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High-order harmonic enhancement using the quasi-phase-matching in laser plasma

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The quasi-phase-matching of the high-order harmonics generating in the multiple plasma jets produced on the surfaces of perforated Ag, V, and Mn targets is demonstrated. The 20× and 30× enhancement factors of quasi-phase-matched harmonics of 804 nm radiation in the range of 35th harmonic in the cases of silver and vanadium plasmas were achieved.

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Quasi-phase matching (QPM) is an attractive approach to resolve the phase mismatch problem for the efficient high-order harmonic generation (HHG) of laser radiation in the extreme ultraviolet (XUV) range, which has been demonstrated in the case of gas media [1–12]. Until now there were no reports on the studies of the QPM for enhancement of the high-order harmonics in the plasma media produced using the laser ablation. In the meantime, high efficiencies obtained with several plasma formations and resonance-induced enhancement of individual harmonics have demonstrated the advantages of using specially prepared plasmas for the HHG.

In this paper, we show the method of perforated target ablation for the formation of periodically separated plasma jets, which allowed implementing the QPM for enhancement of a group of harmonics.

The uncompressed radiation of Ti:sapphire laser was used as a heating pulse (central wavelength $\lambda = 804\,$nm, pulse duration 370 ps, pulse energy up to $E_{\rm hp} = 4\,$mJ) to ablate the targets for extended plasma or multi-jet plasma formation depending on the structure of target. The heating pulse was focused using the 200 mm focal length cylindrical lens inside the vacuum chamber containing an ablating target to create the plasma plume (Fig. 1). The sizes of ablated area were defined by the length of plain targets (5 mm). The focusing of heating pulse on the ablating surface in the case of plain targets produced the line plasma. The intensity of heating pulses on a plain target surface was varied in the range of $(1{-}3)\cdot 10^9\,\mathrm{W\cdot cm^{-2}}$.

The compressed driving pulse from the same laser with the energy of up to $E_{\rm dp} = 5\,$mJ and 64 fs (FWHM) pulse duration was used, after 45 ns delay from the beginning of plasma ablation, for harmonic generation in the plasma plumes. The driving pulse was focused using the 400 mm focal length spherical lens onto the extended plasma from the orthogonal direction, at a distance of $\sim 100\,\mu$m above the target surface, with a confocal parameter of 12 mm. The maximum available intensity of 804 nm driving pulse from this laser at the focus area was $7\cdot 10^{14}\,\mathrm{W\cdot cm^{-2}}$, while most of experiments were carried out at the intensities $\le 5\cdot 10^{14}\,\mathrm{W\,cm^{-2}}$. The harmonic emission was analyzed by an XUV spectrometer.

Ag, V, and Mn were used as the ablating targets. The application of these targets enabled the observation of harmonic generation along a broad spectral range and allowed the study of the variation of phase matching conditions for some groups of harmonics. To create multiple plasma jets, we used the separated plates of 0.5-mm-long Ag, 0.5-mm-long V, and 1-mm-long Mn. The principles of perforated plasma formation are shown in the left inset of Fig. 1, while the image of perforated Mn target and the plasma formation on this target are shown in the central and right insets. We also used the plain 5-mm-long Ag, V, and Mn plates for ablation and comparative studies of the HHG from extended homogeneous and perforated plasmas.

Fig. 2a shows the plateau-like harmonic spectra obtained from the 5-mm-long silver plasma at different energies of the heating pulse. The intensities of harmonics did not change much with the growth of the energy of the heating pulse, while the concentration of plasma (N_p) was increased from $1\cdot 10^{17}\,\mathrm{cm^{-3}}$ (at 1.1 mJ heating pulse) to $2.5\cdot 10^{17}\,\mathrm{cm^{-3}}$ (at 3 mJ heating pulse).

A separation of this extended plasma onto four 0.5-mm-long plasma jets at the conditions of perforated Ag excitation has dramatically changed the shape of generating harmonic spectra (Fig. 2b). The envelopes of harmonic spectra in the case of perforated plasmas were changed from the featureless shape to the significantly

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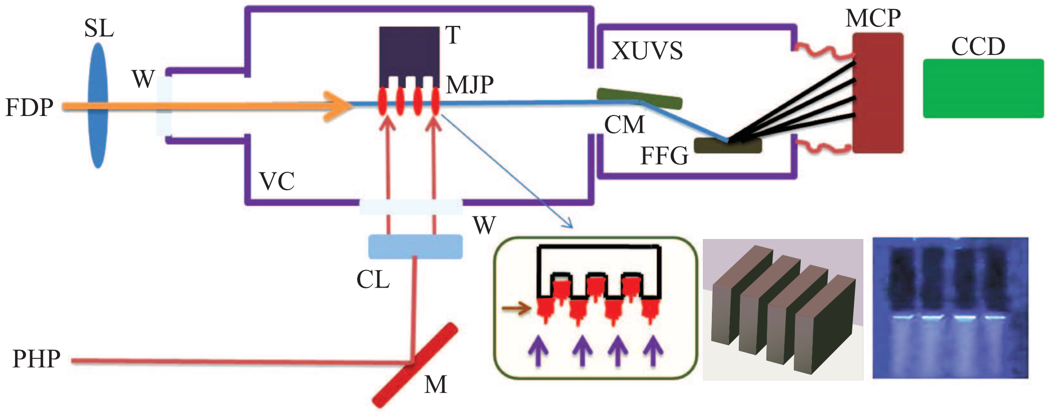

Fig. 1. Experimental scheme for harmonic generation in multiple plasma jets: FDP – femtosecond driving pulse; PHP – picosecond heating pulse; SL – spherical lens; CL – cylindrical lens; VC – vacuum chamber; W – windows of vacuum chamber; T – target; MJP – multi-jet plasma; XUVS – extreme ultraviolet spectrometer; CM – gold-coated mirror; FFG – flat field grating; MCP – micro-channel plate registrar; CCD – charge coupled device camera. Left inset shows the principle of plasma formation at different distances from the axis of propagation of the driving pulse. Central inset shows the image of perforated Mn target. Right inset shows the image of the plasma jets formed on the perforated Mn target

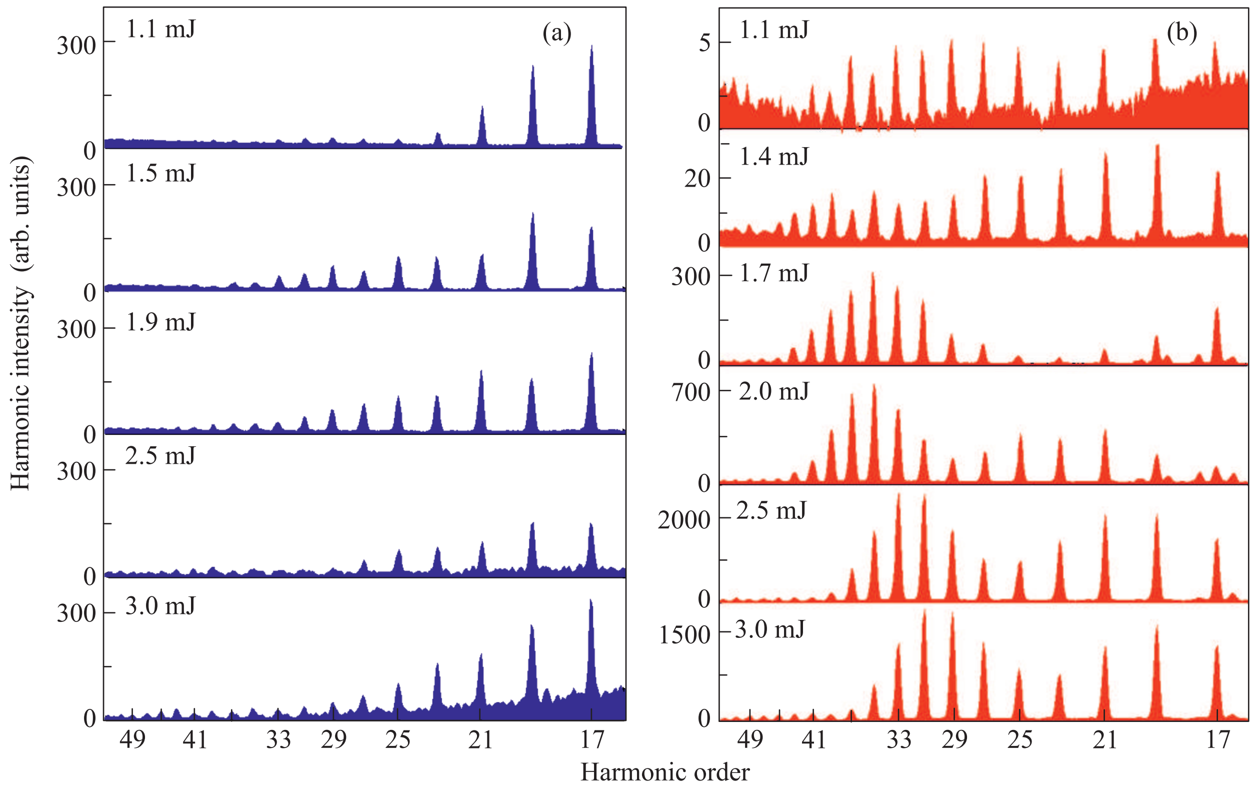

Fig. 2. Comparative harmonic spectra from extended 5-mm-long plasma produced on the plain silver target (a) and four 0.5-mm-long jets formed on the perforated silver target (b) at different energies of heating pulse

modulated pattern. A clear evidence of constructive and destructive interference in different parts of studied spectral range is presented on the panel showing the excitation using the 1.7-mJ pulses.

The influence of plasma effect or, by other words, ionization on the dynamics of HHG could be easily checked by analyzing the dependence of harmonic yield I_H on the length d of extended plasma. We analyzed these $I_{\mathrm{H}}(d)$ dependences for different harmonics in a few plasma plumes and found that they had the slopes in the range of 1.8–2.2, i.e. close to the quadratic dependence. These studies showed that the free electrons appearing during both laser ablation and propagation of driving pulse did not play a decisive role in the variation of this dependence at the optimal excitation of the target.

The comparative measurements of the harmonic yields for extended and perforated plasmas at similar heating fluence showed a 20-fold enhancement of the QPM harmonics. The shortage of the length (compared with extended 5-mm line plasma) should lead to a 6.25-fold decrease of harmonic yield ($(5/2)^2$) in the case of four 0.5-mm-long plasmas. Instead, we achieved the $20\times$ growth of QPM harmonic yield. If one assumes the method of enhancement factor calculations introduced in [7], then we have to multiply this enhancement factor by 6.25. With these assumptions, the achieved enhancement is comparable or higher with regard to those reported for gas QPM ($36\times$ [3], $90\times$ [7], $300\times$ [2]).

The measurements of the absolute values of HHG conversion efficiency were carried out using the technique described in [13]. For the 2 mJ heating pulse exciting target at a 10 Hz pulse repetition rate, the conversion efficiency for the 35th harmonic generating from four-jet Ag plasma was measured to be $1 \cdot 10^{-5}$. This efficiency corresponded to the flux of the 35th harmonic of $5 \cdot 10^9$ photons per pulse. This flux is significantly larger than the reported flux per pulse of the enhanced 41st harmonic in the case of the gas QPM scheme using counterpropagating beams (10^7 photons per pulse [2]).

Here we address the observed optimization of QPM for shorter wavelengths at less excitation of target during the formation of multi-jet plasma (Fig. 2b; compare the spectral maxima of QPM harmonics for the 1.7- and 3-mJ heating pulses). To create the QPM conditions, one has to realize a modulation of the coupling between the driving and converted frequencies, which is periodic along the generation path. For a given distance between the 0.5-mm-long jets (~ 0.5 mm), the QPM for the q-th harmonic could be realized at a fixed product $q \times N_e$ (since $L_{\mathrm{coh}} \sim (q \times N_e)^{-1}$ [14]). Here L_{coh} is the coherence length of generating harmonic and N_e is the electron concentration in plasma. A decrease of N_e (at less excitation of target) should lead to optimization of the QPM for higher q to keep the product qN_e unchanged at the fixed spatial characteristics of plasma jets, which was demonstrated in Fig. 2b.

There are four factors that contribute to phase mismatch: atomic/ionic dispersion, Gouy phase shift, intensity-dependent dynamical phase shift in nonlinear dipole moments, and plasma (actually electron) dispersion. The first one refers to the variation in the refractive index of the atomic/ionic components of plasma, which show a considerably less influence than the dispersion induced by the presence of free electrons (fourth factor) in the plasma. Gouy phase shift could also be excluded from consideration when the confocal parameter of focused driving radiation exceeds the sizes of plasma, which was the case of our experiments (correspondingly 12 and 5 mm). The intensity of used femtosecond radiation in the plasma area did not exceed $5 \cdot 10^{14}\,\mathrm{W \cdot cm^{-2}}$. At these conditions the phase mismatch induced by second factor can be estimated to be $8\,\mathrm{cm^{-1}}$. Finally, the phase mismatch due to plasma electrons was considerably higher ($\sim 50\,\mathrm{cm^{-1}}$ for the thirties harmonics) compared with other components.

There is some another interesting feature in Fig. 2b, which requires further consideration. For increasing electron density a second enhancement peak around the 19th harmonic was observed. Probably, this could be due to a higher-order QPM process. Such a possibility has been analyzed previously in [11]. The study of this effect is under way.

It was shown during previous QPM studies using counter-propagating pulses [6] that QPM is highly selective as the number of contributing zones increases. We irradiated the plain silver target by the modulated heating beam using the multi-slit shield to create the multiple plasma jets. The multi-slit shield was installed after the focusing cylindrical lens in such a manner, that it allowed the division of the extended continuous 5-mm-long plasma onto the set of a few plasma jets. These studies showed that the QPM occurred for the same harmonics (approximately centered at 35th–37th orders) for the 2-, 3-, 4-, and 5-jet plasma structures. Simultaneously, we observed the broadening for the QPM envelope with the growth of number of contributing zones, which points out a decrease of selectivity. Notice that plasma QPM HHG did not depend much on the distance between the zones. This was probably due to the prevailing influence of the modulation of density of particles (both electrons and atoms/ions) over the distance between these modulations.

The signature of QPM is the growth of harmonic yield $\sim n^2$, where n is the number of coherent zones contributing to the signal. We studied the intensity of harmonics as a function of the number of plasma jets. For this we used the multi-slit shield to create the perforated plasma. We shielded step-by-step the number of heating areas on the target surface to change the number of plasma jets. The anticipated featureless shape of harmonic spectra from the single 0.5-mm-long plasma

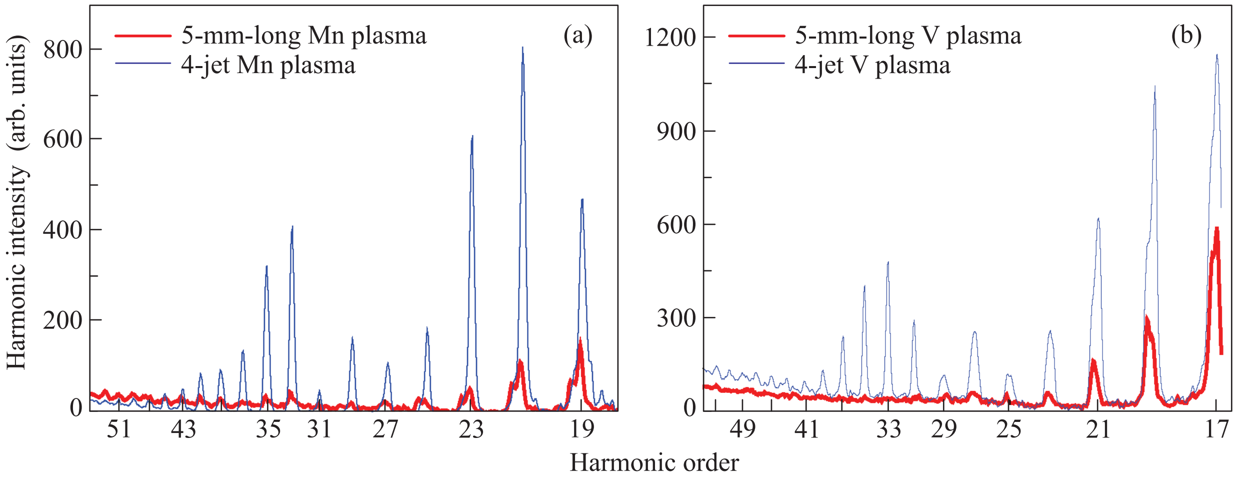

Fig. 3. (a) – Comparative harmonic spectra from extended line plasma produced on the plain manganese target (thick curve) and four 1-mm-long jets of Mn plasma (thin curve). (b) – Comparative harmonic spectra from extended line plasma produced on the plain vanadium target (thick curve) and four 0.5-mm-long jets of V plasma (thin curve)

jet was similar to those observed in the case of 5-mm-long plasma. 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 22 times stronger compared with the case of single jet plasma. The expected growth factor of 25 in the case of five-jet medium was close to experimentally measured enhancement factor.

The HHG in perforated V and Mn plasmas also revealed similar properties. There is a reason for investigation of the plasma QPM using the latter target, which has previously shown a resonance-induced enhancement of 33rd harmonic. In present experiments with 5-mm-long manganese plasma, the enhanced 33rd harmonic was followed with a gradual decrease of higher-order harmonics (Fig. 3a, thick curve), analogously to earlier reported studies [15]. In the meantime, the use of perforated manganese plasma led to the enhancement both in the range of so called second plateau and in the longer-wavelength range (Fig. 3a, thin curve). The QPM-induced enhancement of harmonics in the range of 21st order was related with the use of broader plasma plumes compared with the case of silver plasma (1 and 0.5 mm respectively). The shift of QPM towards the lower-order harmonics corroborates with the $q_{\rm opt} \sim (L_{\rm jet} N_e)^{-1}$ relation for the fixed size of single jet ($L_{\rm jet}$). Once $L_{\rm jet}$ is increased, the optimal harmonic orders ($q_{\rm opt}$) at fixed N_e, for which the QPM is fulfilled, become smaller and move towards the longer wavelength range.

The 5-mm-long vanadium plasma showed very weak harmonics above the 29th order (Fig. 3b, thick curve), with barely seen higher orders. Again, this pattern was drastically changed in the case of 4-jet plasma (Fig. 3b, thin curve). Along with some enhancement of lower-order harmonics, a noticeable appearance of the group of harmonics centered in the range of 33rd order showed a clear evidence of the amendments of this nonlinear optical process due to the constructive interference of harmonics in this spectral region. We measured the 30× enhancement of these harmonics compared with the case of non-perforated 5-mm-long vanadium plasma.

The scalability of this approach to higher photon energies depends on the availability in generation of harmonics from some extended plasma plumes in this spectral region. Such plasmas as Ag, Mn, and B allow generation of up to the sixties harmonics and above (up to the hundredth in the case of manganese). The limits of plasma QPM HHG technique are defined by the properties of plasma plumes. Probably, maximal photon energies at which plasma QPM could be realized at present stage of the developments of plasma HHG in laser-produced plumes would not exceed 130–150 eV.

In conclusion, we have demonstrated, for the first time, the method of quasi-phase-matching of the high-order harmonics generating in the multiple plasma jets produced on the surfaces of perforated silver, vanadium, and manganese targets. The 20× and 30× enhancement factors of quasi-phase-matched harmonics in the cases of silver and vanadium plasmas were achieved. This technique allows the manipulation of the phase difference between the interacting waves in the laser-produced plasmas and the enhancement of a group of harmonics in different spectral ranges.

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