

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 $3 \times 10^{13} \text{ W cm}^{-2}$, 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.

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