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Enhancement of high-order harmonic generation using a two-color pump in plasma plumes

R. A. Ganeev,[1,2,*] H. Singhal,[1] P. A. Naik,[1] I. A. Kulagin,[2] P. V. Redkin,[3] J. A. Chakera,[1] M. Tayyab,[1]
R. A. Khan,[1] and P. D. Gupta[1]
[1]*Raja Ramanna Centre for Advanced Technology, Indore 452013, India*
[2]*Institute of Electronics, Uzbekistan Academy of Sciences, Akademgorodok, 33, Dormon Yoli Street, Tashkent 100125, Uzbekistan*
[3]*Samarqand State University, Samarqand 703004, Uzbekistan*
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The results of enhanced high-order harmonic generation (HHG) in laser-produced plasmas using a two-color laser pump (98% of 800 nm and 2% of 400 nm) are presented. Even (up to the 38th order) and odd (up to the 45th order) harmonics of 800 nm radiation were generated in the case of a two-color orthogonal polarization pump of various plasmas. The comparison between HHGs using parallel and orthogonally polarized 400 and 800 nm radiations showed better conversion efficiency for the latter case. We have also used circularly polarized 800 nm pump for second-harmonic generation and HHG. We also present the results of the influence of the chirp and phase modulation of the fundamental radiation in laser plasma on the spectral properties of odd and even harmonics.

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I. INTRODUCTION

High-order harmonic generation (HHG) is a well-established technique for producing coherent radiation in the extreme ultraviolet (xuv) range. However, the relatively low HHG efficiency (at the level of 10^{-5} or less) still remains an obstacle for any practical application of this radiation. Various methods such as phase matching, quasi-phase-matching, resonance-induced enhancement of HHG efficiency, use of efficient nonlinear media, etc., have been used to increase the harmonic yield during the interaction of strong laser field with atoms and ions [1,2]. The search of new approaches for improvement of the HHG efficiency is an important goal of nonlinear optics. One such approach is to use a two-color pump for the enhancement of harmonics. The two-color pump, using the fundamental and second-harmonic (SH) fields, has become a practical way of harmonic enhancement in gas media [3–12]. It can generate odd harmonics, which are moderately stronger than those with the fundamental field alone. Moreover, it simultaneously generates even harmonics as strong as the odd ones, which leads to not only intensity enhancement but also energy enhancement of the high-order harmonics.

Experiments and theoretical studies on two-color HHG in gas jets and cells have been previously carried out for frequency ratios 2:1 [3–8] or even 3:1 [9] of the two fields, with the strengths of two fields either widely different or comparable. The experimental results have shown that the presence of the second field strongly modifies the harmonic spectrum. The enhancement of the HHG has been experimentally observed both for parallel and perpendicularly linearly polarized two-color fields [6,9,10], while two orders of magnitude enhancement have been predicted theoretically in this HHG configuration for parallel polarization [11,12]. By controlling the relative phase between the two frequency components, conversion efficiency as high as 5×10^{-5} was reported for the 38th harmonic at 21.6 nm [10]. Another interesting feature predicted in that case is the generation of a strong attosecond pulse train for orthogonally polarized two-color pump using the polarization gate schemes even for relatively long laser pulses [3,4,7,8].

Another method of HHG is the creation of specially prepared plasma plume as the nonlinear medium. The application of a two-color pump for the HHG in plasma plume can reveal some new features related with the ionic transitions of medium and specific relations between the phases of two pump waves. In this paper, we present the observation of enhanced harmonic generation in laser-produced plasma plumes using a two-color laser pump when the intensity of 400 nm wave was about 50 times weaker than the intensity of 800 nm wave. At these conditions, we achieved the enhanced even (up to the 38th order) and odd (up to 45th order) harmonic generations in the case of a two-color orthogonal polarization pump of various plasmas. A comparison between harmonic generations using parallel and orthogonally polarized 400 and 800 nm radiations showed better conversion efficiency for the latter case. We demonstrate the application of circularly polarized 800 nm pump for second-harmonic generation and HHG. We also present the results of the influence of the chirp and phase modulation of the fundamental radiation in laser plasma on the spectral properties of odd and even harmonics.

II. EXPERIMENTAL ARRANGEMENTS

Experiments were performed using a 10 TW laser, operated at 3 TW level. To create the plasma plume, a prepulse was split from the uncompressed Ti:sapphire laser (30 mJ, pulse duration $\tau=210$ ps, wavelength $\lambda=800$ nm, and pulse repetition rate of 10 Hz) and was focused on a target placed in a vacuum chamber by using a plano-convex lens (focal length $f=500$ mm) to create a plasma plume (see the inset of Fig. 1). The targets used in these studies were bulk Ag, C, In, and Cr slabs ($\sim5\times5$ mm^2), as well as fullerenes. The intensity of the prepulse on the target surface was varied be-

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[*] Electronic mail: rashid_ganeev@mail.ru

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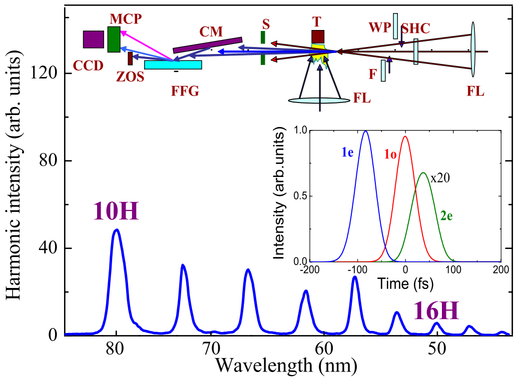

FIG. 1. (Color online) Harmonic spectrum obtained from the chromium plasma plume. Upper inset: Experimental scheme. FL, focusing lenses; SHC, second-harmonic crystal; WP, wave plate; F, filter; T, target; S, slit; CM, cylindrical mirror; FFG, flat-field grating; ZOS, zero-order beam stop; MCP, microchannel plate; and CCD, charge-coupled device. Bottom inset: The calculated temporal distributions of ordinary (solid curve) and extraordinary (dotted curve) fundamental and second-harmonic (dashed curve) pulses at the output of KDP.

tween 2×10^9 and 3×10^{10} W cm^{-2}. After some delay (varying in the range of 6–60 ns), the femtosecond pulse [referred to as the "main pulse," $E=45$ mJ, $\tau=48$ fs, $\lambda=800$ nm central wavelength, and 19 nm full width at half maximum (FWHM)] was focused on the plasma plume from the orthogonal direction using a plano-convex lens ($f=500$ mm). The experiments were performed with main pulse intensity of up to 7×10^{14} W cm^{-2}, above which the HHG efficiency decreased considerably due to various impeding processes in the laser plasma. The high-order harmonics were spectrally dispersed by a homemade xuv spectrograph with a flat-field grating (1200 lines/mm, Hitachi) and cylindrical focusing mirror. The focused xuv spectrum was detected by a microchannel plate and finally recorded using a charge-coupled device (CCD). Details of the experimental setup can be found elsewhere [13].

Our scheme for a two-color pump was extremely simplified compared with previous schemes used in the gas HHG experiments. For SH generation, a potassium dihydrogen phosphate (KDP) crystal (1 mm thick, type-I phase matched) was placed between the focusing lens and plasma plume, so that, after frequency up-conversion in the crystal, the emerging laser field consisted of both the SH and fundamental laser pulses. The intensity of fundamental radiation inside the SH crystal was maintained at such a level that no self-phase modulation (SPM), appreciable chirp, and white light generation were introduced on the residual radiation by the KDP crystal. The SH conversion efficiency at these conditions was measured to be ~2%. The polarizations of SH and fundamental fields were orthogonal. These conditions were used in most HHG experiments with plasma plumes. At these conditions, a sufficient spatial and temporal overlap of the two pulses was achieved. We also used a zero-order wave plate, which acts as a half-wave plate for the fundamental and a full-wave plate for the SH fields, to rotate the polarizations of both pumps in such a manner that, at appropriate angle of wave plate, they coincided with each other. A BG-39 filter was used for elimination of fundamental radiation for experiments with only SH pump.

III. RESULTS AND DISCUSSION

A. Comparison of harmonic generation at single- and two-color pump conditions

There are many differences between the targets used. Most important is the morphological state of C60 powder compared to the bulk targets (Cr, C, Ag, and In). They can also be distinguished from each other by their ionization potentials ($I_{\mathrm{In}}=5.79$ eV, $I_{\mathrm{Cr}}=6.77$ eV, $I_{\mathrm{Ag}}=7.58$ eV, $I_{\mathrm{C60}}=7.6$ eV, and $I_{\mathrm{C}}=11.26$ eV). The plasmas used in these experiments mostly consisted of the neutrals and singly ionized particles. The presence of doubly charged ions led to considerable increase in free electron concentration. At these conditions, various impeding processes (i.e., self-phase-modulation, self-defocusing, and phase mismatch) considerably decreased the HHG conversion efficiency.

The HHG in plasma plumes using single (800 nm) pump led to appearance of the conventional harmonic spectra consisting of only odd orders. The enhancement of single (13th) harmonic in the plateau region was observed in indium plasma, as reported previously in Ref. [14]. Another result is a relatively long plateau in harmonic distribution spectrum in the case of silver plasma. The plasma plumes of these two targets have previously proved to be the most efficient media where the highest HHG efficiency was achieved [15].

The main results of these studies are presented below. As is well known, a single-color driving field yields a spectrum of odd harmonics due to inversion symmetry. The use of a two-color field breaks the inversion symmetry and produces a spectrum containing both even and odd harmonics [16]. Insertion of SH crystal in the beam path after the focusing lens led to generation of the enhanced harmonic yield and appearance of even and odd harmonics with approximately equal intensity. It may be noted that, in some cases, the intensity of low-order even harmonics was stronger than that of the odd harmonics at the same spectral range [see the harmonic spectrum from the chromium plasma (Fig. 1)]. Introduction of the UV short pass filter after the SH crystal led to generation of the fifth, seventh, and, in some cases, ninth harmonics from the 400 nm field (i.e., generation of the 10th, 14th, and 18th harmonics of fundamental radiation), while the intensities of these harmonics were weaker compared to the case of a two-color pump.

It should be noted that at phase-matching conditions for the KDP (type I) the quasistatic interaction length $L_g=\tau/(u_{2e}^{-1}-u_{1o}^{-1})$ is about 0.3 mm at 800 nm. Here τ is the pulse duration and u_{1o} and u_{2e} are the group velocities of the ordinary fundamental and extraordinary second-harmonic pulses, respectively. This value is smaller than the crystal length used in the experiment ($l=1$ mm). At these conditions the process of second-harmonic generation is a transient one. In spite of the influence of group velocity mismatch and walk-off of fundamental and second-harmonic radiations in KDP, the calculations show that the ordinary 800 nm pulse and extraordinary 400 nm pulse have a sufficient overlapping area (see the inset of Fig. 1). In this figure the pulse profiles

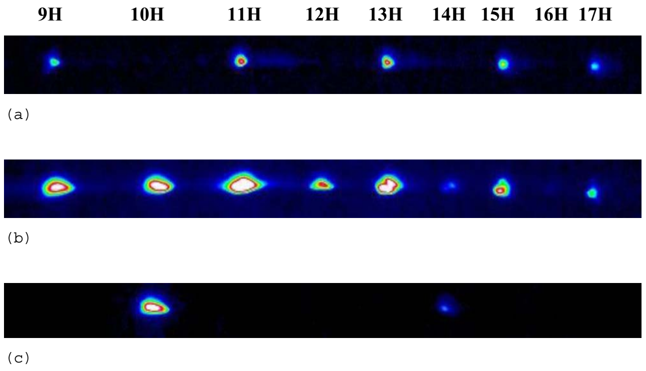

FIG. 2. (Color online) CCD images of the harmonic spectra generated in C_{60} plasma in the cases of (a) single-color fundamental pump (800 nm), (b) two-color pump (800 nm+400 nm), and (c) single-color SH pump (400 nm). The data were collected under similar experimental conditions.

of fundamental (solid curve) and second-harmonic (dashed curve) radiations are presented at the output of 1-mm-thick KDP taking into account the 2% conversion efficiency. The input temporal and spatial intensity distributions are assumed to be the Gaussian ones. At the crystal output the maximum intensity of harmonic radiation is ~14 times less than that of fundamental one.

The SH crystal was inserted in the experimental scheme at appropriate position after the focusing lens, so that no impeding processes (such as SPM, chirp, white light generation, etc.) were observed after propagation of the laser radiation through the crystal. The influence of walk-off effect and group velocity dispersion in KDP did not lead to any significant mismatch of two fields within the nonlinear medium (0.8-mm-long and 0.3-mm-thick laser-produced plasma plume). At these conditions, a considerable enhancement of HHG efficiency compared with single-color pump was observed, together with the simultaneous appearance of even and odd harmonics. Figure 2 shows the CCD images of harmonic spectra from fullerene-contained laser plasma in the cases of single- [(a) 800 nm and (c) 400 nm] and two-color [(b) 800 nm+400 nm] pumps. The enhancement factor for the odd harmonics at the plateau range varied between three times and eight times depending on the harmonic order. In the experiments with single SH pump, only few harmonics were observed [Fig. 2(c)], which was due to small conversion efficiency in the SH wave and weak pump of laser-produced plasma. Note that all these experiments were carried out at conditions when the variations in harmonic spectra were achieved by insertion of the SH crystal, without any additional improvements of the experimental parameters.

B. Influence of chirp and polarization of two-color pump radiation on the harmonic generation efficiency

We now present some specific results of these studies in different plasmas using variable experimental conditions (i.e., chirp of the fundamental radiation, polarization of the two pumps, initial polarization of the fundamental radiation, etc.). It is observed that a dramatic variation in the harmonic spectrum can be achieved despite considerable difference between the two pump intensities (50:1) and without any precise temporal and spatial matching of the two pumps. The results also show that, by appropriate chirping of the 800 nm pump, one can achieve a considerable variation in the odd and even harmonic spectra. Finally, we demonstrate the use of circularly polarized fundamental radiation for generation of the odd and even harmonics.

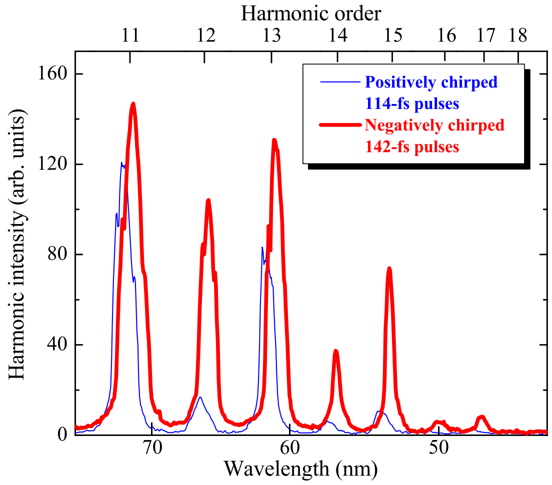

FIG. 3. (Color online) Harmonic spectra obtained from carbon plasma plume using the two-color pump scheme in the case of negatively chirped (thick line) and positively chirped (thin line) pulses of fundamental radiation.

The chirp of the main pulse was varied by adjusting the distance between the two gratings of pulse compressor. Reducing the separation between the gratings from the chirp-free conditions resulted in positively chirped pulses, and an increase in the distance between the gratings provided negatively chirped pulses. At negative chirp, the pulse contains short-wavelength components of the laser spectrum in the leading part of the pulse and vice versa. Preferential conversion of the leading part of spectrum during the nonlinear conversion in laser plasma leads to frequency shift of the harmonics. For example, in the case of single-color positively chirped pulses, redshifted harmonics were observed [14]. In the case of a two-color pump (with chirped fundamental radiation) also, tuning of both odd and even harmonics is observed. It may be noted that the HHG efficiency was better in the case of negatively chirped pulses compared to that with positively chirped radiation (Fig. 3).

Variation in the odd or even harmonic spectrum was also achieved by modulation of the fundamental spectrum. Laser spectrum was modulated during propagation of the strong 800 nm radiation through the plasma. It was clearly seen that the laser spectrum after passage through the plasma shows two peaks (800 and 780 nm), with the overall FWHM of the laser beam increasing from 19 to ~30 nm, instead of the single peak centered near 800 nm in the case of initial femtosecond pulse. It was observed that high-order odd harmonics generated in the two-color scheme by this spectrally modulated pulse are broadened as well.

The harmonic generation by using two-color laser fields with parallel linear polarizations was investigated and compared with the case of orthogonally polarized 800 and 400

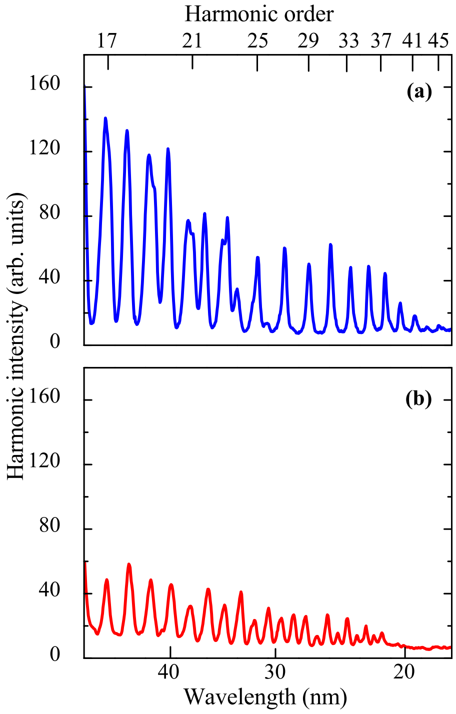

FIG. 4. (Color online) Harmonic spectra obtained in the Ag plasma using the two-color pump with parallel polarization in the cases of (a) negatively chirped 270 fs pulses and (b) positively chirped 324 fs pulses. The spectra were obtained at identical experimental conditions.

nm pulses. For HHG in an orthogonally polarized two-color field, the relative phase between the fundamental and second-harmonic components is a crucial parameter. After optimization of orthogonally polarized two-color field-induced odd and even harmonic generations, a wave plate was inserted after the SH crystal at appropriate angle to achieve a parallel polarization of the two pumps. The overall HHG efficiency in the case of parallel polarization two-color pump scheme was diminished compared to the orthogonal polarization case. This was caused mainly due to the dispersive properties of wave plate leading to the increase in the temporal mismatch between the pump waves in the plasma volume. Nevertheless, the even harmonics were clearly observed even at these unfavorable conditions. Figure 4 shows the harmonic spectra obtained from the Ag plasma for positively and negatively chirped pulses. While the application of negatively chirped pulses led (as in the case presented in Fig. 3) to generation of stronger harmonics compared with positively chirped pulses, the latter generated extended even harmonics (up to the 38th order).

The comparison of harmonic spectra for single- and two-color fields showed different ranges of efficient harmonic generation caused by different phase-matching conditions for these two cases. The single (800 nm) field-induced harmonics from silver plasma proved to create the efficient conditions for 40th–50th orders when the harmonics in the range of 16–20 nm demonstrated a considerable enhancement compared to the lower-order harmonics (see also [17] where this phenomenon was analyzed and attributed to the propagation effects). The application of a two-color pump in plasma HHG led to dramatic change in the phase-matching conditions. In this case, the low-order harmonics at the beginning of the plateau became stronger compared to the higher order ones (Fig. 5). The optimal phase-matching conditions were shifted toward 65–80 nm. It may be noted that the intensity of low-order harmonics for single-pump scheme [Fig. 5(a)] was three to five times less than the intensity of the same harmonics for both orthogonal and parallel polarization two-color schemes [Figs. 5(b) and 5(c)].

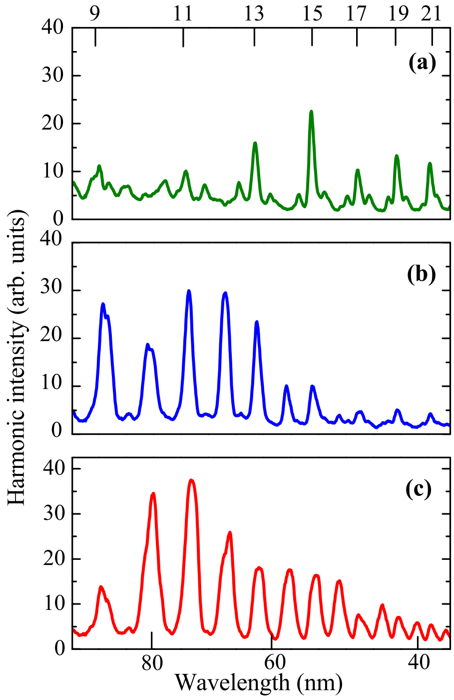

FIG. 5. (Color online) Low-order harmonic spectra from Ag plasma at (a) single-color (800 nm) pump, (b) parallel polarized two-color pump, and (c) orthogonally polarized two-color pump. Side lobes of the odd harmonics on (a) correspond to the second-order diffraction lines of strong high-order harmonics.

Forming resonance conditions to enhance the nonlinear optical response of the medium may be an alternative to the phase-matching technique. The role of atomic resonances in increasing the laser radiation conversion efficiency was actively discussed in the framework of the perturbation theory at the early stages of the study of low-order harmonic generation [18]. While theoretical estimates testified the possibility of an efficient enhancement of individual harmonics and groups of harmonics, experimental works revealed the difficulties encountered in HHG in gases. Therefore, the use of plasma medium could largely facilitate the solution to the problem of resonance harmonic enhancement. Examining a large group of potential targets allowed identifying some of them as suited for demonstrating this process [15]. 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.

As it was mentioned above, a typical harmonic spectrum generated in indium plasmas using single-color (800 nm) pump showed that the 13th harmonic intensity is much stronger than the intensities of the neighboring harmonics. Past

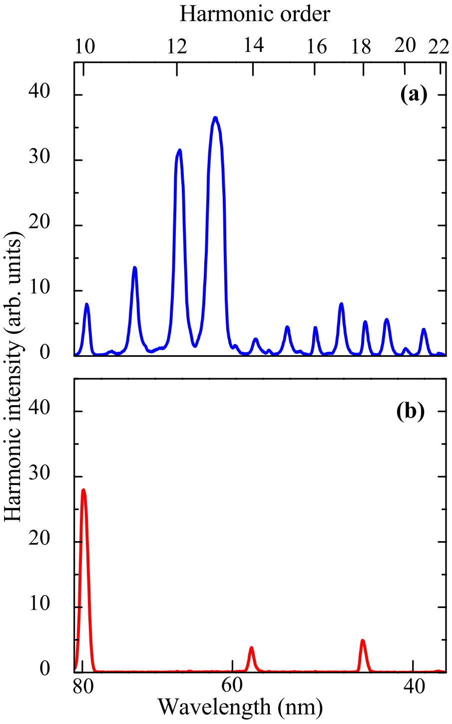

FIG. 6. (Color online) Harmonic spectra from indium plasma for (a) two-color (800+400 nm) orthogonally polarized pump and (b) single-color (400 nm) pump. Note the enhancement in the intensity of the 12th harmonic of indium.

investigations of indium plasmas showed that the bulk of emission in the 40–65 nm wavelength range was due to radiative transitions to the ground state ($4d^{10}5s^2\ {}^1S_0$) and the low-lying excited state ($4d^{10}5s5p$) [19]. These investigations also revealed a strong 62.1 nm line (19.92 eV) corresponding to the transition $4d^{10}5s^2\ {}^1S_0 \rightarrow 4d^9 5s^2 5p(^2D)\ {}^1P_1$. The oscillator strength of this transition was equal to 1.11, which exceeded the corresponding parameter of other transitions in this range by more than a factor of 12. This transition may be tuned to resonance with the 13th harmonic of 800 nm radiation ($\lambda = 61.5$ nm, $E_{\text{ph}} = 20.2$ eV) due to the Stark effect. Application of a two-color pump revealed that, alongside the strong 13th harmonic, a very intense 12th harmonic appears in the plateau range (Fig. 6). One can assume that the enhancement of this harmonic was caused by the closeness with the same ionic transition, which induced the strong 13th harmonic generation. Thus, the use of a two-color pump allowed the generation of a strong resonantly enhanced harmonic, which by a factor of 3–6 exceeded those of neighboring even harmonics.

For each of results presented in Figs. 4–6, the intensity scales are similar. We have compared the relative intensities of harmonics for specific cases [for example, in Fig. 5, we compare the intensities of low-order harmonic spectra from Ag plasma at single-color (800 nm) pump, parallel polarized two-color pump, and orthogonally polarized two-color pump on same intensity scales]. The same can be said about other figures.

Finally, we report the observation of harmonic generation using two-color scheme in the case of circularly polarized 800 nm pump. A quarter-wave plate was inserted in front of the focusing lens to change the polarization of the main laser beam interacting with the plasma plume. In the case of single-color (800 nm) pump, minor deviations from the linear polarization resulted in a substantial lowering of the harmonic intensity, which is typical for HHG in an isotropic medium. In this case, the use of circularly polarized radiation made the harmonics vanish completely, while the plasma radiation spectrum remained unaltered.

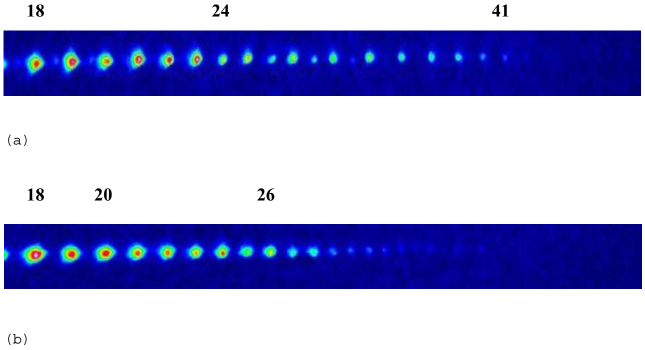

FIG. 7. (Color online) CCD images of the harmonic spectra obtained from the silver plasma using (a) a quarter-wave plate in front of the focusing lens and two-color pump and (b) orthogonally polarized two-color pump.

A completely different pattern was observed when the 800 nm radiation was made circularly polarized before it was incident on the SH crystal [Fig. 7(a)]. In this case, both, the even (up to the 30th order) and odd (up to the 41st order) harmonics were observed. For comparison, Fig. 7(b) shows the harmonic spectrum in identical conditions but without the quarter-wave plate when the polarization of 800 nm radiation was linear. One can see the equal intensities of the odd and even harmonics in the case of orthogonally polarized 800 and 400 nm pumps.

Observation of better harmonics in the case of circularly polarized incident laser light can be explained as follows: circularly polarized light is composed of two orthogonal linear polarizations differing in phase by a quarter-wave. Of these, the component which is ordinary ray for the KDP crystal (type-I phase matched) gets converted to SH and the other orthogonal component, which becomes extraordinary for the KDP crystal, goes through the crystal unconverted (no phase matching). As the refractive indices and group velocities for the ordinary and extraordinary components (of the fundamental) in the KDP crystal are quite different, the two components get separated in time and do not give rise to circularly polarized fundamental but act as independent radiation pulses. So we have a case of SH (extraordinary) and fundamental (ordinary) orthogonal to each other, with the remaining fundamental field (unconverted extraordinary) producing harmonics independently (as it is temporally shifted from the other two beams).

The inset of Fig. 1 shows the temporal distribution of extraordinary fundamental radiation at the output of KDP. In the calculations, the initial intensity of extraordinary and ordinary fundamental pulses was assumed to be equal with each other. From this figure one can see that the extraordinary and ordinary fundamental pulses have a rather small overlapping area after propagation of KDP. As seen earlier in Fig. 5, in the orthogonal two-color field case, one gets more intense harmonics than the single-color case. So having these two cases simultaneously (two-color orthogonal and single color), a higher production of odd-even harmonics is expected, as seen in Fig. 7.

C. Simulation of high-order harmonic generation using a two-color pump of various laser plumes

Below we briefly address some observed features of two-color field-induced laser plasma HHG. According to [10], the tunneling ionization rate, calculated using the Ammosov-Delone-Krainov formula [20], for the two-color field is much larger than that for the fundamental field, i.e., the electron wave packet at the time of ionization is significantly denser. Consequently, orthogonally polarized two-color field can generate harmonics much more strongly than the fundamental field alone. As it was underlined in Ref. [10], strong harmonic generation was possible due to the formation of a quasilinear field, the selection of short quantum path component, which has denser electron wave packet, and high ionization rate. Longer time of flight leads to more divergent harmonics in the two-color pump case, taking into consideration only the short trajectories, because the long ones will not show up in the measurements in a stable way. With suitable control of the relative phase between the fundamental and second-harmonic fields, this particular field significantly enhances the short-path contribution while diminishing other electron paths, resulting in a clean high-harmonic spectrum as well as a strong and regular attosecond pulse train.

The free electron state in the middle of the process offers an opportunity to control electron paths because the motion of a free electron can be easily steered by changing the shape of the applied laser field. Since an elliptically polarized field reduces the electron recombination significantly, the simplest way of changing the shape of the laser field is to add another frequency component whose intensity is comparable to the existing one. By adjusting the intensity, frequency, phase, and polarization of the new component, one can shape the laser field and select an electron path with favorable properties.

Here we present the results of simulation of high-order harmonic generation using a two-color pump of various laser plumes. All computations were performed on the basis of time-dependent density functional theory (TDDFT) [21] with the aid of real-space, real-time OCTOPUS code [22,23]. In the TDDFT approach, the many-body time-dependent wave function $\varphi(r,t)$ is replaced by the time-dependent density $n(r,t)$, which is a simple function of the three-dimensional vector $\mathbf{r}$. $n(r,t)$ is obtained with the help of a fictitious system of noninteracting electrons by solving the time-dependent Kohn-Sham equations,

$$i\frac{\partial}{\partial t}\varphi_i(r,t)=\left[-\frac{\nabla^2}{2}+v_{KS}(r,t)\right]\varphi_i(r,t). \tag{1}$$

These are one-particle equations, so most many-electron problems can be solved with only a linear increase in computational effort with the number of electrons. The density of the interacting system is obtained from the time-dependent Kohn-Sham orbitals,

$$n(r,t)=\sum_i^{occ}|\varphi_i(r,t)|^2, \tag{2}$$

$$v_{KS}(r,t)=v_{ext}(r,t)+v_{Hartree}(r,t)+v_{xc}(r,t). \tag{3}$$

Here $v_{ext}(r,t)$ is the external potential (i.e., laser field), $u_{xc}(r,t)$ is the exchange correlation potential, and $v_{Hartree}(r,t)$ accounts for the classical electrostatic interaction between the electrons,

$$v_{Hartree}(r,t)=\int d^3r'\frac{n(r,t)}{|r-r'|}. \tag{4}$$

The systems under investigation were simplified as follows. Silver atoms and ions were represented by corresponding Hartwigsen-Goedecker-Hutter pseudopotentials [24]. Double-grid technique was used to increase the precision of the application of the pseudopotentials. For all computations, Slater exchange and Perdew and Zunger correlation functionals [25] were used. All systems were studied within the so-called adiabatic local density approximation, assuming that the potential is the time-independent exchange correlation potential.

For taking into account the chirp-induced spectral broadening, the laser pulses were chosen as orthogonally linear polarized continuous waves with frequencies of 0.057 and 0.114 a.u. for the fundamental pulse and its SH, respectively, multiplied by a Gaussian temporal envelope. Peak electric field amplitudes were set to 0.1166 a.u. (4.8×10^{14} W cm^{-2}) and 0.0165 a.u. (9.5×10^{12} W cm^{-2}) for the fundamental pulse and its SH.

The time step was chosen as 0.124 a.u. (0.003 fs) and the time propagation was performed for the first 48 fs. In order not to spoil the symmetry of investigated systems, a spherical integration box was used. Probability density reaching the radial boundary of the integration box at 100 a.u. was replaced by an imaginary potential using the mask technique similar to that in Ref. [26].

High-order harmonic power spectra $P_L(\omega)$ were obtained using the relation

$$P_L(\omega)=\omega^4\frac{Q^2}{4\pi c^3}|d_L(\omega)^2|. \tag{5}$$

Here $d_L(\omega)$ is the discrete Fourier transform of dipole expectation value. To minimize the artifacts of discrete Fourier transform itself induced by sampling finiteness, intermediate values between time steps were added using linear interpolation. The results are normalized to the intensity of the first harmonic and presented on a logarithmic scale.

Almost all high-order harmonic spectra were found in reasonable qualitative agreement with the experimental results, especially in cutoff positions. This indicates properly chosen ionization potentials of the simulated systems and the

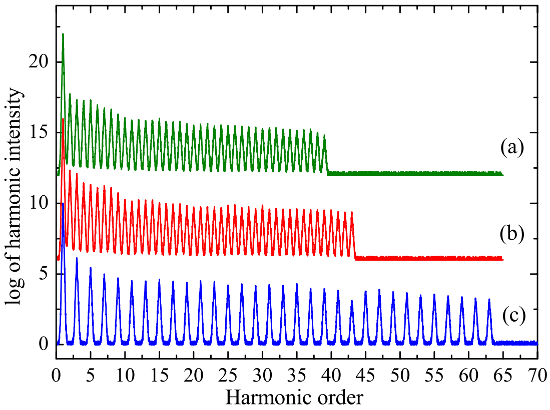

FIG. 8. (Color online) Theoretical results of the HHG in silver plume using various pumps. $\lambda_0=800$ nm. (a) Two-color pump with parallel polarization, (b) two-color pump with orthogonal polarization, and (c) single color (800 nm) pump. Spectra (a) and (b) are shifted upward by 12 and 6 orders of magnitude, respectively, on a logarithmic scale for better visibility.

average laser field intensity. One can see from Fig. 8 that, in the case of a two-color pump and silver ions, the highest harmonic orders are truncated. This effect was not observed in case of lower harmonic orders. This validates the short-path selection hypothesis introduced earlier in this paper. However, it is more likely that adding a collinear SH field spoils the return conditions for electrons. That can be the reason why the cutoff in the case of silver ions and two-color collinear field is lower than for corresponding single field pump.

The comparison of simulated results and experimental data shows a good agreement in the cutoffs in the cases of single- and two-color pumps. In particular, the experimentally observed cutoff for 800 nm pump for the HHG in silver plasma was in the harmonic range of 60s (which is similar to previously reported results), while the cutoffs for a two-color pump using different polarizations were in the harmonic range of 40s (Fig. 4). The simulations show similar cutoffs for these two cases (Fig. 8). The same comparison was made for carbon and fullerene plasmas. In these cases also, we obtained a good agreement between the harmonic cutoffs found in experiment and the ones predicted by the theory.

At the same time, the enhancement of harmonics simulated in the case of a two-color pump was insignificant. This can be seen from the comparison of the pump intensities (e.g., "first" harmonics on the harmonic distribution presented in Fig. 8) and harmonic intensities at the plateau range for two- and single-pump conditions. Further studies are needed for understanding the difference in conversion efficiency for these two schemes of harmonic generation at the conditions of very small intensity of the second field.

IV. CONCLUSIONS

The results presented in this paper show the enhancement of harmonic intensity and energy using a two-color pump of the laser-produced plasma. Some observations (i.e., influence of the polarization, chirp, and phase modulation on the odd or even harmonic yield) point out the effectiveness of this scheme in plasma HHG. The important peculiarity in this case is the appearance of resonantly enhanced 12th harmonic in the In plasma plume. This approach allows for the search of appropriate plasma plumes where the resonance enhancement can be realized for the even harmonics of Ti:sapphire laser, analogously to previously reported observations of resonantly enhanced odd harmonics [17].

Another noticeable difference compared with two-color gas HHG is the cutoff order (Fig. 4). The one-color field gives a high cutoff order (up to the 63rd harmonic) in Ag plasma, compared to the two-color field, which yields a lower cutoff order (~43rd harmonic). Since the cutoff order is inversely proportional to the square of the laser frequency, an increase in the intensity of the second-harmonic field at the expense of decreasing the fundamental field intensity leads to a decrease in the cutoff order. In contrast to the one-color field that drives an electron both in long and short paths, the two-color field generates harmonics through only short electron paths. These short paths do not have sufficient time for acquiring large kinetic energy required for high cutoff orders.

In this paper, first observation of large increase in the harmonic conversion efficiency is reported from plasmas irradiated by an intense two-color femtosecond laser pulse, in which the fundamental field and its weak second harmonic (energy ratio of 50:1) are linearly polarized and orthogonal to each other. In contrast to usual high-harmonic generation with linearly polarized fundamental field alone, a very strong HHG spectrum, consisting of both odd and even orders of harmonics, was generated in the orthogonally polarized two-color laser field even without proper selection of the relative phase between the fundamental and SH fields. Very efficient HHG in a two-color laser field was achieved using various laser-induced plasmas. With optimization of the laser parameters and target conditions, strong harmonics were produced at even orders. Stronger harmonics were obtained when the two-color laser field with orthogonal polarization was applied compared with parallel configuration. In this case also, all integer-order harmonics in the low energy part of the plateau were generated. Stronger odd and even harmonics in two-color scheme were observed for negatively chirped 800 nm pump compared with positively chirped pulses. All the above features were observed using extremely simple configuration when the harmonic enhancement was achieved by insertion of a KDP (1-mm-thick) crystal between the focusing lens and plasma plume. HHG was observed even in the two-color pump scheme when the initial radiation was circularly polarized. In our studies no attempt was made to confirm the circular polarization of the harmonics, but the harmonic intensities were found to be high and the existence of a kind of plateau was established, which is terminated by a cutoff, analogously to the results of HHG with gas medium reported in [27]. We have performed theoretical investigations of two-color HHG in the experimentally studied systems by means of TDDFT with symmetry conservation. Theoretical data in general match the corresponding experimental results.

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[1] E. A. Gibson, A. Paul, N. Wagner, R. Tobey, D. Gaudiosi, S. Baskus, I. P. Christov, A. Aquila, E. M. Gullikson, D. T. Attwood, M. M. Murnane, and H. C. Kapteyn, Science **302**, 95 (2003).

[2] S. Kazamias, D. Douillet, F. Weihe, C. Valentin, A. Rousse, S. Sebban, G. Grillon, F. Auge, D. Hulin, and P. Balcou, Phys. Rev. Lett. **90**, 193901 (2003).

[3] J. Mauritsson, P. Johnsson, E. Gustafsson, A. L'Huillier, K. J. Schafer, and M. B. Gaarde, Phys. Rev. Lett. **97**, 013001 (2006).

[4] D. Charalambidis, P. Tzallas, E. P. Benis, E. Skantzakis, G. Maravelias, L. A. A. Nikolopoulos, A. P. Conde, and G. D. Tsakiris, New J. Phys. **10**, 025018 (2008).

[5] X.-S. Liu and N.-N. Li, J. Phys. B **41**, 015602 (2008).

[6] I. J. Kim, C. M. Kim, H. T. Kim, G. H. Lee, Y. S. Lee, J. Y. Park, D. J. Cho, and C. H. Nam, Phys. Rev. Lett. **94**, 243901 (2005).

[7] T. Pfeifer, L. Gallmann, M. J. Abel, D. M. Neumark, and S. R. Leone, Opt. Lett. **31**, 975 (2006).

[8] Y. Yu, X. Song, Y. Fu, R. Li, Y. Cheng, and Z. Xu, Opt. Express **16**, 686 (2008).

[9] S. Watanabe, K. Kondo, Y. Nabekawa, A. Sagisaka, and Y. Kobayashi, Phys. Rev. Lett. **73**, 2692 (1994).

[10] I. J. Kim, G. H. Lee, S. B. Park, Y. S. Lee, T. K. Kim, C. H. Nam, T. Mocek, and K. Jakubczak, Appl. Phys. Lett. **92**, 021125 (2008).

[11] D. A. Telnov, J. Wang, and Shih-I Chu, Phys. Rev. A **52**, 3988 (1995).

[12] E. Cormier and M. Lewenstein, Eur. Phys. J. D **12**, 227 (2000).

[13] H. Singhal, R. A. Ganeev, P. A. Naik, V. Arora, U. Chakravarty, and P. D. Gupta, J. Appl. Phys. **103**, 013107 (2008).

[14] R. A. Ganeev, H. Singhal, P. A. Naik, V. Arora, U. Chakravarty, J. A. Chakera, R. A. Khan, I. A. Kulagin, P. V. Redkin, M. Raghuramaiah, and P. D. Gupta, Phys. Rev. A **74**, 063824 (2006).

[15] R. A. Ganeev, J. Phys. B **40**, R213 (2007).

[16] M. D. Perry and J. K. Crane, Phys. Rev. A **48**, R4051 (1993).

[17] R. A. Ganeev, Phys. Usp. **52**, 55 (2009).

[18] J. F. Reintjes, *Nonlinear Optical Parametric Processes in Liquids and Gases* (Academic Press, Orlando, 1984).

[19] G. Duffy and P. Dunne, J. Phys. B **34**, L173 (2001).

[20] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).

[21] E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).

[22] M. A. L. Marques, A. Castro, G. F. Bertsch, and A. Rubio, Comput. Phys. Commun. **151**, 60 (2003).

[23] A. Castro, H. Appel, M. Oliveira, C. A. Rozzi, X. Andrade, F. Lorenzen, M. A. L. Marques, E. K. U. Gross, and A. Rubio, Phys. Status Solidi B **243**, 2465 (2006).

[24] C. Hartwigsen, S. Goedecker, and J. Hutter, Phys. Rev. B **58**, 3641 (1998).

[25] J. P. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981).

[26] M. Tafipolsky and R. Schmid, J. Chem. Phys. **124**, 174102 (2006).

[27] H. Eichmann, A. Egbert, S. Nolte, C. Momma, B. Wellegehausen, W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **51**, R3414 (1995).