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High-order-harmonic generation in gas with a flat-top laser beam

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We present experimental and numerical results on high-order-harmonic generation with a flat-top laser beam. We show that a simple binary tunable phase plate, made of two concentric glass plates, can produce a flat-top profile at the focus of a Gaussian infrared beam. Both experiments and numerical calculations show that there is a scaling law between the harmonic generation efficiency and the increase of the generation volume.

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I. INTRODUCTION

With the recent developments in high-power ultrashort pulse-laser technology, many experimental and theoretical works have been devoted to high-order laser-harmonics generation (HHG) by intense lasers interacting with gas [1,2]. HHG provides a promising XUV radiation source with characteristics suitable for a large array of applications [3]. Its ultrashort time duration, in the femtosecond or attosecond domain, has been used in time-resolved studies of ultrafast electron dynamics [4,5]. Its high degree of spatial coherence is used in coherent diffractive imaging [6–8]. Depending on the generation geometry, HHG central frequency can be tuned in a very large spectral domain, from a few electron volts up to the kilo-electron-volts range [9], and presents either a continuous or discrete spectrum [1]. It can consequently be tuned to the desired spectral range for time-resolved or nonlinear studies in diluted, condensed, or plasma phases for instance. Nevertheless, many applications, especially nonlinear studies, suffers from the low conversion efficiency of HHG. It is worth noting that other techniques for high-order-harmonics generation have already provided promising results in both energy range and photon flux, namely, harmonic from solids [10] and harmonic from laser-produced plasmas [11].

Many generation schemes have been explored, most of them trying to improve the key parameter for the coherent macroscopic field construction, phase matching. Inspired by nonlinear visible optics, quasiphase matching solutions have been proposed [12] using a modulated medium [13–15] or a modulated laser intensity [16–18]. Experimentally, slowly varying geometries are easier to implement and have already produced high-photon numbers in gas-filled capillaries [19,20], gas cells [8,21,22], or long gas jets [23]. The main idea here can be explained through a simple discussion between two quantities: the coherence length $L_{coh}$, which is the distance over which the nonlinear polarization and the harmonics field are dephased by $\pi$, and the absorption length $L_{abs}$ over which the harmonic field is absorbed by the generating medium. An optimal conversion efficiency is achieved for $L_{coh} > 5L_{abs}$ [19]. $L_{coh}$ being a local property, this relation must be fulfilled in the whole generating medium, which can be realized for a uniform laser field, or more easily in a slowly varying geometry.

To that end, one focuses the laser beam in the generating medium using a lens with a focal length of several meters. Choosing the right focal length depends on the available laser energy and the desired laser intensity in the medium. If the latter is too high, ionization would create free electrons in the medium, preventing efficient phase matching of the HHG due to electron dispersion. Moreover, free electrons induce propagation effects for the infrared field, often leading to a reduced laser intensity. The laser intensity must then be less than the saturation intensity, which depends on the generating gas (typically in the $10^{14}$ W/cm$^2$ range for rare gases). This presents a serious constraint when using a high-energy laser. For instance, focusing a typical 50 mJ, 50 fs laser ($w = 16$ mm, $M^2 = 2$) up to argon saturation intensity ($2.5 \times 10^{14}$ W/cm$^2$) requires a 16-m-long focal length. It is quite easy to create an optical line using a few mirrors to reduce the size of the infrared part (before the generating medium) of the setup, but mirrors in the XUV range are difficult to manufacture and have low reflectivity, therefore counterbalancing the gain from the loose focusing geometry. Taking advantage of the divergence of a focused beam, it is possible to reach the same intensity using a shorter focal length by moving the generating medium away from the focus position. Using the previous beam parameters, but shifting the medium at the Rayleigh distance from the focus, lowers the focal length to 11 m. However, this requires a laser beam of very high spatial quality.

The other possibility to take advantage of high-energy laser in HHG is to enlarge the generation volume, not only in the longitudinal but also in the transverse direction. Actually, when one increases the focal length, one also enlarges the beam profile at focus, and thus plays not only with phase matching but also with the number of emitters. Energies of $7 \mu J/$pulse for the 11th and $4.7 \mu J$ for the 13th harmonics have been reported associated with loose focusing in a long Xe cell [24,25], whereas an energy of $1.9 \mu J/$pulse for the 15th harmonic generated in a long Xe gas jet was obtained in similar beam geometry [23]. We recently reported $0.25 \mu J/$pulse for the 25th harmonic generated in argon using a 5.5-m-long focal lens

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and a 8-cm-long gas cell [26]. However, as we just mentioned, current laser technology asks for a new geometry to go beyond the limits of the loose focusing. Several works already reported attempts to shape the laser intensity distribution in the focus vicinity. Roos et al. [27] used a birefringent lens to create two foci separated by 6.2 mm along the beam propagation axis, and thus increased the longitudinal size of the generating volume, increasing the efficiency of HHG in neon by a factor of 4 compared to a single focus under the same conditions. Diffractionless beams, like Bessel-Gauss beams [28–30], have also been proposed. Also, more advanced schemes relying on deformable mirrors and genetic algorithms have been demonstrated [31–33].

In the present work we use another approach. Through beam shaping we want to increase the beam radius in the focus area while using a reasonable focal length lens. Moreover, we want to create a uniform intensity distribution in the radial direction—a so-called flat-top profile—in the generating volume, slowly evolving along the propagation axis, to enhance phase matching and create uniform conditions of generation. This intensity profile was also theoretically shown to allow for the generation of isolated attosecond pulses with long driving pulse [34,35]. Using ionization, one can achieve transient phase matching [19,22] and temporally confine the harmonics generation. Because the beam intensity is constant spatially, there is no temporal enlargement of the macroscopic response due to spatial effect (with standard Gaussian beam, XUV emission does not occur at the same time at the center or at the outer part of the beam enlarging the macroscopic response as compared to the short atomic response [36]) ensuring a uniform ionization gating in the medium.

The paper is divided as follows: Sec. II presents, numerically as well as experimentally, the phase-shaping technique we used to create a flat-top profile. Section III details both our HHG experimental setup and our numerical code. In Sec. IV we finally discuss our experimental results and compare them with the simulations.

II. PHASE-SHAPING AND LASER-PULSE PROPAGATION

One simple way to generate a flat-top beam profile has already been proposed and used in high-order-harmonics generation [37]. Toma et al. put an aperture in the beam path and imaged the flat-top profile thus created by diffraction after the iris. By imaging this profile in the generating medium, they could study intensity effects in HHG without spatial averaging effect. The main drawback of this technique is that it implies a large number of optics for the imaging system, which introduce aberrations and are an additional source of large losses. Chaloupka and co-workers used a coaxially segmented wave plate to introduce a $\pi$ phase shift between the central and the outer parts of a beam, creating a electron trap at focus [38]. Boyko et al. showed later that a similar annular intensity profile could be generated with a deformable mirror [39]. Fromager et al. [40] proposed the use of a circular binary object dephasing the central part of the beam by $\pi$ to create a flat-top profile close to the focus position.

The setup we used, also described in [34,35], combines the techniques of the phase plate from [40] and the versatility of the segmented wave plate from [38]. A silica plate, cut in two concentric parts, is inserted in the beam path just before the focusing lens. The dephasing is controlled by the rotation of the inner plate around the vertical axis, which changes the effective thickness of the inner phase plate, while the ratio between the inner and outer part radii ($r_2/r_1$) is controlled by an iris (see the inset in Fig. 1). In Fig. 2 we present the simulated intensity and phase distributions of a shaped beam using the following parameters: focal length $f = 2$ m, dephasing $= 0.8 \pi$, inner plate radius $r_1 = 18$ mm, and ratio $r_2/r_1 = 1.36$. The beam radial profile has a flat-top shape, with a radius of 110 μm (2.5 times larger compared to what is expected for the exact same parameters but no phase shift). Overall, in the volume presented in Fig. 2, the difference between the beam profile and an equivalent real flat-top profile is small (5% rms). The spatial phase stays almost flat over a 2 mm distance. Experimentally we tested two sets of phase plates (hereafter called small plate and intermediate plate with, respectively, 9 and 12 mm, 18 and 24.5 mm of inner and iris diameters) on the LUCA laser from CEA Saclay (France) ($w = 24$ mm, 55 fs, $\lambda = 800$ nm, 20 Hz repetition rate, and a pulse energy up to 16 mJ in this experiment). The laser profiles at focus in Fig. 3 were measured using the small [Figs. 3(a) and 3(b)] and intermediate [Fig. 3(c)] plates with the camera depicted in Fig. 1. Using the small plate, the beam radius was increased by a factor of 2.1 (1.5 with the intermediate plate), close to the expected value. The difference may be due to the degradation of the laser wavefront with increasing radius. Indeed, when we used the whole beam with a set of larger plates, we could not recognize any shape in the beam profile: the quality of the laser spatial phase on the edges of the beam radial profile prevented any efficient shaping.

III. HIGH-ORDER-HARMONIC GENERATION

We focused the shaped beam with a 2 m focal length lens in a 1-mm-long pulsed gas jet. We used argon and xenon, with pressure of the order of 100 Torr. The relatively short medium length was chosen to keep the flat-top profile in the whole interaction region. The harmonic beam was then sent to a spectrometer (constituted by a grazing incidence gold coated toroidal mirror with a 50-cm-long focal length, mounted in a $2f$–$2f$ imaging configuration, and a highly diffractive grating) and detected by a photomultiplier. We concentrated our study on the 13th harmonic generated in argon. Changing from argon to xenon showed the same overall behavior, as did studying other plateau harmonic orders. A variable attenuator before the phase plate allowed us to control the laser energy between 2 and 16 mJ without changing the other beam parameters. In our experimental conditions, the maximum intensity we could achieve with a tailored beam using the small plate...
was limited to $3 \times 10^{13}$ W/cm$^2$. This value is far below the saturation intensities for harmonics generation in argon and xenon (around $8 \times 10^{13}$ W/cm$^2$). Consequently, in the following, our study will be restricted to the intermediate plate (resulting in a maximum intensity below $2 \times 10^{14}$ W/cm$^2$).

In parallel to the experiment, we ran simulations of the HHG by a shaped laser beam. The numerical model we used has been described in [30,41]. Briefly, the two-dimensional code in cylindrical geometry solves the coupled propagation equations of the fundamental and harmonic fields in the paraxial and slowly varying envelope approximations. The ionization by the infrared field is calculated following the Ammosov-Delone-Krainov (ADK) model [42]. The resulting depletion of the medium and free-electron dispersion are included in the propagation equations, as well as the atomic dispersion and the absorption of the harmonic field. Because we compared HHG from Gaussian and flat-top beams with the same initial energy, we had to consider intensities well above the gas saturation intensity. Because of the modifications of the laser field properties and of the dispersion of the harmonic field induced by the free electrons in the medium, an accurate description of the total number of electrons was needed. Using the ADK model we calculated the tunneling rates for higher ionization states (up to four); for a laser energy of 16 mJ, the maximum intensity at focus for our generation conditions reached $8 \times 10^{15}$ W/cm$^2$. Actually, because of refraction effects in the gas medium, such high intensities are never reached and lower ionization states could be enough. The dephasing of the inner part of the field was added to the laser super-Gaussian profile before the focusing lens. The complex spatial field distribution was then calculated at the entrance of the generating medium, where we started the calculation grid. The beam and the phase plates characteristics were the same as for the experiment. We considered a medium with a truncated Lorentzian density profile with a full width at half maximum of 1 mm. The gas pressure was 120 Torr, resulting in an absorption length below 100 $\mu$m for the harmonic orders we considered.

**IV. RESULTS AND DISCUSSION**

First, we studied the harmonics generation when the focus was at the center of the gas jet. The observed behavior depends on the initial laser energy. At low energy, the harmonic signal is higher when the beam profile is Gaussian (around $\Phi = 0$ in Fig. 4) compared to the flat-top profile case (around $\Phi = \pi$). On the contrary, at high-laser energy, the harmonic signal is about twice as high for a shaped beam profile. The simulations reproduce well this behavior, for both laser energies. A simple explanation can be given considering the infrared intensity at focus in both cases. When the laser energy is low, due to the losses and the larger beam waist, the maximal intensity of the flat-top beam is too low to generate harmonics efficiently.
When the energy is increased, the flat-top intensity reaches the saturation value, while the Gaussian beam gets above it: defocusing due to the free electrons and depletion of the neutral population then prevent HHG. However, the conversion efficiency, in the Gaussian case, can be improved by changing the relative position of the focus with respect to the generation medium [43]. This is illustrated in Fig. 5. For a given laser energy (11 mJ in this example) we varied the focus position. In the Gaussian case, the beam profile evolves accordingly to Gaussian beam propagation. The shaped beam however has a well known profile only at focus ($z_{jet} - z_{lens} = 0$). At this given position, as expected, the harmonic signal is much higher compared to what can be generated by a Gaussian beam. However, if the lens is moved away from the focus, the signal increases up to the same level. When the lens is moved 2 cm away from the jet, the beam waist is enlarged and argon saturation intensity is reached. Moreover, the generation volume is enlarged, increasing the number of emitters. The harmonic generation efficiency is thus optimized, and the measured signal is equal to what is generated with a flat-top beam. This behavior is well reproduced by the simulations (shown in Fig. 5 for the same laser energy). For every pulse energy we considered, we scanned the focus position with respect to the jet and recorded the harmonic signal. The optimal measured harmonic signal with respect to the energy is reported in Fig. 6(c) (opened symbols). The position of the focus was fixed in the jet for the shaped beam, in order to keep the flat-top profile. For each energy, it was possible to find a position where the harmonic signal from the Gaussian beam was at least equal to the signal generated by the flat-top profile. This tendency is well reproduced by the simulations [filled symbols in Fig. 6(c)]. The harmonics generation with a Gaussian beam becomes more efficient at high energy because in the flat-top case argon saturation intensity is reached above 10 mJ, while it is possible to move the focus further away from the jet to enlarge the focal spot with a Gaussian beam. In order to use that much energy or more, a larger flat-top profile has to be created.

Experimentally, we measured the Gaussian beam radius at different $z$ positions with the camera represented in Fig. 1 at very low energy. Consequently, this does not take into account any refraction effects which can happen in the generating gas at high intensities. Therefore, we do not have any measurement of the laser radius in the jet for a Gaussian beam, and we have to rely on the simulations to estimate the infrared intensity. This explains why in Figs. 6(a) and 6(b) we only reported the radii and intensities obtained from the calculations. We can however use those numbers together with the measured harmonic signal to check whether using a flat-top profile results in an enhancement of the phase matching. Comparing the harmonic signal generated by a Gaussian beam and a flat-top beam for the same intensity (e.g., $1.1 \times 10^{14}$ W/cm²), one
can see that the increase of the signal between a Gaussian beam at low energy and the flat-top beam is equal to the simultaneous increase of the generation volume. Moreover, the signal generated by a Gaussian beam at higher energy (but same intensity) also increases proportionately to the volume. This behavior is well reproduced by the simulations.

It is striking to see that here the phase matching conditions do not seem to play a significant role in the efficiency of the process as only the radius of the beam seems important. This is expected when phase matching is automatically achieved as it is transiently the case here. This transient phase matching has already been described [19,44] and relies on the fact that the phase mismatch $\Delta k$ changes with ionization and that harmonic is efficient provided that $\Delta k L$ is smaller than $\pi$. As $L$ is small (here $10^{-3}$ m), $\Delta k$ can be as large as $10^3 \pi$ and lead to phase matched harmonic emission. For the loose focusing conditions and the low-order harmonics considered here, the influence of the geometrical phase advance is small (and comparable for the Gaussian beam and the spatially shaped beam as shown in [35]). Moreover, for a pressure of 120 mbar (atomic density $= 2.7 \times 10^{18}$ at/cm$^3$), $\Delta k$, initially close to zero for neutral atoms, remains below $10^3 \pi$ until the ionization rate reaches 4%. It implies that phase matching is achieved in the generating conditions we used and further justifies why the volume effect is the only effect observed here.

V. CONCLUSION

In summary, we presented a simple way to efficiently generate high-order harmonics from interaction of noble gases with shaped laser beams. A tunable binary object was used to introduce a variable phase shift in the center part of the beam in order to create a flat-top profile at a lens focal point, enlarging the beam radius by a factor close to 2, and thus the harmonic generation volume by a factor of 4. Both the intensity and phase of the infrared field remain almost constant over 1 mm across the focus position. We confirmed a scaling of the number of photons generated in argon with the generating volume by comparison of HHG from a Gaussian beam and a flat-top beam at similar intensities. Two-dimensional numerical calculations, propagating both the infrared and the harmonic field and taking into account spatial effects due to several orders of ionization, confirmed those results.

We observed that shaped flat-top beams and Gaussian beams can lead to similar efficiencies under optimal conditions. We explained this observation in terms of volume effect and transient phase matching. This observation is also consistent with the fact that with shaped flat-top beams the macroscopic response is not enlarged by spatial averaging effect and should be shorter than for the Gaussian beam case. Using higher laser energy, we think that this technique should help decreasing the focal lens length to a more reasonable size, allowing for easier experimental procedures and designs. Transmission phase plates are not usable at high energy due to damages or nonlinear effects during propagation inside the material, but they can advantageously be replaced by phase steps working in reflection or specifically designed mirrors [46].

Finally, we think this technique could also find its use in any nonlinear interaction device where phase matching plays an important role, such as sum-frequency generation in a crystal, as well as in many other strong-field phenomena that are intensity dependent and suffer from spatial averaging when Gaussian beam are used.

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