

Chapter 2

HHG in Short-Length Plasmas

Here we discuss some most recent advantages in the developments of the HHG in the narrow (0.3–0.5 mm) plasmas. Among them are the spatial coherence measurements of non-resonant and resonant high-order harmonics generated in narrow laser ablation plumes, resonance processes in plasma plumes, and peculiarities of the high-order harmonics from different narrow plasmas generating at 1 kHz repetition rate.

2.1 Modern History and Perspectives of Harmonic Generation in Narrow Plasma Plumes

A search for the ways for increasing the notoriously low HHG efficiency in the extreme ultraviolet region has long been (and still is) among the most topical problems of nonlinear optics. In the majority of cases, the conversion efficiency of the high-order harmonics generating in the gaseous and plasma media turns out to be insufficient for using them as the 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.

First experiments using narrow over-excited plasma plumes were carried out in 1990s [1–6] and have shown the frustrating results. Nevertheless, there was a reason to hope that harmonic intensities may be substantially increased and efficient shorter-wavelength coherent radiation could be achieved using properly produced plasmas. There are no fundamental limitations here; it only remained finding the “optimal” conditions for formation of a plasma plume to serve as the 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 minimized, as it was underlined in early studies of plasma HHG [1, 3, 5].

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 plasma length, density of ions, electrons, and neutral particles, and degree of their excitation. The use of any elements of the periodic table (Fig. 2.1) [7], as well as thousands of complex samples that exist as solids, may largely extend the range of materials employed, whereas only a few noble gases are typically available for the 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 material science.

In several cases, this method furnishes an opportunity to realize the quasi-resonance conditions and increase the efficiency of single harmonic generation due to the effect of ion transitions on the nonlinear response in different spectral regions, thus allowing the studies of those transitions possessing large oscillator strengths. This effect (Fig. 2.2a) [8] could be hardly observed in the 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, studies of resonance-induced modification of the harmonic spectra generated in gases have demonstrated the enhancement of the narrow parts of harmonics due to the influence of Fano resonances and Stark shift [9].

The modern history of harmonic studies using narrow plasmas has started in 2005 [10]. A substantial increase in the highest order of generating 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, realization of the resonance enhancement of individual harmonics, efficient harmonic enhancement from the plasma plumes containing clusters of different materials, and other properties revealed in [11–16] have demonstrated the advantages of using optimally prepared laser-produced plasmas for the HHG. The orders of harmonics obtained in plasma media to date range into the sixties and seventies [17, 18]. The harmonics up to the 101st order have been

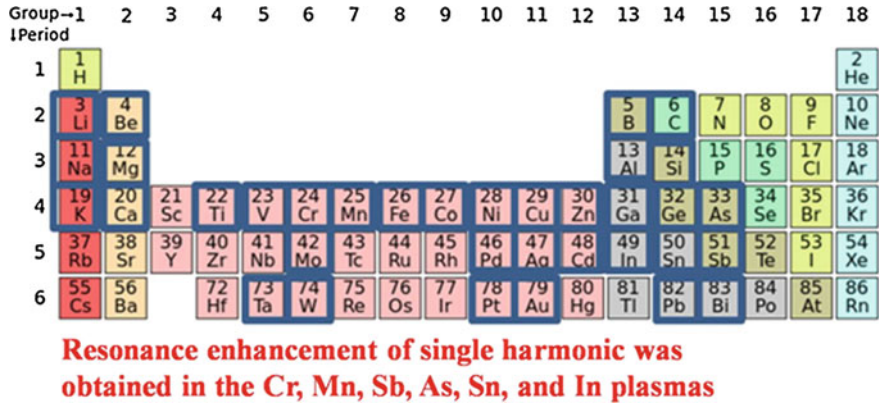


Fig. 2.1 The elements of periodic table (in *thick squares*), which were studied for the HHG in the ablation plumes. Reproduced from [7] with permission from Optical Society of America

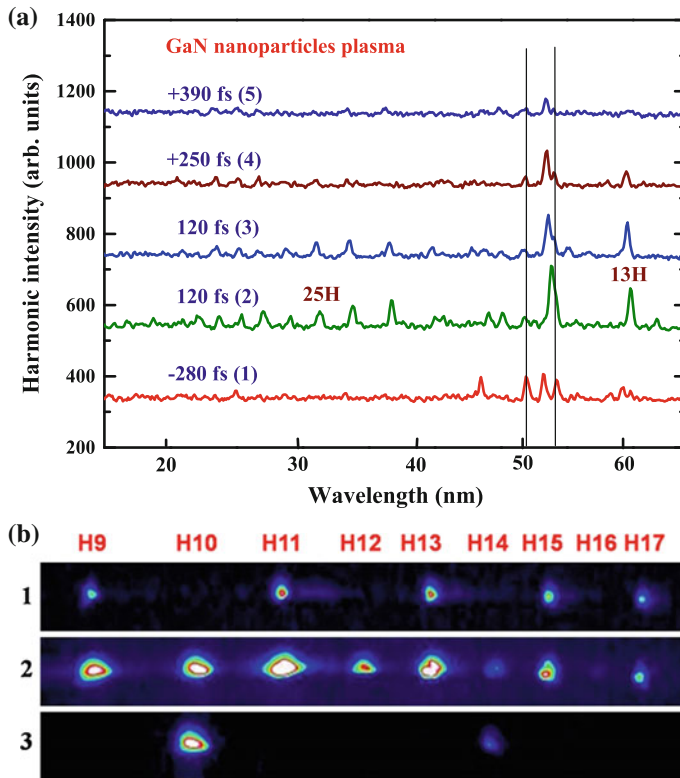
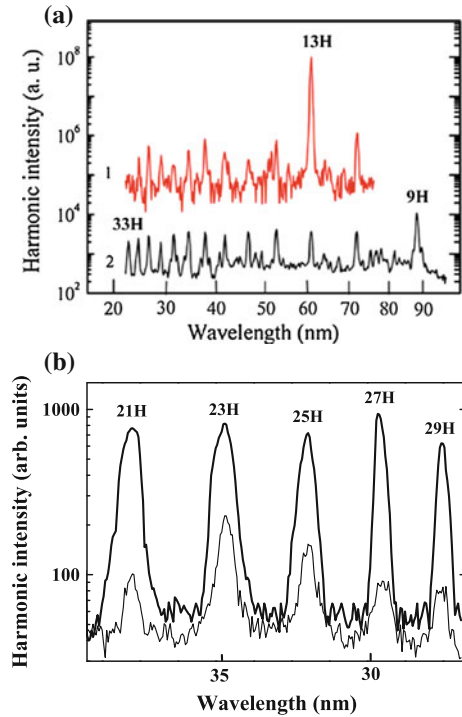


Fig. 2.2 **a** Harmonic spectra obtained from the GaN nanoparticles-contained plasma using the pulses of different chirp and duration. Reproduced from [8] with permission from American Physical Society. **b** CCD images of the harmonic spectra between the 9th and 17th orders generated in the C_{60} plasma in the cases of (1) single-color fundamental pump (800 nm), (2) two-color pump (800 + 400 nm), and (3) single-color second harmonic pump (400 nm). The data were collected under similar experimental conditions. Adapted from [23] with permission from American Physical Society. Copyright 2009

demonstrated during generation in the thin manganese plasma [19] and currently can be routinely achieved using the 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 [20–22]. Application of two-color pump led to enhancement of the odd harmonics, as well as to the appearance of strong even orders (Fig. 2.2b) [23]. In addition to that, the conversion efficiency towards a resonantly enhanced high-order harmonic was almost two orders stronger compared with the neighboring harmonics (Fig. 2.3a) [24].

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

Fig. 2.3 **a** HHG spectra obtained from the (1) indium and (2) silver plasmas. Reproduced from [24] with permission from Optical Society of America. **b** Harmonic distribution in the mid-plateau region in the cases of the plasma produced on the surface of bulk Ag target (*thin line-out*) and the Ag nanoparticles-contained plasma (*thick line-out*). Reproduced from [28] with permission from IOP Publishing. All rights reserved



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 high oscillator strength by wavelength tuning of the driving laser [14], as well as by varying the chirp of laser radiation [11, 13, 16]. Many new features of plasma harmonics, which were emerged during last time, allow expecting further extension of our knowledge of the material properties using this new tool of nonlinear spectroscopy. The advantages of harmonic studies in the thin plasma plumes were summarized in the monographs [25–27].

Among the achievements emerged during following years one can admit the application of the ablated nanoparticles and clusters (Fig. 2.3b) [28], definition of the high-order nonlinearities of fullerenes [29], single sub-femtosecond harmonic generation in manganese plasma using few-cycle pulses (Fig. 2.4a) [30], comparative research of plasma and gas media for efficient HHG [31], temporal characterization of plasma harmonics [32, 33], generation of continuum plasma harmonics [34], stabilization of harmonic yield over one million laser shots on the rotating targets [35], generation of high-order harmonics using picosecond driving pulses [36], various applications of 1 kHz lasers for plasma HHG to increase the average power of converted radiation [37, 38], demonstration of the quantum path

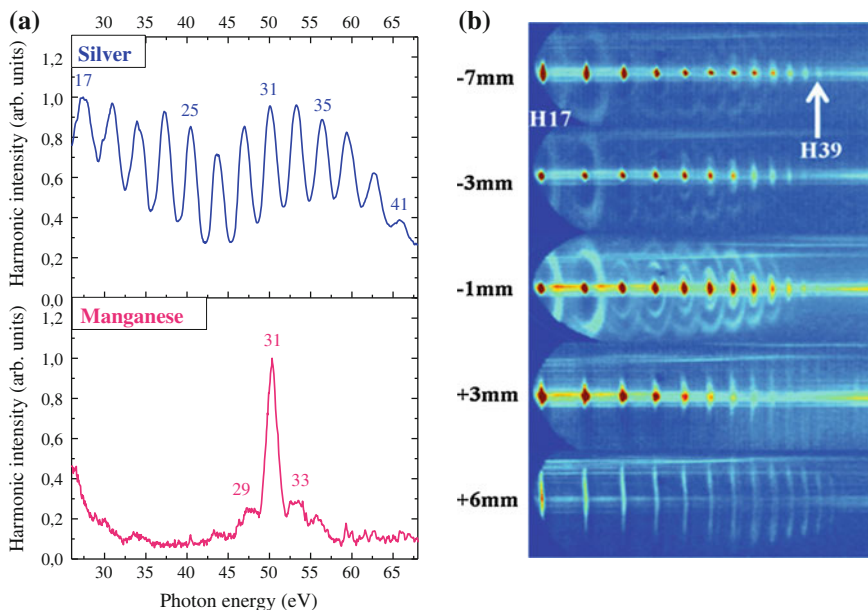


Fig. 2.4 **a** Harmonic spectra from the silver plasma (*upper curve*) and manganese plasma (*bottom curve*). Reproduced from [30] with permission from Optical Society of America. **b** Dependence of harmonic spectra on the position of aluminum plasma with respect to the focus of femtosecond radiation (negative values on the pictures correspond to focusing after plasma plume). Reproduced from [39] with permission from American Physical Society. Copyright 2011

interference of the long and short trajectories of electrons in plasma HHG experiments (Fig. 2.4b) [39], etc. All those findings substantially pushed ahead our knowledge of the peculiarities of plasma media through the analysis of their high-order nonlinear optical characteristics.

It follows from the above that investigations in this area of nonlinear optics are making rapid strides and may bring new success in the nearest future. Plasma harmonics became an important part of the studies carrying out in various laboratories worldwide [32–34, 40–46]. The achievements of present-day plasma harmonic studies motivate for further development of this technique. Recent reviews of various aspects of plasma harmonics [7, 22] made it clear that this field of nonlinear optics rapidly develops towards the applications of plasma harmonic spectroscopy, as well as further amendments of harmonic yield.

I hope that, with this short review, I convinced the reader that the 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. In the following sections of this chapter we will discuss some most recent advantages in the developments of the HHG in the narrow (0.3–0.5 mm) plasmas.

2.2 Spatial Coherence Measurements of Non-resonant and Resonant High-Order Harmonics Generated in Narrow Laser Ablation Plumes

The high coherence of the drive laser beam can be transferred to the harmonic radiation, which can thus exhibit near full spatial coherence [47]. This makes high-order harmonics an attractive table-top, short-wavelength source for applications, such as diffraction imaging [48], holography [49, 50], and, more generally, for short-wavelength interferometry using wavefront division [51].

Studies of the spatial coherence of harmonic radiation have proved useful in helping to elucidate the underlying physics of the HHG process [52], particularly in identifying mechanisms that can degrade the coherence. Previous measurements in gas targets showed that, for laser intensities high enough to cause significant ionization of the neutral medium, the rapid production of free electrons can degrade the coherence [53–55]. The refractive index of the free electrons imparts a rapidly varying phase on the harmonic. This can lower its spatial coherence if the rate of formation of free electrons varies at different points across the focused beam [54], leading to decorrelation of the time-dependent fields. This can be caused by density or laser-intensity variations across the laser focus.

Ablation targets for HHG differ from conventional (neutral) gas targets in a number of ways. Firstly, they are usually preformed plasmas with a density distribution that is determined by the ablation dynamics and subsequent evolution of the plasma, which depend on the target composition and ablation pulse parameters in a complex way. Secondly, the HHG process can be resonantly enhanced in the plasma [24]. In the four-step model of the resonant enhancement [56], the third step (recombination) of the conventional three-step model of HHG is partitioned into two steps: the capture of a laser-accelerated electron into an autoionizing state of the parent ion followed by the radiative relaxation to the ground state with emission of the harmonic photon. For HHG in gas targets, the harmonic phase is largely determined by the phase accumulated by the electron wavepacket during its quasi-free motion in the laser field (step 2), which is the same in both the three-step and four-step models. However, in the four-step model, the phase added during the fourth step (radiative relaxation) is not negligible and can be very sensitive to the detuning from the resonance. Recently, phase distortion of resonantly enhanced harmonics in Sn ablation plumes has been investigated using the RABBIT technique for different amounts of detuning of the 17th harmonic radiation from the Sn II plasma transition at 26.27 eV [33].

These differences between ablation and conventional gas target for HHG raise interesting questions about the degree of spatial coherence of high harmonics from ablation plumes. First study of the spatial coherence of the high order harmonic radiation generated by the interaction of 45 fs Ti:sapphire laser beam with carbon (graphite) plasma plume has been reported in [45]. It was observed that the spatial coherence varies with harmonic order, laser focal spot size in plasma plume, and peaks at an optimal spot size. It has also been reported that the spatial coherence is

higher when the laser pulse is focused before the plasma plume than when focused after the plume, and it decreases with increase in the harmonic order. The optimum laser parameters and the focusing conditions to achieve good spatial coherence with high harmonic conversion have been identified, which is desirable for practical applications of the harmonic radiation.

It is known that these harmonics are emitted as low divergence, near Gaussian beams [around 2 mrad full-width-at-half-maximum (FWHM)]. Below we discuss direct measurement of their spatial coherence [57]. The reviewed studies have shown that the spatial coherence of both non-resonant and resonant harmonics from carbon, zinc, and indium ablation targets generated with few-cycle pulses is reasonably high (in the range of 0.6–0.75) and somewhat higher than for harmonics generated in argon gas under similar experimental conditions. This finding confirms that high-order harmonics from ablation plumes can be used in applications requiring high spatial coherence. For such applications, other features of the harmonic radiation from ablation targets may be useful, such as the resonant enhancement of particular harmonic orders that give rise to quasi-monochromatization of the radiation, thus reducing the requirements for spectral filtering.

The experimental set-up for the measurements of the coherence of plasma harmonics is shown in Fig. 2.5. A Ti:sapphire laser was used in these experiments and provided pulses of 30 fs duration and energies of up to 2.5 mJ at a repetition rate of 1 kHz. These pulses were focused into a 1-m-long differentially pumped hollow core fiber (250 μm inner core diameter) filled with neon [58]. The spectrally

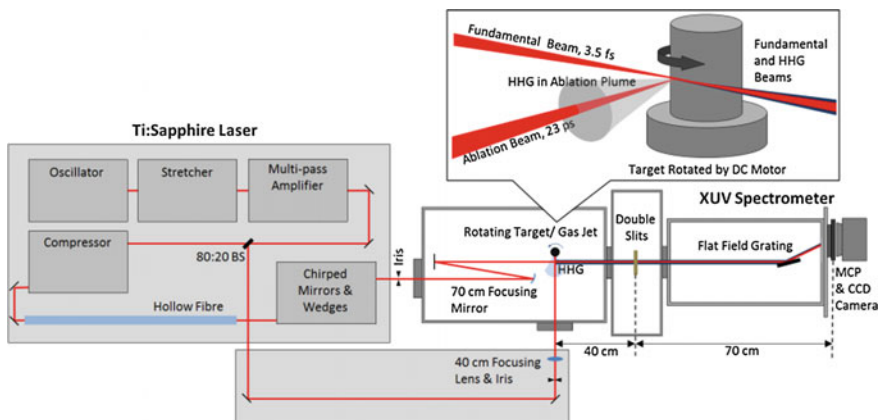


Fig. 2.5 Experimental setup for measuring the spatial coherence of the harmonics generated in ablation plasma plumes and gas targets. A chirped pulse amplification system with a hollow fiber pulse compressor was used to produce the few-cycle pulses to drive HHG. A beam-splitter was used to pick off part of the stretched laser pulse for ablation. A rotating target set-up allowed operation at 1 kHz pulse repetition rate. The harmonics were analyzed using a spatially-resolving XUV spectrometer with micro-channel plate detector. Double slits could be introduced into the harmonic beam to produce an interference pattern on the detector, from which the coherence of the radiation could be determined. Reproduced from [57]. Copyright 2014. AIP Publishing LLC

broadened pulses at the output of the fiber system were compressed by 10 chirped mirrors. High-intensity few cycle pulses (775 nm central wavelength, 0.5 mJ, 4 fs) were typically obtained. The compressed pulses were characterized spatially and temporally with a spatially encoded arrangement for spectral shearing interferometry for direct electric field reconstruction [59]. A part of the uncompressed beam of the chirped pulse amplification Ti:sapphire laser (central wavelength 800 nm, pulse energy 120 μ J, pulse duration 23 ps, pulse repetition rate 1 kHz) was split from the beam line prior to the laser compressor stage and was directed into the vacuum chamber to create an ablation plume from the target. The picosecond pulses were focused by a 400 mm focal length spherical lens to the intensity on the target surface of $I_{\text{ps}} = 8 \times 10^9 \text{ W cm}^{-2}$. Ablation plumes from carbon, zinc, and indium were produced in these experiments. The 15 mm diameter rods of these materials rotated at 30 rpm, which allowed a considerable improvement of the stability of harmonics compared with fixed targets in the case of high pulse repetition rate (1 kHz). In particular, it was shown that once the target rotation is stopped, the harmonic efficiency from the plasmas decreased by more than one order of magnitude within 1–2 s [35]. Harmonics were generated with 4 fs laser pulses, which were focused into the ablation plumes using a 700 mm focal length spherical mirror. The delay between the picosecond and the femtosecond pulses was set to 33 ns. The focal position of the femtosecond pulse with respect to the ablation plume was chosen to maximize the harmonic signal, and the intensity in the ablation plume was estimated to be $I_{\text{fs}} = 3 \times 10^{14} \text{ W cm}^{-2}$. This estimate was consistent with the 70 eV high-order harmonic cut-off that was observed in neon. Neon provides a more reliable corroboration of intensity than the HHG cut-off in argon due to the Cooper minimum in argon at 48 eV that masks the real cut-off position.

The harmonics were analyzed using an XUV spectrometer with a grazing incidence flat-field grating and an imaging micro-channel plate (MCP) detector coupled to a phosphor screen [60]. The spatially resolved spectra of the generated harmonics on the phosphor screen were recorded with a CCD camera. The experimental setup allowed the removal of the ablation target and generation of harmonics in a gas target (Ar or Ne, density $\sim 10^{18} \text{ cm}^{-3}$, target length $L = 1.5 \text{ mm}$) placed at the same position. Under these conditions, the spatially resolved HHG spectra are dominated by the low-divergence short trajectory contribution.

The spatial coherence of the harmonics was measured using double slit interference. A pair of slits was mounted on a translation stage and placed 40 cm from the targets and 70 cm from the MCP. The slits were made in a tungsten foil and had 50 μm spacing, 6 μm width, and were 10 mm long. Each interference pattern (“frame”), measured in this experiment, was integrated over 1000 laser shots. A series of measurements were made for different targets (laser-produced plasmas and Ar gas) in the following energy ranges: 15–30 eV for C, Zn, and In and 15–40 eV for Ar. The relatively low cut-off observed for C is in agreement with earlier work [44]. For each fringe pattern, the fringe visibility was determined through the relation $V = (I_{\text{max}} - I_{\text{min}})/(I_{\text{max}} + I_{\text{min}})$, where I_{max} and I_{min} are the maximum and minimum intensities of the interference pattern. With equal intensity at both slits, as

was the case in those measurements, the fringe visibility at the centre of the fringe pattern was equal to the modulus of the complex coherence factor.

Figures 2.6, 2.7, 2.8 and 2.9 show the double-slit interference data for harmonics from C, Zn, and In plasmas, and from Ar gas targets, respectively. In each figure, the spatially resolved HHG spectrum is shown in part (a), the spatially integrated spectrum is shown in part (b), and a spatial lineout of a single harmonic (interference pattern) is shown in part (c). Taking an average of the visibility at the centre of the fringe pattern over 20 frames, the measured visibilities for the different targets and harmonic orders are as follows: $V = 0.63$ for C_{H13} (i.e. 13th harmonic generated in the carbon plasma); $V = 0.66$ for In_{H13} ; $V = 0.74$ for Zn_{H11} ; and $V = 0.47$ for Ar_{H15} . The HHG spectra from Zn and In have shown resonantly enhanced harmonics due to the overlap of harmonic radiation with plasma resonance lines. In the case of Zn (Fig. 2.7), the emission spectrum was dominated by a peak at ~ 18 eV, which was identified as the partial overlap of H11 (photon energy ~ 17 eV, FWHM bandwidth ~ 1 eV) with the 18.3 eV transition $3d^{10}-3d^9(^2D)4p$ of Zn III, which has an oscillator strength significantly greater than other lines in

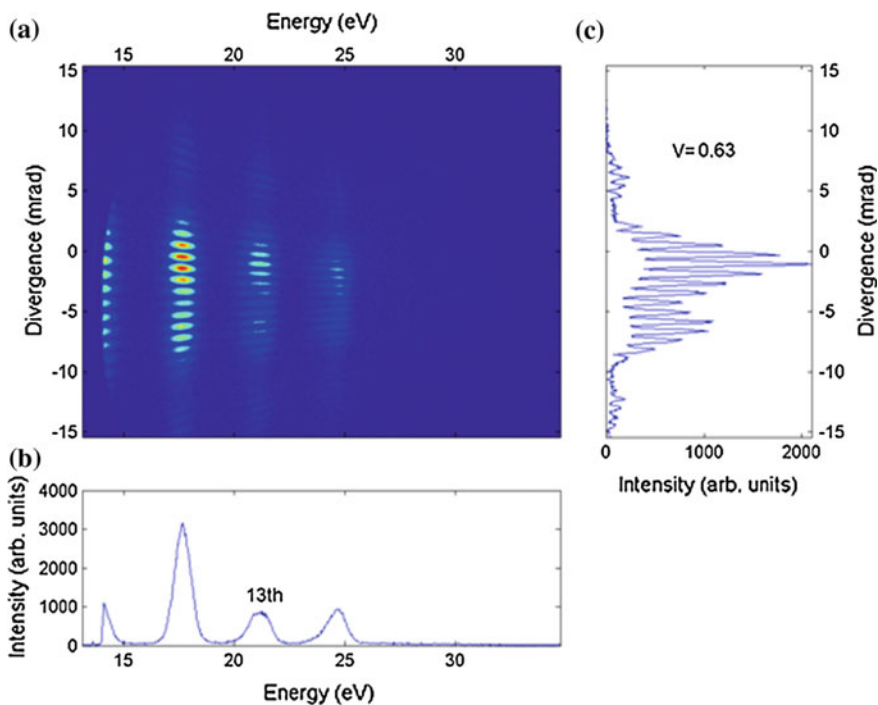


Fig. 2.6 Non-resonant harmonics generated in a carbon plasma plume. **a** Spatially resolved HHG spectrum showing interference fringes from the double slits, **b** spectral line-out, **c** spatial line-out of the interference pattern for the 13th harmonic. The average visibility of fringes near the centre of the pattern was $V = 0.63$. Reproduced from [57]. Copyright 2014. AIP Publishing LLC

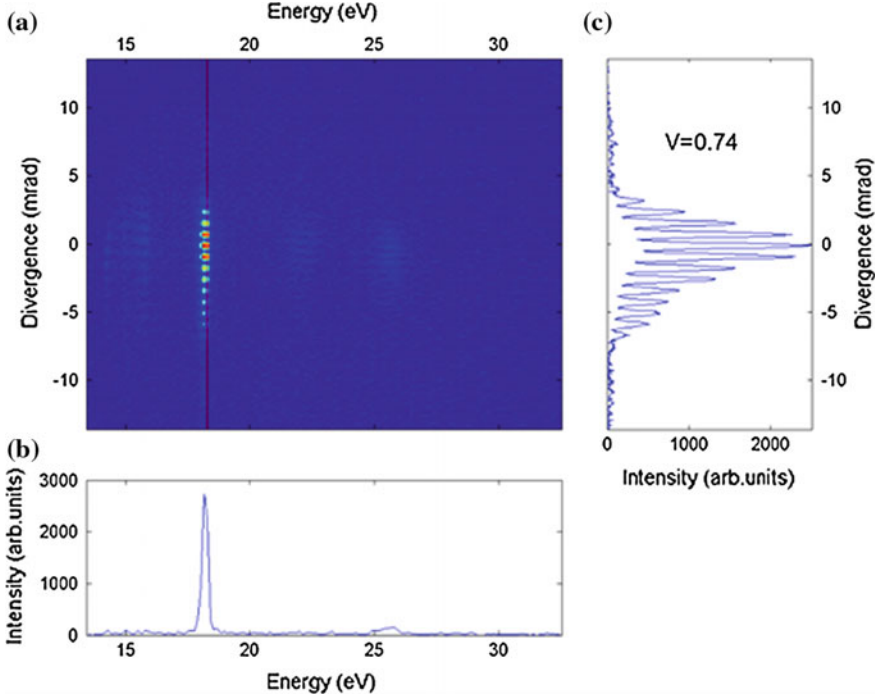


Fig. 2.7 Resonantly enhanced harmonics generated in a zinc plasma plume. **a** Spatially resolved HHG spectrum showing interference fringes from the double slits, **b** spectral line-out showing enhancement of the 11th harmonic due to its overlap with a plasma transition line in Zn^+ at 18.3 eV, **c** spatial line-out of the resonant harmonic. The average visibility of fringes near the centre of the pattern was $V = 0.74$. Reproduced from [57]. Copyright 2014. AIP Publishing LLC

this spectral region [61]. For In (Fig. 2.8), an enhanced feature at ~ 20 eV was observed and attributed to the overlap of H13 (photon energy ~ 20 eV, FWHM bandwidth ~ 1 eV) with the 19.9 eV ground to autoionizing state transition $4d^{10}5s^{21}S_0-4d^95s^25p(^2D)^1P_1$ in In II, which has an oscillator strength (gf) $\sim 10 \times$ larger than other transitions [62]. The variation of HHG conversion efficiency in the vicinity of resonance lines of indium has been reported in [24].

Attempts were made to calculate the visibility of the nonresonant harmonics of Zn and In; however, the signal to noise ratio was too low to accurately analyze the fringe patterns. The maximum visibility measured in those experiments ($V = 0.74$ for the Zn target) was lower than reported visibilities of gases in some earlier work [47] but is consistent with a small departure from full spatial coherence for the driving laser field. In the absence of other effects that can degrade the coherence, the visibility of the fringe pattern for the q th harmonic is approximately given by $V_q \approx 1 - q(1 - V_1)$, where V_1 is the visibility for the driving laser field [54]. Hence, a value of $V_1 = 0.98$ for the laser beam would already completely account for the visibility $V_{H11} = 0.74$ measured for the 11th harmonic from the Zn target. Though

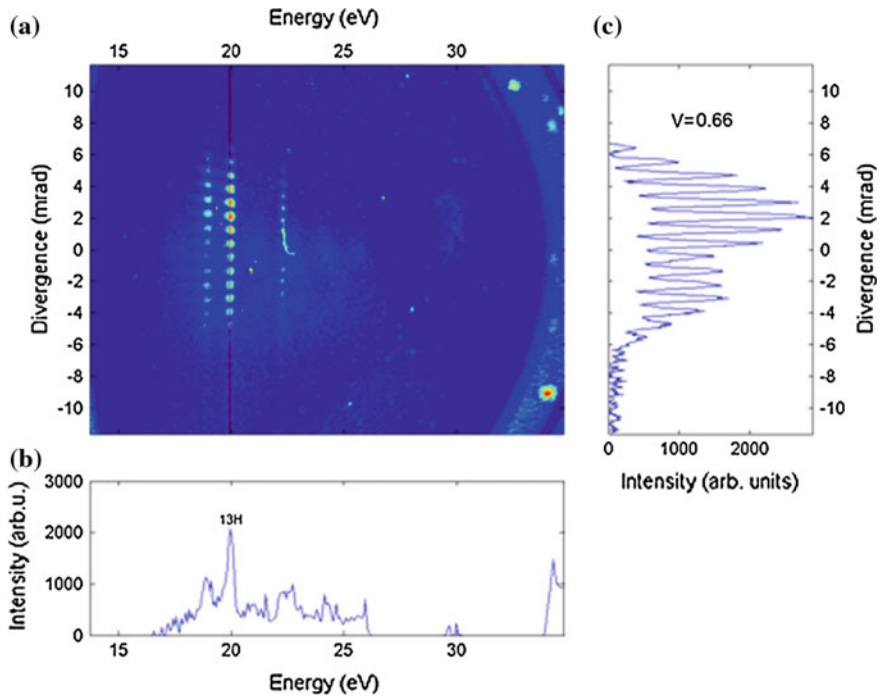


Fig. 2.8 Resonantly enhanced harmonics generated in an indium plasma plume. **a** Spatially resolved HHG spectrum showing interference fringes from the double slits, **b** spectral line-out showing enhancement of the 13th harmonic due to its overlap with a plasma transition line in In^+ at 19.9 eV, **c** spatial line-out of the resonant harmonic. The average visibility of fringes near the centre of the pattern was $V = 0.66$. Reproduced from [57]. Copyright 2014. AIP Publishing LLC

the femtosecond laser beam exhibited essentially full spatial coherence, one cannot rule out such a small decrease in coherence of the radiation delivered to the target, for example, due to nonlinear propagation effects in air and the vacuum chamber window coupled with a small intensity asymmetry of the transverse beam profile.

The significantly lower visibility ($V = 0.47$) observed for harmonics from Ar gas, compared to the plasma targets (average visibility 0.68), was attributed to increased free electron production during the propagation of laser pulse through the Ar gas compared to the other targets. One should note that only rapid variation of the free electron density during the HHG process can degrade the harmonic spatial coherence. Pre-existing free electrons in the ablation plumes should not degrade the coherence, since their density is effectively static on the timescale of the few-cycle pulse.

The tunnel ionization of the targets by a 4 fs laser pulse was simulated using ionization rates from Ammosov-Delone-Krainov (ADK) theory [63]. In the absence of details of the ionic composition of the ablation plasmas, it was assumed that the plasma targets are fully singly ionized. ADK rates are known to be reasonably accurate for rare gases such as Ar, but for multielectron atoms they significantly

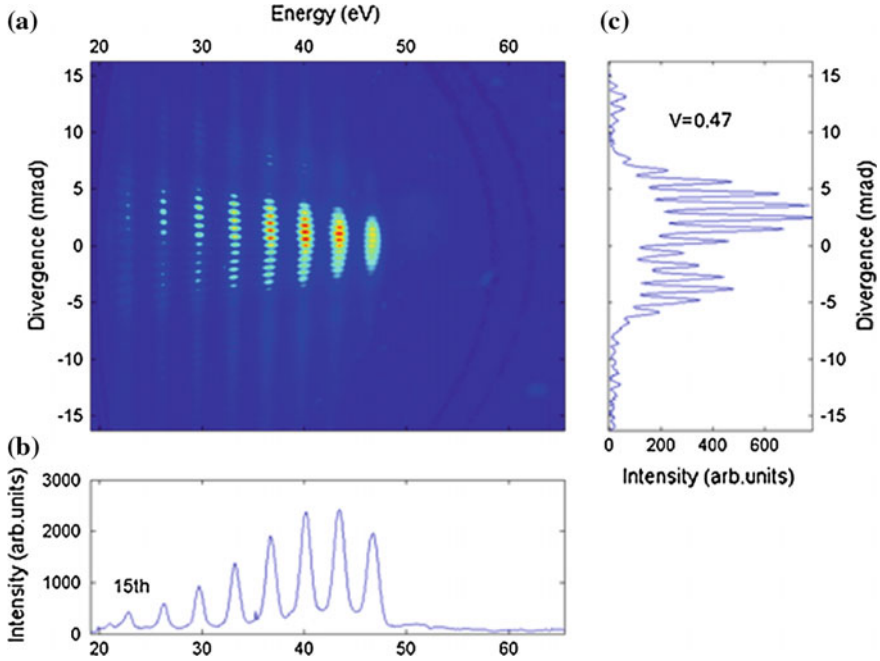


Fig. 2.9 High-order harmonics generated in argon gas. **a** Spatially resolved HHG spectrum showing interference fringes from the double slits, **b** spectral line-out, **c** spatial line-out of the 15th harmonic. The average visibility of fringes near the centre of the pattern was $V = 0.47$. Reproduced from [57]. Copyright 2014. AIP Publishing LLC

overestimate the ionization rate [64]. Therefore, the calculations for C, Zn, and In provide an upper limit for the ionization fraction in those atoms. For a peak intensity of $3 \times 10^{14} \text{ W cm}^{-2}$, the ionization fraction at the peak of the pulse was calculated to be ~ 0.1 for Ar ($I_p = 15.75 \text{ eV}$) compared with $\sim 10^{-5}$ for C II ($I_p = 24.4 \text{ eV}$), 0.004 for In II ($I_p = 18.87$), and ~ 0.01 for Zn II ($I_p = 17.96 \text{ eV}$). Using the target densities given earlier, this implies a free electron density of $\sim 10^{17} \text{ cm}^{-3}$ produced during HHG in the Ar target, which is at least 3 $\times$, 8 $\times$, and 3000 $\times$ higher than for Zn, In, and C targets, respectively. The measured visibility for the case of Ar is consistent with earlier work in which free electron densities in the region of 10^{17} cm^{-3} produced during harmonic generation were shown to lead to a reduction in harmonic visibility to the range of 0.4–0.6 for similar harmonic orders [52, 55]. One can note that for carbon, an intensity of $3 \times 10^{14} \text{ W cm}^{-2}$ is close to the ionization saturation intensity. Therefore, a small fraction of neutral carbon atoms in the ablation plume ($I_p = 11.26 \text{ eV}$), as seen in earlier work [65], would increase the electron density significantly compared to the value calculated above. This could explain the lower visibility for harmonics generated in carbon compared to the other plasma targets.

2.3 Resonance Processes in the Plasma Plumes

2.3.1 *Resonant and Non-resonant High-Order Harmonic Generation in the Plasmas Produced by Picosecond and Femtosecond Pulses*

In order to increase the conversion efficiency into high-order harmonics at kHz repetition rate various routes can be followed. New methods of plasma formation could be an attractive way to improve the harmonic yield. A straightforward implementation is the replacement of the plasma generating laser beam with several tens to hundreds of picosecond duration by one which features only a few tens of femtosecond duration. Thereby, one can elucidate the importance of the laser intensity or the laser fluence for the formation of the plasma, which shows the optimal harmonic yield. Closely related is the investigation of an enhancement of specific harmonics in the spectral vicinity of ionic transitions of the plasma constituents [24, 38]. Below we analyze the results on the application of femtosecond and picosecond laser pulses for plasma formation on metal targets at 1 kHz repetition rate and particularly take attention to the resonance enhancement of single harmonics in these plasmas [66].

A chirped-pulse amplified Ti:sapphire laser with preamplifier and cryogenically cooled booster amplifier [67] delivering pulses with 40 fs duration at a central wavelength of 800 nm and at 1 kHz repetition rate was used as the main laser driving source. Part of the uncompressed radiation was split off from the beam line prior to the compressor stage (800 nm, 0.8 mJ, 12 ps) and used for ablation of the solid targets placed in a vacuum chamber (Fig. 2.10a). This beam was loosely focused onto the target with a spot size of about 300 μm diameter. The resulting intensity of the heating pulse on the target surface of up to $E_{\text{hp}} = 9 \times 10^{10} \text{ W cm}^{-2}$ created a weakly ionized plasma plume. After a variable optical delay in the range from 5 to 75 ns, required for formation and expansion of the plasma plume away from the target surface, the compressed driving laser pulse (800 nm, 0.6 mJ, 40 fs) was focused into the plasma by a 500 mm focal length lens to generate the high-order harmonics. Intensities up to $4 \times 10^{14} \text{ W cm}^{-2}$ have been employed. The position of the focus relative to the plume was chosen to maximize the harmonic output. HHG experiments were also performed using the femtosecond pulse with 40 fs duration as ablating pulse. In that case a beam splitter was placed outside the laser and divided the femtosecond beam into two parts of equal energy of 0.6 mJ for the heating and the harmonic driving pulses (Fig. 2.10a).

The harmonic emission was analyzed using an XUV spectrometer consisting of a flat field grating and MCP detector coupled to a phosphor screen. The images of harmonics were captured using a cooled CCD camera. The XUV spectrometer allowed the registration of the 11th to 23rd harmonics of 800 nm radiation. Various targets (Al, Cu, Ag, In, and Sn) prepared as 15 mm thick rods were used in these experiments. To minimize overheating and damage of the surface from repeated laser shots the rods rotated at slow revolutions, thus ensuring more stable ablation

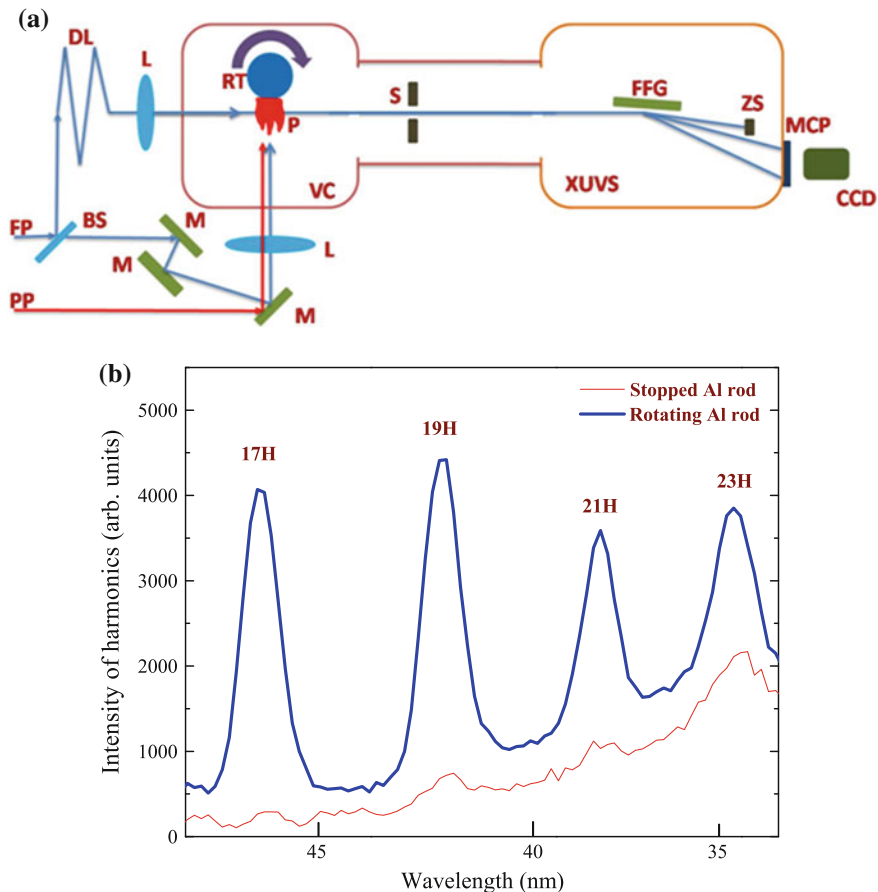


Fig. 2.10 **a** Experimental setup for plasma harmonic generation using picosecond and femtosecond heating pulses. PP, picosecond pulses; FP, femtosecond pulses; BS, beam splitter; M, mirrors; DL, delay line; L, focusing lenses; VC, vacuum chamber; RT, rotating target; P, plasma plume; S, slit; XUVS, extreme ultraviolet spectrometer; FFG, flat field grating; ZS, zero-order diffraction stop; MCP, micro-channel plate; CCD, CCD camera. **b** Harmonic yields from the plasma produced on the rotating (*thick blue curve*) and stopped (*thin red curve*) aluminum rods. Reproduced from [66] with kind permission of The European Physical Journal (EPJ)

conditions analogous to the technique of plasma harmonics stabilization [35]. After moving to a new spot, the previously irradiated target area cooled down and became available again (one rotation later) for further ablation with approximately the same harmonic yield.

The stability of the harmonics from plasma plumes is an important issue for any application of coherent short-wavelength radiation. The cylindrical metallic rods were rotated with different revolutions per minute. A variation from 10 to 300 rpm did not considerably influence the harmonic yield. At the given pumping laser spot

size a rotational speed of 50 revolutions per minute would yield a new surface area for each laser pulse. It seems that this is not necessary for a stable operation. Once the target rotation is stopped, the harmonic efficiency from the plasma plume was significantly decreased. Figure 2.10b shows the line-outs of the harmonic spectra generated from the plasmas produced during ablation of rotating (thick blue curve) and stopped (thin red curve) aluminum rods. One can see the drastic difference between these two spectra collected at identical experimental conditions of target ablation. The harmonics (17th–23rd orders) decreased by a factor of greater than 10 within a few seconds once the target stopped to rotate. The reason of such a drastic decrease is the melting of the surface of the fixed target, as has been reported in [35].

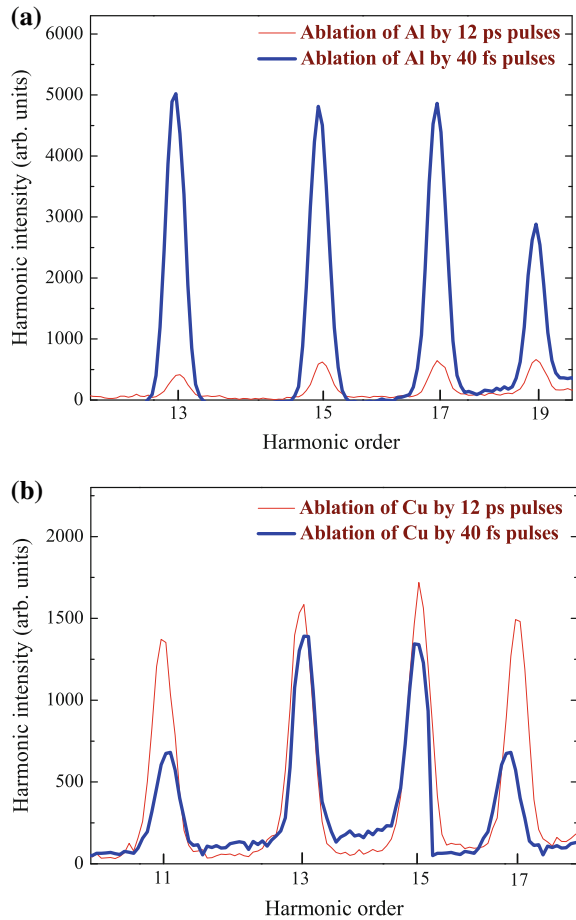
The conversion efficiency in the Al plasma in the range of 17th–23rd harmonics was estimated from the comparison with the harmonics generated in the silver plasma during a same set of experiments. The conversion efficiency from the latter medium at similar conditions of experiments has been reported in [20] to be 8×10^{-6} . One can point out the difficulty in carrying out the experiments at absolutely the same conditions. The efficiency of HHG depends on the density of the target, the laser pulse parameters (intensity, pulse duration, contrast, focusability, etc.), background ionization, composition of the medium, etc. These parameters cannot exactly be reproduced in different experimental setups with different driving laser sources. However, previous experiments show that the ratio between the harmonic yields from different plasmas remains approximately the same even when being studied in different laboratories. The density of the plasma was optimized in both cases to provide a maximum harmonic yield. The contrast does not play any role during plasma HHG. The same can be said about the pulse duration. The intensity defines the cut-off harmonic rather than the harmonic yield being in the saturation conditions. Focusability plays role in the case when the Rayleigh length becomes shorter than the size of nonlinear medium.

Background ionization was optimized in all plasma harmonic experiments prior to the measurements of the conversion efficiency. The composition of the medium was the same (neutrals and singly ionized particles of Al and Ag with approximately the same ionization rate). So, the optimization of HHG in both plasmas allows a rough estimation of the HHG efficiency. The efficiencies of harmonics from these two plasmas were rated as 1:4.

After achieving stable harmonic generation using picosecond ablating pulses, comparative studies were carried out on the plasma formation using pulses of different duration (12 ps and 40 fs). In these two cases approximately the same fluence of the heating pulses was maintained ($\sim 1 \text{ J cm}^{-2}$) by using the same focusing conditions and heating pulse energy, while the intensities of heating 12 ps and 40 fs pulses were 9×10^{10} and $2.5 \times 10^{13} \text{ W cm}^{-2}$. Note the ~ 300 times increase of the intensity of the heating pulse on the target surface in the case of femtosecond pulses compared to picosecond pulses.

One of the aims of these studies was to define which parameter, fluence or intensity of the heating pulse, plays the dominant role in the formation of the suitable plasma for efficient HHG. Figure 2.11 shows two examples of harmonic

Fig. 2.11 Harmonics from the **a** Al and **b** Cu plasmas produced by 12 ps (*thin red curves*) and 40 fs (*thick blue curves*) heating pulses. Reproduced from [66] with kind permission of The European Physical Journal (EPJ)



spectra obtained from (a) aluminum and (b) copper plasmas using the picosecond (thin red curves) and femtosecond (thick blue curves) pulses. One can notice comparable harmonic yields from the Cu plasmas in both regimes of plasma formation, while in the case of Al plasmas, in the fluence range investigated, some advantage of plasma formation using femtosecond pulses for efficient HHG was observed. The use of relatively long or short heating pulses may drastically change the dynamics of formation and spreading of the laser plasma. First attempt to address this issue in the case of plasma plumes was reported in [68].

The observed prevalence of femtosecond pulse produced plasma over picosecond pulse produced plasma could be related with the dynamics of the plasma formation on the aluminum surface. One can assume that, in the case of short pulses, the ablation of aluminum could lead to the formation of some amount of nanoparticles in the plasma plume. These particles may cause the enhancement of the harmonic yield compared with the neutrals. To prove this assumption one has to

analyze the morphology of debris from the ablation of the Al target by the femtosecond pulses as well as to use mass spectrometry of the plasma plume. Probably, the ablation of aluminum was carried out at the conditions when the “optimal” plasma formation coincided with the conditions for the formation of aluminum clusters. Another situation was in the case of other targets. The over-excitation of targets leading to the formation of nanoparticles caused the appearance of a significant amount of free electrons, which led to a phase mismatch of the interacting waves.

The most important parameter for plasma formation turned out to be the fluence rather than the intensity of the pumping radiation. When the fluence was maintained in the range of $0.8\text{--}1.2\text{ J cm}^{-2}$, the harmonics from the plasma showed a reasonably good stability using either picosecond or femtosecond heating pulses. An increase of this parameter to greater than 2.3 J cm^{-2} resulted in a disruption of the harmonic spectra, the appearance of strong ionic lines, and a phase-mismatch between driving and harmonic waves due to the presence of abundant free electrons. One may conclude from the above results that, at the used experimental conditions, laser ablation using pulses of different duration at optimal and equal fluences of the ablating radiation leads to the formation of comparable atom and ion concentrations in both plasmas. These plasma bursts reached the position of the femtosecond driving pulse at about the same time and contained approximately the same number of particles (ions and neutrals) per volume unit.

Below we discuss the resonant enhancement of some specific harmonics generated at different conditions of target ablation (i.e. picosecond and femtosecond pulse induced plasma formation). It has previously been shown that a resonance-induced enhancement of HHG can be achieved in various plasma media at specific single harmonics. Since the first observation of this phenomenon in indium plasma [24], some new media (GaAs [13], Sb [69], Cr [16]) were identified for the potential enhancement of single harmonics due to their spectral closeness with the ionic transitions possessing strong oscillator strengths. Analogous attempts for noble gases, both theoretical and experimental, were reported, which, however, were less successful to demonstrate, until recent time, a resonance enhancement of a single harmonic, even in cases where such an enhancement was expected [70–73]. This fact can be attributed to the availability of a much wider range of target materials for plasma HHG compared to the few commonly used gases, which increases the possibility of the resonance of an ionic transition in the plasma media matching one of the harmonic wavelengths.

The resonant enhancement of HHG was observed in the tin plasma using the picosecond heating pulses (Fig. 2.12a). A strong 17th harmonic ($h\nu = 26.35\text{ eV}$) was observed analogous to those reported in previous studies of this plasma medium [14, 38, 74]. However, in the present case a stronger enhancement of the 17th harmonic is observed, with an enhancement factor of about $10\times$ compared with neighboring harmonic orders. Further, the 13th harmonic ($h\nu = 20.15\text{ eV}$) generated in an indium plasma was studied at different ablation conditions. The experiments with indium plasma produced by laser ablation using picosecond pulses showed an about 70-fold enhancement of the 13th harmonic relative to the

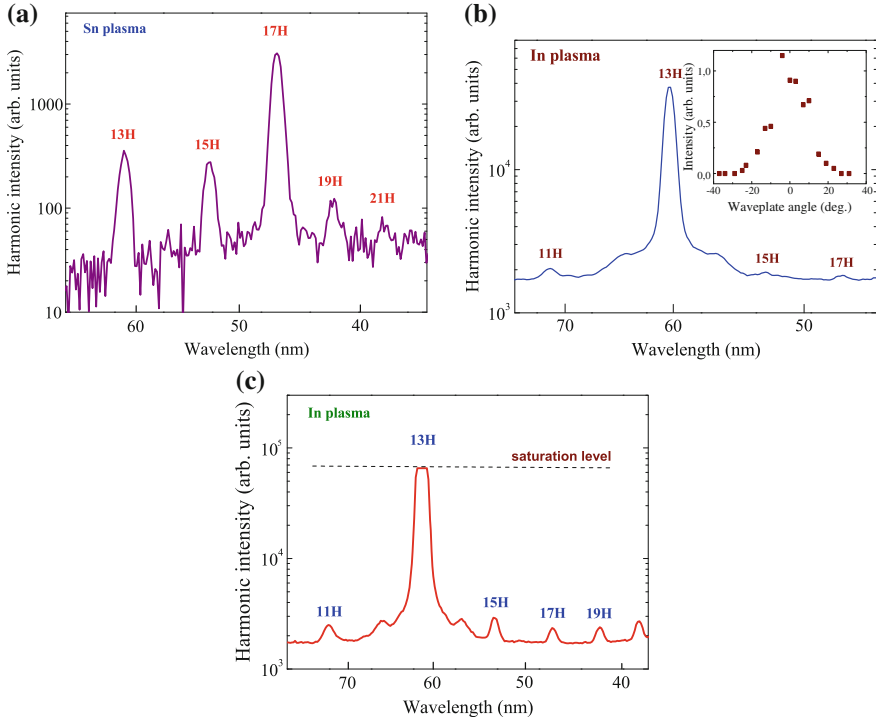


Fig. 2.12 **a** Harmonic spectrum from tin plasma. The resonant (17th) harmonic was ~ 10 times stronger than neighboring ones. **b** Indium harmonic spectrum in the case of plasma produced by 12 ps pulses. The ratio between the 13th and other harmonics in this region was ~ 70 . *Inset* Polarization dependence of 13th harmonic yield. **c** Harmonic spectrum from the indium plasma produced by 40 fs pulses. The 13th harmonic is saturated. The enhancement of resonance harmonic over the plateau harmonics exceeded 10^2 . Reproduced from [66] with kind permission of The European Physical Journal (EPJ)

neighboring ones (Fig. 2.12b). Polarization measurements of the 13th harmonic yield showed the expected decrease of its intensity with the change of the polarization of the driving radiation from linear to elliptical and circular, which is a characteristic feature of this nonlinear optical process (see inset in Fig. 2.12b).

The application of femtosecond pulses for ablation of the rotating indium target showed an even stronger 13th harmonic generation, with pronounced neighboring harmonics (Fig. 2.12c). The saturated 13th harmonic is presented in this figure together with the 11th to 21st harmonics. In this case the enhancement exceeded a factor of $100\times$. Note the logarithmic scales of the harmonic intensities in Fig. 2.12. The two side lobes visible in the spectral vicinity of the 13th harmonic actually also belong to this harmonic and can be traced to the quantum path interference ring between the short and long trajectories of accelerated electrons. The intensity of this ring is comparable with those of neighboring harmonics, as the line-outs in Fig. 2.12b, c suggest.

Below we discuss the concentration properties of the plasmas produced by the pulses of different duration studied in [66]. The molecular dynamics based laser ablation simulation for Al and Cu targets was carried out using optimal experimental parameters of laser radiation by means of the code ITAP IMD [75]. One can take the advantage of short heating pulses (12 ps and 40 fs), which allowed a direct simulation of the ablation process at equal fluence (1 J cm^{-2}). However, the optimal delay between the beginning of ablation and the interaction of targets with the driving pulse is of the order of tens of nanoseconds, which cannot be easily simulated directly. In addition, little is known about the heating of already ablated particles. Therefore, for a correct treatment of the delay, which is an important optimization parameter, one should use the following calculation model. After leaving the surface of the sample all particles were considered as non-interacting with the field and were removed from further simulation time steps. For all particles, which left the surface after a given simulation time step, only those with kinetic energies sufficient to be in the interaction volume ($\sim 200 \text{ }\mu\text{m}$ above the surface) exactly after the supposed delay (5, 30, 40 and 75 ns) were chosen for calculation.

In order to calculate the relative concentration of neutrals and ions the single atom ionization probabilities were calculated by time-dependent density functional theory using the Octopus package [76, 77]. The ionization probability was computed as the occupation of all but the 10 lowest states. The simulations for the 40 fs ablating pulse were performed directly. For 12 ps ablation pulses a series of calculations over 50 fs with constant intensities was performed, which allowed the definition of ionization probability over the envelope of the ablating pulse, which is then approximated by a least-squares fit to yield a continuous time dependent function of the ionization probability. Then, at every time step of the molecular dynamics simulation, the number of particles with velocities sufficient to reach the interaction region after a given delay was multiplied by this ionization probability. This approach yielded the concentration of ionized particles in the interaction region with the fundamental HHG pulse. As a result, the relative number of ionized species was slightly different for different delays, however, the absolute difference was no larger than 5 %. Calculations were performed for Al and Cu for two ablating pulse durations. The concentrations of neutral and ionized particles obtained are presented in Table 2.1. The most important parameter for plasma concentration turned out to be the fluence rather than the intensity of the heating radiation.

One may see from the results presented in table that, at the used experimental conditions, all concentrations arising from ablation by the picosecond and femtosecond pulses of the same fluence are close to each other. It is seen that for Al the largest concentration of neutral particles was obtained for 30 ns delay for both heating pulses, while for Cu better results were found for 40 ns delay. One should note that, at given HHG conditions (driving pulse is not ultrashort, plasma is not very dense, no resonant absorption is present), the plasma concentration influences the HHG yield mainly in two ways. It increases the number of particles participating in HHG, and thus HHG intensity grows with the increase of the concentration, and further it increases the number of the free electrons, which influence HHG efficiency in a rather complicated way.

Table 2.1 Concentrations of neutral and ionized atoms in the interaction volume (in 10^{17} cm^{-3}) for two different durations of the ablation pulse on the Al and Cu targets and at different delays between heating and driving pulses

	Al				Cu			
	40 fs		12 ps		40 fs		12 ps	
Delay (ns)	Neutral	Ion	Neutral	Ion	Neutral	Ion	Neutral	Ion
5	1.53	0.24	1.67	0.11	1.51	0.26	1.43	0.12
30	3.47	0.21	2.79	0.15	2.77	0.23	2.65	0.14
40	2.58	0.26	2.61	0.13	3.15	0.31	2.88	0.16
75	1.73	0.27	1.45	0.10	1.61	0.24	1.51	0.11

Pulse energies of 0.6 and 0.8 mJ were assumed for 40 fs and 12 ps ablating pulses, respectively. Reproduced from [66] with kind permission of The European Physical Journal (EPJ)

2.3.2 Resonance Enhancement of the 11th Harmonic of 1064 nm Picosecond Radiation Generated in Lead Plasma

Here we analyze the results of studies of the HHG in a lead plasma using the relatively long (picosecond) driving pulses [78]. The two-stage amplification of a single pulse of Nd:YAG laser (wavelength 1064 nm, pulse duration 38 ps) was followed by splitting of this pulse into two parts, one (pump pulse) with the energy of 5 mJ, which was used for plasma formation on the target, and another (probe pulse) with the energy of up to 28 mJ, which was used, after some delay, for frequency conversion in the prepared plasma. The pump pulse was focused inside the vacuum chamber containing an ablating target to create the plasma plume. The intensity of the probe pulse at the focus was $4 \times 10^{13} \text{ W cm}^{-2}$. The delay between these two pulses during most of experiments was maintained at 50–70 ns, which is optimal for efficient harmonic generation in laser-produced plasmas. The pulse delay was changed by introducing variable path for the converting pulse. The harmonic radiation was analyzed using the vacuum monochromator and was detected using the luminescent absorber (sodium salicylate) and photomultiplier tube (PMT).

The Pb, Sn:Pb alloy, and Sn were used as the targets during these experiments. A three-coordinate manipulator made it possible to change the targets and to control the zone of interaction of the probe radiation with the plasma relative to the target surface. Various gases were inserted inside the vacuum chamber containing plasma plumes to analyze the influence of the dispersion properties of these gases on the variations of harmonic spectra. The chamber allowed the insertion of the gases and was connected to the vacuum monochromator, thus the gas was also inserted in this monochromator. The absorption of the resonance-enhanced 11th harmonic of 1064 nm radiation at the used densities of gases was insignificant.

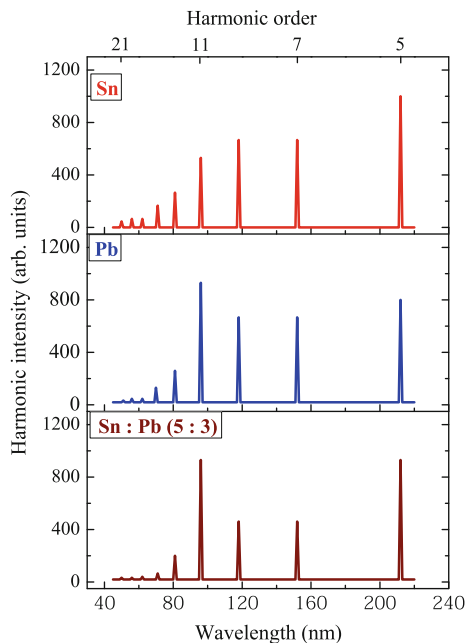
Below we present some specific features of lead and alloy plasmas as the media of HHG using the picosecond laser pulses. The goal of these studies was to define

whether the components of Pb-containing plasma influence the resonance-enhanced harmonic generation efficiency. Changing a composition of plasma from pure lead was accomplished using an alloy of this material, which consisted of tin and lead (at a volume ratio of $\sim 5:3$). Plasma harmonic spectra from pure lead and Pb:Sn alloy with those from pure tin were compared during these studies. To analyze the influence of the propagation effect the variations of harmonic spectra from lead plasma were studied while adding different gases in the vacuum chamber, thus changing the dispersion inside the plasma plume.

Figure 2.13 shows the HHG spectra obtained in the Sn, Pb, and Sn:Pb alloy plasma plumes. The harmonics from tin plasma (Fig. 2.13, upper panel) showed a gradual decrease of each next order, which is a common feature in the case of most of plasma HHG experiments [13]. Contrary to that, a lead plasma demonstrated the enhanced 11th harmonic (Fig. 2.13, middle panel), which was stronger than the lower orders. This peculiarity of lead harmonics was maintained at different conditions of experiments by varying the confocal parameter, plasma length, and so forth. The concentration of plasma ($\sim 2 \times 10^{17} \text{ cm}^{-3}$) was insufficient for the absorption of lower-order harmonics to create the conditions of the stronger 11th harmonic compared with lower ones.

The harmonics from the mixture of lead and tin plasmas comprised both harmonic spectra while maintaining the enhanced 11th harmonic (Fig. 2.13, bottom panel). It is seen that the role of other plasma components (i.e., the tin in the case of ablating Sn:Pb alloy) was insignificant and did not lead to the considerable variation of the envelope of harmonic spectra compared with the pure Pb plasma plume.

Fig. 2.13 Harmonic spectra from the Sn, Pb, and Sn:Pb alloy (5:3) plasmas. Reproduced from [78] with permission from Optical Society of America

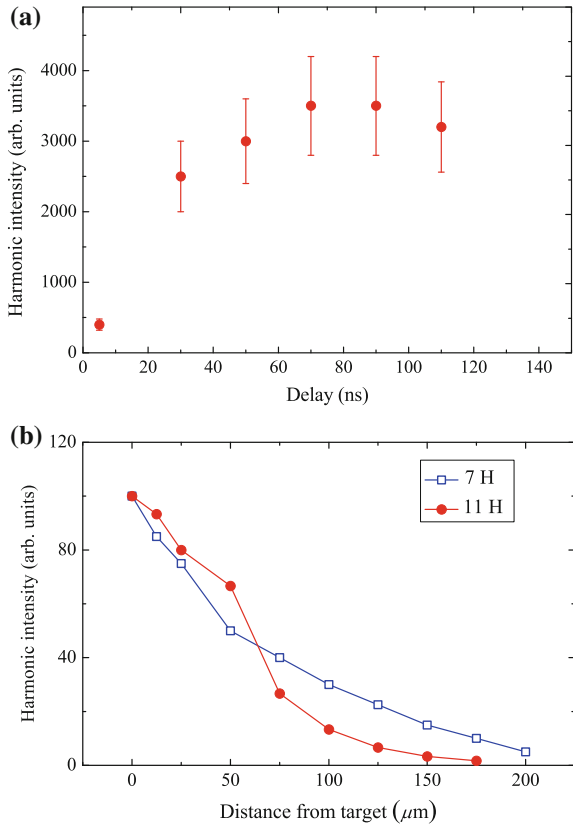


There are no specific reasons in different enhancement factors of the 11th harmonic in the pure Pb and Sn:Pb plasmas. The ratio I_{11H}/I_{5H} from shot to shot was close to 1, or slightly larger, while in both plasmas stronger 11th harmonic was observed with regard to the 9th and 7th orders. This observation points out the insignificant influence of the dispersion properties of additional plasma component on the relation between the phases of the 11th harmonic and 1064 nm radiation.

The comparative studies of harmonic generation efficiency were carried out by moving the translating stage up and down, thus allowing to measure the HHG conversion efficiency (in arbitrary units) in different plasmas without changing the conditions of experiments (delay between pulses, intensity of the heating and converting pulses, distance from the target to the axis of propagation of the converting radiation, confocal parameter of the radiation, and so forth). These studies allowed comparing the efficiencies of harmonics from various metal plasmas during one set of measurements just by changing the targets at similar conditions. The meaning of “arbitrary units” refers to the voltage signal measured from the PMT registering the luminescence from the sodium salicylate. No absolute measurements of HHG conversion efficiency were carried out for each set of studies, since it was unpractical due to necessity in the changes of the conditions of experiments. However, in some cases the absolute values of conversion efficiency were measured by using the technique described in [20]. The conversion efficiency of HHG was defined using the following procedure. At the first step, the fourth harmonic signal was measured using a “monochromator + sodium salicylate + PMT” detection system using the calibrated energy of the 4th harmonic of 1064 nm radiation generating in the nonlinear crystals. This allowed the calibration of monochromator and registration system at the wavelength of 266 nm. Since the quantum yield of sodium salicylate has the same value along a broad range between 30 and 350 nm, the calibration of the registration system at 266 nm allowed the calculation of the conversion efficiency along the whole spectral range of harmonic measurements.

The conversion efficiency of the 11th harmonic from the Pb plasma was measured to be 3×10^{-6} . The enhanced harmonic from lead plasma was analyzed at different experimental conditions. The delay between heating and converting pulses is crucial for optimization of the HHG. A typical dependence of the 11th harmonic intensity on the delay between pulses is presented in Fig. 2.14a. At the initial stages of plasma formation the concentration of particles (neutrals and singly charged ions) in the interaction area is insufficient, since the particles possessing the velocities of $\sim 5 \times 10^5 \text{ cm s}^{-1}$ do not reach the optical axis of probe beam propagation ($\sim 100 \text{ }\mu\text{m}$ above the target surface). The increase of delay allowed the appearance of plasma particles along the path of the probe pulse, which caused the growth of HHG efficiency. Further increase of delay led to saturation of the HHG at $\sim 70 \text{ ns}$ and gradual decrease of conversion efficiency at longer delays ($>110 \text{ ns}$). The influence of the distance between the target and the optical axis of propagation of the probe radiation at a fixed delay is shown in Fig. 2.14b. This distance was varied by a manipulator, which controlled the position of the target relative to the waist of the probe picosecond radiation.

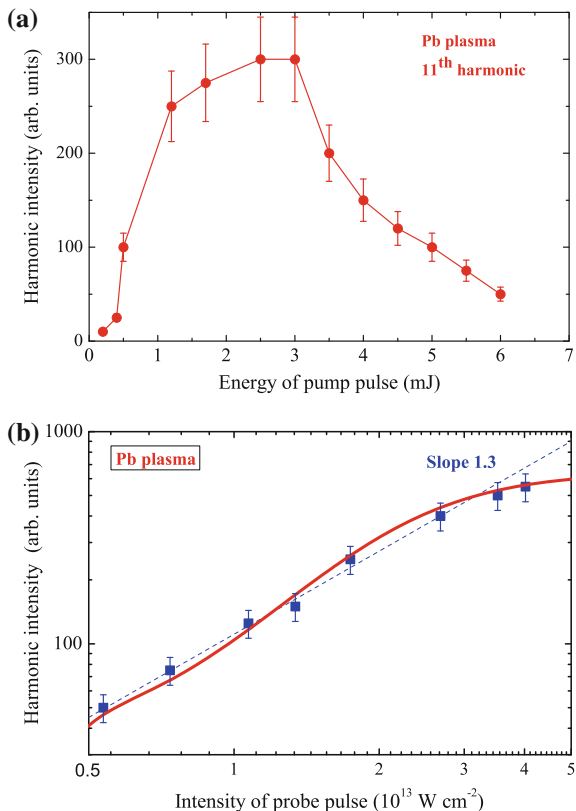
Fig. 2.14 **a** The dependence of the 11th harmonic intensity on the delay between the pump and probe pulses. **b** Harmonic intensity as a function of the distance between Pb target surface and probe beam axis for the 7th (*squares*) and 11th (*circles*) harmonics. Reproduced from [78] with permission from Optical Society of America



The common feature of these studies was an abrupt decrease of harmonics at the irradiation of targets above the optimal level. The term “optimal level” refers to the conditions of plasma ablation when the components of ablation do not cause the growth of the impeding processes, which restrict, or even entirely cancel, the harmonic generation. Those include (a) the excess of free electron concentration leading to strong variation of the dispersion properties of the nonlinear medium and to the phase mismatch between the interacting waves, (b) the excess in absorption of the XUV radiation, and (c) the intense emission from the plasma. The origin of these processes is related with the formation of highly ionized plasma, which leads to the appearance of the abundance of free electrons. The latter species cause a phase mismatch between the waves of probe and harmonic fields. This effect is especially important for the lower-order harmonics. One can note that a decrease of harmonic efficiency in highly ionized plasma has been reported for higher-order harmonics as well [1, 4, 5, 25], though this effect was less abrupt than that observed in the discussed studies. Figure 2.15a shows the influence of the energy of the heating pulse on the intensity of the 11th harmonic generating in the lead plasma.

Fig. 2.15 a The dependence of the harmonic intensity on the pump pulse energy for the 11th harmonic generating in the lead plasma.

b Comparison of experimental (squares, dashed curve) and calculated (solid curve) dependences of the 11th harmonic yield on the converting pulse intensity. Reproduced from [78] with permission from Optical Society of America



Harmonic intensity increased up to the heating pulse energy of 3 mJ. Further growth of the pulse energy led to a decrease of harmonic intensity.

Lead plasma is not the first plasma medium where the enhancement of some specific harmonic was reported. It is just a sample showing such feature while using the 1064 nm radiation. Mn, In, Te, As, Sn, and Cr were among the plasma plumes where the enhancements of single harmonic of 800 nm radiation of a Ti:sapphire laser were reported [79]. Notice that no enhancement of the single harmonic was observed from these metal plasmas using the Nd:YAG laser radiation, and vice versa, the 800 nm femtosecond lasers were not able to produce the enhanced single harmonic from the lead plasma. All previous studies point out a decisive role of the properties of the target material, such as the ionic resonances possessing large oscillation strengths, on the resonance enhancement of single harmonic. The closeness of the ionic transition possessing high gf and the enhanced harmonic has been clearly shown in the case of indium plasma, where the 13th harmonic of 800 nm radiation was considerably (up to a hundred times) stronger than all (even lower-order) neighboring harmonics [24] (see also previous section).

There are various theoretical studies offering the explanations of the resonance-induced enhancement of harmonics [56, 70–72, 80–86]. Particularly in [56], this process was described in the frames of the four-stage model. However, this model could be applied when the resonance level is an autoionizing state, which is not a case of the low-lying states of Pb ions presumably involved in the observed enhancement of 11th harmonic. Probably, the model developed in [71] is more suitable for the discussed conditions. Strongly coupled, low-lying states are a feature of many atomic and molecular systems with ionization potentials substantially lower than those of noble gases. Their calculations showed that the resonance width of the enhancement can be smaller than the laser bandwidth, which means that this is an explicitly multiphoton process. Note that the experimental evidence of resonantly induced single harmonic enhancement was mostly reported in the case of plasma HHG studies. It is obvious that the probability of such occasional coincidence of the wavelengths of ionic transition and harmonics is extremely small among a few used gases, while most of the materials exist in a solid state and thus could be ablated to produce a plasma plume.

The analysis of the influence of the probe intensity on the 11th-harmonic yield from the lead plasma was carried out as well (Fig. 2.15b). The slope of this dependence was close to 1.3 (dashed line). As the frequency shift from the 11th photon resonance between ground and excited states $6s^26p\ ^2P_{1/2}-6s^28d\ ^2D_{3/2}$ (96.72 nm) of Pb II was only 4.5 cm^{-1} , the analysis of harmonic yield was carried out using the resonant approach [87–89]. In this approach, the squared module of the off-diagonal elements of the density matrix defines the resonant harmonic intensity. The calculated $I_{11H}(I_{\text{probe}})$ dependence using the fittings of AC Stark shift, relaxation time, and generalized matrix elements is presented as the solid line in Fig. 2.15b. The influence of resonances in the enhancement of the single harmonic is defined by both the oscillator strength of the nearby transition and the detuning between the wavelengths of harmonic and transition. There is no need for exact coincidence of the resonance transition and harmonic. The optimal detuning defines how strong the nonlinear optical response is expected to be. Indeed, the coincidence of 11th harmonic ($\lambda = 96.73\text{ nm}$) and 96.72 nm transition may be the reason for the insignificant enhancement of this harmonic due to self-absorption, as was observed in those studies. Probably, another reason for a weak enhancement is a small gf of this transition. One can note that all previous observations of resonantly induced harmonic enhancement were obtained at different detuning of harmonics with regard to the exact positions of ionic transitions. Thus, the exact coincidence of these wavelengths may cause some restrictions in the harmonic enhancement due to the influence of self-absorption.

The role of atomic resonances in the growth of the laser radiation conversion efficiency has been actively discussed in the framework of the perturbation theory at the early stages of the study of low-order harmonic generation (see [88] and the references therein). In the case of HHG, the increase in the efficiency of generated harmonics due to resonance processes has come under consideration almost two decades ago, and this approach appears to have considerable promise with the use of ionic and, in some cases, atomic resonances.

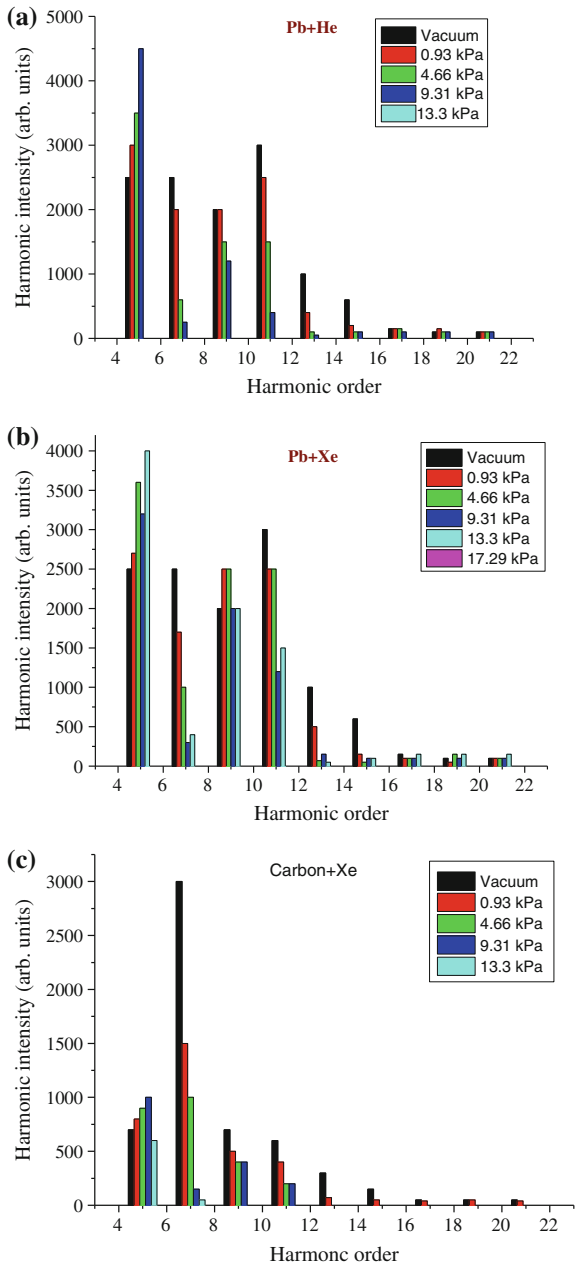
The resonance HHG studies comprised both the theoretical analysis of this process and the first attempts to form the resonance conditions in experiments. While theoretical estimates testified the possibility of an efficient enhancement of individual harmonics, experimental works revealed the difficulties encountered in gas harmonics. Therefore, the use of plasma media could largely facilitate the solution of the problem of resonance harmonic enhancement. Examining a large group of potential targets allowed identifying some of them as suited for demonstrating this process. The advantages of plasma HHG over gas HHG were amply manifested in this case because the number of possible media in the former case is far greater than in the latter case. The observation of enhanced harmonic in Pb plasma in the vicinity of singly charged ionic transition (96.72 nm) just another proof of the concept.

The role of resonances on the high-order nonlinear optical response of medium could be monitored by two methods: tuning of the driving radiation wavelength and changing the phase relations of the interacting waves. The former method was often used in the case of resonance-enhanced harmonics of the broadband Ti:sapphire lasers allowing both direct tuning of radiation wavelength and manipulation of the harmonic wavelength through the chirp variation. This approach cannot be applied in the case of the Nd:YAG laser, since it is impossible to tune the wavelength of this narrowband laser. The only clear proof for the involvement of the resonance on the yield of the 11th harmonic could be the variation of the phase mismatch between the two waves (1064 and 96 nm) in the vicinity of this ionic transition. Below we discuss the experiments allowing the variation of the refractive index of medium by adding different gases in the plasma. We would like to remind that this approach in the studies of the resonance enhancement is not new. It was used in the early stage of low-order harmonic and parametric generation in gases, which showed the attractiveness of the manipulation of the phase relations in the vicinity of resonances by introducing the variable ratios of positive and negative dispersions using different gases [88].

The insertion of gases in the vacuum chamber containing targets led to variations of harmonic spectra compared with the case of pure lead plasma. Four gases (He, Ar, Kr, Xe) possessing different dispersion, as well as different absorption, in the 50–250 nm region were used in the discussed studies. Figure 2.16a, b shows the Pb harmonic spectra in the presence of light (He) and heavy (Xe) gases. The former gas possesses weak absorption until 65 nm, and thus its influence on the variations of harmonic spectra could be attributed mostly to the optical dispersion properties of the gas changing the phase matching conditions for resonant and out-of-resonance harmonics.

The He pressure was varied up to 13.3 kPa, above which the optical breakdown of the gas (without ignition of the metal plasma) was observed. One can see the abrupt decrease of 11th harmonic with the increase of He pressure. Meanwhile, some harmonics, in particular the 9th one, showed less decrease, and the 5th harmonic became even stronger compared with the plasma formation at vacuum conditions. Approximately same features were observed in the case of insertion of the xenon gas, though a decrease of the 11th harmonic was not so pronounced.

Fig. 2.16 Variations of plasma harmonic spectra generated at different pressures of the gases inserted in the target chamber. **a** Pb plasma, He gas, **b** Pb plasma, Xe gas, **c** carbon plasma, Xe gas. Reproduced from [78] with permission from Optical Society of America



The influence of different gases on the harmonic spectra from the plasma media showing the enhancement of specific orders was compared. Recently, the plasma harmonics from graphite ablation using 1064 nm radiation were studied and a

strong 7th harmonic of Nd:YAG laser radiation (152 nm) was observed [90]. The enhancement of this harmonic was attributed to the closeness with the ionic transition of carbon, which led to the formation of phase-matching conditions in this particular case. Figure 2.16c shows the spectra of carbon harmonics at different pressures of xenon. One can see that a decrease of the strong 7th harmonic with the growth of gas pressure resembles the one shown in the case of the 11th harmonic generating in the lead plasma (Fig. 2.16a). The comparable variations of Pb and C harmonic spectra may lead to the following conclusions. The enhancement of harmonics at purely plasma conditions (i.e., without insertion of the gases) in both these cases was originated from better phase matching of the converting and harmonic waves (11th and 7th harmonics from the lead and carbon plasmas, respectively). The addition of the medium possessing positive dispersion worsens the phase matching between the waves and correspondingly decreases the intensities of those harmonics. The role of the gas on other harmonics is also defined by the positive addition to the refractive index of the plasma–gas mixture. However, in that case the dispersion properties of the gas play a less decisive role compared with the resonance-enhanced harmonics.

The role of gas absorption on the observed properties of the resonant harmonic was also analyzed in those studies. At the most unfavorable conditions (2-m-long optical path inside the monochromator, He gas pressure 7 kPa) the absorption starts at 23 eV [91], which corresponds to the 19th harmonic of 1064 nm radiation, while the transmittance for lower-order harmonics was equal to 1 (1.17 eV corresponds to the energy of 1064 nm probe photons). No significant absorption of the 11th harmonic in Ar and Ne was observed at these experimental conditions. In the cases of Ne and Ar the absorption starts at 19 and 14.5 eV, respectively, which should lead to the suppression of the 17th and 13th harmonics but not the 11th one ($E_{11H} = 12.9$ eV). The insertion of Xe led to the 30 % absorption of the 11th harmonic [91], since the transmission in this gas started to decrease at ~ 12 eV (for Kr this value is 13.8 eV, and this gas does not absorb the 11th harmonic).

The reviewed studies showed that the insertion of gas led to the change of the influence of the propagation effect on the harmonic generation. Probably, the combination of micro-processes (i.e., closeness of harmonic wavelength and ionic transition, absorption processes) and macro-processes (propagation effect comprising the joint influence of Gouy phase, dispersion of plasma close to the resonant transitions, and dispersion of neutral gas) cause the observed variations of harmonic spectra from the plasma plume at different gas pressures.

The concentration of lead particles in the area of interaction with the probe pulse ($\sim 2 \times 10^{17} \text{ cm}^{-3}$) becomes comparable with the one of gases $[(1-10) \times 10^{17} \text{ cm}^{-3}]$ inside the plasma volume. At the early stages of gas harmonic studies, the inclusion of an additional gaseous component allowed the enhancement (or decrease) of generating harmonics in the gas mixtures in the vacuum ultraviolet range due to achievement of the phase matching conditions between the interacting waves [92–94].

2.4 Peculiarities of the High-Order Harmonics from Different Narrow Plasmas Generating at 1 kHz Repetition Rate

Each plasma component can cause constructive or destructive action on the overall nonlinear optical response of a whole ensemble of plasma particles. Some components, e.g., free electrons, large nanoparticles or aggregates and microparticles, generally cause a decrease of the HHG conversion efficiency due to some impeding processes originating from the nature of these particles. Free electrons, for instance, enhance the phase-mismatch between driving and harmonic fields, which has a negative influence on the conditions of efficient HHG. Their negative impact on HHG has long been known starting from the very beginning of the studies of gas harmonics [95–97]. To minimize their influence on HHG the intensity of laser radiation is maintained at conditions where multi-photon and tunneling ionization do not create large amount of free electrons.

The presence of free electrons in plasma plumes is unavoidable due to the nature of laser-induced ablation of solid targets. However, even in that case one can limit their influence so that HHG does not suffer greatly from the difference between the phases of interacting waves. Singly charged ions are the main emitters, which are responsible for achieving extended spectral cut-offs and relatively high efficiencies of the HHG process [25]. Neutral atoms also generate strong harmonics; however, in that case the cut-off occurs at lower photon energies than in the case of ions. Small nanoparticles may considerably enhance the intensities in HHG [26]. Their properties as efficient emitters of harmonics were analyzed during both gas [98–101] and plasma [102–106] HHG studies. Theoretical simulations predicted a considerable increase of the harmonic conversion efficiency compared to the corresponding monoatomic media [107–110].

The HHG conversion efficiency considerably depends on the relative phases of harmonics from different emitters within the plasma plumes, realized as constructive or destructive interference. A few studies were reported on the interference of the HHG emitted by successive sources or mixtures of different gases [111–114]. A suppression of harmonic efficiency in a mixture of Ag and Au nanoparticles compared with the separated ingredients was reported in [115]. Modifications of harmonic spectra were also observed for separated boron and carbon targets in comparison with their mixtures. In that case a destructive interference was observed, too, similar to the mixtures of Ag and Au nanoparticles. The HHG efficiency considerably decreased for both higher and lower harmonics compared with the case of separately ablated boron and graphite targets [115].

Another interesting issue is a comparative analysis of harmonic generation from gases and plasmas. Recent studies have demonstrated superior properties of the carbon plasma, which allowed the generation of lower-order harmonics with reported five [116] and ten [34] times higher efficiency than in the case of an argon gas medium. A morphological analysis of the carbon plasma debris revealed an abundance of carbon nanoparticles in the plasma. The increase in harmonic yield

was attributed to the presence of those carbon nanoparticles in the ablation plume which can enhance the nonlinear optical response of the medium. Those studies were carried out using a few-cycle pulses of 3.5 and 8 fs duration, respectively. For a more general application of plasma harmonics it is interesting to generate carbon plasma harmonics and compare their yield with argon gas harmonics using longer driving pulses as well.

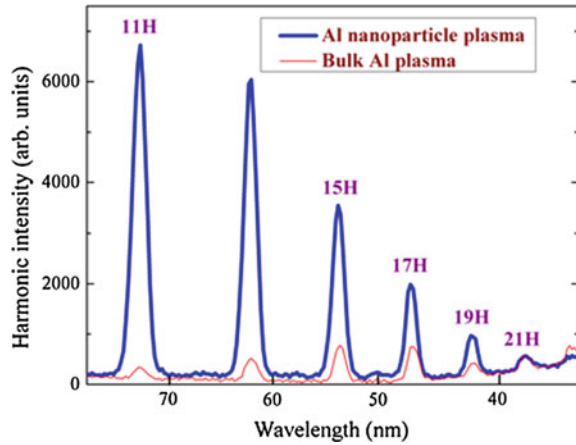
Here we discuss the above issues, which were studied using a femtosecond laser operated with the pulse duration of 40 fs. We analyze the reported results of harmonic generation from mono- and nanoparticle- containing plasma plumes, compare the harmonic yields from Zn, Cu, and brass alloy to address the issue of interference of harmonic generation from species of different origin, and finally compare the HHG efficiencies from gaseous (argon) and plasma (carbon) media at comparable conditions [117].

A 1 kHz repetition rate chirped pulse amplification Ti:sapphire laser was used as the laser driving source. The scheme of HHG was similar to the one described in Sect. 2.3.1. The delay between heating and driving pulses could be variably adjusted with respect to each other between $\Delta t = 0$ and 100 ns to allow for the formation and expansion of the plasma plume away from the surface of the target before propagation of the driving pulse. It should be noted that for an increasing delay the optical pathway of the femtosecond pulse had to be increased up to 30 m. Therefore, for each delay setting the dispersion in air had to be pre-compensated, and the other beam parameters had to be kept constant. For lighter atoms a short delay of, e.g. $\Delta t = 25$ ns for C, and for heavier atoms a longer delay, e.g. $\Delta t = 70$ ns for Ag proved to be optimal.

Various targets (Al, Cu, Zn, brass, graphite, and Ag) as well as nanoparticles were used for these experiments. For a comparison of the conversion efficiency into harmonic radiation the solid target could be replaced by a gas cell (length $L = 0.7$ mm) operated with a continuous Ar flow at an optimal pressure of 100 mbar for gas HHG [118, 119] without changing other experimental parameters, in particular the laser properties.

In Fig. 2.17 the HHG spectra recorded for the 11th to 23rd harmonics generated in the plasmas produced on bulk and nanoparticle aluminum targets are presented. The ablation conditions with respect to pulse energy and fluence of the ablation pulse, as well as of the driving pulse, remained identical for these two samples. The same holds for other parameters of the experiment, like the distance between the target surface and the driving beam ($\sim 100 \mu\text{m}$), the pulse duration of the ablation pulse (12 ps), and the delay between the ablation and driving pulses ($\Delta t = 30$ ns). Figure 2.17 shows the line-outs of the harmonic spectra generated in the spectral range between $\lambda = 35$ and 80 nm for nanoparticle (thick blue line) and bulk (thin red line) Al targets. For the two lowest orders (11th and 13th) of the displayed harmonics, the intensity ratio $I_{\text{nano}}/I_{\text{bulk}}$ was found to be approximately 30 and 15, respectively. This ratio decreases for higher harmonics until above the 23rd order when the harmonics from the nanoparticle target entirely disappear, while the harmonics from the bulk Al target show similar intensities over the whole spectrum. This observation is a clear experimental evidence for the low degree of ionization of

Fig. 2.17 Harmonic spectra from plasmas produced on the bulk Al (*thin curve*) and Al nanoparticles contained targets (*thick curve*). Reproduced from [117] with permission from Springer Science+Business Media



the plasma obtained from glued nanoparticles, because the harmonics, which can be generated from ions only, are suppressed. The harmonic cut-off in the spectra from bulk Al was beyond the short-wavelength limit of XUV spectrometer ($\lambda = 35$ nm), while in accordance with earlier observations it should be in the region of 20 nm (41st harmonic, [120]). Further, the ablation of the pure glue alone, without nanoparticles, did not lead to harmonic generation, which confirms that the emitters of harmonics were associated with the aluminum nanoparticles. The harmonics generated from the nanoparticle plasma target showed a larger divergence than those from the bulk target. This larger divergence could arise from a stronger influence of the long trajectories of accelerated electrons on the yield of the harmonic radiation. This observation can also be related to a growth of the recombination cross section when returning electrons moving on the long trajectory have a higher probability to recombine with larger particles. The observation of only relatively low-order harmonics from the Al nanoparticle-containing plasma could be related with the origin of nanoparticle-induced HHG, which is assumed to occur from neutral particles. Additionally the ionization potential of Al clusters containing more than about 15 atoms falls below the ionization potential of a single neutral Al atom (5.98 eV) and further decreases with increasing particle number (~ 5.2 eV for $n = 50$) [121]. This also leads to a lower cut-off in the harmonic spectra.

In a molecular dynamics simulation the evolution of the kinetic energy of ablated Al atoms from bulk and nanoparticle targets was studied based on the ITAP IMD molecular dynamics code. The calculations were performed for two different durations of the ablating pulse (12 and 28 ps). When comparing the two targets it is evident that in the case of a nanoparticle target (Fig. 2.18; blue triangles and black squares) the ablated atoms have an average kinetic energy, which is lower by a factor of two compared to atoms ablated from the corresponding bulk target (pink triangles and red dots) over the entire pulse duration of the ablating pulse. In order to gain further insight from these results one may consider two scenarios. If an atom is ablated instantly from the target, then its kinetic energy will be relatively

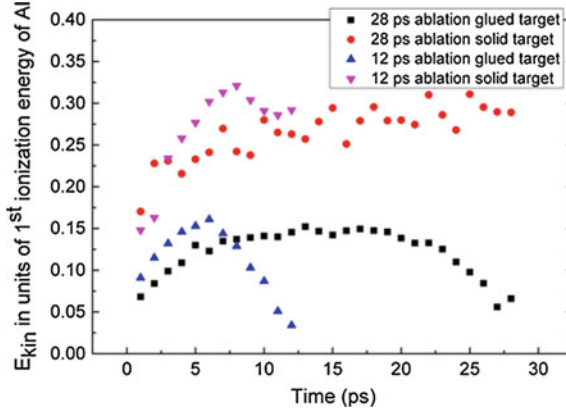


Fig. 2.18 Molecular dynamics simulation of the average kinetic energy of aluminum atoms ablated from $5 \times 5 \times 5 \text{ nm}^3$ Al nanoparticles (blue triangles and black squares) and a solid Al target (pink triangles and red dots). Two different durations of the ablating laser pulse of 12 and 28 ps have been assumed in the simulation. The energy scale is normalized to the first ionization potential of aluminum (5.98 eV). Reproduced from [117] with permission from Springer Science+Business Media

high. However, if an atom gains the energy sufficient to break a bulk bond via energy exchange with neighboring atoms, its kinetic energy will be low and very unlikely exceed the doubled value of the binding energy. The kinetic energy of the ablated atoms is thus a good indicator of the nature of the ablation process. In the discussed case the significantly larger kinetic energy of ablated atoms from the solid target have shown that the ablation process occurs instantly for atoms near the surface and becomes more stable with surface melting. In the case of nanoparticle ablation the process is different. The strong decrease of the kinetic energy near the end of the pulse indicates that the nanoparticle is completely ablated. Otherwise the energy exchange between neighboring atoms would lead to a stable average kinetic energy. The simulations together with these considerations lead thus to the conclusion that the nanoparticles are evaporated more uniformly and are ablated completely before the pulse ends.

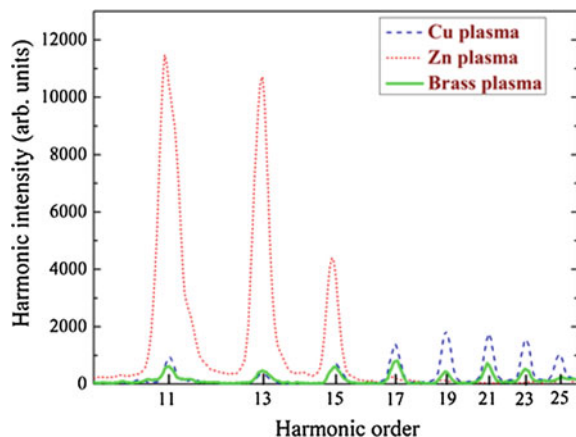
Below we discuss the case when the mixed materials participate in HHG. The influence of two sources of emission on the intensities of HHG is a rather complicated phenomenon. HHG in mixed gases has been reported in several studies [11, 114, 122, 123] in which a suppression of the harmonics above the cut-off of one of the gases was observed. Constructive interference due to quasi-phase matching can be achieved when these gas sources are located at appropriate distances from each other [112, 124–126]. Analogous features were reported for mixed boron and carbon plasmas, which separately showed considerably different cut-offs [115]. Also in that case a destructive interference occurred over the whole observed spectrum of harmonic generation up to 26 eV.

Figure 2.19 shows high-order harmonic spectra observed from the plasmas produced on bulk Zn, Cu, and brass targets. The HHG in a zinc plasma shows strong low-order harmonics up to the 15th order (dotted red curve), while the copper plasma (dashed blue curve) allowed harmonic generation extending towards shorter wavelengths out of the registration range of the used XUV spectrometer. It is interesting to note that the strong Zn III resonances at $\lambda = 67.8$ and 71.4 nm do not enhance the 11th harmonic at $\lambda = 72.7$ nm. This indicates that the comparatively cold plasma is formed with a low concentration of electrons and of multiply charged ions. The brass target contained a mixture of Zn and Cu at a ratio of 35:65, and the plasma produced on this target contained therefore both Zn and Cu atoms and ions. It should be noted, however, that the evaporation of brass with a high concentration of Zn is a complicated process due to the initially preferable evaporation of the Cu, which prevents an efficient evaporation of Zn [127]. Therefore in the brass plasma the concentration of Zn atoms is different from that of the bulk Zn target.

The harmonics from this brass plasma (solid green curve) on the other hand show a yield diminished over the whole spectral range of harmonic generation from the 11th to the 25th order, being lower than those from both Zn and Cu plasmas. While for zinc the decrease amounts to a factor of about five, a decrease of a factor of two is observed for copper. A decrease of the intensity without a sign of constructive interference was observed over the measured HHG spectrum ($\lambda = 35\text{--}80$ nm, $h\nu = 15.5\text{--}35.5$ eV) in the mixed plasma. The dispersion of the low-density laser plasma must therefore be weak in this spectral range, maintaining the relative phases of the two emitters.

The origin of this phenomenon is yet clear and further studies are needed for a clarification of the influence of a low cut-off component on the harmonic intensity from another component showing a cut-off at higher photon energies. The goal of the discussed experiments was to study the HHG produced in a macroscopically uniform and isotropic two-component plume. Since HHG is a coherent process, interference can considerably influence the harmonic efficiency. Considering the

Fig. 2.19 Harmonic spectra from Zn (dotted red curve), Cu (dashed blue curve), and brass (solid green curve) plasmas. Reproduced from [117] with permission from Springer Science+Business Media



contributions from two different induced dipole moments the interference is related to the different dipole phases. In that case, the interference mechanism should occur even on a microscopic scale, since the medium is isotropic and uniform. These studies have shown that the relative dipole phase interference is the proper explanation of the observed results.

The delay between the ablation and the driving pulses is crucial for an optimization of HHG. A typical dependence of the harmonic intensity on the delay between these pulses is presented in Fig. 2.20 for the carbon-containing plasma produced on graphite target (black squares) and the silver plasma (blue squares). At short delays of $\Delta t = 10$ ns even low-Z species such as carbon with velocities of typically 1×10^4 m s⁻¹ do not reach the optical axis of propagation of the driving beam at about 200 μ m distance from the target surface with sufficient density. An increase of the delay to $\Delta t = 25$ ns led to a strong increase of the harmonic yield in a carbon plasma, because now a high concentration of particles was presented within the beam path of the driving laser. At longer delays a gradual decrease of the yield follows. Corresponding results for plasma from a silver target are shown as blue squares. A strong rise of the high harmonic signal at about 55 ns delay is followed by a plateau in the range between 60 and 80 ns delays, while for larger delays the harmonic intensity started to drop. This demonstrates that there are different optimal delays for harmonics generated in low-Z and high-Z plasmas. Although the velocities of the ablated silver particles are now reduced by about a factor of three, the kinetic energies of active particles from the heavier target (silver, $m = 107.87$ amu) and those from the lighter target (carbon, $m = 12.01$ amu) are about the same.

To directly compare the high-order harmonics generated from plasma media and gas a gas cell (length 0.7 mm, $p_{\text{opt}} = 100$ mbar) and a solid carbon target were

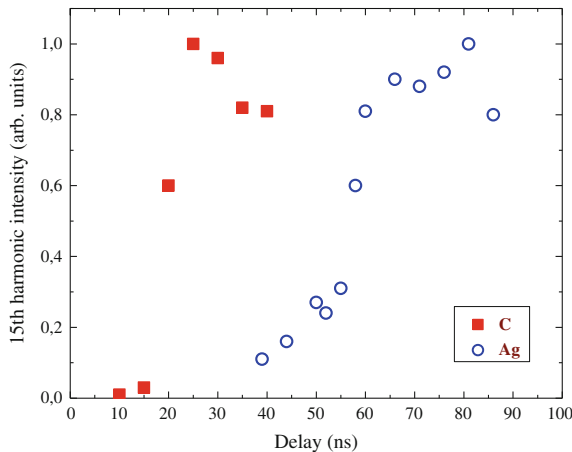


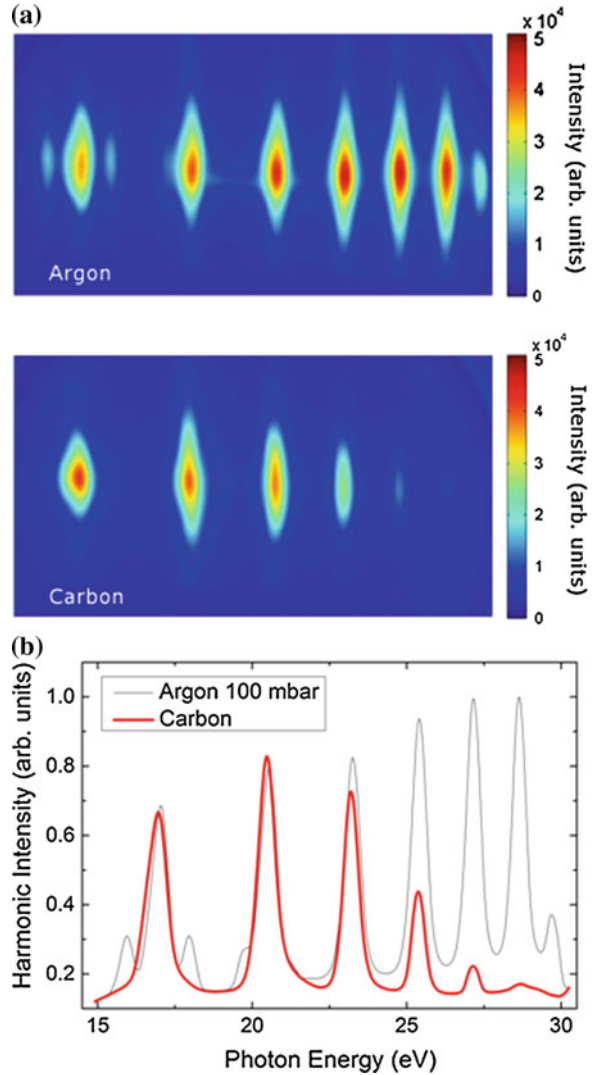
Fig. 2.20 Dependence of the 15th harmonic yield on the delay between the heating and driving pulses in the cases of carbon (*filled squares*) and silver (*empty circles*) plasmas, respectively. The distance between target and driving pulses was maintained at 100 μ m. Adapted from [117] with permission from Springer Science+Business Media

placed in the same vacuum chamber. Previously, a major complication with plasma harmonic experiments compared with gas harmonic experiments was a rapid evaporation and cratering of the solid target surface [25, 34]. This caused the target to deform, thus changing the conditions of plasma formation. The rotating rod system allowed a considerable reduction of these influences on the stability of HHG. The debris from the ablation also coats the window from which the picosecond ablating pulse enters the vacuum chamber, thus reducing the fluence of heating radiation on the target with time. To exclude this effect a protective microscope slide was installed between the target and the entrance window which could be easily replaced from time to time.

The efficiency and characteristics of high-order harmonics from the carbon plasma were compared with those from the argon gas cell, using the 40 fs laser as the main driving pulse. The two upper panels in Fig. 2.21a show the raw images of harmonic spectra optimized with respect to the peak intensity generated in Ar gas and in carbon plasma at equal acquisition times. Under these conditions, the energies of the driving pulse were 0.6 mJ in case of harmonics generated from the carbon plasma plume and 0.9 mJ from gaseous argon, respectively. In both media comparable divergences of the harmonics were observed. In the case of HHG from the carbon plasma an increase of the driving pulse energy led, at the given focusing conditions, to a significant growth of the harmonic divergence. This may indicate that for higher driving pulse energies the energy is converted more preferably by the longer electron trajectory. It further supports the interpretation that these plasma plumes contain nanoparticles, as mentioned above. At driving pulse energies above 0.8 mJ the blue side maxima emerged in the harmonic profile. The decrease of harmonic intensity suggested that the harmonics originated from long and short trajectories do not interfere constructively. It should be noted that due to the larger beam profile it is still possible to generate higher photon fluxes, however, with a higher proportion of the long trajectory contributing to the photon flux. Additionally, when comparing the intensities of Ar and C harmonics it should be kept in mind that the active lengths of the media are estimated to be 0.7 and 0.3 mm for the gas and plasma medium, respectively. Further, the densities are estimated to be about 2.5×10^{18} and $2 \times 10^{17} \text{ cm}^{-3}$, respectively. Integrated intensities of harmonics for both targets are given in Fig. 2.21b.

The relative photon flux of 13th harmonic was determined with a MCP detector and the absolute photon flux with a calibrated photodiode. In both cases a Sn filter was used to suppress the fundamental Ti:sapphire radiation. At a thickness of $d = 300 \text{ nm}$ a pure Sn shows a transmission of $T = 9.9 \%$ [128], and a filter oxidized by a 4 nm thick layer had the transmission $T = 8.1 \%$ [128, 129]. It should be noted that these filters do not withstand the fundamental radiation for a long time without damage, making the usage of a slit of 0.9 mm in front of XUV spectrometer width mandatory. The MCP measurements yielded an intensity ratio of the 11th–15th harmonics generated in C plasma and in Ar gas of 2.5:1.5. For the determination of the conversion efficiency and the photon flux by the calibrated photodiode all three harmonics—the 11th, 13th, and 15th harmonic—which are transmitted by the Sn filter have been measured in the direct beam, because the diffraction efficiency of

Fig. 2.21 **a** Raw images of the 11th to 23rd harmonic spectra from argon gas and carbon plasmas, taken at 0.9 and 0.6 mJ femtosecond pulse energy, respectively. **b** Comparison of the integrated line-outs of harmonic spectra from argon gas (*thin black curve*) and carbon plasma (*thick red curve*). Reproduced from [117] with permission from Springer Science+Business Media



the grating was not known. The relative intensities of the harmonics in this direct beam were taken from the dispersed spectra. The conversion efficiencies of $\eta = 3.2 \times 10^{-7}$ for Ar and $\eta = 5.3 \times 10^{-7}$ for the C plasma in the case of 13th harmonic were defined. This compares well with the relative intensities measured by the MCP. Taking an average fundamental power of 600 mW at 1 kHz repetition rate a photon flux of 9.8×10^{10} photons s^{-1} generated in the C plasma and a flux of 6.0×10^{10} photons s^{-1} generated in the Ar gas were estimated. The measured intensities compare well with the harmonic intensities reported in the literature

Table 2.2 Conversion efficiencies and photon fluxes in the case of HHG in Ar gas and C plasma

Harmonic	Diode sensitivity [A/W]	Argon gas		Carbon plasma	
		Efficiency	Photon flux [s ⁻¹]	Efficiency	Photon flux [s ⁻¹]
11	0.10	2.6×10^{-7}	5.7×10^{10}	2.6×10^{-7}	5.8×10^{10}
13	0.13	3.2×10^{-7}	6.0×10^{10}	5.3×10^{-7}	9.8×10^{10}
15	0.16	3.4×10^{-7}	5.5×10^{10}	4.0×10^{-7}	6.4×10^{10}

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[65, 119, 130, 131]. The corresponding results for the 11th, 13th and 15th harmonics are summarized in Table 2.2.

2.5 Concluding Comments

The main goal of the description of HHG in narrow plasmas was to acquaint the reader with most advanced results of this technique achieved so far using the 0.3–0.5 mm long plasma plumes. While presenting the overview of previous results of plasma harmonic studies, we also showed most recent developments in this field.

Particularly in this chapter, we have discussed the measurements of the spatial coherence of the high-order harmonics generated in laser-produced narrow plasma plumes in the cases of resonant and non-resonant harmonics. Reasonably high visibilities in the range of ~ 0.6 – 0.75 were measured for C, Zn, and In targets for harmonics in the range of 15–25 eV. These results have confirmed the assumption that the harmonics from ablation plasmas can be used for applications requiring high spatial coherence such as diffraction imaging. The higher visibility for the plasma harmonics compared with an argon gas target in these experiments was attributed to a reduced production of free electrons during the propagation of femtosecond driving pulse through the preformed plasmas.

We analyzed the application of femtosecond and picosecond laser pulses for plasma formation on the metal targets for non-resonant and resonant high-order harmonic generation using a 1 kHz laser. Efficient HHG was achieved by using a rotating target for plasma formation. These studies revealed that the harmonic yields from the plasmas produced by picosecond and femtosecond pulses of the same fluence were comparable, despite the significantly different intensities of the heating pulses. Further, the resonance enhancement of single harmonics in Sn and In plasmas using picosecond and femtosecond ablating pulses was discussed. The optimization of single harmonics with enhancement factors of $10\times$ and $100\times$ for these harmonics compared to the neighboring ones was achieved. The concentrations of neutrals and ions of Al and Cu at different heating pulse durations were defined using the code ITAP IMD.

The discussed studies also included the demonstration of the generation of high-order harmonics from the Pb plasma using the mode-locked picosecond laser.

The important peculiarity of those studies was the observation and analysis of the enhanced 11th harmonic ($\lambda = 96.7$ nm), which originated from the resonance-induced growth of the nonlinear optical response of lead plasma at the wavelength of the 11th harmonic lying close to the strong ionic transition of lead (Pb II, $\lambda = 96.72$ nm). The conversion efficiency of this harmonic was measured to be 3×10^{-6} . A strong departure from enhanced yield to suppressed state of 11th harmonic in the case of addition of the He in the area of plasma formation clearly indicated the involvement of the propagation effect, which spoiled the favorable conditions for the 11th harmonic generation. The resonance effect of the enhancement of the 11th harmonic in lead was not as strong as in the cases observed in indium and other plasmas, which obviously can be attributed to both the weaker oscillator strength of Pb resonance and better conditions for the observation of the resonance enhancement in the case of the broadband femtosecond radiation of Ti:sapphire lasers. The clear evidence for the availability to monitor the involvement of resonances in HHG through the modulation of phase mismatch was also presented in the case of carbon plasma. The similarity in the behavior of resonance harmonics in the cases of addition of the gas with positive dispersion in the carbon and lead plasmas was an additional proof of the influence of C and Pb ionic transitions on the nonlinear optical response and particular harmonic yields.

We have analyzed the superior harmonic intensity from an aluminum cluster contained plasma compared to atomic and ionic Al plumes. The analysis of HHG in the mixtures of two emitters (Zn and Cu) revealed the destructive interference of the two harmonic sources. Different plasmas were studied at various delays between the ablation and the driving pulse to analyze the role of the atomic mass of different plasma components in the HHG yield. Finally, conversion efficiencies and photon fluxes have been determined for Ar gas and carbon plasma in the case of comparable orders of harmonics. Those results have shown that harmonic generation in a carbon plasma compares favorably with that in an Ar gas.

Concluding this chapter, we would like to stress that the HHG in partially ionized narrow plasmas was actively developed during last few years in different laboratories worldwide. The developments in this area allow us to expect further amendments to this technique of coherent short-wavelength radiation generation. The extended plasma medium is the example of such an amendment of the high-order nonlinear optical processes in the plasma plumes. Chapters 3–7 contain the evidence of this assumption.

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