Low-Order Harmonic Generation of 1064 nm Radiation in Long Plasma Plumes

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Abstract

The advanced properties of long (~5 mm) plasma plume has been demonstrated for harmonic generation compared with the short (~0.5 mm) lengths of plasmas used in previous studies. The studies of the third and fourth harmonics generation using single color and two color pumps of the long plasmas produced on various metal surfaces are presented. Then the role of heating pulse characteristics on the improvement of harmonic yield from extended plasmas has been analyzed.

Keywords

Laser Plasma; Low-Order Harmonics

Introduction

Harmonic generation in laser plasmas has long been considered as the one demonstrating lower conversion efficiency compared with that in gases. From the very beginning of the studies of high-order harmonic generation (HHG) in plasmas (Akiyama et al, Kubodera et al, Wahlström et al, Theobald et al, Ganeev et al 1997, Krushelnick et al), the superiority of gas HHG was demonstrated with extended harmonic cutoff, higher conversion efficiency, appearance of the plateau-like distribution of harmonic intensities in the latter case, while the application of plasma plumes allowed generation of harmonics in the range of tens to twenties orders, without the plateaulike distribution and of a low efficiency. The over-excitation of targets followed with highly ionized particles formation, large free electron densities and phase mismatch between the driving and harmonic waves led to creation of "inefficient" plasma plumes. This term refers to the worsened conditions for HHG.

The advantages of plasma HHG were realized when low-excited, low-ionized plasma was applied for harmonic generation. It immediately followed with the observation of considerably higher conversion efficiencies which became comparable with those reported in gas HHG studies. Moreover, a substantial increase in the highest order of generated harmonics, emergence of a plateau in the energy distribution of harmonics, high efficiencies obtained with several plasma formations, resonance-induced enhancement of individual harmonics, efficient harmonic enhancement from the plasma plumes containing clusters of different materials, and other features have demonstrated the advantages of plasma HHG (Ganeev).

All those studies were carried out using the narrow plasma plumes (≤0.5 mm). The sizes of plasma plumes were defined by the focusing conditions of the spherical lenses used for ablation of the targets. In the meantime, it can be expected that application of longer plasmas would further enhance the conversion efficiency due to quadratic dependence of the nonlinear optical response of medium on the length of laser-matter interaction. To create such a long media, one has to carefully chose the conditions of plasma formation. The absorption of extended plasma can prevent the enhancement of harmonic yield. Presence of free electrons and excited ions in extended plasma plumes can cause the growth of some impeding processes, such as phase mismatch and Kerr effect, during propagation of laser pulse through extended medium.

To elaborate and analyze the restriction processes, one has to start with the lowest harmonic generation in long plasmas. The easiness of third harmonic (3H) generation in laser-produced plasmas allows defining various impeding processes with better accuracy, thus revealing best conditions for lowest-order harmonic generation in extended plasmas, which could be useful for optimization of higher-order harmonic generation.

3H generation from plasma plumes has been a subject of thorough analysis from the very beginning of the studies of frequency conversion of radiation in laser-produced plasma plumes (Gladkov et al). An analysis of low-order (third (Fedotov et al, 1991) and fifth (Fedotov et al, 1995)) harmonic generation in such plasmas produced by a heating pulse on the surface of solid target allowed formulating several recommendations as regards further advancement towards the shorter wavelengths.

Notice that all these early studies of low-order harmonic generation in plasma plumes, as well as some recent studies (Oujja et al, López-Arias et al, 2012(1), López-Arias et al (2012(2)), were performed using the ablation beams producing narrow plasma plumes (~0.5 mm). In this paper, the properties of long (~5 mm) plasma plumes for low-order harmonic generation are analyzed, and third and fourth harmonics generation are studied using single color and two color pumps of the long plasmas produced on various metal surfaces.

Experimental Arrangements

A single pulse of Nd:YAG laser (wavelength 1064 nm, pulse duration 38 ps, 2 Hz pulse repetition rate) was divided into two parts. First (heating) pulse with the energy of up to E_{hp} = 15 mJ was used for extended plasma formation on the target, and second (driving) pulse with the energy of up to E_{dp} = 28 mJ was used, after some delay, for frequency conversion in the long plasma. The heating pulse was focused using the 30 cm focal length cylindrical lens inside the vacuum chamber containing ablating target to create the extended plasma plume (Fig. 1). The intensity of heating pulse on the target surface was 3×10^{10} W cm^{-2}. The driving pulse was then focused by using the 30 cm focal length lithium fuoride spherical lens possessing small chromatic aberration on the prepared plasma from the orthogonal direction, parallel to the target surface. The distance between driving beam and target surface was maintained at ~50 μm. The confocal parameter of focused driving radiation was 8 mm. The intensity of driving pulse at the focus area was 1×10^{13} W cm^{-2}. The delay between these two pulses during most of experiments was maintained at 30 ns, which was optimal for efficient harmonic generation in laser-produced plasmas. 3H radiation was analyzed using the fiber spectrometer (HR4000, Ocean Optics).

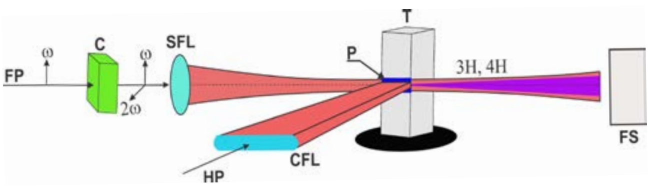

FIG. 1 EXPERIMENTAL SCHEME. FP, FUNDAMENTAL DRIVING PULSE; HP, HEATING PULSE; C, NONLINEAR CRYSTAL (KDP); SFL, SPHERICAL FOCUSING LENS; CFL, CYLINDRICAL FOCUSING LENS; T, TARGET; P, PLASMA; FS, FIBER SPECTROMETER (HR4000).

To analyze the fourth harmonic (4H) generation, the two-color pump scheme was utilized. Part of the driving pulse was converted to the second harmonic in the nonlinear crystal (KDP, type I, crystal length 10 mm). The conversion efficiency of second harmonic was 6%. Driving and second harmonic beams were focused inside the plasma plume. The polarizations of these two pumps were orthogonal (Fig. 1). The confocal parameter of second harmonic radiation was 6 mm. Two pump beams were overlapped in the plasma plume area, which was confirmed by generation of the fourth harmonic.

The targets were made of various metals (aluminum, tin, and copper). The sizes of targets where the ablation occurred were 5 mm. A three-coordinate manipulator was allowed to move the target and control a zone of interaction of the driving radiation with the plasma relative to the target surface.

Results and Discussion

Third Harmonic Generation Studies in Long Plasmas

The important issue of these studies is the role of the length of plasma plume (*d*) in variation of the nonlinear optical response of this medium. Various scenarios can be expected in the case of presence of the free electrons, which can drastically change the phase matching conditions between the driving and harmonic waves. The exceeding of the length of plasma over the coherence length of 3H generation should lead to saturation and decrease of harmonic yield. The unsaturated 3H yield should follow the quadratic dependence on the length of nonlinear medium ($I_{3H} \propto d^{l}$, where l=2 (Reintjes)).

These dependences were analyzed in a few plasma plumes. Figure 2 presents these results in the case of aluminum, tin, and copper plasmas. Then the length of plasma was changed using the slit placed after the cylindrical lens. In the case of Al plasma (Fig. 2a), this dependence had a slope (l) close to 2.2 up to the d ~ 2.5 mm, with further decease of the slope (l=1.8). Nevertheless, one can see a significant increase of 3H yield with the growth of plasma length, meaning that free electrons, absorption of plasma, phase mismatch, Kerr effect, etc., did not play a decisive role in the variation of this dependence (at optimal excitation of the target).

3H generation in Sn (Fig. 2b) and Cu (Fig. 2c) plasmas showed analogous behawior, with less slope in the former case (l=1.7) compared with the copper plasma (l=2). Thus the tendency to follow the quadratic rule of harmonic-length dependence seems a reasonable feature at the conditions of our experiment (plasma concentration ~1×10^{18} cm^{-3}, heating pulse duration 38 ps, fluence on the surfaces ~0.7 J cm^{-2}).

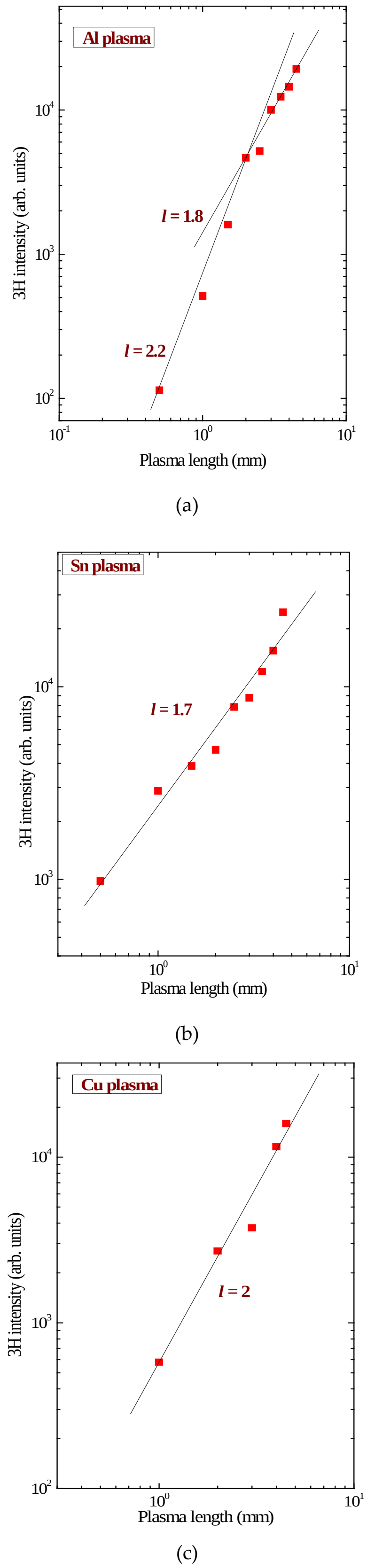

FIG. 2 DEPENDENCES OF THE 3H INTENSITY ON THE LENGTH OF (a) Al, (b) Sn, AND (c) Cu PLASMAS.

Another important parameter for optimization of low-order harmonic emission is an energy density of the heating pulse on the target surface. The dependences of 3H efficiency were measured on the energy of heating pulse on the surfaces of Al and Sn, while maintaining the same geometry of ablation beam on the targets (Fig. 3). The common feature of these studies was an observation of clearly defined maximum of the $I_{3H}(E_{hp})$ dependence (E_{hp}~2.5 mJ for both cases) with the following gradual decrease of harmonic yield during irradiation of targets using stronger heating pulses. The reason of these observations is related with over-excitation of the target, which led to appearance of the abundance of free electrons in the plasma plume. Notice that in the case of 3H this decrease of conversion efficiency with the growth of heating pulse intensity was not as abrupt as in the case of higher-order harmonics (Ganeev et al 2012 (1)).

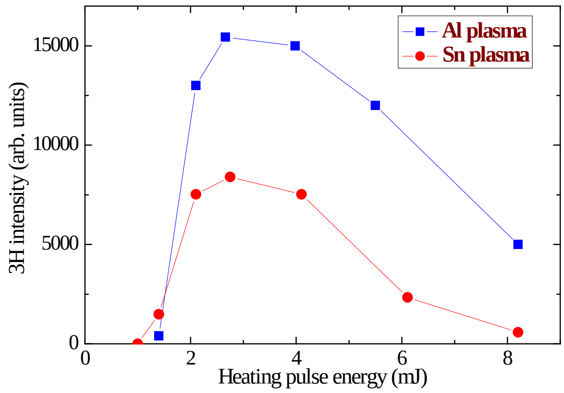

FIG. 3 DEPENDENCES OF THE 3H INTENSITY ON THE HEATING PULSE ENERGY IN THE CASES OF Al (FILLED SQUARES) AND Sn (FILLED CIRCLES) EXTENDED PLASMAS. THE INTENSITY OF DRIVING PULSE WAS MAINTAINED AT 1×10^{13} W cm^{-2}.

The measurements of the absolute value of 3H conversion efficiency were carried out using the following procedure. In the first step, the 3H was measured by a fiber spectrometer using the known energy of the 3H of 1064 nm radiation generating in the nonlinear crystals. This allowed the calibration of fiber spectrometer at 355 nm taking into account the apparatus function of our experimental setup for plasma harmonic generation. Following this calibration, the 3H conversion efficiencies in the tin and aluminum extended plasmas were estimated to be 3.5×10^{-4} and 8×10^{-4}.

Fourth Harmonic Generation in Long Plasma Using Two-Color Orthogonal Pump Scheme

4H generation in isotropic medium using orthogonal driving fields has previously been reported, to our best knowledge, only in Ref (Ganeev et al 2010) where a simplified scheme has been proposed for efficient 200nm pulse generation during two-color filamentation in air using the 50 fs, 800 nm radiation. A systematic study of the influence of the laser intensity, polarization, chirp, and pulse duration on the 4H output from the filaments was carried out and the 4H conversion efficiency was estimated to be of the order of 10^{-4}. The use of two-color pump may lead to an increase in the low-order harmonic generation efficiency similar to previously reported studies of the HHG in gases (Kim et al 2005, Mauritsson et al, Pfeifer et al, Charalambidis et al, Kim et al 2008) and plasmas (Ganeev et al 2012 (1)).

The insertion of KDP crystal in front of spherical focusing lens led to second harmonic generation with the conversion efficiency of 6%. The ratio of second harmonic (532 nm) and main (1064 nm) driving pulses (1:15) was sufficient for generation of the 4H in the extended plasma. Notice that 4H generation was excluded from observation in the case of two-color pump when the plasma was produced using the spherical focusing of heating pulse. This observation pointed out the importance of extended isotropic media for efficient 4H generation.

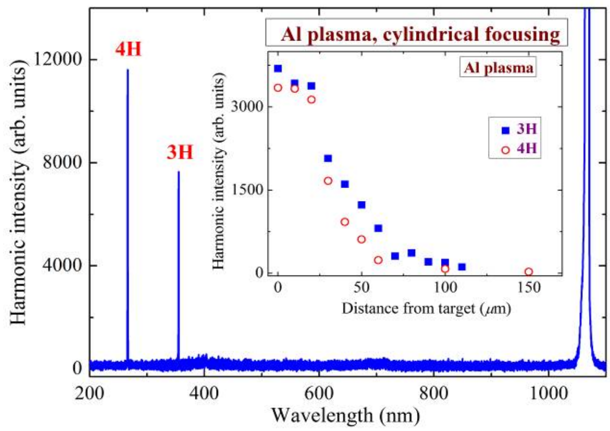

FIG. 4 SPECTRA OF RADIATION GENERATING IN EXTENDED Al PLASMA DURING TWO COLOR PUMP. INSET: DEPENDENCES OF THE 3H AND 4H INTENSITIES ON THE DISTANCE BETWEEN THE TARGET SURFACE AND DRIVING BEAM.

Figure 4 shows the typical spectral distribution of low-order (third and fourth) harmonics in the case of the two-color pump of aluminum plasma. Measurements of the absolute value of the 4H conversion efficiency were implemented using the procedure analogous to the one described for 3H calibration. For this, a fiber spectrometer HR4000 was calibrated at 266 nm using the 4H generating in the nonlinear crystals. In the case of Al plasma, 4H was stronger than 3H, though the ratio between the intensities of these harmonics was unstable, due to some instabilities of laser intensity and plasma formation. The conversion efficiencies of 3H and 4H in these two-color experiments were measured to be 3×10^{-5} and 5×10^{-5}. Harmonic generation in Sn and Cu plasmas showed less efficient 4H compared with the 3H.

The 3H and 4H yields considerably depended on the distance between the optical axis of the driving beams and the target surface (see inset in Fig. 4). This dependence was caused by the plasma density distribution above the target surface at a fixed delay between the heating and driving pulses.

The polarizations of 3H and 4H followed the polarization of driving 1064 nm beam, while the polarizations of two driving pulses (1064 nm and 532 nm) were orthogonal to each other.

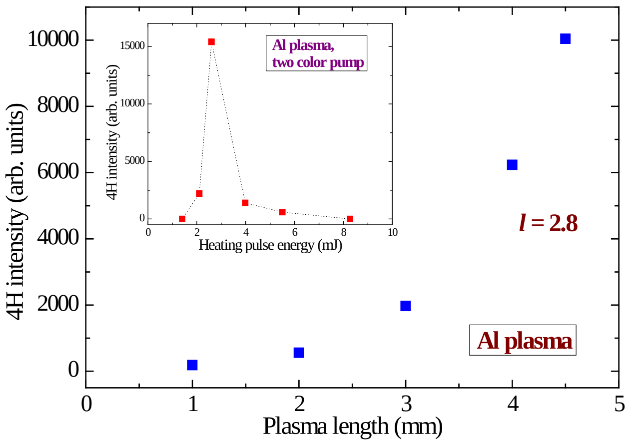

FIG. 5 DEPENDENCE OF THE 4H INTENSITY ON THE LENGTH OF Al PLASMA. INSET: DEPENDENCE OF THE 4H INTENSITY ON THE HEATING PULSE ENERGY IN THE CASE OF Al PLASMA.

The nonlinear dependence of the 4H yield on the length of plasma medium was analyzed using the Al plasma plume (Fig. 5). The slope of $I_{4H} \propto d^l$ dependence was steeper compared with the 3H case (l = 2.8 and 2.2 respectively). The reasons for the increase of a slope of this dependence in the case of 4H generation are not clear, since both the 3H and 4H generations are related with the four-photon processes.

The dependence of the 4H efficiency on the energy of heating pulse on the surface of aluminum target is presented in the inset in Fig. 5. A sharp increase of 4H efficiency was observed at E_{hp}=2.7 mJ with following steep decrease of harmonic yield, contrary to the case of 3H (Fig. 3). The same strong $I_{4H}(E_{hp})$ dependence was observed in the Sn plasma.

Discussion

The difference in $I_{harm}(E_{hp})$ dependences for 3H and 4H may have the same origin as above-described $I_{harm} \propto d^{\,l}$

dependences. Particularly, the propagation processes which are related with the phase difference between the waves of two pumps and 4H can play important role in this process. Some inexact spatial overlap of the two pump beams (1064 and 532 nm) in the plasma area can vary the two above dependences and create the conditions when the optimization of 4H occurs at a narrow range of the energies of heating pulses.

The insignificant difference in the 3H efficiency in Al, Sn, and Cu plasmas demonstrates that these samples have no specific features (neither strong absorption, nor resonance-induced enhancement) in the near ultraviolet range, meaning that plasma harmonic generation can be considered as a tool for the nonlinear spectroscopy analysis of the atomic physics and structure of materials, which has been also proved during the analysis of the resonance-induced enhancement of single harmonic during HHG using different driving laser pulses (Ganeev).

One can clearly distinguish the difference between the micro-processes (i.e. processes related with the nonlinear optical response of single emitter, when the harmonic yield can be significantly changed, particularly, in the case of resonance-induced enhancement of the low-order nonlinear susceptibility of these emitters) and macro-processes (so called propagation effects, which could be manifested through the optimization of the Gouy phase, addition to the nonlinear medium of the species possessing positive or negative dispersion for achieving phase matching, application of quasi-phase matching conditions, particularly using few gas or plasma jets in the case of isotropic media, laser beam aperturing, etc. (Reintjes, Ferray et al, Miyazaki et al, Christov et al 1997, Tempea et al, Pirri et al, Hergott et al)) governing harmonic generation. In the case of absence of the influence of resonance-induced single particle response, the efficiency of harmonic generation can be related with three length parameters. For efficient harmonic generation, the length of nonlinear medium L_{med} should be (a) comparable with the coherence length $L_{coh} = \pi/\Delta k$, which is defined by the phase mismatch between the fundamental and harmonic fields ($\Delta k = k_q - qk_0$ where k_q and k_0 are the harmonic and fundamental wave vectors, respectively) and depends on density and ionization conditions, and (b) smaller than the absorption length of the medium $L_{abs} = 1/\varrho\sigma$, where ϱ is the atomic density and σ is the absorption cross-section. Since, in the case of low-density plasma, the absorption in the spectral range of interest does not plays any significant role, the optimal length of the medium directly depends only on the phase relations between the fundamental wave and third harmonic (in the case of single-color pump) or between the fundamental and second harmonic waves and sum frequency wave (in the case of two-color pump).

One of unexpected finding of these studies is a stronger fourth harmonic yield compared with the third harmonic yield at equal conditionsof plasma formation on the aluminum surface. Notice that observation of stronger even harmonic compared with odd harmonic has already been reported in the case of the HHG induced by femtosecond pulses in some plasmas, while maintaining approximately same ratio between the fundamental and second harmonic driving fields (25:1, (Ganeev et al 2012 (2))).

As an explanation for the enhanced fourth harmonic generation in aluminum plasma, one can consider a sum mixing process ($4\omega = \omega+\omega+2\omega$), which can have other mismatch function with regard to the case 3H generation. The difference between these mismatch functions governs the ratio between the yields of third and fourth harmonics. The important parameter here is the intensity of the second pump field. In our case, with the ratio between two pumps (fundamental and second harmonic) of the order of 15 (94%:6%), one can expect rather weaker fourth harmonic yield compared with the third harmonic.

The assumption which explains the observed peculiarity of strong yield of 4H from Al plasma could be related with the following consideration. Recent observations of surface plasmon resonance (SPR) of aluminum nanoparticles in the UV range (210 nm central wavelength and 100 nm bandwidth for 8 nm nanoparticles) (Martin et al) suggest that, in the case of the plasmas containing small clusters, some nonlinear optical processes could be resonantly enhanced. The wavelength of 4H (266 nm) coincides with the shoulder of the SPR of aluminum nanoparticles and thus the harmonic in that case could be resonantly enhanced.

The presence of small nanoparticles in the plasma plume during ablation of Al targets using short (picosecond) laser pulses is a reasonable assumption, since such cluster formation has frequently been reported earlier during laser-matter interaction experiments. Currently, nanoparticle formation in vacuum during laser ablation of solid-state targets using short laser pulses is a well-developed technique (Amoruso et al, Preuss et al, Götz et al, Zergioti et al). Previously, a comparison of the morphology of debris after nanosecond and picosecond ablation unequivocally showed the advantages of short-pulse ablation for the preparation of nanoparticles [30]. It can be assumed that, in present experiments, we had the plasma plume containing some amount of aluminum nanoparticles.

The problem of consideration of the sum harmonic generation in the frames of perturbative theory is related with the orthogonal polarization of the two pump (ω and 2ω) waves. At these conditions, two pumps could hardly be considered as interacting waves. In this connection, the consideration of second wave as a source, which removes the restriction in generation of even harmonics related with the symmetry of the medium, could be an alternative approach in explanation of the observed peculiarities. The intensity of fundamental wave (1×10^{13} W cm^{-2}) was above the barrier suppression intensity of the aluminum atoms (6×10^{12} W cm^{-2}) with their first ionization potential of 6 eV. In that case, one can consider the same scenario of low-order harmonic generation, as in the case of HHG, when the trajectory of accelerated electron can be affected by the weak orthogonally polarized second harmonic field, which leads to the removal of the symmetry of plasma medium and correspondingly allows even harmonics generation alongside with the odd ones.

Below the following advantage of using the extended plasma media for harmonic generation has been addressed. The application of long plasmas can give the opportunity in observation of the quasi-phase matching (QPM) between the waves of driving and harmonic pulses. For this one has to use a bunch of plasma plumes of the sizes of coherence length. The coherence length is significantly depends on the dispersion of plasma, which can be manipulated by the heating pulse fluence, which leads to the variation of free electron concentration. Moreover, once the long plasma-induced low-order harmonic generation becomes optimized (as shown in present work), the following studies in the shorter wavelength range could reveal some attractive properties of the higher-order harmonics generating at these conditions, such as the QPM for some groups of harmonics in the extreme ultraviolet range. The application of relatively long, picosecond pulses at these conditions allows obtaining further ways for enhancement of the high-order harmonic yield.

As it has already been mentioned, all previous plasma harmonic studies were performed using the narrow plasma plumes. In the case of extended plasmas, the phase mismatch can cause a deterioration of the optimal relations between the phases of interacting waves. Once the coherence length of this process becomes less than the sizes of plasma, the reverse down-conversion of harmonics can compete with the up-conversion. Across a distance equal to the L_{coh}, a phase mismatch of π grows and causes destructive interference between the pump and harmonic waves. This process is one of the major limitations of the HHG conversion efficiency, especially in the case of abundant presence of free electrons in the medium. The phase mismatch varies as a laser pulse passes through a plasma plume due to further ionization of the nonlinear medium. Notice that the phase mismatch due to free electrons is considerably larger than the mismatch caused by atoms and singly charged ions.

Quasi-phase matching is a well-known approach for resolving this phase mismatch problem, which has previously been implemented in the case of gas media. QPM for HHG has started to be studied as early as in the mid of nineties (Shkolnikov et al,). A feasible structure for HHG was proposed to improve the conversion efficiency by a periodically modulated hollow waveguide (Paul et al, Christov et al 2000,) , and enhanced high harmonics have been generated by 15 fs driving laser pulses (Christov et al 2000,). Another method of QPM was demonstrated in (Zhang et al) where the use of a train of counterpropagating pulses allowed enhancing high-harmonic emission. This technique uses interfering beams to scramble the quantum phase of the generated short-wavelength light, to suppress emission from out-of-phase regions. Additionally, QPM was realized by using multiple gas jets whose pressure and separation were properly controlled (Geissler et al, Seres et al, Pirri et al). Moreover, it was shown in (Tosa et al) that multi-jet systems, beyond enhancing the harmonic photon flux, allow efficient control of the spectral and temporal structure of the harmonic field generated by a few-cycle laser pulse. It was demonstrated that QPM provided by a specially designed multi-jet system is able to enhance selected harmonics generated within a sub-femtosecond time window. This approach opens the door for the generation of single attosecond pulses with enhanced flux and tunable photon energy.

Recently, it has been proposed that the same procedure could be applied for the plasma harmonics using a simple method to fabricate numerous plasma jets tailored for the HHG, relieving technical restraints on the dimensions of the gas jets and their periodicity (Sheinfux et al). In their scheme, the jets were produced by ablation of a microlithographic periodic stripe pattern. Cylindrical plasma jets formed by ablation extension of the lithographic pattern into the space above the target, creating a row of narrow plasma jets of different material composition. The efficiency of HHG has been demonstrated to vary considerably with the plasma composition, and the periodic change in this efficiency enables QPM of HHG. However, until now there are no reports on the experimental studies of QPM for enhancement of harmonics in the plasma media.

Among the approaches for plasma harmonic QPM, one can consider the following techniques: (a) use of microlithografic targets, analogously to those proposed in (Sheinfux et al), (b) interference of two heating beams on the target surface with variable distance between the maximums of interference pattern, (c) use of the perforated targets allowing a separation in time of the plasma jets to reach the axis of propagation of the driving beam, (d) use of meshes with different distance between the wires as the shields before the ablating targets, which allows achieving the djustable separation between the plasma jets. All these approaches yet analyzed experimentally, whilst their application will allow the optimization of active areas of plasmas to match with the coherence lengths for different harmonics.

Additional attractiveness of plasma QPM is related with adjustment of the electron concentration in plasma by appropriate excitation of the targets. variation of electron concentration allows the adjustment of coherence length of harmonic generation in different spectral ranges. For HHG in the extreme ultraviolet range and beyond, dispersion in the medium can be mostly attributed to the free electrons generated by laser ionization of the medium. The manipulation of electron density could be easily accomplished at plasma harmonic generation compared with the gas harmonic generation, since the heating pulse could be properly adjusted to achieve a required density of free electrons.

Conclusions

In conclusion, the advanced properties of long (~5 mm) plasma plume for low-order harmonic generation have been demonstrated compared with the short (~0.5 mm) lengths of plasmas used in previous studiesanalysed; then the third and fourth harmonic generation have been analysed using the single-color and two-color pumps of the long plasmas produced on various metal surfaces. The fourth harmonic yield exceeded the one of 3H. The 3H conversion efficiencies in the tin and aluminum extended plasmas were measured to be 3.5×10^{-4} and 8×10^{-4}. Our studies revealed the conditions of efficient low-order harmonic generation in extended plasma plumes, which allowed the application of this technique for the analysis of the quasi-phase matching plasma harmonics.

ACKNOWLEDGMENTS

We thank The World Academy of Sciences (TWAS) for partial support of these studies (TWAS Research Grant).

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