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NONLINEAR
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Two-Color Pump of Laser Plasmas for Harmonic Generation[1]

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Abstract—Various laser-produced metal plasmas were used for high-order sum and difference harmonic generation using the mixing of tunable mid-infrared pulses from optical parametric amplifier and 810 nm ultrashort pulses. We show that, regardless of non-optimal spatio-temporal overlap of two sources of radiation in the plasmas, the application of proposed technique allows a significant growth of harmonic yield and broadening of the number of different combinations of interacting waves in the extreme ultraviolet region, which could be useful for observation of the resonance induced enhancement of some frequency components.

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INTRODUCTION

One of the goals of high-order harmonic generation (HHG) of ultrashort laser pulses using different approaches is the growth of emitting components at different wavelengths of extreme ultraviolet (XUV) region. The commonly used way here is the application of the 800-nm-class lasers and second-order harmonics leading to doubling of generated harmonics from odd to odd and even orders. A strong harmonic generation in the case of two-color pump (TCP) is possible due to the formation of a quasi-linear field, selection of a short quantum path component, which has a denser electron wave packet, and higher ionization rate compared with the single-color pump (SCP). There is an extensive body of the studies dealing with TCP in gas phase, which consider many aspects of this approach, including macroscopic effects, polarization-related processes, and commensurate versus incommensurate TCP [1–14].

Notice that most previous TCP schemes used the so-called commensurate waves, i.e., waves representing some integers from each other, particularly 800 nm radiation and its second harmonic (H2). Meanwhile, application of incommensurate waves for TCP of gases showed the advantages both in harmonic yield and realization of individual attosecond pulses generation from the multi-cycle sources [11, 13, 15, 16]. The attractiveness of this approach was related with the availability of such multi-cycle sources in many laboratories and attempts to overpass the restrictions in generation of single attosecond pulses by the few-cycle radiation.

In the meantime, another method of harmonic generation utilizing ablation of solids with further propagation of femtosecond pulses through the plasma plumes has recently shown various attractive properties, which distinguish this method of short-wavelength coherent sources formation from gas harmonics. Among them are the resonance enhancement of single harmonic [17], quasi-phase matching in extended plasmas [18], and application of ablated nanoparticles for enhancement of harmonic yield [19]. In some cases, the efficiencies/photon numbers observed in plasma HHG studies were comparable to gas HHG [20–22]. Two-color pump of plasmas using conventional scheme (800 nm + H2) was analyzed in [23] and has shown the notable enhancement of harmonic yield compared with SCP.

In this paper, we demonstrate high-order sum and difference harmonic generation using the mixing of mid-infrared (MIR) pulses from optical parametric amplifier (OPA) and 810 nm ultrashort pulses in extended laser-produced metal plasmas. We analyze high-order parametric and harmonic generation in palladium, zinc, lead, silver, and cadmium laser-produced plasmas (LPP) and show a significant enhancement of harmonic yield using the MIR + 810 nm pump scheme compared with the SCP utilizing MIR pulses.

[1]The article is published in the original.

EXPERIMENTAL ARRANGEMENTS

Experimental setup comprised the Ti:sapphire laser, traveling-wave OPA, and HHG scheme using propagation of two pulses (amplified signal or idler radiation from OPA and 810 nm radiation) through the extended LPP (Fig. 1). Part of amplified uncompressed radiation of pump laser with pulse energy of 5 mJ was separated from a whole beam and used as a heating pulse for homogeneous extended plasma formation using the 200-mm focal length cylindrical lens installed in front of the extended targets placed in the vacuum chamber. The intensity and fluency of the heating pulses on the target surface were varied up to 4×10^9 W cm^{-2} and 1.4 J cm^{-2}, respectively. We used 5-mm-long Pd, Pb, Ag, Zn, and Cd samples as the targets for ablation and formation of extended homogeneous plasma.

The compressed radiation of Ti:sapphire laser (pulse energy 8 mJ, pulse duration 64 fs, 10 Hz pulse repetition rate, pulse bandwidth 17 nm, central wavelength 810 nm) pumped the OPA (HE-TOPAS Prime, Light Conversion). Signal (S) and idler (I) pulses from OPA allowed the tuning along the 1200–1600 nm and 1600–2600 nm ranges, respectively. Most of experiments were carried out using the mixture of 1 mJ, 70 fs signal pulses and 3 mJ, 64 fs, 810 nm pulses in metal plasmas, while some studies of harmonic emission were carried out using the idler radiation mixed with the 810 nm pulses. The intensity of focused 1310 nm pulses inside the LPP was 2×10^{14} W cm^{-2}. The harmonic emission was analyzed using an XUV spectrometer.

Most of experiments were carried out using the TCP pump of metal LPP. The reason for using the TCP instead of SCP was related with the small energy of driving MIR signal pulses. The $I_H \propto \lambda^{-5}$ rule (I_H is the harmonic intensity and λ is the driving field wavelength [24]) led to a significant decrease of harmonic yield in the case of longer-wavelength sources compared with 810 nm pump and did not allow the observation of strong harmonics even from the 1310 nm pulses, which had the highest energy in the 1200–1600 nm region. Because of this in most of following studies we used the TCP scheme comprising mid-infrared pulses and Ti:sapphire laser radiation, alongside the conventional TCP using the H2 radiation as a second field. In the latter scheme, the 0.5-mm-thick BBO crystal (type I, $\theta = 21°$) was installed inside the vacuum chamber on the path of focused signal pulse (Fig. 1). The conversion efficiency of H2 pulses was 20%.

RESULTS AND DISCUSSION

The addition of 810 nm wave to the MIR pulses led to appearance of strong high-order sum and difference frequencies alongside the harmonics attributed to the 810 nm pulses. Figure 2a shows the example of this enhancement compared with SCP in the case of zinc plasma. The application of 1310 nm signal pulses (filled curve) allowed generation of a few weak harmonics, especially in the spectral region below 80 nm. The use of the TCP (MIR + 810 nm, thick blue curve) at similar experimental conditions caused a dramatic growth of harmonic yield (3×–10×, depending on harmonic order). Odd harmonics of 810 nm radiation appeared alongside the sum and difference frequencies ($H_{odd} \pm$ MIR) along the whole spectral range of observation. The intensities of sum and difference frequency components were almost similar to the odd harmonics of 810 nm pump, in spite of significant difference of the pulse energies of MIR and Ti:sapphire laser radiation (1 mJ and 3 mJ, respectively) and abovementioned rule of wavelength-dependent yield of harmonics. The cutoff energies of generated harmonics in the case of TCP and SCP schemes were notably different from each other (29 and 18 eV, respectively). Similar amendment and extension of

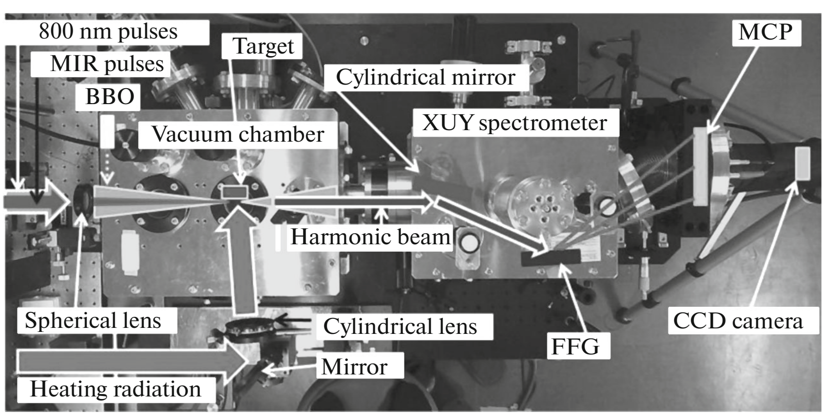

Fig. 1. Two-color pump experiments using MIR and Ti:sapphire laser radiation. FFG, flat field grating; MCP, micro-channel plate; BBO, nonlinear crystal for second harmonic generation of the signal pulse.

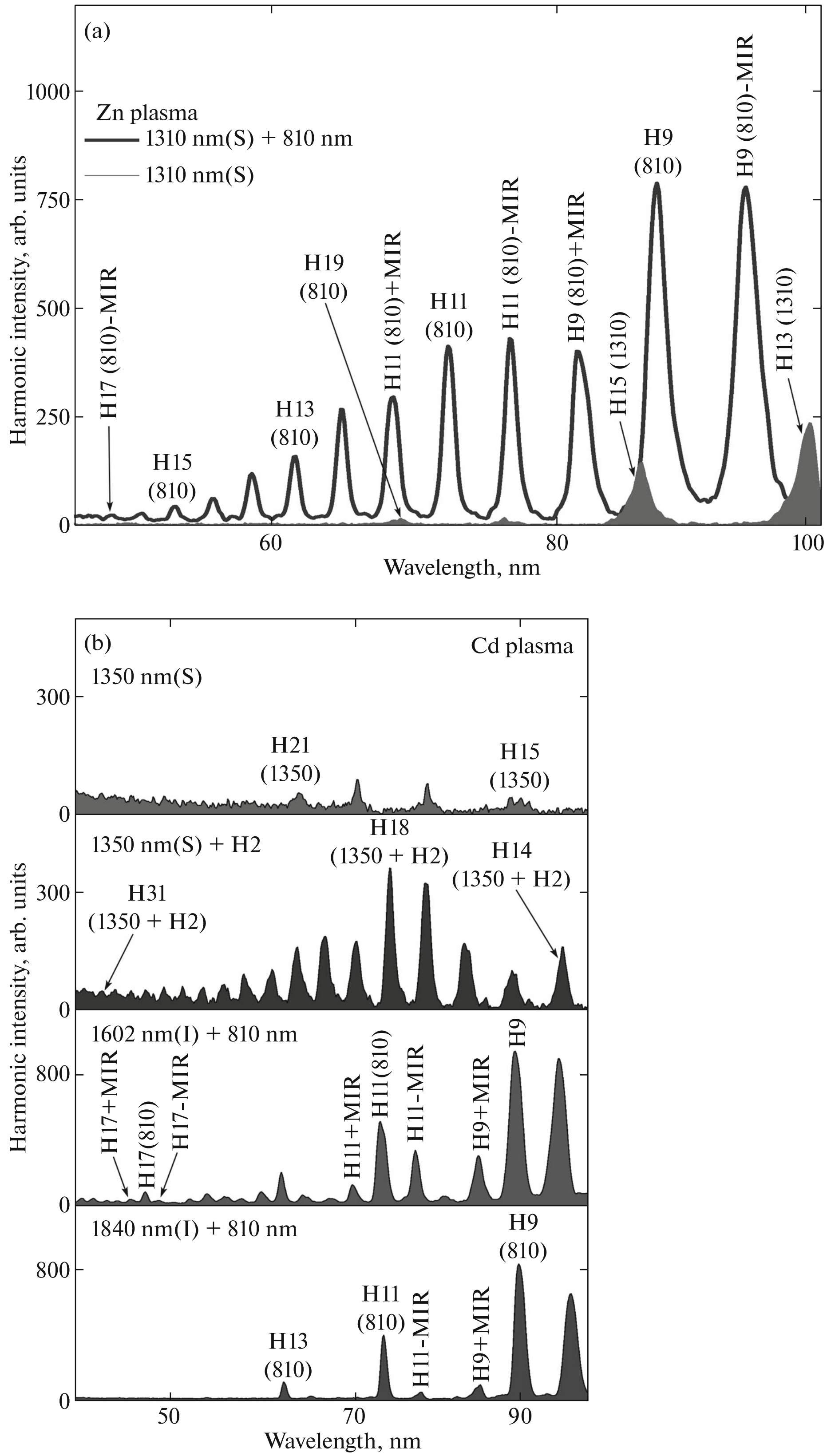

Fig. 2. (a) Single-color (MIR) and two-color (MIR + 810 nm) pump induced harmonic spectra from zinc plasma. (b) Single-color (MIR) and two-color (MIR + H2) pump induced harmonic spectra from cadmium plasma using signal pulses (two upper panels). Application of idler pulses for the TCP (MIR + 810 nm) of cadmium plasma (two bottom panels).

harmonic cut-off were observed in the case of other plasmas.

Previous gas HHG studies have demonstrated that application of two-color field either through fundamental plus second harmonic, or two incommensurate waves (via frequency conversion in OPA), led to the growth in harmonic yield and extension of harmonic cut-off. Our studies showed that HHG in laser-produced plasmas behaves in the same way. The conventional TCP scheme also allowed the extension of harmonic cutoff. Particularly, one can compare two upper panels of Fig. 2b showing harmonic generation in the cadmium plasma using SCP (MIR) and TCP (MIR + H2). The generated odd and even harmonics of 1350 and 675 nm waves in the case of TCP were notably stronger compared with the odd harmonics generated using SCP when only 1350 nm radiation was utilized for HHG. Notice a similarity of experimental conditions for both TCP and SCP schemes when the second field was introduced by either removing a dichroic mirror separating 810 nm and parametric waves in OPA or inserting the BBO crystal on the path of MIR radiation. The comparative studies of two TCP schemes showed that both of them allow extension of harmonic cut-off and significant growth of harmonic yield while providing different frequency components in a whole generating spectrum.

The application of weak idler MIR pulses in the case of TCP led to preferable generation of the harmonics associated with 810 nm pulses (two bottom panels in Fig. 2b). However, even at these unfavorable conditions when the ratio of MIR and 810 nm pumps was 1 : 6 (in the case of 1602 nm pulses) and 1 : 8 (in the case of 1840 nm pulses) we were able distinguishing high-order sum and difference frequency components, especially in the longer wavelength range. Note that the abovementioned $I_H \propto \lambda^{-5}$ rule should lead to a 60-fold decrease of harmonic yield in the case of 1840 nm SCP compared with 810 nm pump.

The characteristic feature of these TCP studies using shorter and longer wavelength MIR pulses was the appearance of different number of frequency components between each pair of the odd harmonics of 810 nm pump. In the case of S + 810 nm pump, there were two components between each pair of odd harmonics (Fig. 2a, thick curve), while for the I + 810 nm case we observed three frequency components (third panel of Fig. 2b). The third peak appeared between two MIR harmonics (particularly H9 and H11) could be attributed to the $E_{\mathrm{H11(1602\,nm)}} - E_{\mathrm{806\,nm}}$ difference frequency generation and/or $E_{\mathrm{H9(1602\,nm)}} + E_{\mathrm{806\,nm}}$ sum frequency generation.

The application of two MIR pulses (signal and idler) for mixing with 810 nm radiation in LPP allows realizing three-color pump conditions. Figure 3 (two upper panels) presents such an interaction of three waves in silver plasma using nondegenerate [1310 nm (S) + 2075 nm (I) + 810 nm] and degenerated [1605 nm (S) +1607 nm (I) + 810 nm] regimes of OPA operation. The harmonics generated in silver plasma using former three-waves interaction showed a difference with regard to the 1310 nm + 810 nm pump (Fig. 2a). One can clearly see the appearance of additional frequency components between each pair of the odd harmonics of 810 nm radiation (Fig. 3, upper panel). In the degenerate conditions of OPA the MIR pulses represent the wavelengths, which almost exactly match with the half of 810 nm photon energy. In that case the number of frequency components between odd harmonics of 810 nm radiation (Fig. 3a, middle panel) remained same as the one shown in the third panel of Fig. 2b irrespective on which MIR pulses, either signal or idler, participate in the frequency mixing in LPP. Note a difference in the yields of the frequencies associated with odd harmonics of 810 nm and sum and difference harmonics associated with the involvement of MIR pulses in these two cases. Contrary to this observation, the use of MIR + H2 pump allowed generation of almost equal odd and even harmonics of one of MIR pulses (1310 nm; see bottom panel of Fig. 3).

These observations show the involvement of three waves in the parametric generation of coherent XUV radiation. The three-color pump induced harmonic generation significantly depends on the relative phases of interacting waves, delays between them, and spatial overlap. Two MIR pulses have some delay between each other due to the group velocity dispersion in the last amplifier of OPA. Particularly, the longer wavelength MIR component (I) always slightly delayed with regard to the shorter wavelength one (S) during propagation through the BBO amplifier of OPA.

During most of these studies we used the orthogonally polarized pumps (signal waves from OPA and 810 nm pulses) since the signal waves were stronger compared with the idler ones (Fig. 2a; Fig. 2b, second panel; Fig. 3, third panel). The example of parallel polarized I + 810 nm pump is presented in the case of Cd plasma (Fig. 2b, two bottom panels). The application of parallel polarized pumps was also analyzed using the TCP when the weak idler pulses were mixed with the 810 nm radiation in the Pd and Pb plasmas. Figure 4a shows two spectra in the case of the 1602 nm idler pulses mixed with the 810 nm pulses to pump the palladium plasma at different temporal overlaps of interacting waves. The energies of idler pulses were weaker compared with signal waves, whilst the appearance of additional harmonic components in the case of temporally overlapped TCP using former pulses was qualitatively similar to the application of stronger orthogonally polarized signal pulses (upper panel).

The delay of 810 nm pump with regard to seeding pulses in the last amplifier of OPA was commonly optimized to generate strongest signal pulses rather than idler ones. At these conditions, the signal pulses were insufficiently overlapped with 810 nm pulses in

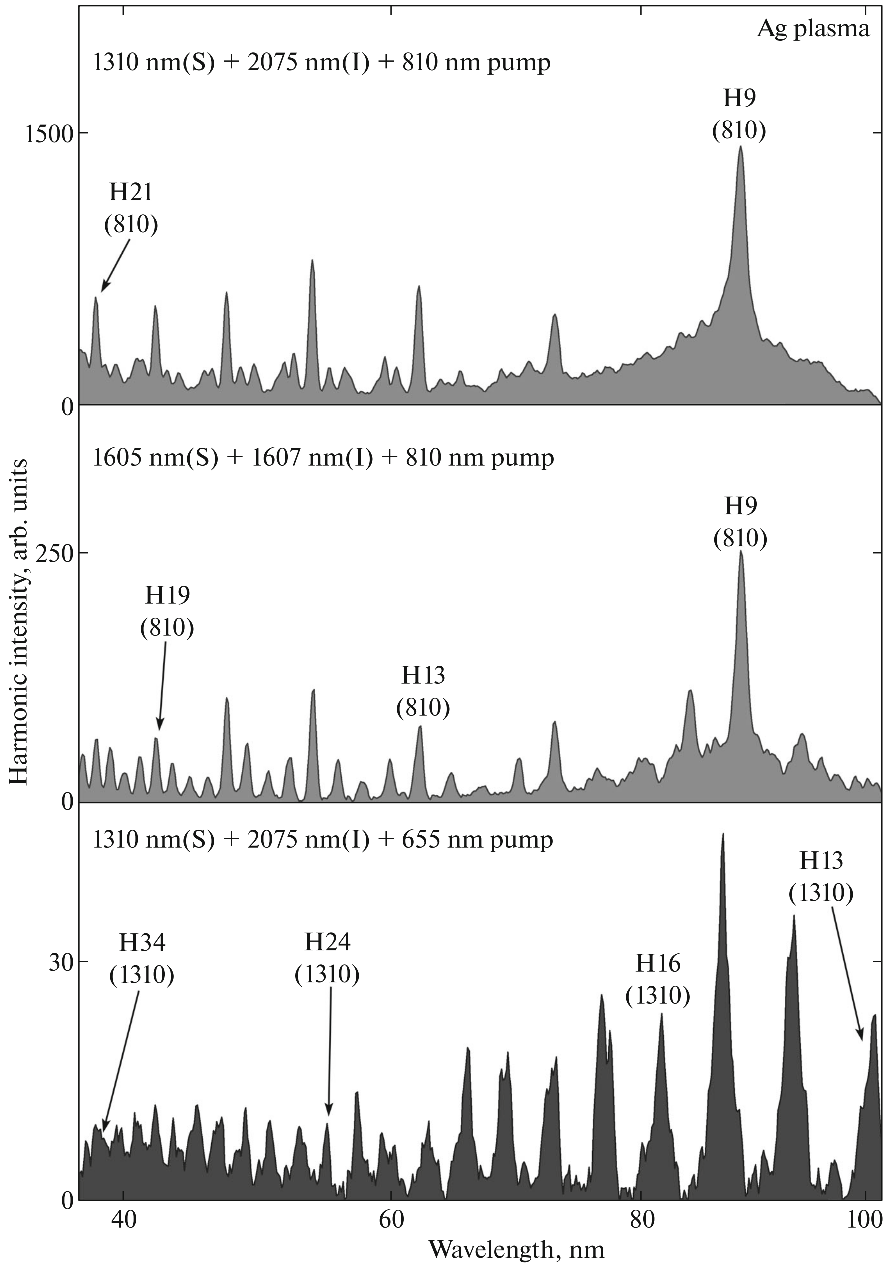

Fig. 3. Harmonic spectra from silver plasma in the case of joint influence of signal, idler, and 810 nm pulses (two upper panels). Bottom panel shows the harmonic spectrum during MIR (1310 nm) + H2 pump of silver plasma. In that case, no influence of idler pulse on generating harmonic spectrum was observed.

the plasma area compared with the idler pulses due to polarization- and wavelength-dependent dispersion in the BBO amplifier of OPA, as well as the dispersion of other optics, such as focusing lens and input MgF_2 window of vacuum chamber. The temporal tuning of 810 nm pump allowed generation of sum and difference frequency components during mixing with idler pumps. At worse conditions of the overlap of idler and 810 nm pulses, particularly at 40 fs separation between these pulses (Fig. 4a, bottom panel), only odd harmonics of 810 nm pump were generated.

The high-order sum and difference harmonic generation requires maximal spatial overlap of two pump beams in extended plasma as well. Once we tuned the focal planes of idler and 810 nm waves with regard to the extended plasma area the generating harmonic spectrum was changed from showing large amount of frequency components (see upper panel of Fig. 4b in the case of HHG in lead plasma when the idler beam was spatially overlapped with the 810 nm beam) towards the generation of mostly odd harmonics of

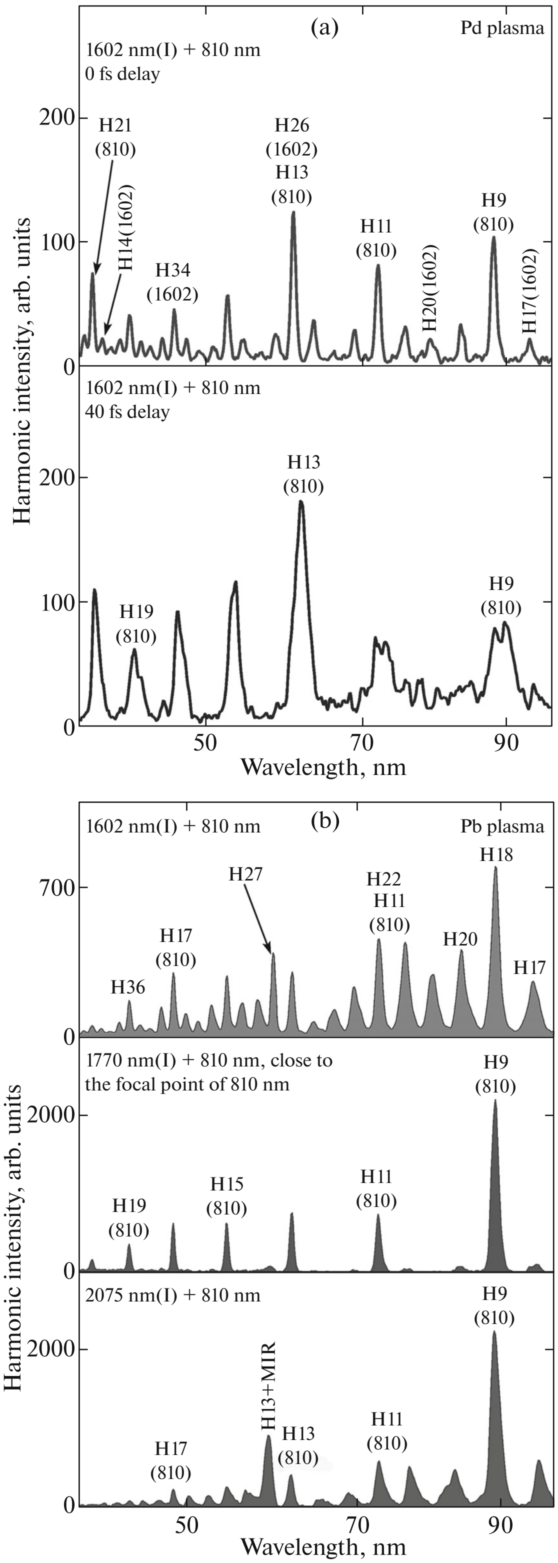

Fig. 4. (a) Application of idler pulses for the TCP of palladium plasma at minimal delay between pumps (upper panel) and 40 fs delay (bottom panel). (b) TCP (I + 810 nm) of lead plasma at maximum spatial overlap of MIR and 810 nm beams in LPP (upper panel) and at the conditions when the focal plane of MIR pulses was shifted out of the center of plasma area (middle panel). Bottom panel shows the harmonic spectrum using TCP (2075 nm + 810 nm) of Pb plasma at optimum spatio-temporal overlap of two pumps.

810 nm radiation at lesser spatial overlap of these two beams (middle panel).

Application of incommensurate two-color sources for HHG allows significantly increasing number of sum and difference frequencies of interacting waves, which in turn may lead to the observation of resonance-induced enhancement of single component of those multi-harmonic sources. This process mostly related with the ionic transitions of various materials possessing high oscillator strengths. From this point of view, the application of ablated materials with further use of the TCP comprising OPA and 810 nm pulses during propagation through the LPP may offer some advantages over MIR+H2 scheme, since the former method allows the formation of better conditions for resonance enhancement.

The example of such resonance-induced enhancement of sum component is shown in the bottom panel of Fig. 4b. One can see that H13 (810 nm) + MIR component ($\lambda = 59.8$ nm) was stronger compared with the H13, which is unusual assuming a significant difference of the energies of used pumps (0.4 and 3 mJ in the case of 2075 nm idler pulses and 810 nm radiation). Some strong ionic transitions of Pb in the 55–60 nm spectral region could be responsible for the enhancement of nearby frequency component. Similar enhancement is seen in the upper panel of Fig. 4b.

Comparison of TCP schemes shows a difference of relative intensities of the second (assistant) wave with regard to the original one. In the case of TCP using second-harmonic generation followed with the odd and even orders generation the ratio of second-harmonic and fundamental waves of Ti:sapphire lasers was in the range between 0.05 and 0.2. This ratio in the case of second-harmonic emission of MIR pulses was larger, but maintained at approximately same value (0.1–0.3). As it was mentioned, the second-harmonic photons were considered as the assistant field, which is sufficient to improve the ionization rate of emitters, as well as to amend the recombination cross-section of accelerated electrons. In the case described in present paper, the second wave (810 nm) was notably stronger compared with the signal waves from OPA. The 810 nm/MIR ratio of pulse energies was in the range between 3 and 10, thus one can rather consider MIR pulses as the assistant field to the strong 810 nm pulses.

These studies showed that efficiency of TCP induced high-order sum and difference harmonics did not depend on the relative polarizations of interacting waves, but rather on the spatio-temporal overlap and phase difference of MIR and 810 nm pulses. The comparative studies of two TCP schemes showed both advantages [stronger yield of the MIR-related components of generated XUV spectrum, absence of additional nonlinear medium (BBO) for formation of second field, and denser emission spectrum] and disadvantages (less harmonic cut-off) of the MIR + 810 nm pump compared with the MIR + H2 scheme. The former scheme based on two-color laser field synthesis [15] has previously been proven to be suitable for nonlinear pump/probe experiments in gases that demonstrated the capability of time-resolved attosecond dynamic processes using individual attosecond pulses generated in gaseous media [16, 24]. The implementation of similar technique for plasma harmonics generation may reveal new interesting features including the peculiar properties of individual emitters thus paving the way for laser ablation induced time-resolved HHG spectroscopy. Plasma harmonic approach allows using thousands solids as targets for ablation, contrary to gas harmonic approach, thus creating the conditions for material studies.

CONCLUSIONS

In conclusion, we have analyzed various laser-produced metal plasmas for high-order sum and difference harmonic generation using the mixing of tunable mid-infrared signal and idler pulses and 810 nm ultrashort pulses. We have shown that the two-color (MIR + 810 nm) pump allows significant broadening of the frequency combinations of generating pulses in the extreme ultraviolet range. The comparative studies of two TCP schemes have shown that both of them allow significant growth of harmonic yield while providing different additional frequency components in a whole generating spectrum. We have also shown that, regardless of the non-optimal spatio-temporal overlap of two pumps in the plasmas, the application of proposed technique allows the increase of the number of sum and difference frequency harmonics in the extreme ultraviolet region, which could be useful for observation of resonance induced enhancement of some of those harmonic components.

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