Quantum Electronics **45** (9) 785–796 (2015)

REVIEW

PACS numbers: 42.65.Ky; 52.38.Mf; 42.65.Re
DOI: 10.1070/QE2015v045n09ABEH015574

Why plasma harmonics?

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Abstract. **We discuss the emergence of interest in the high-order harmonic generation (HHG) of ultrashort pulses propagated through laser-produced plasmas. It is shown that, during the last few years, substantial amendments of plasma HHG allowed in some cases the characteristics of gas HHG to be surpassed. The attractiveness of a new approach in coherent extreme ultraviolet radiation generation is demonstrated, which can also be used as a tool for laser-ablation-induced HHG spectroscopy of a giant class of solids. We present general ideas and prospects for this relatively new field of nonlinear optics.**

Keywords: *plasma high-order harmonics, laser ablation, ultrashort laser pulses.*

1. Introduction: sources of coherent extreme ultraviolet radiation

The question, posed in the title of this review, is closely related with the directions of further developments in one of branches of nonlinear optics. Indeed, why plasma harmonics? Do we need plasma harmonics if we have a well-elaborated technique of frequency conversion of laser radiation towards a short-wavelength range through nonlinear optical high-order harmonic generation (HHG) in gaseous media? One can recall that there is also another technique of HHG, i.e., surface harmonic generation. Both of them have a relatively long history of developments and applications for various needs and seem to fully satisfy the scientific community as sources of coherent extreme ultraviolet (XUV) radiation.

From the conventional point of view, the developments in our lives should follow logical principles of simplicity and economy. Ptolemy stated [1]: “We consider it a good principle to explain the phenomena by the simplest hypothesis possible.” This philosophical vision of everyday life was later developed in the medieval period as a principle of Ockham’s razor, which is also called law of parsimony, economy, or succinctness used in logic and problem-solving. It states that “one should proceed to simpler methods until simplicity can be traded for greater explanatory and usefulness power” [2]. One should note that the simplest available method or theory need not be the most accurate. Moreover, science itself is an example of Ockham’s razor principle, since new, more sophisticated ideas and hypotheses, which initially contradicted the commonly accepted and simplest explanations and approaches in the description of the basics of our universe, have finally completely changed our vision of the surrounding world.

However, let us return from the heights of philosophical heavens back to our Earth. Here we will consider a new approach in optics, which has no aim to replace the existing methods of the development of coherent short-wavelength sources, but just feels good enough to find its own niche in this field of physics. Is the principle of Ockham’s razor (“It is futile to do with more things that which can be done with fewer”) applicable in science, and particularly, in nonlinear optics, and more particularly, in the methods of coherent short-wavelength generation through the nonlinear interaction of light and matter? To answer this question, as well as the question posed in the title of this review, let us first recall the history of gas and surface HHG, as well as the best achievements of those techniques. We also briefly discuss here other methods of generation of the coherent XUV light.

The earliest observations of the HHG in gases were reported during the second half of the 1980s using a picosecond Nd : YAG laser at 1064 nm [3], as well as an excimer laser at 248 nm [4]. The harmonics from different gases up to the 21st and 33rd orders of 1064 nm radiation were reported at an intensity of 3×10^{13} W cm^{-2}, which led to an enormous growth of interest in this field of nonlinear optics. Those studies demonstrated that the application of gases as the nonlinear media can be used as an advanced method for generation of the coherent XUV radiation using picosecond driving pulses. Those early developments were further transformed towards the field of gas HHG spectroscopy when femtosecond lasers became involved in this area of studies. Currently, the harmonics up to the 5000th orders have been obtained, while some recent gas HHG studies are related with the development of attosecond sources of laser radiation, analysis of structural features of some gaseous molecules through the study of harmonic spectra from these species, as well as applications of gas harmonics for surface science, biology, medicine, physics and chemistry. The attractiveness of this method is based on the availability of femtosecond lasers in many laboratories worldwide and the simplicity in handling the gas HHG technique.

The surface HHG, which is based on another physical mechanism, is less popular due to necessity of advanced equipment required for its implementation. Extremely high fluences and intensities (above 10^{18} W cm^{-2}) and, most important, very high contrast ratios between a driving pulse and a prepulse already existing in any laser system are the main requirements for this technique. A few of laboratories can

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Received 1 June 2014; revision received 9 December 2014
Kvantovaya Elektronika **45** (9) 785–796 (2015)
Submitted in English

afford these requirements for the surface HHG. The extended cut-offs (above the 3000th harmonic) and high conversion efficiencies for the lowest orders of harmonics were achieved using this technique.

Additional approaches in coherent XUV generation include X-ray lasers [5–7] and free-electron lasers [8, 9], which are based on other principles than the HHG of laser radiation [10, 11]. The gas HHG seems very attractive from the point of view of availability of moderate energy/intensity lasers in many laboratories and less expenditure for the everyday use compared with two above approaches. Other disadvantages of X-ray lasers are their low pulse repetition rate, poor spatial coherence and divergence. As regards free-electron lasers that generate radiation in the short-wavelength spectral range, there are only a few of sources available so far. Furthermore, the application of these lasers is largely limited by their high cost.

The main concern for coherent XUV pulses is the typically rather weak strength and low generation efficiency. Many applications require substantially stronger XUV brightness. To achieve high intensity, laser-plasma interaction has been exploited as a promising alternative because surface-based over-dense plasma can withstand high laser fields driving the harmonics. Two distinct generation mechanisms have been identified to contribute to HHG from over-dense plasmas: coherent wake emission [12–15] and relativistic oscillating mirror process [16–19]. Both mechanisms emit XUV radiation in the reflected direction through nonlinear conversion processes at the plasma front surfaces. Another mechanism called coherent synchrotron emission by dense electron nanobunches can generate coherent XUV pulses in the transmitted direction [20, 21].

XUV lasers and systems based on high-order harmonics are being developed in many laboratories worldwide. For its part, the semiconductor industry is investing heavily in incoherent plasma sources. Compared with XUV lasers, the first offers a shorter pulse width, wavelength tunability and higher repetition rates. The second can provide magnitudes more power. However, both alternatives also suffer from disadvantages when compared with XUV lasers. High-harmonic sources produce lower energy and spectrally broader pulses. Plasma sources are limited in their peak brightness. Thus different XUV sources have their advantages and disadvantages.

Today's advanced coherent XUV sources produce, at best, a few tens of microwatts of pulse power. In the meantime, the 13.5-nm XUV lithography being pursued by the semiconductor industry [22] requires hundreds of watts of in-band power. Thus, available coherent XUV sources cannot be considered for micro-lithography for many years to come.

Though not applicable for micro-lithography, coherent XUV sources could be useful elsewhere in semiconductor manufacturing because they enable high-resolution imaging. The short wavelength of these sources offers advantages, particularly by focusing the laser into a small spot, on the order of 50 nm. This makes it possible to ablate a sample, such as a microorganism, and study its chemical composition at different spots for the goals of biology and medicine.

Other coherent XUV research applications include patterning, imaging, photochemistry and microscopy. Particularly, to work in single-shot mode, such microscopes need an adequate photon flux, such as recently developed new class of XUV lasers [23–35]. These devices fire laser pulses onto a solid target. The initial ones ablate the target and create plasma, and they are followed by a main driving pulse, which arrives at an angle, producing a wave that travels through the plasma at a speed calibrated to improve the XUV production efficiency. For imaging, one area of attention is on photons in the 3 nm region, where there is a water window created by differential absorption of oxygen and carbon. This allows a natural contrast between a biological specimen and the water that surrounds it.

Returning back to the HHG approaches, one has to note that there are many good reports showing the developments of gas HHG (for example, review [36]). The mechanisms of odd and even harmonics generation in the reflection of laser radiation from over-dense plasmas were also frequently discussed in the literature (see monograph [37] and review [38]). These HHG techniques are beyond the scope of present review paper. The aim is to acquaint the reader with most recent approaches of harmonic generation in plasma plumes and to discuss the future developments of this field.

The motivation for writing this review paper containing some important missing parts not discussed in other reviews is to show the most recent findings of various new schemes of the high-order harmonic generation in laser-produced low-ionised, low-dense plasma plumes, which I simply dubbed as 'plasma harmonics'. Some incorrectness of this term is probably compensated for by the shortness and attractiveness of this definition. Indeed, since the first experiments on HHG in gaseous media (e.g. gas jets and gas cells) the terms 'gas harmonics' and 'gas HHG' appeared in the scientific literature, alongside with the term 'surface harmonics' referring to another process of HHG during the specular reflection of strong laser fields from the surfaces of various materials. Since most of studies on the harmonics generation in specially prepared plasma plumes during last time have been performed with my participation, I feel the privilege to dub this field of nonlinear spectroscopy as plasma harmonics.

So again, why plasma harmonics, when gas harmonics is an elaborated technique, which, on the one hand, allows the generation of coherent XUV pulses and, on the other hand, gives the opportunity in analysis of the features of some molecular gases and provides the generation and application of attosecond pulses? This question arose in the first half of the nineties, when first studies of high-order harmonics from plasma plumes were performed. The frustration caused by the unimpressive results shown in those studies was the reason to ask oneself on the necessity in the plasma harmonic experiments. In the following section I will discuss those results and reasons of why the proposed technique was forgotten for almost ten years. This give up in plasma harmonics coincided with the impressive achievements in creation of the sources of coherent XUV radiation through generation of gas harmonics, as well as the first reports on the generation of attosecond pulses. Probably, Ockham's principle was a sufficient reason for a drop of interest in plasma harmonic studies, since no ideas were disseminated among the laser community on how to overcome the obstacles of notoriously low conversion efficiency of this process, as well as low harmonic cut-offs compared with the achievements of gas HHG.

It seems too early to talk about the novelties reported during the first successful HHG experiments with plasma plumes in 2005. Let us follow the logic of the historical developments in this field. In the following section, I discus some advantages and disadvantages of plasma harmonics observed by researchers at the beginning of those studies. I also was among them, and one can imagine my frustration in those results,

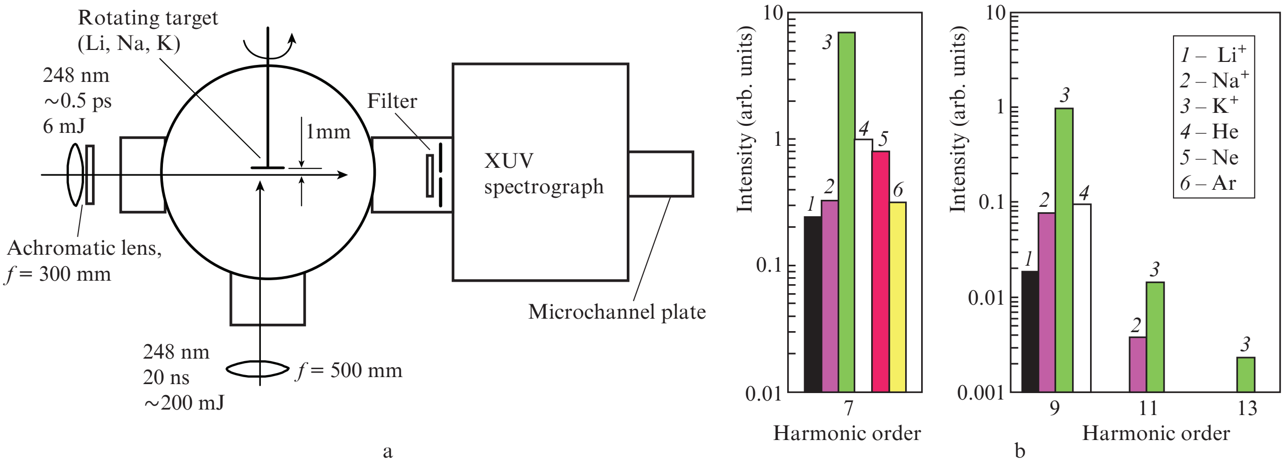

Figure 1. (a) Schematic diagram of the experimental setup and (b) relative intensities of harmonics in the rare gases and alkali-metal ions (adapted from [39] with permission from the American Physical Society; copyright 1992).

which were far behind of our expectations and best achievements of gas harmonics.

2. Early stage of plasma harmonic studies: hopes and frustrations

First studies of plasma HHG were aimed to extend the harmonic cut-off by using particles with high ionisation potentials, which seemed very logical from the consideration of the micro-processes driving harmonic generation. From this point of view, single-charged alkali metal ions (Li^+, Na^+ and K^+) should be a good choice, because they are isoelectric to neutral rare gases and have higher ionisation potentials than those of rare gas atoms. Akiyama et al. [39] used sub-picosecond KrF laser pulses (0.5 ps, 6 mJ, 248 nm) for harmonic generation in plasmas containing alkaline metal ions produced by 20 ns, 200 mJ, 248 nm radiation (Fig. 1a). The most extended harmonic observed had the 13th order in the K^+-containing plasma; the harmonics up to the 11th order in Na^+ and the 9th order in Li^+ were also reported. Among the nonlinear optical media used, the K^+ ions provided the highest intensity for all orders of harmonics (Fig. 1b).

The same team, in their following research, used other ions in laser-produced plasmas for the HHG in order to minimise the influence of ionisation processes [40]. The highest observed order was the 21st harmonic generated in lead ions (Fig. 2a). The highest harmonics in various ions and neutral gases were found to be proportional to the ionisation potentials of those species. The experiments were carried out using an extremely high intensity of a subpicosecond KrF laser (0.5 ps, 3×10^{17} W cm^{-2}), while an intensity of a 20 ns heating pulse was in the range of 1×10^9 W cm^{-2}. Six solid targets with various atomic numbers were chosen: boron ($Z = 5$), carbon (6), titanium (22), copper (29), tungsten (74) and lead (82). Figure 2b compiles the harmonic distributions using these six elements. For each species a dominant charge state was evaluated using a collisional–radiative model.

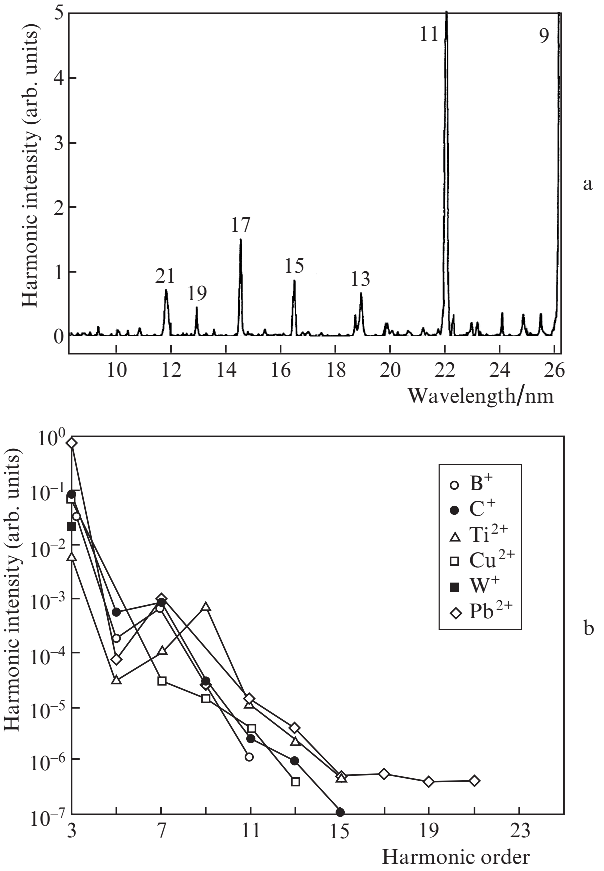

Figure 2. (a) Time-integrated high-order harmonic spectrum observed in a lead plasma (the spectral peaks are labelled by the harmonic orders) and (b) high-order harmonic intensity distributions of six ionic species (reproduced from [40] with permission from the American Physical Society; copyright 1993).

Wahlström et al. [41] also studied the HHG in laser-produced alkaline (Na^+ and K^+) ions using 150 fs, 794 nm driving pulses and 100 ps, 1064 nm heating pulses. The highest observed orders were not as high as expected from the estimates based on the saturation intensities for those ions and on the focused intensities in the absence of defocusing (Fig. 3). The spatial far-field distribution of the harmonic radiation was found to exhibit the ring structures, which were varied with the focusing conditions. The researchers assumed that the absence of a plateau-like harmonic distribution may be a consequence of ionisation-induced defocusing of the high-power laser beam, reducing the peak intensity obtained

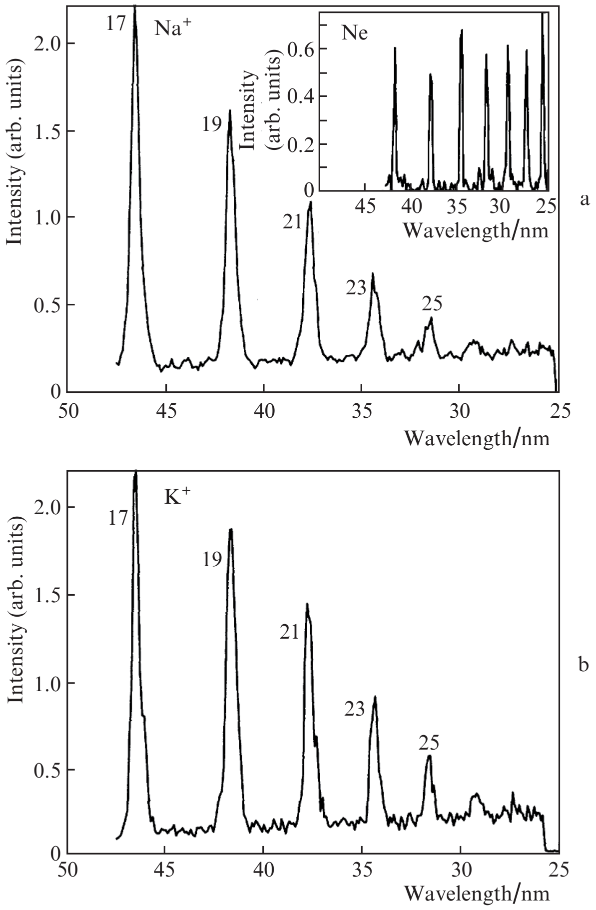

Figure 3. Harmonic spectra obtained in (a) Na$^+$ and (b) K$^+$. The inset in (a) is a part of spectrum obtained under similar conditions, but with the laser produced plasma replaced with a pulsed nozzle producing a jet of neutral Ne atoms (reproduced from [41] with permission from the American Physical Society; copyright 1995).

in the medium. This defocusing is expected to be diminished by using a short-wavelength laser for harmonic generation.

The propagation effects in the HHG (up to the 13th order) of short-pulse KrF laser radiation in carbon vapour and low charged carbon plasma was studied in [42]. It was found that, under used experimental conditions, high-order harmonics were generated mainly from neutral atoms. The authors of [42] also assumed that, in plasma, the sum-frequency generation of high-order harmonics was suppressed by an unfavourable positive phase-mismatch. In their work, the noncollinear phase-matched HHG by difference-frequency mixing in plasmas was discussed. They suggested that the observed anomalous growth of the fifth harmonic intensity at longer plasma lengths originates from the noncollinear phase-matched difference-frequency mixing and defocusing of the laser radiation.

In [43], the possibility of optimisation of the HHG in plasma when only one laser is used as a heating and driving source has been demonstrated. An analysis of plasma excitation showed that the double-beam scheme has some advantages due to possibility of optimisation of the excitation level. It was demonstrated that the picosecond pulses can serve as an efficient tool for both generation of plasma and harmonic generation. In those experiments, the harmonics up to the eleventh order of Nd : glass laser radiation (λ = 96 nm) in a low-dense plasma were studied. A maximum efficiency (10^{-3}) was obtained in the case of the third harmonic, while the efficiency of highest (eleventh) harmonic was considerably lower ($\sim 10^{-8}$).

Krushelnick et al. [44] analysed the HHG from preformed under-dense plasmas using subpicosecond ultraviolet laser pulses focused to intensities up to 10^{18} W cm^{-2}. The generation of the seventh and ninth harmonics from aluminium plasma was reported as well as the harmonics up to the nineteenth order from lithium–fluorine plasmas. The harmonic generation efficiency measured in those experiments was less than that from the gas HHG experiments. In their studies harmonic generation was limited by the presence of the abundant amount of free electrons in the nonlinear medium, which caused dispersion and phase mismatch. Nonetheless, it was suggested that the harmonic generation from ions is observable in plasmas if the polarisation of the ion is sufficiently large and the electron density is not too high. The emission of the seventh and ninth harmonics from these plasmas was recorded at the laser intensities of 5×10^{17} W cm^{-2}, an intensity at which the efficiency of harmonic emission from initially neutral helium gas targets was similar. At low intensities, however, harmonic emission from neutral-gas targets was still significantly stronger than that from preformed plasma targets. The observation of the seventh and ninth harmonics from the aluminium plasmas was probably due to a nonlinear polarisation of the plasma ions. In fact, as the high-intensity laser pulse propagates through the plasma, the process of tunnelling ionisation may act to inhibit the production of harmonics if the generation mechanism can be regarded as a three-step process [45]. The effect of ionisation may also cause refractive defocusing, which prevents the laser beam from achieving high intensities inside the plasma. This process can also create heterogeneities in the plasma that could then contribute to the phase mismatch.

Those first experiments of the HHG in the passage of laser radiation through a plasma produced by laser ablation of a solid target turned out to be much less successful compared with gas harmonic studies. Above-presented data were obtained with the use of highly excited plasmas containing multiply charged ions. The studies revealed several limiting factors, which did not permit harmonic generation of reasonable efficiency and sufficiently high orders. The investigations only demonstrated relatively low-order harmonics (see Fig. 4 [46] that summarises the best results achieved during early stages of plasma HHG studies).

These disadvantages, as well as the low conversion efficiency, led to erosion of interest in this plasma harmonic technique, especially in comparison with the achievements involving gas HHG sources. From 1997 till 2005, there were no publications on plasma harmonics, since nobody believed in success in this field. All efforts of those groups, who initially tried to achieve some interesting results using the plasma harmonic technique, were turned towards gas HHG. The plasma harmonics were pronounced dead.

3. From frustration to success: light at the end of the tunnel

In the late 2004, I studied at the Institute for Solid State Physics (Japan) the properties of boron target ablation and excitation using both strong (femtosecond) and relatively weak (picosecond) laser pulses. Ablation was performed using 300 ps pulses, and then the plasma plume was irradiated

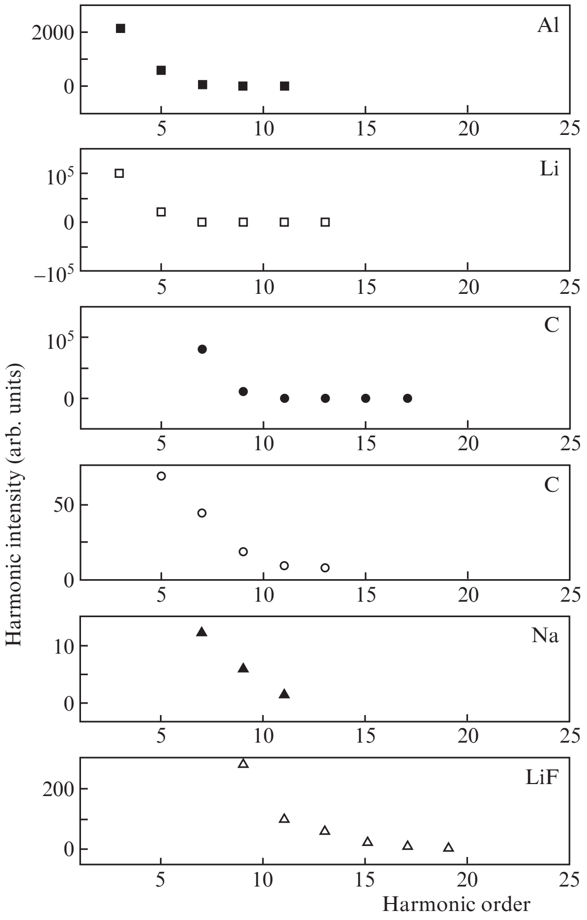

Figure 4. High-order harmonic spectra obtained from various laser-produced plasmas at the early stage of research (1992–1997): Al [43], Li [39], C [40], C [42], Na [39] and LiF [44] (reproduced from [46] with permission from Wiley-VCH; copyright 2012).

using a broad beam of the second harmonic of a Ti:sapphire laser ($\lambda \sim 400$ nm) to obtain shaddowgrams. The aim of those studies was to measure a velocity of plasma wavefront expansion in the case of strong ablation of the targets for explanation of the backscattered three-halved harmonic generation observed earlier in our experiments [47].

Our equipment allowed the observation of plasma spectra in the XUV range. We often analysed the UV and XUV spectral characteristics of the plasmas produced by pulses of different duration. The plasma spectra during ablation of boron (Fig. 5) at a 40-ns delay between a 300 ps and 110 fs pulse were collected. Unlike a conventional experiment, we analysed what happened when the plasma is produced at low-dense, low-ionised conditions during interaction of the picosecond pulses with the target, without the influence of the femtosecond pulse. No plasma lines in the XUV, as expected, were observed in that case. However, once femtosecond pulses were focused inside this plasma, a set of multiple narrow lines resembling the harmonic spectral distribution accompanied with a few lines of plasma emission was observed directly on the phosphor screen of a microchannel plate with naked eye, without a CCD amplifier (Fig. 6a). The increase or decrease in the fluence of a heating picosecond pulse on the target surface above or below some level led to a disappearance of harmonic spectra. In the case of over-excitation of the boron target, strong ionic lines attributed to singly, doubly and triply charged particles, as well as the second and above orders of diffraction of extremely strong 6.03 and 4.86 nm emissions were dominated in the XUV spectra (Fig. 6b). In the meantime, at the optimal conditions of excitation of the boron target, the harmonics were spread along the whole area of the microchannel plate. The calibration of our XUV spectrometer confirmed that those emissions correspond to the odd integers of the wavelength of 800 nm driving radiation. The harmonics were extended up to the sixtieth orders demonstrating a plateau-like shape of the intensity distribution (Fig. 6c), analogously to the gas harmonics.

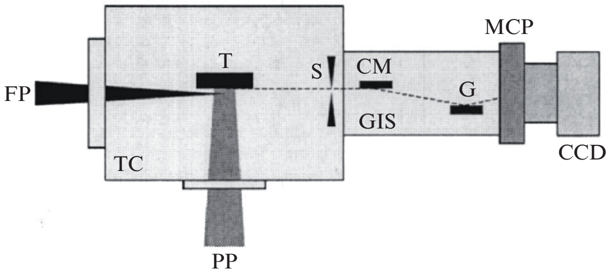

Figure 5. Schematic of the experimental setup for HHG: (TC) target chamber; (T) target; (S) slit; (GIS) grazing incidence spectrometer; (CM) gold-coated cylindrical mirror; (G) grating; (MCP) micro-channel plate; (CCD) charge-coupled device; (FP) femtosecond pulse; (PP) picosecond pulse (reproduced from [48] with permission from Optical Society of America).

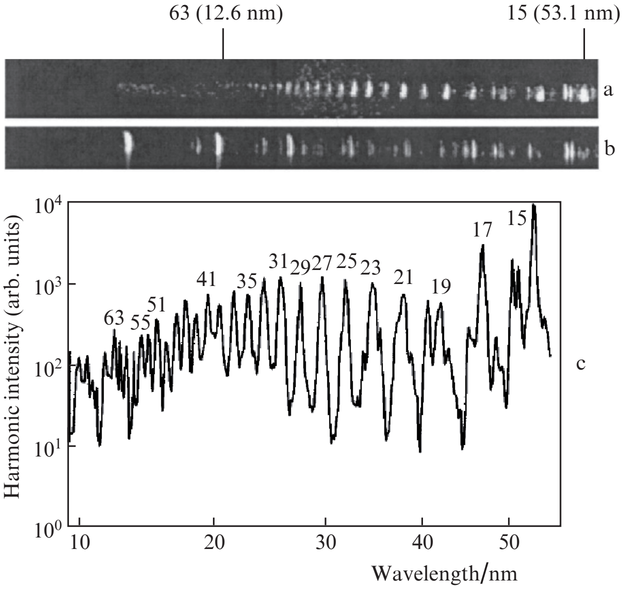

Figure 6. (a) Raw images of boron harmonic spectra obtained at optimal ablation of the target and (b) boron plasma emission appearing during strong ablation; (c) lineout of the harmonics generated from the boron plasma (reproduced from [48] with permission from Optical Society of America.

Thus, the occasional observation of the high-order nonlinear optical process in the boron plasma at the specific conditions of target ablation allowed formulating the main requirements for efficient HHG in this medium. Those results were published in 2005 [48]. It was the beginning of the studies of various plasma formations and their high-order nonlinear optical properties. From then, plasma HHG has opened new doors in many unexpected areas of light–matter interaction. Apart from an alternative method for generation of coherent XUV radiation, it proved to be a powerful tool for various spectroscopic and analytical applications. The application of doubly charged ions for high-order harmonic generation showed a promising extension of the cut-off photon energy of plasma harmonics, without having to rely on few-cycle driving pulses. As was shown in the case of low- and high-order harmonics, the conversion efficiency can be strongly enhanced in the vicinity of the resonances of atomic or ionic systems. This enhancement of a single harmonic was demonstrated during the experiments using the indium plasma. For laser-generated plasmas, a large variety of materials can be employed, thereby increasing the chance to match such resonances with the radiation of Ti : sapphire lasers. Furthermore, it was shown in gas HHG studies that the two-colour pump profitably enhances the high-order harmonic intensity and significantly influences the output and properties of the harmonic spectrum generation in noble gases. For plasma harmonics, where a two-colour pump technique has recently been introduced, this is a new approach for the nonlinear spectroscopy of the numerous ionic transitions of ablating materials possessing high oscillator strengths [49].

Those studies have shown that the enhanced high-order harmonics can also be generated from ablated nanoparticles, which opens the prospects for applications of the local field enhancement, use of broad plasmonic resonances in the XUV and efficient recombination processes for plasma HHG. As a highly interesting perspective an increase in the harmonic output by quasi-phase-matching (QPM) in specially prepared plasmas may be considered.

Thus the plasma harmonics have proven to be useful for producing an efficient source of short-wavelength, ultra-short pulses for various applications and studies of the properties of harmonic emitters. The laser-ablation-induced high-order harmonic generation spectroscopy is a new method for the studies of materials and one of most important applications of plasma HHG.

4. Capabilities of plasma harmonics: proposals and realisations

A search for the ways of increasing the notoriously low HHG efficiency in the XUV spectral range has long been (and still is) among the most topical problems of nonlinear optics. In the majority of cases, the conversion efficiency of high-order harmonics turns out to be insufficient for using them as reliable coherent short-wavelength radiation sources in biology, plasma diagnostics, medicine, microscopy, photolithography, XUV coherent diffraction imaging and time-resolved measurements, to mention few of them. The feasibility of increasing the intensities of high-order harmonics generating in gas jet sources by using atomic and ion resonances has long been studied primarily by theoretical methods [50, 51]. The results of those and other calculations suggested that the intensity of a single harmonic may be substantially increased when this harmonic is at resonance with the transitions of the atomic and ion spectra of gases. This approach may be an alternative to the method of phase matching for harmonics and laser radiation [52, 53].

As was shown in Section 2, first experiments using over-excited plasma plumes yielded frustrating results. Nevertheless, there was a reason to hope that harmonic intensities may be substantially increased and efficient shorter-wavelength coherent radiation may be obtained using properly produced plasmas. There are no fundamental limitations here; it is only needed to find the 'optimal' conditions for formation of a plasma plume to serve as an efficient medium for the HHG. Laser-produced plasma may be validly used for this process if the effects of the limiting factors (self-defocusing, self-phase modulation and wave-phase mismatch of the harmonics and the radiation being converted) are minimised [39, 43, 44].

Among the special features of HHG in laser-produced plasmas, one can note a wide range of medium characteristics, which can be tuned by varying the conditions of ablation on the surface of a solid. This applies to such parameters as the plasma length, the density of ions, electrons and neutral particles, and the degree of their excitation. The use of any elements of the periodic table, as well as thousands of complex samples that exist as solids largely extends the range of materials employed, whereas only a few rare gases are typically available for gas HHG. Thus the exploration of practically any solid-state material through the nonlinear spectroscopy comprising laser ablation and harmonic generation can be considered as a new tool for materials science.

In several cases, this method furnishes an opportunity to realise the quasi-resonance conditions and increase the efficiency of single harmonic generation due to the effect of ion transitions on the nonlinear response in the spectral range in question, thus allowing the studies of those transitions possessing large oscillator strengths [54–58]. This effect could be hardly observed in gas HHG because of a low probability for the coincidence of the atomic transition frequencies of a few gases and the frequencies of the single harmonics of laser sources. In the meantime, recent studies of the resonance enhancement of the harmonics generation in the gases have demonstrated the enhancement of the narrow parts of harmonics due to the influence of Fano resonances and Stark shift [59, 60].

A substantial increase in the highest order of generated harmonics, observation of a long plateau and emergence of a second plateau in the energy distribution of highest-order harmonics, high efficiencies obtained with several plasma formations, realisation of the resonance enhancement of individual harmonics, efficient harmonic enhancement from the plasma plumes containing clusters of different materials, and other properties revealed in [61–66] have demonstrated the advantages of using specially prepared plasmas for the HHG. The orders of harmonics obtained in plasma media to date range into the sixtieth and seventieth [48, 67, 68]. The highest harmonics (101st order) have been demonstrated during ablation of manganese [69] and currently can be routinely achieved using moderate level (60 fs, 3 mJ) laser pulses.

The plasma HHG conversion efficiency in the plateau region amounted to 10^{-5} was demonstrated in the case of the laser ablation of silver targets [70–72]. Application of a two-colour pump led to enhancement of the odd harmonics, as well as to the appearance of strong even orders [73–75]. In addition to that, the conversion efficiency towards an individual (resonantly enhanced) high-order harmonic approached 10^{-4} [54] (Fig. 7a). The use of extended plasma [76] allowed the implementation of the QPM concept for the plasma harmonics [77].

The quest for new plasma media that would favour the enhancement of an individual harmonic allows further improvement of the harmonic conversion efficiency. The pro-

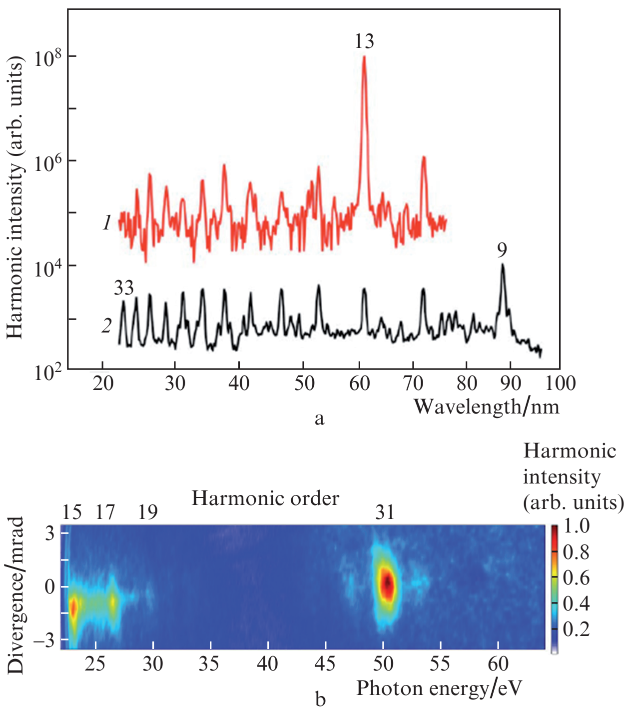

Figure 7. (a) High-order harmonic spectra from (*1*) In and (*2*) silver plasma plumes (reproduced from [54] with permission from Optical Society of America) and (b) raw image of the harmonic spectrum from manganese plasma in the case of 3.5 fs driving pulses (adapted from [82] with permission from Optical Society of America).

duction of a single high-intensity harmonic (rather than a group of harmonics of equal intensity in the plateau region) would open up the way to the practical application of these coherent short-wavelength sources. Resonantly enhanced harmonics observed in several plasma media allowed expecting that similar conditions will be discovered using other plasma formations. The generated harmonic wavelength may then be tuned to the transitions with a high oscillator strength by wavelength tuning of the driving laser [54, 65], as well as by varying the chirp of laser radiation [62, 64]. The yield of harmonics in the XUV can be also enhanced by application of the ablated nanoparticles and clusters, currently widely available for various purposes. Many new features of plasma harmonics, which have recently emerged, allow expecting further extension of our knowledge of the material properties using this new tool of nonlinear spectroscopy. The advantages of plasma harmonic studies were summarised in monographs [49, 78, 79].

Among the achievements emerged during recent years one can admit the comparative theoretical and experimental studies of silver plasma harmonic cut-off [80], new findings in fullerenes' high-order nonlinearities [81], single sub-femtosecond harmonic generation in manganese plasma using few-cycle pulses (Fig. 7b, [82]), comparative research of plasma and gas media for efficient HHG [83], temporal characterisation of plasma harmonics [84, 85], generation of continuum plasma harmonics [86], stabilisation of harmonic yield over one million laser shots on the rotating targets (Fig. 8a, [87]), generation of high-order harmonics using picosecond driving pulses [88], various applications of 1 kHz lasers for plasma HHG to increase the average power of converted radiation [89,90], analysis of coherence properties of plasma harmonics [91], use of double-pulse technique for plasma HHG [92], demonstration of the quantum path interference of the long and short trajectories of electrons in plasma HHG experiments (Fig. 8b, [93]), etc. All those findings substantially pushed ahead our knowledge on the peculiarities of plasma media through the analysis of their high-order nonlinear optical characteristics.

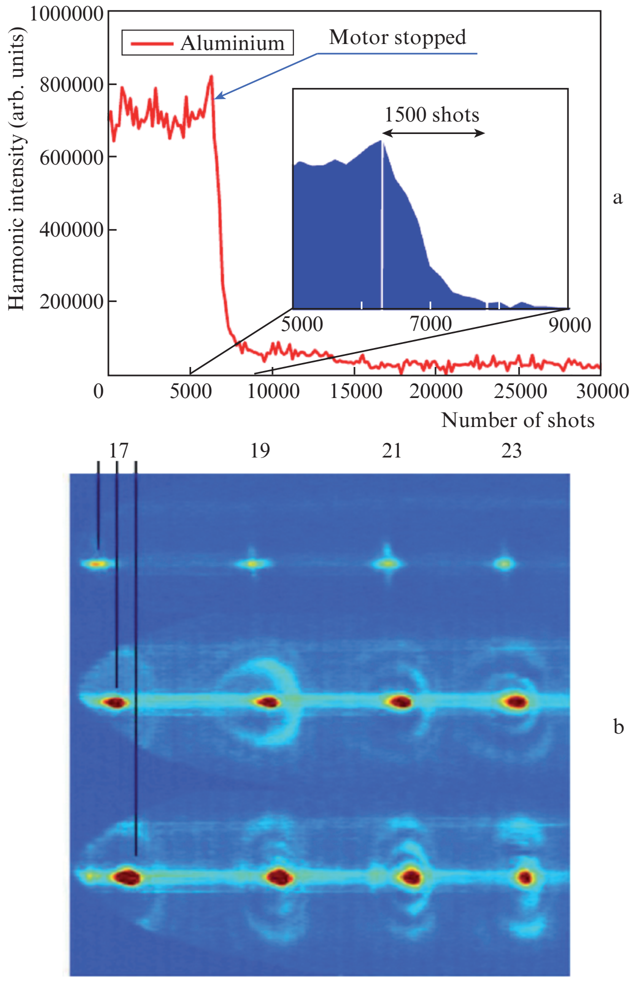

Figure 8. (a) Stabilisation of harmonic yield for over 1.2 millions shots and decay of the harmonics from aluminium plasma after stopping the rotation of the target (reproduced from [87] with permission from Optical Society of America) and (b) dependence of quantum path interference on the chirp of laser radiation during HHG in an aluminium plasma in the cases of positively chirped 93 fs pulses (upper panel), chirp-free 35 fs pulses (middle panel) and negatively chirped 62 fs pulses (bottom panel) (reproduced from [93] with permission from the American Physical Society; copyright 2011).

5. Future of plasma harmonics: new ideas and approaches

The new method of the analysis of plasma formations using the laser ablation-induced HHG spectroscopy can be considered as an advanced approach in material science. This method will open new opportunities of the optimisation of HHG in the laser-produced plasma plumes, develop tools for large molecules and clusters studies in the ablated conditions, and significantly broaden the subjects of studies compared with the HHG in gases. Thus, besides considering as an alternative method for generation of coherent XUV radiation, this approach can be used as a powerful tool for various spectroscopic and analytical applications. A few of them have emerged during recent years and have been discussed in the previous Section.

One can improve plasma HHG through the double excitation of laser-produced plasmas, optimisation of the longitudinal harmonic generation schemes in the laser plume, use of multi-component plasma plumes, optimisation of the ablation of nanostructured targets, formation of QPM conditions in complex extended and perforated plasmas, provision of a regime of waveguide pump propagation through the plasma medium, use of mid-IR pulses for harmonic cut-off extension, stabilisation of plasma and harmonic characteristics at high pulse repetition rates using rotating and moving targets, search for the attosecond pulse generation by achieving the continuum in the harmonic emission near the cut-off region, use of a polarisation gating technique for shortening the plasma harmonic pulses, etc.

One can expect that in the nearest future various approaches for further amendments of plasma HHG will be examined. Among them the following can be mentioned: harmonic generation in plasma using the two-colour pump in the case of commensurate and noncommensurate wavelength sources in the mid-IR and ultraviolet ranges, studies of HHG from various clusters appearing *in situ* during laser ablation of bulk targets, analysis of the influence of molecular orientation on the harmonic output from molecular plumes, development of ablation-induced HHG spectroscopy of various organic materials, analysis of orientation-induced nonlinear optical response of large ablated molecules and clusters, and the time-resolved pump-probe analysis of the complex plasmas containing various molecules.

The joint implementation of above-mentioned new and old (i.e. resonance-induced harmonic enhancement, application of the clusters with controllable and variable sizes, two-colour pump-induced enhancement of the odd and even harmonics, search for the influence of the multi-electron dynamics of complex clusters, such as fullerenes and nanotubes, on the plasmon resonance-induced growth of a few harmonics in the XUV range, etc. [94–107]) techniques will allow further establishment of this method of materials science. The development of the advanced QPM schemes in multi-jet plasma plumes, studies of the time-dependent dynamics of aggregation and disintegration of clusters through their nonlinear optical response, comparative analysis of gas and plasma HHG, joint application of gas and plasma HHG for the studies of the interacting gaseous and ablated species, application of extended plasma plumes for HHG, etc. are also among the future goals of plasma harmonic studies. Particularly, the application of spatially modulated heating pulses [108–110], or perforated targets [111], could be an alternative to the formation of multiple plasma jets on the surfaces of microlithographic targets for the QPM, which was proposed in [112].

Among the advantages of the quasi-phase-matching of plasma harmonics are the simplicity in regulation of the electron concentration in the multi-jet plasmas, the high enhancement factors of harmonics and the availability of the express analysis of electron density. Two former advantages were demonstrated in [77, 108–111]. The reported conversion efficiency for the enhanced 33rd harmonic of a Ti:sapphire laser (~24 nm) generated in the Ag plasma jets (~2×10^{-5}) [108] is among the highest values achieved so far in this spectral region using both gases and plasmas (Fig. 9). As for the latter advantage, the definition of the maximally enhanced harmonics at the fixed sizes of separated plasma jets allows the calculation of the electron density, which has also been demonstrated during recent studies. Electron density was defined using the QPM relations. The results of measurements of the electron density in the region of 10^{16} cm^{-3} using a multi-jet silver plasma produced for the HHG are presented in [108]. The proposed method allows the analysis of the variations of the electron density in low-ionised plasmas. Particularly, the variation of the distance from the ablating target and the delay between heating and driving pulses allow analysing the dynamics of the electron density along the low-ionised medium.

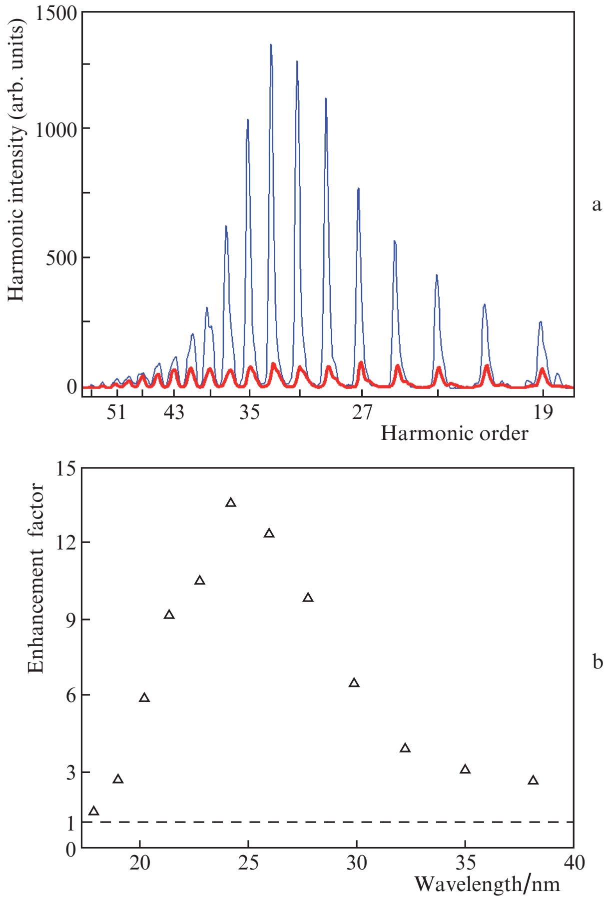

Figure 9. (a) Normalised spectra from the 6-mm-long plasma (thick line) and five 0.5-mm-long plasma jets (thin line) as well as (b) harmonic enhancement factors for the five-jet medium along the 18–38 nm spectral range (reproduced from [108] with permission from the American Physical Society; copyright 2014).

It would be interesting to analyse the application of mid-IR (2000–5000 nm) radiation to study the dynamics of the nonlinear optical response of ablated molecular structures compared with commonly used Ti : sapphire lasers for plasma HHG, including the studies of extended harmonics at a comparable conversion efficiency with shorter wavelength laser sources, and a search for new opportunities in improvement of the HHG conversion efficiency in the mid-IR range, such as the application of clustered molecules.

Future developments also include the analysis of the plasmonic properties of clusters produced during laser ablation at the conditions of resonance-induced enhancement of harmonics; application of graphene- and ring-like structures, multicomponent molecules, nanofibers and other recently emerged clusters for plasma HHG [113]; and the analysis of plasmonic properties of carbon nanoparticles in the XUV and the studies of the indirect involvement of clusters in HHG (when they do not directly participate as the harmonic emitters, but rather enhance the local field, analogously to the experiments and calculations using gold nanostructures enhancing gas HHG [114–116]).

These ideas of plasma harmonic developments were partially borrowed from the gas harmonics. There are no fundamental restrictions in the implementation of gas HHG approaches in the plasma harmonic generation. Moreover, some peculiarities of the latter medium may offer the advantages in the new developments using such sources. Particularly, QPM in periodically modulated gas structures led to the growth of conversion efficiency for some groups of harmonics [117–120]. Recent implementation of a similar approach in the plasma harmonic studies has demonstrated even a larger ratio between QPM-enhanced and conventional plateau-like harmonics [108, 110]. Further, such methods as two-colour pump and use of long-wavelength lasers have recently been applied to various plasma samples using few-cycle laser pulses [81, 83, 89], which led to enhancement of odd and even harmonics, as well as to extension of harmonic cut-offs. Moreover, single harmonic generation from such sources was reported in the case of manganese plasma [82]. Finally, the developed method has already allowed the analysis of the structural properties of complex systems, such as DNA components (thymine and uracil, Fig. 10) [121, 122].

It is worth noting that, currently, the nonlinear spectroscopy involving gas harmonics is trying to deal with such problems as extension of the HHG spectroscopy based attosecond structural technique to image the nuclear re-arrangements induced by localised hole excitations, application of strong-field ionisation to create localised hole excitations and study their attosecond dynamics in polyatomic molecules, search for the selective imaging of hole dynamics induced by the removal of e.g. inner electrons using the XUV initiated HHG technique, and development of multi-dimensional HHG spectroscopy capable of following energy flow between different molecular modes over femtosecond time-scale. The application of plasma harmonics allows adding some important impetus to those studies.

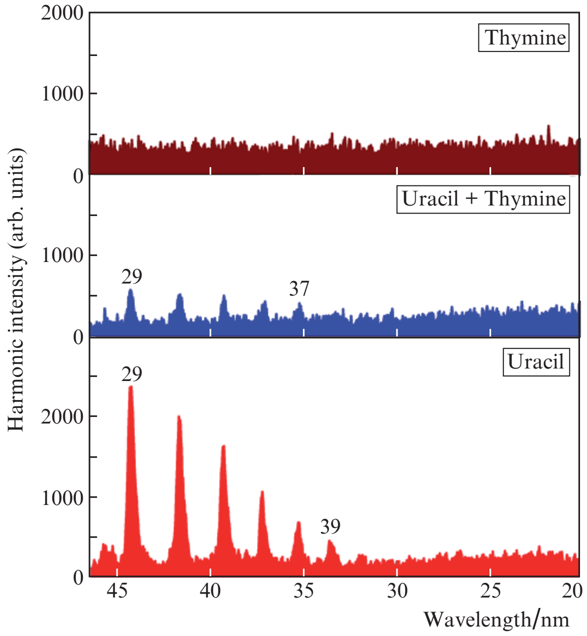

Figure 10. XUV spectra from uracil, thymine and a target made of a 50 : 50 mixture of uracil and thymine in the case of 1300 nm probe pulses and 780 nm, 160 ps ablating pulses ($I = 2 \times 10^{10}$ W cm^{-2}) (reproduced from [121] with permission from the PCCP Owners Society).

The important topics of future plasma HHG studies will be the applications of ablation plumes for attosecond science. Measurement of physical processes with a temporal resolution approaching 10^{-16} s has emerged in the last few years as one of the most exciting frontiers in physical science. Such studies make accessible the ultrafast dynamics of correlated electronic motion that underpin the first moments of a wide range of physical and chemical processes, such as photochemical reactions, radiation damage in biomolecules and conversion of light energy into chemical energy. This science demands the most advanced technology and in particular suitable light sources of exceptionally high bandwidth (>10 eV) to support the ultra-high temporal resolution. Presently, the primary technique to do this is a harmonic generation in gases. Nevertheless the capabilities for present-day attosecond measurements are severely limited by several factors including low photon yield in the generation of isolated attosecond pulses, and the limited range of molecules that can be obtained in gas phase at the densities sufficient for HHG.

First attempts to analyse the temporal characteristics of plasma harmonics were discussed in [123] where the pulse duration of 11th–17th harmonics generated from chromium plasma were measured to be ~300 as. The studies of plasma harmonic pulse duration were also reported in [84, 85]. The goal of the future research consists in the fact that ultrashort pulse generation through plasma harmonics seems efficient for attosecond sciences. One can expect that present expertise in this field has the possibility to make a step-change advance in plasma harmonics induced attosecond science. For this one has to test if, as expected from calculation, the resonance-enhanced HHG in some ablation plumes leads to isolated sub-femtosecond pulses using the attosecond streaking technique; to utilise the isolated sub-femtosecond pulses from those metal plasmas in the pump-probe measurements at surfaces and in molecules; to develop optimal conditions for ablation plumes of high density for HHG studies of various organics, such as intact ribonucleic acid and deoxyribonucleic acid bases; and to allow the attosecond investigation of electron's dynamics in these molecules.

Among other possible developments in the application of plasma HHG technique one may anticipate such areas as seeding of plasma resonance harmonics in the XUV free-electron lasers, application and analysis of endohedral fullerenes using plasma HHG, analysis of molecular structures through the study of harmonic spectra from oriented molecules in plasmas using the gating and pump-probe techniques [121, 123, 124].

Investigations in this area of nonlinear optics are making rapid strides in various laboratories worldwide [84–86, 123, 125–131]. So, the answer to the question on why the plasma harmonics are need and why they become more and more popular seem clearly defined in this review paper. The achievements of present-day plasma harmonic studies motivate for further development of this technique. Recent reviews of various aspects of plasma harmonics [75, 132, 133] made it clear that this field of nonlinear optics develops towards the applications of plasma HHG spectroscopy, as well as further amendments of the harmonic yield.

It has become obvious that plasma HHG is not simply another method for generation of coherent XUV light but rather a new technique for the analysis of various features of the harmonic emitters appearing in the plasma plumes during laser ablation of solids. Because of this, Ockham's principle is applicable to this new method, assuming that it gives new knowledge, which could not be acquired by gas and surface harmonic techniques.

Acknowledgements. I would like to expresses my gratitude to numerous colleagues from various laser centres with whom I have enjoyed the observation of new phenomena during plasma harmonics studies. Special acknowledgments are due to H. Kuroda, P.D. Gupta, P.A. Naik, T. Ozaki, J.P. Marangos, J.W.G. Tisch, H. Zacharias, M. Castillejo, M. Suzuki, H. Singhal, L.B. Elouga Bom and C. Hutchison for the cooperation and fruitful discussions of different aspects of the high-order harmonic generation in laser-produced plasmas.

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