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Dependence of high-order harmonic intensity on the length of preformed plasma plumes

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An experimental study has been carried out on high-order harmonic generation from the interaction of 45 fs Ti:sapphire laser pulses with low-excited silver plasma plumes. The dependence of high-order harmonic intensity (I_H) on the plasma plume length ($L_{\rm med}$) shows slower than quadratic scaling $I_H \sim L_{\rm med}^p$ with "p"≈0.9, 0.8, and 0.7 for 21st, 33rd, and 41st harmonics, respectively. The harmonic intensity is observed to decrease with increasing harmonic order, and this decrease is faster for longer plasma plumes. The results are explained from physical considerations of phase mismatch between the driving laser pulse and the harmonic radiation during propagation, and reabsorption of the harmonic radiation in the plasma plume.

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I. INTRODUCTION

High-order harmonic generation (HHG) from the interaction of ultrashort laser pulses with underdense gaseous media is an area of active interest [1–3] due to its potential application as a coherent extreme ultraviolet (xuv) source [4,5], and for the generation of attosecond pulses [6–8]. Gas jets [9–11], static gas cells [12–14], and gas filled hollow core fibers [4,5] have been commonly used as the nonlinear medium for harmonic conversion. An important aim of such investigations is to increase the harmonic intensity for utilization of the harmonic radiation for practical applications. Since HHG is a nonlinear process, use of higher intensity may be utilized to increase the intensity of harmonic radiation. However, the use of laser intensity beyond a limit (saturation intensity) [11] results in the reduction of harmonic intensities. The ionization of the gas atoms (i.e., the harmonic emitters), and the relativistic drift of the electrons at high laser intensities, are the main reasons for the reduction in harmonic intensity [15]. Various other techniques have been explored to increase the intensity of the high-order harmonics. One such method is to enhance the harmonic intensity through the atomic (ionic) resonances [16–18].

HHG can be understood in terms of emission of high energy photons through nonlinear interaction of laser radiation with gas atoms [19,20]. It has been commonly explained from a three-step model [19]. When an atom is placed in a laser field, its potential gets distorted. A bound electron can tunnel out from this lowered potential barrier. The tunneled electron will then accelerate in the laser field. Depending on the laser phase at the time of tunneling, the free electron may or may not return to the parent atom. The one which returns may recombine with the atom and emit a photon. Depending on the laser phase at the instant of ionization, the energy of the recombining electron can be up to 3.17 times the quiver (or ponderomotive) energy (U_p). Hence the energy of the recombination photon can take on a substantial range above the ionization energy [ionzation potential (I_P)+kinetic energy of the electron]. However, due to the periodic occurrence of this process, and due to the inversion symmetry of the atomic potential, only photons of odd harmonic frequency will survive and emerge out of the interaction zone. In harmonics generated from gaseous targets, the intensity of harmonics is observed to decrease rapidly for the first few orders followed by a long intensity plateau of harmonic orders having a well defined cutoff [1,2,11,20]. The observed cutoff energy is the same as that given by the three-step model discussed above, as [19]

$$h\nu_{\rm max} = I_P + 3.17U_p. \tag{1}$$

The harmonic radiation is coherent since the process of HHG is essentially phase locked with the driving laser field. Harmonic intensity builds up during its copropagation with the laser pulse in the medium. However, any phase mismatch between the laser field and the harmonic radiation due to dispersion in the medium may decrease the conversion efficiency [21]. Hence, it is important to optimize medium parameters to achieve phase-matching conditions as much as possible. For instance, in experiments performed using gas jets, a better phase matching is achieved by positioning the gas jet *before* the beam waist of the focused laser beam [21], rather than *at* the beam waist. To achieve higher conversion efficiency, the length of the medium can also be increased, provided the phase mismatch between the laser field and the harmonic radiation can be kept low. The maximum possible medium length may also be limited by the laser focusing conditions. In gas cells, large interaction lengths (of ~6 cm) [12] have been achieved by self-guiding of the laser beam. The use of gas filled hollow core fibers is another method to increase the interaction length [4,5]. However, it may be pointed out that, apart from a growing phase mismatch with an increase in medium length, the reabsorption of the harmonic radiation may also limit the conversion efficiency [13]. Kazamias *et al.* [22] have reported conversion efficiency of $\sim 1-3\times10^{-5}$ for harmonics in the plateau region under phase matching conditions in semi-infinite gas cells. Recently, weakly ionized, low excited underdense plasma plumes of solids have been used for HHG [3,23,24]. The HHG in an intensity plateau region up to 61st harmonic order was observed from these targets [23]. The plasma plumes were generated by focusing a low intensity laser prepulse ($I\sim10^{10}$ W/cm^2) on a solid target surface, before the main femtosecond pulse, which on interaction with these pre-

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formed plasma plumes generated high-order harmonics. These low-excited plasma plumes mainly consist of neutral and singly charged particles, and behave like gaseous media for HHG [16,23]. These targets show conversion efficiencies comparable to the gas targets. Ganeev *et al.* [25] have reported conversion efficiency of $\sim 8\times 10^{-6}$ for harmonics in the plateau region in preformed silver plasma plumes. However, their use is especially promising for the resonance enhancement of the intensity of particular harmonic orders [16–18]. The use of resonance enhancement can increase the intensity of particular harmonics by 1–2 orders of magnitude. For example, in the case of an indium plasma plume, the intensity of 13th order harmonic is enhanced by a factor of ~ 200 compared to the neighboring harmonics in the plateau region [16]. Recently, a theoretical explanation has been proposed for the resonance enhancement of a particular harmonic order or a bunch of harmonic orders [17]. Accordingly, if the transition frequency ($\Delta\omega$) of an atomic or ionic transition is matched with a certain harmonic wavelength, then that particular harmonic or collection of harmonics around it gets enhanced. Due to the availability of a large number of target materials, the probability of the overlapping of these transitions with harmonics increases considerably. Moreover, the partial tunability of the harmonic wavelengths through the chirp control of the laser has been utilized in the tuning of harmonics into resonance for their intensity enhancement [26].

In the case of preformed plasma plumes, the intensity of HHG has been increased by proper optimization of various parameters of laser and plasma plume, such as the intensity of the prepulse [23], delay between the prepulse and the main pulse, intensity of the main pulse [16], and focal position of the laser in the plasma plume [27]. Further, the intensity of HHG in plasma plumes has been enhanced by the use of structured targets for plasma formation [3]. As has been done for the gaseous media, harmonic intensity from the plasma plumes may also be increased by increasing the plume length. However, no such study for plasma plumes has been reported. Such an investigation is desired to gain a better understanding of the various factors affecting the phase-matching conditions during propagation through the plasma medium, and in optimizing the medium parameters to achieve high conversion efficiency.

In this paper, we present an experimental study on the variation of harmonic intensity with the length of the plasma plume. Silver is used as the plasma medium as it does not have any known atomic or ionic resonances with the harmonics produced, to facilitate the analysis of the results in terms of various factors contributing to the phase mismatch during propagation through the medium. It is observed that the harmonic intensity (I_H) scales with medium length (L_{med}) as $I_H \propto L_{\text{med}}^p$ with the scaling exponent "p" smaller than 2, for lengths longer than 0.8 mm. The intensity of harmonics decreases with increasing harmonic order, and this decrease is steeper for longer plasma plumes. The experimental results are phenomenologically explained by considering various factors contributing to the phase mismatch, and reabsorption of the harmonic radiation. Ionization in the low-excited plasma plume is noted to be the dominant reason limiting the medium length for growth of harmonic radiation to $\sim 2-3$ mm.

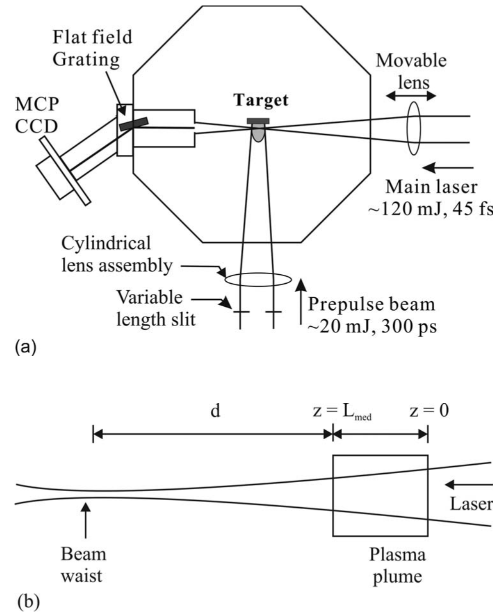

FIG. 1. (a) Schematic of the experimental setup. (b) The laser propagation geometry inside the plasma plume.

II. DESCRIPTION OF THE EXPERIMENT

The laser used in this study was a chirped-pulse amplification-based Ti:sapphire laser system (Thales Lasers S.A., France) operating at a 10 Hz repetition rate. A schematic of the experimental setup is shown in Fig. 1(a). A part of the uncompressed laser beam (pulse energy of ~ 20 mJ, pulse duration of 300 ps, central wavelength at 793 nm) was split from the main beam by a beam splitter. This beam was line focused at normal incidence by an assembly of two crossed cylindrical lenses of focal lengths 450 and 550 mm, on a planar silver strip of 2 mm width, kept in a vacuum chamber evacuated to 10^{-5} mbar. This beam (referred to as the prepulse beam) created a plasma plume to serve as the medium for harmonic generation. Focal-spot size of this prepulse beam at the target surface was 2 mm$\times$300 μm [full width at half maximum (FWHM)]. The peak laser intensity of the prepulse beam on the target was $\sim 10^{10}$ W/cm^2, so that the plasma plume produced predominantly consisted of neutrals and singly charged ions [24]. At this intensity, the target surface does not degrade much and many laser shots can be fired at the same place. This can be helpful in the HHG at a high repetition rate. The density of the plasma plumes generated under similar conditions was estimated to be $\sim 2\times 10^{17}$ cm^{-3} in our earlier experiment [23]. After a time delay of 60 ns, the main laser pulse, compressed to 45 fs (energy: 120 mJ), propagating parallel to the target surface, was focused in the above preformed plasma plume at a distance of ~ 100 μm away from the target surface, using a spherical lens of 500 mm focal length. The Rayleigh range for this focusing geometry was ~ 320 μm. The beam waist was located at a distance of 6.5 mm *after* the plasma plume center, as shown in Fig. 1(b). The femtosecond peak laser intensity at the center of the plasma plume was $\sim 2.5\times 10^{15}$ W/cm^2. The delay between the prepulse and the main laser pulse was optimized to achieve good harmonic

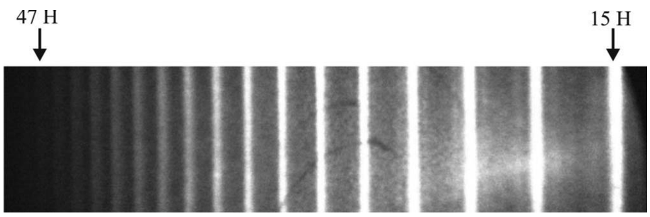

FIG. 2. Spectrum of high order harmonics generated from the silver plasma plume.

conversion. In our previous studies [16], it was observed that the harmonic intensity increases rapidly up to a delay of ~30 ns and thereafter remains approximately constant up to the maximum delay of ~60 ns used in the experiment.

The high-order harmonics were analyzed by a flat-field grazing-incidence xuv spectrograph based on a 1200 grooves/mm variable line spacing grating (Hitachi). The spectrum was recorded on a multichannel plate (MCP) detector, the output of which was imaged onto a 12 bit digital charge-coupled device (CCD) camera (SamBa, Sensovation) connected to a PC. To study the effect of medium length on harmonic emission, the length of the plasma plume was varied by inserting a slit of variable width in the path of the prepulse laser beam before the cylindrical lens assembly. The length of the plasma plume was varied in the range of 0.8–2 mm with step size 0.4 mm.

III. RESULTS AND DISCUSSION

High-order harmonics from silver plasma plumes of different lengths were recorded. A typical harmonic spectrum for a plasma plume length of 2 mm is shown in Fig. 2. Odd harmonics from 15th to 47th order are seen covering a spectral range of 52.9–16.8 nm. The dependence of the harmonic intensity on the length of the plasma medium was studied. Figure 3 shows the variation of 21st, 33rd, and 41st harmonic intensity with the length of plasma medium. The harmonic intensity (I_H) increases with the medium length (L_{med}), and it shows a length scaling of $I_H \propto L_{\text{med}}^p$, where the

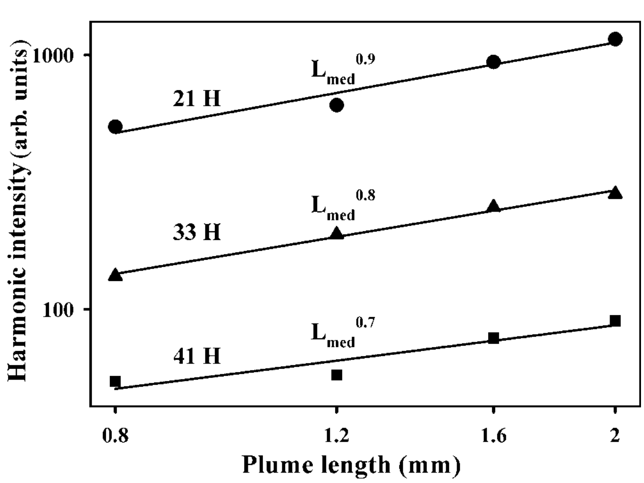

FIG. 3. Variation of the harmonic intensity with plume length.

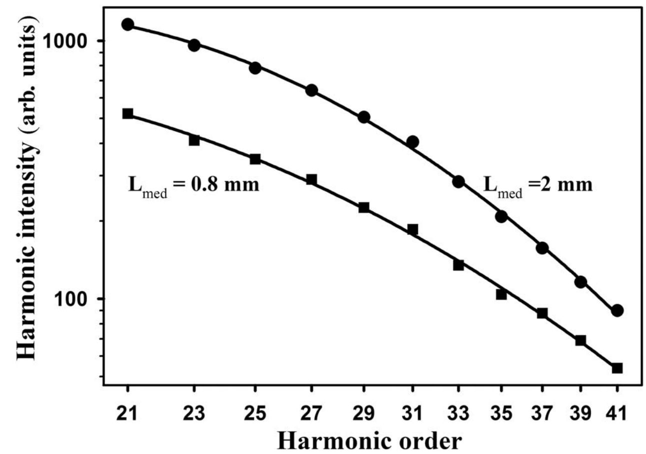

FIG. 4. Variation of the harmonic intensity with harmonic order for two different plasma plume lengths.

scaling exponent p is ~0.9, 0.8, and 0.7 for 21st, 33rd, and 41st harmonics, respectively.

Figure 4 shows the variation of harmonic intensity with harmonic order for medium lengths of 0.8 and 2 mm. The intensity of harmonics decreases with increasing harmonic order. For instance, for the case of the plasma plume length of 2 mm, the intensity of the 41st order harmonic is ~13 times smaller than that of the 21st order harmonic. However, the decrease is slower for a shorter plume length; the ratio of the intensity of the 21st harmonic to the 41st harmonic is reduced to ~9.5 in the case of 0.8 mm plume length. Nevertheless, in both cases, the harmonic emission deviates from the plateaulike emission behavior observed in earlier studies with gas jets.

We now examine the above experimental results from physical considerations involved in HHG and the growth of harmonic intensity with propagation length in the medium. Since harmonic generation is a nonlinear coherent process, the growth of harmonic intensity is governed by any phase mismatch between the driving laser pulse and the harmonic radiation produced during propagation through the plasma medium. The field amplitude of the harmonic radiation after propagation in a medium of length L_{med} may be written as

$$E_q = \int_0^{L_{\text{med}}} N_0 d(q\omega_L) dz, \tag{2}$$

and $d(q\omega_L)$ is the nonlinear dipole moment of the emitters for the qth harmonic of laser frequency ωL. Considering the phase mismatch between the harmonic radiation and the laser field, and the reabsorption coefficient (α) of harmonic radiation in the plasma medium, Eq. (2) is modified to become [13]

$$E_q = \int_0^{L_{\text{med}}} N_0 d(q\omega_L) \exp(i\Delta kz) \exp[-\alpha(L_{\text{med}} - z)] dz, \tag{3}$$

where Δkz represents the phase mismatch at a location z measured with respect to the entrance edge of the plasma plume. The term $\exp[-\alpha(L_{\text{med}} - z)]$ in Eq. (3) represents the reabsorption of harmonics generated at position z in the remaining plasma plume, where α is the absorption coefficient. For the sake of simplicity, we may assume the plasma plume to be of uniform density through its length so that Δk and α are not functions of z. This is reasonable since the electron density in plasmas produced during interaction of subnanosecond laser pulses scales with laser intensity as $I_L^{1/3}$ [28]. Thus the maximum density variation from the entrance (exit) edge of the plasma plume with respect to its center will be ~15% only. Next, in order to visualize the effect of the phase mismatch Δk and absorption coefficient α with respect to the plasma length (L_{med}), the above parameters may be expressed in terms of appropriate length parameters. We may write

$$L_{\text{abs}} = \frac{1}{2\alpha}, \quad L_{\text{coh}} = \frac{\pi}{\Delta k}, \tag{4}$$

where L_{abs} is the absorption length defined as the length for which the harmonic intensity is reduced to its $1/e$ value due to absorption during propagation, and L_{coh} is the coherence length defined as the length for which a phase mismatch of π is introduced between fundamental and harmonic radiations during propagation. Equation (3) can now be expressed as

$$E_q = \int_0^{L_{\text{med}}} N_0 d(q\omega_L) \exp\left(\frac{i\pi z}{L_{\text{coh}}}\right) \exp\left[-\frac{(L_{\text{med}} - z)}{2L_{\text{abs}}}\right] dz. \tag{5}$$

Since L_{coh} and L_{abs} are taken to be independent of z in the plasma plume, one can integrate Eq. (5) and obtain

$$E_q = N_0 d(q\omega_L) \exp\left[-\frac{L_{\text{med}}}{2L_{\text{abs}}}\right] \int_0^{L_{\text{med}}} dz \exp\left[\left(\frac{i\pi}{L_{\text{coh}}} + \frac{1}{2L_{\text{abs}}}\right) z\right]$$
$$= N_0 d(q\omega_L) \exp\left[-\frac{L_{\text{med}}}{2L_{\text{abs}}}\right] \frac{\exp\left(\frac{i\pi}{L_{\text{coh}}} + \frac{1}{2L_{\text{abs}}}\right) L_{\text{med}} - 1}{\frac{i\pi}{L_{\text{coh}}} + \frac{1}{2L_{\text{abs}}}}. \tag{6}$$

The number of photons of qth harmonic order can be written as

$$N_q \propto |E_q^2|$$

or

$$N_q \propto N_0^2 |d(q\omega_L\ |^2 \frac{4(L_{\text{abs}} L_{\text{coh}})^2}{L_{\text{coh}}^2 + (2\pi L_{\text{abs}})^2} \times \left[1 + \exp\left(-\frac{L_{\text{med}}}{L_{\text{abs}}}\right) - 2\cos\left(-\frac{\pi L_{\text{med}}}{L_{\text{coh}}}\right) \exp\left(-\frac{L_{\text{med}}}{2L_{\text{abs}}}\right)\right]. \tag{7}$$

It follows from the above expression that for given values of L_{coh} and L_{abs}, the harmonic intensity can be optimized with respect to the length of the plasma medium. Moreover, since in general, L_{coh} and L_{abs} vary with plasma density and harmonic order, one can make appropriate choices of the plasma density and medium length to achieve maximum harmonic intensity for different orders of harmonics.

There are mainly four factors that contribute to phase mismatch [21]: (1) atomic dispersion [11,21], (2) Gouy phase shift [29], (3) plasma dispersion, and (4) intensity dependent dynamical phase shifts in nonlinear dipole moments [21]. Atomic dispersion refers to the variation in the refractive index of the gaseous medium for the fundamental and harmonic radiation. The corresponding phase mismatch Δk_a due to atomic dispersion can be written as

$$\Delta k_a = \frac{q\omega_L}{c}(n_q - n_0), \tag{8}$$

where n_0 and n_q are the refractive indices for the laser and the qth harmonic radiation, respectively, and c is the speed of light. Generally, n_q for harmonic radiation in the xuv range is close to 1. On the other hand, the refractive index n_0 for radiation in a visible-ir region for underdense media may be expressed as

$$n_0(\omega) = 1 + (1-\eta) N\pi c^3 \sum_k \frac{g_{2k}}{g_{1k}} \frac{A_k}{\omega_k^2(\omega_k^2 - \omega^2)}, \tag{9}$$

where η is the ionization fraction, N is the atomic density of the plume, ω_k is the transition frequencies ground state transitions near laser frequency ω, g_{2k}/g_{1k} is the ratio of statistical weights of the kth state to the ground state, and A_k are Einstein's coefficients for the kth level. Atomic phase mismatch between the laser and the qth harmonic order can be written as

$$\Delta k_a = -(1-\eta) N\pi c^2 q\omega_L \sum_k \frac{g_{2k}}{g_{1k}} \frac{1}{\omega_k^2(\omega_k^2 - \omega_L^2)} A_k. \tag{10}$$

It could be easily seen that the atomic dispersion is negative and increases with laser frequency, plasma density, and harmonic order. Next, the plasma dispersion arises due to the presence of free electrons in the plasma. The phase mismatch due to plasma electrons for harmonic order $q \gg 1$ can be written as [27]

$$\Delta k_{\text{pl}} = \frac{\eta N e^2 q}{c\omega_L \varepsilon_0 m}. \tag{11}$$

Another term contributing to the phase mismatch is the Gouy phase shift. This arises due to the phase change in the focusing laser radiation during its propagation. Referring to Fig. 1(b), the Gouy phase for a Gaussian laser beam at any point z in the plasma plume can be written as [29]

$$\varphi_G(z) = \tan^{-1}\frac{d + L_{\text{med}} - z}{z_R}, \tag{12}$$

where z is the distance of any point from the laser entrance end of the plasma plume, L_{med} is the length of the plasma medium, and d is the distance of laser focus from the laser exit end of the plasma plume. A similar phase shift would occur for the harmonic radiation during propagation. If the laser is focused at a distance "d" away from the laser exit end of the plasma plume [Fig. 2(b)], the Gouy phase mismatch between the laser beam and the qth harmonic during propagation through the medium length L_{med} can be written as

$$\Delta k_G(z) = \frac{q-1}{L_{\mathrm{med}}}\left[\tan^{-1}\frac{d+L_{\mathrm{med}}}{z_R} - \tan^{-1}\frac{d}{z_R}\right]. \qquad (13)$$

Finally, the last term contributing to phase mismatch is the intensity dependent dynamical phase shift (IDP), which can be understood from the three-step model of HHG [20]. The energy of emitted harmonic photons depends on the energy of electron at the time recombination. This recombination energy is a function of laser phase at the time of tunneling of the electron. It can be shown that for every recombination energy at least two laser phases of tunneling are possible. The electron that tunnels out earlier in phase will recombine with the atom at a later instant (long trajectory) compared to the one which tunnels out later in phase (short trajectory). Due to laser action on the electron wave packet during tunneling and the recombination period, it accumulates an additional phase difference from the driving laser pulse. This phase difference is proportional to the laser intensity and is known as IDP [20,21]. IDP can be written as [13]

$$\varphi_{1,2} = -C_{1,2}I_L, \qquad (14)$$

where the subscripts 1,2 denote short and long trajectories, respectively, $C_1 = 1\times10^{-14}\ \mathrm{cm^2/W}$ and $C_2 = 24\times10^{-14}\ \mathrm{cm^2/W}$ [13]. Due to a variation in laser intensity along the propagation direction, these phases result in a phase mismatch between harmonics generated at two points. The phase mismatch between harmonics emitted between two ends of the plasma medium can be written as

$$\Delta k_{\mathrm{IDP}} = -\frac{1}{L_{\mathrm{med}}}C_{1,2}[I_L(d+L_{\mathrm{med}}) - I_L(d)]. \qquad (15)$$

Overall phase mismatch Δk can be written as

$$\Delta k = \Delta k_a + \Delta k_G + \Delta k_{\mathrm{pl}} + \Delta k_{\mathrm{IDP}}. \qquad (16)$$

The coherence length L_{coh} may be estimated from overall phase mismatch as $L_{\mathrm{coh}} = \pi/\Delta k$.

In gaseous media, the phase matching over the entire interaction length was realized by using the geometry of laser interaction such that different contributing factors of phase mismatch Δk cancel each other [11,21]. In HHG experiments using low intensity (($I \sim 10^{14}\ \mathrm{W/cm^2}$) lasers and underdense gaseous media, atomic dispersion and plasma dispersion due to free electrons are small [21]. Hence proper phase matching conditions can be achieved by adjusting the focal spot inside the gas jet in a way such that the Gouy phase shift and intensity dependent dynamical phase shifts cancel each other. In experiments with self-guided beams and hollow core fibers, Gouy and IDP do not vary along the propagation length due to constant intensity profile. In these cases, phase matching can be achieved by balancing the negative plasma dispersion due to partial ionization of gas, with positive atomic dispersion [4,11].

For short medium lengths, where the reabsorption can be neglected, and for optimum phase-matching conditions, the harmonic intensity increases as the square of medium length [11]. Therefore, increasing the medium length may be used to enhance harmonic intensity. However, quadratic increase in harmonic intensity does not continue if the medium length becomes comparable to or more than the coherence length or absorption length. In fact, when the medium length exceeds the coherence length L_{coh}, harmonic intensity shows oscillations due to phase mismatch [14]. Reabsorption of harmonics in the gas medium may result in damping of these oscillations [12].

The above referred studies were performed on the propagation of high-order harmonics in un-ionized or weakly ionized media like gases, where the plasma dispersion effects are negligible. However, in our case, where HHG is studied from preformed plasma plumes, due to the presence of free electrons, plasma dispersion cannot be neglected. It can be noted that all phase-mismatch factors, except the IDP shifts, are proportional to harmonic order, and hence the coherence length decreases with increasing harmonic order. This will result in the decrease of harmonic intensity with increasing harmonic order, as observed in our study (Fig. 4). Moreover, this decrease in the intensity of harmonic orders should be faster for longer plume length, as phase mismatch accumulated by higher harmonic orders with increasing length is more.

The absorption length of xuv photons due to photoabsorption in neutral silver for the typical plasma plume density $\sim 2\times10^{17}/\mathrm{cm^3}$ [23] is calculated to be ~2.0, 2.3, 2.5 mm for the 21st, 33rd, and 41st harmonic orders, respectively [30]. Coherence length L_{coh} of the plasma plume for different harmonic orders can be calculated from $\pi/\Delta k$, where different components of Δk are given in Eq. (16). Atomic phase mismatch can be determined from Eq. (10). For silver, the major transitions contributing in refractive index are ${}^2P_{3/2} \rightarrow {}^2S_{1/2}$ and ${}^2P_{1/2} \rightarrow {}^2S_{1/2}$ of $4d^{10}5P \rightarrow 4d^{10}5s$ having transition wavelengths of 328, and 338.3 nm, with corresponding spontaneous emission rates $A_k = 1.47\times10^8\ \mathrm{s^{-1}}$ and $1.35\times10^8\ \mathrm{s^{-1}}$, and ratios of statistical weights $g_2/g_1 = 2$ and 1, respectively [31]. In low excited plasmas used for HHG, the degree of ionization is typically in the range of 5–10%. Taking an average value of 7.5% for a plasma plume of density $2\times10^{17}\ \mathrm{cm^{-3}}$, the atomic phase mismatch Δk_a can be estimated from Eq. (10) as −1.3, −2.0, and −2.5 rad/mm for the 21st, 33rd, and 41st harmonic orders, respectively. Similarly, the plasma phase mismatch Δk_{pl} is also estimated from Eq. (11) as 1.5, 2.4, and 3.0 rad/mm for the 21st, 33rd, and 41st harmonic orders, respectively.

Next, the Gouy phase is calculated from Eq. (13) as 0.15, 0.25, and 0.3 rad/mm for the 21st, 33rd, and 41st harmonic orders respectively. Further, the intensity dependent phase shifts Δk_{IDP} can be estimated from Eqs. (14) and (15). The value of Δk_{IDP} is calculated assuming that short trajectory is dominating electron trajectory for the generation of high-order harmonics as it has accumulated less phase mismatch. Although the peak laser intensity at the plume center is $\sim 2.5\times10^{15}\ \mathrm{W/cm^2}$, the laser intensity used to calculate Δk_{IDP} is the effective laser intensity, governed by saturation effect [11]. The electron quiver energy corresponding to the maximum order of harmonic (47th) observed in our experiments is ~16 eV. Using Eq. (1), the saturation intensity is estimated to be $\sim 2.7\times10^{14}\ \mathrm{W/cm^2}$. Assuming this as the effective intensity at the plume center, for a Gaussian beam of geometry shown in Fig. 1(b), the change in laser intensity from the entrance to the exit of the plasma plume $[I_L(0) - I_L(L_{\mathrm{med}})]$ is $\sim -1.8\times10^{14}\ \mathrm{W/cm^2}$. The phase mis-

TABLE I. Estimates of various factors contributing to phase mismatch, coherence length, and absorption length, for different harmonic orders, to calculate the intensity scaling exponent.

Harmonic order	Δk_G (rad/mm)	Δk_{IDP} (rad/mm)	Δk_a (rad/mm)	Δk_{pl} (rad/mm)	Δk (rad/mm)	L_{coh} (mm)	L_{abs} (mm)	Scaling exponent
21	0.15	0.9	−1.3	1.5	1.25	2.55	2.0	1.1
33	0.25	0.9	−2.0	2.4	2.3	2.00	2.3	0.9
41	0.30	0.9	−2.5	3.0	2.5	1.85	2.5	0.8

match Δk_{IDP} is calculated using Eq. (15) as ~ 0.9 rad/mm. As stated earlier, this value is independent of the harmonic order.

The overall phase mismatch Δk, on adding up all four contributions, comes out to be 1.25, 1.55, and 1.7 rad/mm and is the same as shown in Table I. The corresponding coherence lengths (L_{coh}) are 2.5, 2.0, and 1.9 mm for the 21st, 33rd, and 41st harmonic orders respectively. The absorption lengths (L_{abs}) were estimated to be 2.0, 2.3, and 2.5 mm for the 21st, 33rd, and 41st harmonic orders, respectively. Using these values of L_{abs} and L_{coh} in Eq. (7), the harmonic intensities for different plume lengths were calculated and the least square fit of these values gives the length scaling exponent p to be 1.1, 0.9, and 0.8 for the 21st, 33rd, and 41st harmonic orders, respectively. These values are close to the experimentally observed scaling exponents and show a similar decreasing trend with harmonic order. The estimated values of the scaling exponent will depend on the value of the atomic density (N) and the degree of ionization (η) of the plasma plume. The small difference in calculated scaling exponents with experimentally observed value may be due to the choice of number density used for calculation. Moreover, the plasma plume will also have a density variation. In principle, detailed analytical calculations or computer simulations would be required for the exact quantitative comparison of the experimental data with theoretical predictions. However, the simple estimations carried out here are able to reasonably illustrate the observed scaling of harmonic intensity with medium length for different harmonic orders.

The value of the scaling exponent p for all three harmonic orders is smaller than 2, expected for a completely phase-matched media. In principle, one should expect a quadratic scaling of the harmonic intensity with the length of the plasma plume. However, this could not be studied in our experiment. We had used cylindrical focusing of the laser beam to irradiate the target on a focal spot of a transverse dimension of 0.3 mm and a variable length from 0.8 to 2 mm. Since three-dimensional expansion of the plasma would become significant for lengths comparable to the transverse dimension, shorter plume lengths were not used. Further, the decrease in p occurs as the medium length approaches towards the coherence length. Since the coherence length is smallest for the 41st harmonic order, the value of p is also the smallest for this harmonic order. The above calculations also illustrate the faster decrease of harmonic intensity with harmonic order for the longer plasma plume length. Since the coherence length decreases with increasing harmonic order (Table I), the phase mismatch accumulated during propagation through a longer plasma plume will increase with harmonic order at a higher rate.

The contributions of different factors to phase mismatch, listed in Table I, clearly show that the largest contribution is from IDP for the conditions of the present experiment. As stated earlier, this phase mismatch term is also independent of the harmonic order. Contribution of this term can be reduced by reducing the intensity variation along the plasma plume, which can be achieved by increasing the Rayleigh range (z_R) through the use of a large f number of focusing optics.

In conclusion, we have experimentally studied the scaling of the harmonic intensity (I_H) on the medium length. It is observed that the harmonic intensity shows a scaling $I_H \propto L_{\mathrm{med}}^p$, where the scaling exponent p is smaller than 2, for all the harmonic orders. Moreover, the value of the exponent p decreases with increasing harmonic order. The results have been explained from phase mismatch and harmonic absorption considerations. The scaling laws computed for typical values of medium density and ionization are in good agreement with our experimental observations.

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