the nonlinear medium. The harmonics of laser radiation result from a three-stage process [1–3] that comprises the ionization of an atom (or ion), the electron acceleration in the electromagnetic field, and the subsequent recombination with parent particle and emission of harmonics. This process is periodically repeated every half cycle of the electromagnetic wave. The highest-order harmonics are due to the electron acceleration at the instant of ionization at the peak intensity of the



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*laser pulse. Therefore, the generation of highest-order harmonics in isotropic media* results from the interaction of a high-intensity light field with atoms [4–6], atomic clusters [7, 8], molecules [9, 10], and ions [11–16].

A characteristic feature of the three-stage HHG is a rapid decrease in the intensity of low-order harmonics followed by a long plateau where the intensities of high-order harmonics differ only slightly from one another, and an abrupt decrease in the intensity of the highest-order harmonics generated (the so-called harmonic cut-off $H_c$). The position of $H_c$ is determined by the ionization potential $I_i$ of the particles participating in harmonic generation (of atoms in the case of HHG in gases and of ions in the case of HHG in a plasma) and by the ponderomotive potential, which defines the accelerated electron energy and depends on the intensity of femtosecond radiation and it wavelength ($U_p \approx 9.3 \times 10^{-14}\, I_{fp}$ [W cm$^{-2}$] $\lambda^2$ [μm$^2$]). The highest photon energy of the harmonics generated is defined by the relation $E_c \approx I_i + 3.17U_p$.

The main HHG properties may be characterized with the aid of a semiclassical model [1–3], although a more accurate description calls for a consistent quantum mechanical treatment. In the semiclassical model, an electron detached from an atom is considered a free particle, and the effect of all bound states, with the exception of the ground state, is assumed to be negligible. These assumptions are satisfied in the tunnel ionization regime, which is characterized by the inequality $\gamma < 1$ for the Keldysh parameter $\gamma = \omega_L(2I_i)^{0.5}E^{-1}$ where $\omega_L$ is the laser radiation frequency and $E$ is the magnitude of the electric vector of the electromagnetic wave. High-power ultrashort laser pulses are best suited for the fulfillment of these conditions.

Along with the microscopic consideration of the processes occurring in the interaction of high-power ultrashort laser pulses with atoms and ions, account should also be taken of macroscopic processes such as the effect of transmission through a medium and group effects. These effects primarily include dephasing, absorption, and defocusing. These processes are analyzed in [17].

The cut-off law clarifies that the maximum harmonic frequency achievable from the HHG process is strongly linked to the ponderomotive potential $U_p$ and thus to the field amplitude and the wavelength of the fundamental laser light. The maximum applicable field amplitude is limited, because, for very high intensities of the driving laser well above $10^{16}$ W cm$^{-2}$, the magnetic component of the laser field becomes strong enough to induce a lateral acceleration, hence deflecting the electron, reducing the overlap between the electronic and the nuclear wave packets and thus preventing efficient harmonic generation. However, the cut-off law states that the maximum harmonic frequency in the HHG spectra will increase for longer wavelengths of the driving laser field. A shift of the cut-off frequency towards the water window, or even the keV range of photon energy, by using driving lasers in the few μm range was demonstrated at experimentally relevant harmonic flux in [18, 19].

Note that not only the cut-off law, but also some other interesting limits on the HHG process are explained by the above model. For instance, HHG will only occur if the driving laser field is linearly polarized. Electrons in an elliptically polarized laser field fly in spirals and therefore miss the parent nucleus. In terms of quantum mechanics, the overlap of the nuclear and the electron wave packets is reduced upon return. This has been observed in experiments, where the intensity of harmonics has decreased rapidly with increasing ellipticity [20]. However, it is possible to generate elliptically polarized harmonics with linearly polarized driving laser fields by using aligned molecules as non-linear media for harmonic generation, for example laser aligned $N_2$ molecules as demonstrated in [21].

The conclusions on the coherence properties of the HHG radiation can also be drawn from the above model. The electron has to be considered as a quantum mechanical wave packet, which undergoes a transition from a bound state to a continuum state at a certain time $t_i$, evolves in the laser field and finally descends to the bound state again under radiation of the kinetic energy gained while propagating through the continuum. This quantum wave packet oscillates with its own frequency, however the total phase of the electron at recombination and therefore the phase of the occurring XUV radiation is strongly linked to the time of ionization and to the strength of the fundamental laser. Thus the phase of the electronic wave packet at recombination and therefore the phase of the XUV light are locked to the phase and amplitude of the fundamental laser beam. This influences the collective behavior in the spatial domain, since coherence properties of the irradiating laser are transferred to the harmonic emission, hence forth HHG is a spatially coherent process.

The total emitted field in a macroscopic medium is given by a sum over the emissions from many atoms. Thus not only the single atom response, but also collective effects as phase matching or re-absorption of the XUV light determine the intensity of the generated harmonics. Phase matching is given, if the radiation generated by different atoms at different positions in the medium interferes constructively at the exit of the medium. For a perfect match of phases, this condition can be presented as

$$\Delta k = k_q - qk_0 = 0, \tag{1.1}$$

where $\Delta k$ denotes the mismatch between the wave vectors $k_q$ of harmonic $q$ and $k_0$ of the fundamental. Approximate phase matching is achieved for

$$\Delta k L_{med} < \pi, \tag{1.2}$$

where $L_{med}$ describes length of the medium. The dependence of the harmonic phase $\varphi_q$ on the laser intensity at the position of emission can be written as $\varphi_q = \alpha_q I$, where $\alpha_q$ is linked to the $q$th Fourier component of the atomic polarization and proportional to the atomic dipole moment and density. An additional wave vector $k_I = -\nabla \varphi_q$ enters the phase mismatch, leading to a generalized phase matching condition for HHG [22, 23]:

$$\Delta k = k_q - qk_0 + k_I. \tag{1.3}$$

The transversal intensity profile and wavefront shape play an important role for optimizing the HHG yield. Here we consider the case when the Gouy phase shift becomes insignificant during propagation of the loosely focused fundamental radiation through the extended medium. The condition for this consideration is related with the relation

$$L_{\mathrm{med}} < b. \tag{1.4}$$

Here $b$ denotes the confocal parameter of the focused radiation. This relation indicates that the focused radiation propagates through the medium at the conditions of plane wave when the phase of wave front becomes unchanged along the beamwaist. More details on the phase matching conditions will be discussed in Chap. 4.

## 1.2 Overview of the Applications of Long Gaseous Media for the HHG

The most common experimental setup of HHG comprises (apart from laser system and diagnostic apparatus) a gas puff target. Such a target is basically a gas valve injecting a portion of a gas at desired pressure. The valve is repetitively opens and the laser pulses interact with gas in proximity of valve exhaust. Typical repetition rate of the gas puffs is <100 Hz. The valves may have either circular symmetry or could be elongated. Elongated valves provide higher XUV beam outputs but limiting factor is re-absorption in a gas. Thus long valves are used for wavelengths >20 nm (also because of the longer coherence length for longer wavelengths). On the other hand, circular (e.g. 0.5 mm diameter) gas puff valves are used in shorter-wavelength HHG.

High-order harmonics are also generated in extended gas cells. A gas cell is a simple container filled with a noble gas at moderate pressure (few tens of mbar). Arrangements with gas cells are very comfortable to work with because, compared to the gas puff targets, there are fewer parameters to optimize to maintain the phase matching. In this case the phase matching is obtained by tuning only longitudinal position of a cell and gas pressure in it. The gas cells can be as long as desired (due to technical ease of construction compared to gas puff valves). This setup is also favorable when maximization of interaction length is wanted.

Another possible geometry of HHG involves hollow fibers filled with a conversion gas. In such a fiber laser pulses are propagating even meters long resulting in efficient transfer of driving field energy into XUV beam. This geometry is also popular due to ease of control of phase-matching by the fiber parameters.

Below we briefly review the advantages of long media over the narrow ones in the case of the HHG in gases. Initially, HHG was studied using the narrow gas jets, with the sizes of the active medium of the order of 0.3–0.5 mm to decrease the residual pressure in the XUV spectrometers. The gradual increase of the sizes of