

Quasi-phase-matching of high-order harmonics in multiple plasma jetsR. A. Ganeev,^{1,2,*} M. Suzuki,¹ and H. Kuroda¹¹*Ophthalmology and Advanced Laser Medical Center, Saitama Medical University, Saitama 350-0495, Japan*²*Voronezh State University, Voronezh 394006, Russia*

(Received 16 October 2013; published 12 March 2014)

Harmonic generation of laser radiation from ionized particles in the extreme ultraviolet range at the conditions of the phase matching between the femtosecond driving field and harmonics is an advanced concept for the development of efficient coherent sources in this spectral region. Here we present experimental evidence of quasi-phase-matched high-order harmonic generation in multiple plasma jets produced during laser ablation of the silver target. We observed both the enhancement of some groups of harmonics along the plateau range and the variation of maximally enhanced harmonic order; additionally we analyzed the influence of the number of plasma jets on the harmonic yield. The enhancement factor of 13 was achieved for the 33th harmonic of Ti:sapphire laser using the five-jet plasma configuration.

DOI: [10.1103/PhysRevA.89.033821](https://doi.org/10.1103/PhysRevA.89.033821)

PACS number(s): 42.65.Ky, 42.25.Bs, 42.65.Wi, 52.35.Mw

I. INTRODUCTION

Quasi-phase-matching (QPM) is an attractive approach to resolving the phase-mismatch problem for the high-order harmonic generation (HHG) of laser radiation in the extreme ultraviolet (XUV) range, which has been demonstrated in the case of gas media [1]. QPM was studied both theoretically and experimentally by using multiple gas jets whose pressure and separation were properly controlled, as well as by using other methods of the gaseous medium modulation [2–12].

However, the QPM in multijet gases did not achieve further amendments since the first reports of this phenomenon, probably due to the insignificant absolute values of enhancement and the difficulties in the implementation of the QPM technique. Contrary to that, the realization of the QPM in the laser-produced plasmas during high-order harmonic generation (QPM-PHHG) may offer a few prospective applications: easy manipulation of the characteristics of multijet plasmas (e.g., jet sizes, distance between jets, electron concentration, etc.); definition of the electron concentration through the measured values of maximally enhanced harmonics and coherence lengths (corresponding to the distance between the jets at which the group of enhanced harmonics was observed); modification of the QPM-PHHG using the perforated ablation beams, perforated targets, or interfering heating pulses; easy tuning of maximally enhanced groups of harmonics; amendments in absolute fluences of converted radiation for the small group of harmonics, which allows the use of metal filters without the dispersive elements to separate a few harmonics and use them for various applications. Presently, none of these prospective applications were demonstrated using the multijet gas media.

The attractiveness of QPM-PHHG could be related with the adjustment of the electron concentration (N_e) in the periodically modulated plasmas by optimal excitation of the ablating targets. The variation of N_e allows the adjustment of the coherence length (L_{coh}) for the efficient generation of harmonics in different spectral ranges. The adjustment of N_e could be easily accomplished during the plasma harmonic

generation, compared with the gas harmonic generation, through the proper variation of the fluence of an ablating beam on the target surface to achieve a required concentration of the free electrons in the plasma plumes.

In this paper, a demonstration of the QPM concept in the laser-produced plasmas in the case of the HHG of femtosecond laser radiation is presented. We analyze the properties of the multiple plasma jets produced by shielding some parts of the target surface from the laser irradiation. We show both the enhancement of some groups of harmonics along the plateau range and the tuning of maximally enhanced harmonic order at different regimes of multijet plasma formation.

II. EXPERIMENTAL ARRANGEMENTS

The uncompressed radiation of the Ti:sapphire laser operating at 10 Hz pulse repetition rate was used as a heating pulse (central wavelength $\lambda = 804$ nm, pulse duration 370 ps, pulse energy up to $E_{\text{hp}} = 4$ mJ) for extended plasma formation. The heating pulse was focused using the 200-mm focal-length cylindrical lens inside the vacuum chamber containing an ablating target to create the extended plasma plume above the target surface [Fig. 1(a)]. The focusing of the heating pulse on the target surface in the case of the absence of a multislit shield produced a line extended plasma. The intensity of the heating pulses on a plain target surface was varied up to 3×10^9 W cm⁻². The compressed driving pulse from the same laser with the energy of up to $E_{\text{dp}} = 5$ mJ and 64 fs pulse duration was used, after 45 ns from the beginning of ablation, for the harmonic generation in the plasma plume. The driving pulse was focused using the 400-mm focal-length spherical lens onto the prepared plasma from the orthogonal direction, at a distance of ~ 100 μm above the target surface. The confocal parameter of the focused driving beam was 12 mm. The intensity of the driving pulse at the focus area was varied up to 5×10^{14} W cm⁻². The harmonic emission was analyzed by an XUV spectrometer containing a gold cylindrical mirror and a 1200 grooves/mm flat field grating (FFG) with variable line spacing. The spectrum was recorded on a microchannel plate detector with the phosphor screen, which was imaged onto a charge-coupled device (CCD) camera.

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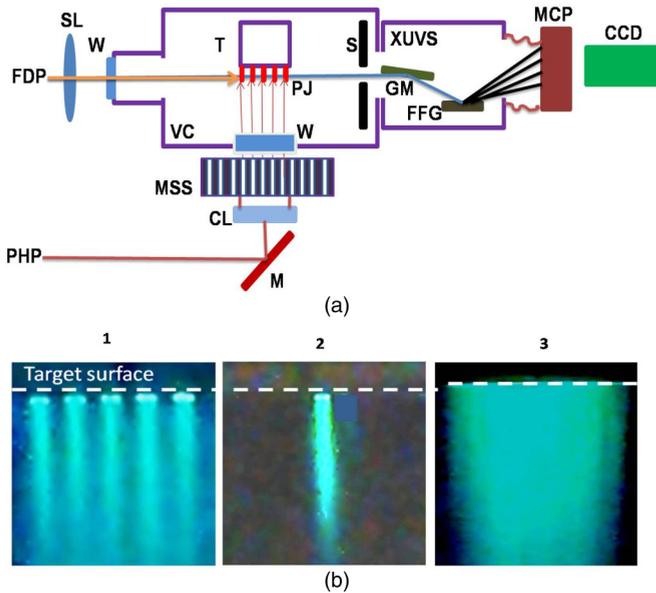


FIG. 1. (Color online) (a) Experimental scheme for the harmonic generation in the multiple plasma jets. FDP, femtosecond driving pulse; PHP, picosecond heating pulse; SL, spherical lens; CL, cylindrical lens; MSS, multislit shield; VC, vacuum chamber; W, windows of vacuum chamber; T, target; PJ, plasma jets; S, slit; XUVS, extreme ultraviolet spectrometer; GM, gold-coated cylindrical mirror; FFG, flat field grating; MCP, microchannel plate detector; CCD, charge coupled device camera. (b) Images of plasma formations. (1) Five-jet plasma formed on the silver target. (2) Single-jet plasma produced by shielding all but one open area between the strips of the multislit shield. (3) Extended plasma plume produced by removing the multislit shield from the pass of heating radiation.

We used silver as the ablating target. The size of the Ag target where the ablation occurred was 6 mm. To create

multiple plasma jets we used a thin glass plate with 0.5-mm-long aluminum strips separated from each other at a distance of 0.5 mm. This multislit shield was installed between the focusing cylindrical lens and target in such a manner that it allowed the division of the extended continuous 6-mm-long plasma into five 0.5-mm-long plasma jets with the distance between them ~ 0.5 mm. The shapes of five- and single-jet plasma formations, as well as of the extended plasma plume, are shown in Fig. 1(b). We were able to change the number of plasma jets by shielding the open parts of the multislit shield.

III. RESULTS

The important issue of these studies is the role of the length of plasma plume (d) in the variation of the harmonic yield from plasma medium. At the conditions of moderate heating of the target (i.e., at the intensity of $2 \times 10^9 \text{ W cm}^{-2}$ and the fluence of 0.4 J cm^{-2}) the harmonic yield should follow the quadratic dependence on the plasma length. At this fluence of heating radiation, the $I_H \sim d^2$ dependence did not show the saturation up to the maximal lengths of plasma [Fig. 2(a)].

The application of periodic plasma jets assumes a decrease of a whole length of active plasma medium compared with the ablation without shielding the target surface by a multislit shield. The division of extended plasma into the set of separated jets leads to a decrease of the effective length of the plasma plume from 6 mm (ablation without the shield) to 2.5 mm (ablation with the shield creating five 0.5-mm-long jets). Our measurements showing a significant decrease of harmonic yield with the shortening of plasma length at suitable ablation conditions [Fig. 2(a)] may predict a fall of conversion efficiency for the perforated plasma, the effective length of which was more than two times shorter than the length of the plasma produced on the target without the shielding

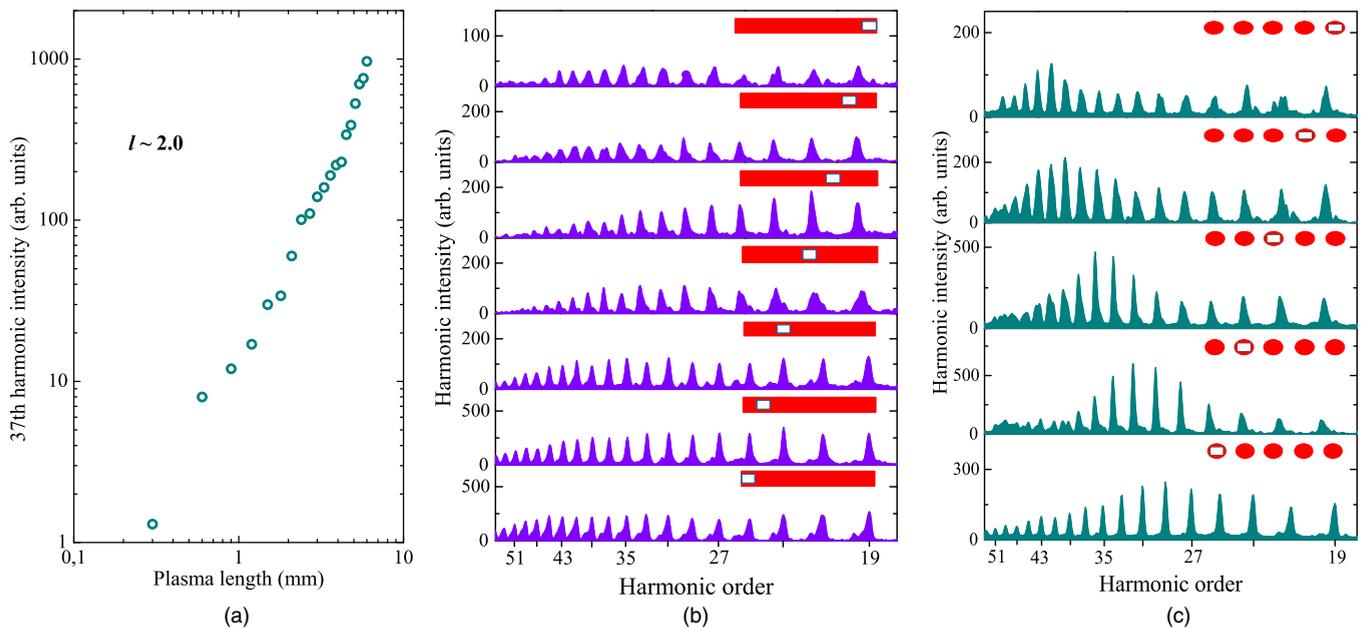


FIG. 2. (Color online) (a) Harmonic yield in the plateau range (37th order) as a function of the length of extended plasma. (b),(c) Silver harmonic spectra in the cases of (b) extended line plasma and (c) five plasma jets. Filled parallelepipeds and circles in the insets to each spectra indicate the spatial shapes of plasma plumes. Empty squares show the positions of the focal plane of driving beam.

of heating radiation. One may expect almost a sixfold decrease of harmonic emission in the case of perforated silver plasma. However, our studies of HHG in multijet plasma formations demonstrate a considerable deviation from this assumption. Moreover, we observed the enhancement of the parts of harmonic spectra, as well as a significant variation of the envelope of harmonic distribution in the plateau range, which unambiguously point out the involvement of the QPM on the structure and yield of coherent XUV radiation.

The harmonic spectra from the extended line plasma in the case of tuning of the focal plane position of driving radiation with respect to the plasma are shown in Fig. 2(b). The filled boxes at the right side of each spectrum show the 6-mm-long plasma plume, while the small empty boxes inside the filled ones indicate the position of the focal plane of the focused driving beam. We observed the featureless distribution of the harmonics extended up to the 53rd order, which did not change much at different positions of the focal plane of the driving field. However, in the case of the multiple plasma jets, a drastic change of the envelope of the harmonic spectra was observed. Additionally, these variations of harmonic structure showed different spectral areas where the maximum harmonic yield was achieved. Figure 2(c) shows the harmonic spectra obtained at different positions of the focal plane with respect to the five plasma jets. One can clearly see a significant departure from almost plateau-like distribution of harmonics, which was a characteristic of the HHG in the extended line plasma [Fig. 2(b)]. The maximums of the spectral envelope were tuned from the 29th harmonic towards the 43rd harmonic at different positions of the focal plane of the driving beam.

A significant enhancement of the groups of harmonics was due to involvement of the multiple plasma jets in the harmonic generation. The enhancement factor of 13 in the case of five 0.5-mm-long plasma jets compared with the 6-mm-long line plasma was achieved for the 33rd harmonic. Figure 3(a) shows the comparative intensities of harmonics in the cases of line plasma (thick line) and five jets (thin line) at equal experimental conditions, while the enhancement factors for the five-jet medium along the 18–38 nm spectral range are shown in Fig. 3(b).

The comparative studies of the harmonic spectra generated from the five-jet structure at different fluences of heating 370-ps pulses on the target surface were accomplished by changing the energy of heating radiation using the calibrated filters. We observed a tuning of the maximum of the spectral envelope towards the higher-order harmonics with a decrease of the fluence of heating radiation [Fig. 4(a)]. This result seems unexpected since one can assume a decrease of electron concentration in plasma for the smaller fluences of heating radiation and a corresponding decrease of the role of phase mismatch, which should lead to the disappearance of a maximum in the spectral distribution of harmonics.

Here we address the observed optimization of QPM-PHHG for shorter wavelengths at less excitation of the target during the formation of multijet plasmas [Fig. 4(a), upper panel]. To create the QPM conditions, one has to maintain a coupling between the driving and harmonic waves. For a given size of individual plasma jet (~ 0.5 mm), the QPM for the q th harmonic could be maintained at a fixed product $q \times N_e$ (since

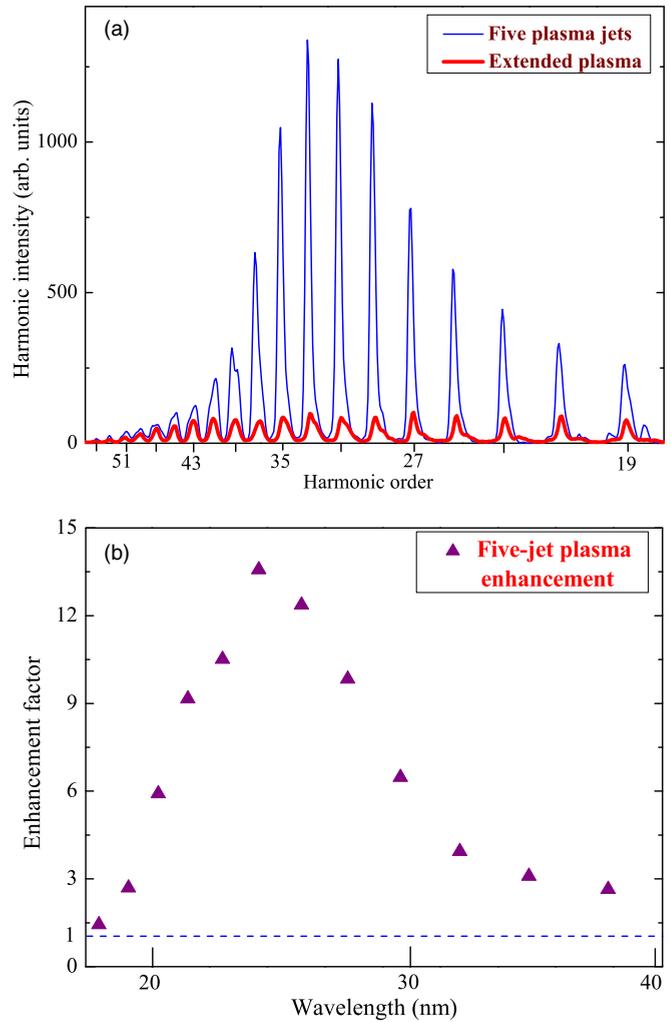


FIG. 3. (Color online) (a) Normalized spectra from the 6-mm-long plasma (thick line) and five 0.5-mm-long plasma jets (thin line). (b) Harmonic enhancement factors for the five-jet medium along the 18–38 nm spectral range.

$L_{\text{coh}} \approx 1.4 \times 10^{18} (q \times N_e)^{-1}$ [13,14]). A decrease of N_e at weaker excitation of the target should lead to the optimization of the QPM for higher q to keep the product $q \times N_e$ unchanged at the fixed spatial characteristics of plasma jets, which was demonstrated in Fig. 4(a).

The signature of QPM is a quadratic growth of harmonic yield with the growth of number (n) of coherent zones contributing to the signal. To prove the role of QPM in the observed peculiarities of harmonic spectra it would be straightforward to investigate the intensity (I_H) of harmonics as a function of the number of plasma plumes. This experiment has also confirmed the involvement of the coherent accumulation of harmonic yield along the whole length of divided nonlinear optical medium. We shielded step-by-step the number of heating areas on the target surface to create different numbers of plasma jets. Figure 4(b) shows the results of these measurements carried out at similar conditions of target excitation. Notice the equal Y axes of those spectra, which allow us to show the variations of the spectral shape of harmonic distribution at different numbers of jets and to demonstrate the growth of

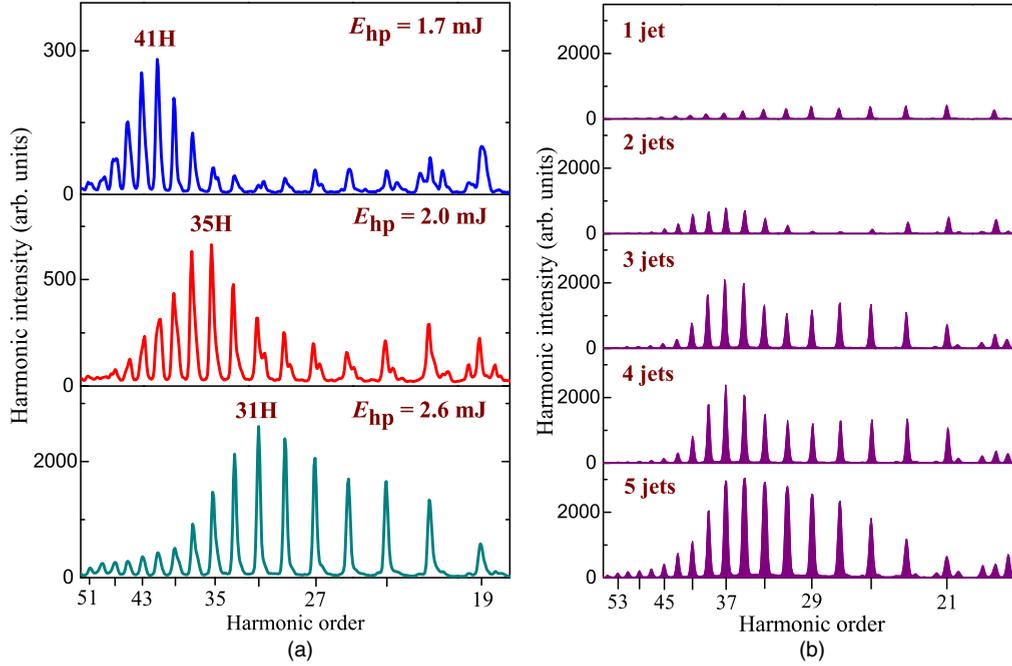


FIG. 4. (Color online) (a) Harmonic spectra from the plasma jets at different energies of the heating pulses. (b) Harmonic spectra generated from the plasma formations containing different number of jets.

QPM-enhanced harmonics during the addition of plasma jets (from single jet to five jets).

The anticipated featureless shape of harmonic spectra from the single 0.5-mm-long plasma jet was similar to those observed in the case of 6-mm-long plasma [compare the upper panel of Fig. 4(b) and the harmonic spectra shown in Fig. 2(b)]. With the addition of each next jet, the spectral envelope was drastically changed, with the 37th harmonic intensity in the case of five-jet configuration becoming almost 20 times stronger compared with the case of single-jet plasma. One can expect the n^2 growth of harmonic yield for the n -jet configuration compared with the single jet once the phase mismatch becomes suppressed [4,7], which gives the expected growth factor of 25 in the case of five-jet medium, which was close to the experimentally measured enhancement factor of 20. Notice that the maximum enhancement factor may decrease from the ideal value of n^2 at the conditions when absorption processes are turned on, or in the case of unequal properties of the jets, which can arise from the heterogeneous excitation of the extended target.

One can see that the QPM occurred for the same harmonics (approximately centered at the 35th–37th orders) for the two-, three-, four-, and five-jet plasma structures. We also observed the broadening of the envelope of QPM-enhanced harmonics with the growth of the number of contributing zones, which points out a decrease of selectivity induced by the above discussed reasons. Notice that the QPM-PHHG did not depend much on the distance between the zones. This was probably due to the prevailing influence of the density modulation of the particles (both electrons and atoms or ions) rather than the distance between the jets.

We calculated the plasma (N_p) and electron concentrations of silver ablation for the heating pulses of 370-ps duration at the fluences of 0.34, 0.4, and 0.52 J cm⁻². This simulation

was carried out by means of the code ITAP IMD [15]. The calculations of N_p and N_e for the three pulse energies presented in Fig. 4(a) show the following values at the distance of 100 μ m above the target surface 45 ns after the beginning of ablation: 1.4×10^{17} cm⁻³ and 2×10^{16} cm⁻³ ($E_{hp} = 1.7$ mJ, fluence 0.34 J cm⁻²), 1.7×10^{17} cm⁻³ and 2.7×10^{16} cm⁻³ ($E_{hp} = 2.0$ mJ, fluence 0.4 J cm⁻²), and 2.0×10^{17} cm⁻³ and 3.1×10^{16} cm⁻³ ($E_{hp} = 2.6$ mJ, fluence 0.52 J cm⁻²). The corresponding ratio between the N_e at $E_{hp} = 1.7$ and 2.6 mJ is equal to ~ 0.64 . The approximately same ratio between the most enhanced harmonic orders is obtained for these two cases [$q_{QPM}(2.6 \text{ mJ})/q_{QPM}(1.7 \text{ mJ}) \approx 0.7$]. Similar ratios of $q_{QPM}(x \text{ mJ})/q_{QPM}(y \text{ mJ})$ and $N_e(y \text{ mJ})/N_e(x \text{ mJ})$ where x and y are the different combinations accounting for the three values applied (1.7, 2.0, and 2.6 mJ) were obtained for all these pulse energies. It means that the simple scaling rule regarding the constant value of the product of $q_{QPM} \times N_e$ is fulfilled at our conditions of plasma excitation.

IV. DISCUSSION

There are four factors that contribute to the phase mismatch: atomic dispersion, Gouy phase shift, intensity-dependent dynamical phase shift in nonlinear dipole moments, and plasma dispersion. The first one refers to the variation in the refractive index of the neutral components of plasma, which shows considerably less influence than the dispersion induced by the presence of free electrons (fourth factor) in the plasma in the spectral ranges far from the resonance transitions, especially in the case of low-density plasma medium, thus allowing us to exclude the influence of atomic dispersion on the phase mismatch. Gouy phase shift could be also dismissed when the confocal parameter of the focused radiation exceeds the sizes of plasma, which was the case of our experiments

(correspondingly 12 and 6 mm). The Gouy phase mismatch [16] at our conditions was 0.6 cm^{-1} . The intensity of the femtosecond radiation used in the plasma area did not exceed $5 \times 10^{14} \text{ W cm}^{-2}$. At these conditions the phase mismatch between the harmonics emitted at different ends of the 6-mm-long plasma medium can be estimated as 8 cm^{-1} [17]. Finally, the phase mismatch due to plasma electrons [18] was considerably higher ($\sim 50 \text{ cm}^{-1}$ for the 33rd harmonic) compared with other components. Thus, at our conditions, the only component influencing the phase mismatch is related with the presence of electrons in the laser-produced plasma. Our studies confirmed that the above-mentioned formula for the coherence length can be used at the conditions of low-density plasma, long confocal parameters, and relatively small intensities of the driving pulse.

The influence of plasma effect or, in other words, ionization on the dynamics of HHG could be easily checked by analyzing the dependence of the harmonic yield on the length of the extended plasma. We analyzed these $I_H(d)$ dependences for different harmonics generated in the silver plasma, as well as in a few other extended plasma formations (Zn, Mn, and Cr) using the moderate excitation of the targets, and found that they had slopes in the range of 1.8–2.2, i.e., close to the quadratic dependence similar to that presented in Fig. 2(a). These studies showed that the free electrons appearing during both laser ablation and propagation of the driving pulse did not change this dependence at the optimal excitation of the target.

The measurements of the absolute values of HHG conversion efficiency were carried out using the technique described in [19]. For the target excited by the 2-mJ heating pulse the conversion efficiencies for the 33rd harmonic ($\lambda = 24 \text{ nm}$) generated from the extended and five-jet plasmas were measured to be $\sim 1.5 \times 10^{-6}$ and $\sim 2 \times 10^{-5}$. The corresponding energy of the 33rd harmonic pulses generating in the perforated plasma was estimated to be $0.1 \mu\text{J}$ (at the 5-mJ energy of the 64-fs driving pulse). This corresponds to the flux of the 33rd harmonic of $\sim 10^{10}$ photons per pulse. This flux is significantly higher than the reported flux of the enhanced 41st harmonic in the case of the gas QPM scheme using counterpropagating beams ($< 10^8$ photons per pulse [2]).

Regarding the comparison of our results with the best achievements of gas harmonics we would like to remind the reader that, presently, with the most common driving wavelength of 800 nm (Ti:sapphire laser) and targets (rare gases), the HHG efficiency in the plateau range of harmonic distribution is of the order of 10^{-6} . Conversion efficiencies of about 10^{-5} in the 30 nm [20] and 10^{-7} in the 13 nm [21,22] regions were obtained using high-power laser pulses

and a loose focusing geometry. The highest harmonic yield has been reported while using the semi-infinite gas cell, which had an estimated conversion efficiency of $\sim 10^{-4}$ for the single-order (27th) harmonic [23]. However, as the authors stressed, this estimate was greatly influenced by any errors in their assumptions of the number of ions, the molecular density, the size of the harmonic beam, the interaction volume, and the ionization cross sections.

Among the advantages of the QPM-PHHG approach are the simplicity in regulation of the electron concentration in the multiple plasma jets, the high enhancement factors of harmonics, and the availability for the express analysis of N_e . Two former advantages were demonstrated throughout this paper. The achieved conversion efficiency for the enhanced 33rd harmonic ($\sim 2 \times 10^{-5}$) is one of the highest reported so far for the harmonics in the plateau range, for both the gas and plasma HHG. As for the latter advantage, the definition of the maximally enhanced harmonics at the fixed sizes of separated plasma jets allows the calculation of N_e from the presented formula. Thus the potential of the proposed approach is, particularly, related with the analysis of the plasma characteristics, alongside with the amendments of harmonic yields in different spectral ranges. It is well known how difficult it is to define the dynamically changing electron concentration in the plasma plume. Our QPM-PHHG method allows resolving this problem by defining the strongest harmonic order and plasma jet sizes. In the case of laser-produced plasma, which has already been proven to be a competing medium for efficient harmonic generation [24–27], it is quite natural to explore the advantages of ionized medium for adjustment of the phase relations between the interacting waves, which was demonstrated in this study.

V. CONCLUSIONS

In conclusion, we presented a demonstration of the quasi-phase-matching concept using the multiple plasma jet formation and high-order harmonic generation. We have demonstrated the advanced properties of the subdivided plasma plumes over the extended line plasmas for the HHG and obtained the enhancement factor of 13 for the 33rd harmonic of 800-nm radiation using the five-jet configuration.

ACKNOWLEDGMENTS

This work was partially supported by JSPS KAKENHI (Grant No. 24760048). The authors thank P. V. Redkin for calculations of plasma characteristics.

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