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Generation of extreme ultraviolet radiation with a Bessel–Gaussian beam

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The generation of few high-order harmonics in the extreme ultraviolet is enhanced by focusing the fundamental femtosecond laser beam with a combination of lens and axicon in an infinite gas cell. We show that this combination leads to an improvement in the phase-matched generation of harmonics in the cutoff region, with a higher photon flux and a better spatial beam profile. A high atom density through the use of a high gas pressure can be used and the absorption limit of the harmonic generation is obtained for an interaction length of a few millimeters. © *2009 American Institute of Physics*. [doi:10.1063/1.3240404]

High-order harmonic generation (HHG) of intense femtosecond laser pulses interacting with gaseous media can provide a table-top source of extreme ultraviolet (XUV) radiation with relatively high photon flux which can be easily implemented for many applications. HHG sources have been used in time-resolved studies of ultrafast dynamics in atomic and molecular systems enabling the imaging of molecular orbitals,[1] surface dynamics,[2] and lensless diffractive imaging.[3,4] However, many applications in imaging and plasma physics require a high brightness source, which is difficult to implement with a small photon intensity generated per harmonic per pulse. The efficiency of the HHG process depends on the density of atoms, the phase matching between the harmonic and the fundamental field, and the absorption of harmonics by the medium. Several configurations for HHG including a hollow fiber,[5,6] a gas cell,[7] and a gas jet[8] have been used for increasing the photon flux and the experimental results show that the laser intensity cannot exceed a saturation limit resulting from a balance between the high nonlinear polarization of the neutral atoms and molecules and their ionization in the laser field. The optimization or improvement of phase matching,[6] or the use of quasiphase matching,[9–11] are suitable ways for improving the photon flux.

A phase mismatch of harmonic generation can be caused by dispersion in the nonlinear medium, which induces a phase difference between the atomic or molecular polarization and the generated field that is very sensitive to the atomic or molecular density, the laser intensity and the interaction length. Focusing of the fundamental laser beam also leads to phase mismatch which depends on the configuration of the gas medium. A hollow waveguide can be used for phase matching[5,6] with the fundamental laser pulse at 800 nm and using argon gas, but this application is limited to low gas pressures and can only achieve a small phase contribution. Also, coupling to the waveguide limits the maximum pump energy and energy loss is unavoidable. A long focal length lens[12] and a semi-infinite gas cell[7] have been used for phase matching and also to enlarge the cross section but control of the process is very difficult because the plasma dispersion and atom dispersion are strongly dependent on each other. Thus, phase-matching techniques for HHG are still an open and debated topic.

Use of a non-Gaussian or a modified Gaussian beam for generation of high-order harmonics has recently attracted attention.[13–15] Nonlinear processes in the waist of a Gaussian beam occur in a collinear excitation geometry whereas in the case of a Bessel beam they originate in a noncollinear geometry. A Bessel beam that is generated by an axicon or a suitable hologram can be considered as a superposition of an infinite number of equally weighted plane waves whose wave vector lies on a conical surface centered on the propagation axis. The theory predicts that the Bessel beam can provide an effective phase mismatch.[13] A Bessel–Gauss (BG) beam can be used for slowly varying the geometry of the laser beam around a focal point[13] that will enhance the HHG emission. Bessel–Gaussian beams have also been used for enhancement of the phase matching in the generation of very low harmonic orders.[16]

Due to its coherent nature, HHG radiation is increasingly being used for coherent diffractive imaging (CDI).[3,4] Instead of traditional CDI in which a monochromatic source with $\lambda/\Delta\lambda \sim 1000$ Ref. 16 is used we have performed multiple-wavelength CDI by using XUV radiation with several harmonic orders and shown that it is possible to use multiple wavelengths to obtain high-resolution imaging.[3] When a few harmonic orders are used a remarkably higher photon flux can be achieved because the total photon flux can be used and no additional spectral selection, such as with the use of a spectrometer or small bandwidth mirror, are needed and therefore the exposure time can be reduced to a few seconds for high-angle diffraction imaging. Thus, the generation of just a few harmonics which have high flux and are highly coherent is essential for this application.

In this paper, we report the generation of a few high-order harmonics in an infinite gas cell by focusing the fundamental laser pulse using a combination of a lens and an axicon. We show that such a combination leads to an enhancement of the phase-matched generation of harmonics in the cutoff region, with a higher photon flux and a better spatial beam profile. The influence of the lens plus axicon combination, the gas pressure, the laser intensity, and the absorption on the HHG process are investigated.

In the experimental setup, we use 30 fs, 2.5 mJ laser pulses, generated by an 1 kHz amplified Ti:sapphire laser

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system, which operates at a center wavelength of 805 nm. The laser pulses are focused by a combination of a 300 mm focal length lens and a 179° cone axicon into a 150 mm long gas cell with a glass window at the entrance and a 150 μm pinhole at the exit. Details of the experimental setup have been described elsewhere,[7] with the lens now replaced by a lens plus axicon system. The distance between the lens and axicon can be varied over the range 10–80 mm. The effective peak intensity at the focus is approximately 1×10^{14}–5×10^{14} W/cm^2. The cell is operated at a pressure from 50 to 100 Torr of argon (Ar) gas. A grazing incidence spectrometer (GIMS #4—Setpoint) is used for spectral analysis where the harmonic spectrum is detected by scanning a 300 grooves/mm grating. An aluminum metal filter is inserted in front of entrance slit to separate the harmonics from the fundamental laser beam. The far-field beam profile is then detected along the height of the exit slit of the spectrometer.

When atoms interact with a high laser field a p-order nonlinear polarization is induced, which acts as a source for the generation of a high-order field. The intensity of the qth harmonic is given by[17]

$$I_q \sim (b^3\tau_p n_0^2/h)|d(q\omega)|^2|F_q|^2, \qquad (1)$$

where b is the laser confocal parameter, τ_p is the time-integrated profile of the pth power of the laser, $d(q\omega)$ is the atomic dipole moment induced by the fundamental laser pulse, n_0 is the atom density, and F_q is the phase-matching factor. When the confocal parameter is longer than the interaction length l the phase-matching factor can be written,[18]

$$|F_q|^2 \sim (ql^2/b^2)\sin c^2(\Delta kl), \qquad (2)$$

where $\sin c(x)=\sin(x)/x$. The phase mismatch Δk on the axis can be expressed as

$$\begin{aligned}\Delta k = {} & 2\pi q(1-\eta)\Delta n/\lambda - P\eta N_{at}r_e(q\lambda - \lambda/q)\\ & + (\text{geometric term}) + (\text{atomic dipole phase}) \qquad (3)\end{aligned}$$

where λ is the laser wavelength, P, η, and r_e are the gas pressure, ionization fraction, and classical electron radius, respectively, $\Delta n = n_{\text{laser}} - n_q$, and n_{laser} and n_q are the refractive indices for the laser light and the harmonic, respectively. In this expression for the phase mismatch, the first (positive) term is related to the medium dispersion and the second (negative) term is due to the plasma dispersion. The geometric term is positive for a focused Gaussian beam and negative in a waveguide or self guide.[6] For a focused Gaussian beam, the geometrical phase shift around the focal point is due to the Gouy shift. The sign of the atomic dipole phase term is not fixed since this phase varies with the intensity of the laser field. When the phase is matched the HHG intensity will scale as $I_q \sim n_0^2 l^2$.

Figure 1 shows the HHG spectrum for three different distances between the axicon and the lens. A remarkable increase in the intensity in the cutoff region <32 nm (harmonic order $H>25$) is achieved compared with when only the focusing lens is used (solid line in Fig. 1). For this measurement, the gas pressure, aperture diameter, pulse energy, and focus position of the lens and axicon system, which is placed close to the exit pinhole and inside of cell, are optimized for maximum total flux of all harmonics. The laser intensity is chosen to be below the saturation intensity so that the high harmonics are emitted at the peak of the laser pulse.

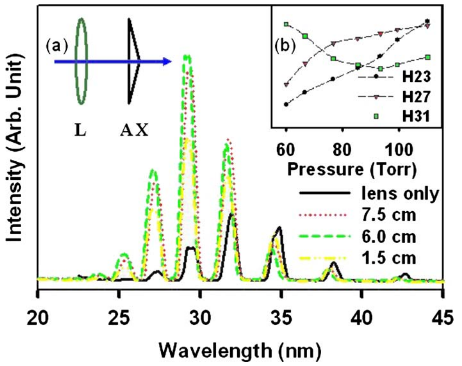

FIG. 1. (Color online) HHG spectra for three different distances between the axicon and the lens (7.5, 6.0, and 1.5 cm). The solid line shows the HHG spectrum for the case without the axicon. The inset shows the intensity of different harmonic orders vs gas pressure when the distance between the axicon and the lens is 6.0 cm.

The HHG intensity increases when the distance between the axicon and the lens increases from 1.5 to ~6 cm and then remains constant.

Figure 2 shows the dependence of the intensity of the 27th harmonic on the position of the laser focus relative to the exit of the Ar gas cell at pressures of 60 Torr (dots) and 95 Torr (triangles). For this measurement, first the aperture diameter is optimized for maximum flux and the position of the laser focus is set to $z=0$ (close to the exit and inside the gas cell). Then the laser focus position is varied, while all other parameters are kept constant. Positive values of the interaction length indicate that the focus is inside the gas cell. When a Gaussian beam with diameter 6.25 mm is focused by a 300 mm lens, the Rayleigh length is ~7 mm. The Rayleigh length is expected to be longer when a combination of lens and axicon is used.[13] For increasing values of the interaction length l, the focus position moves from outside into the gas cell, ($l < -0.75$ mm for 90 Torr and $l < -1.5$ mm for 60 Torr) and the intensity increases quadratically with interaction length, as shown by the green and black lines in Fig. 2. This indicates that for these interaction lengths, which are smaller than the Rayleigh length, the harmonic emission is phase matched [see Eq. (1)]. The phase matching is better when the higher gas pressure is used. The ratio of increasing slopes for the two pressures is ~1.5, which is close to the ratio of the gas pressures (95/60

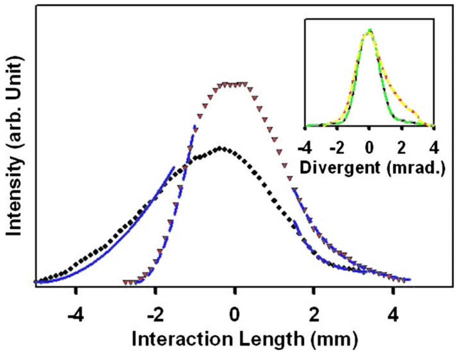

FIG. 2. (Color online) Intensity of the 27th harmonic (H27) vs focus position of the lens and axicon system for an argon gas cell at pressures of 60 Torr (dots) and 95 Torr (triangles). Positive values of the interaction length indicate that the focus is inside the gas cell. The inset shows the beam profile of the HHG emission for the lens only (dashed line) and for the lens plus axicon system (solid line).

$=1.58$). It has been shown[8] that the efficiency of the HHG process is dependent on the absorption of the gas even when the coherence length is infinite. For a longer interaction length ($z \geq 0$), the HHG intensity increases slowly and then decreases due to reabsorption in the medium. The influence of absorption in the medium can be seen in the inset of Fig. 1 where the intensity of H23 decreases with increasing gas pressure while the intensity of H31 is still increasing as a result of the absorption for H23 in Ar being an order of magnitude larger than for H31. For $l > 1.5$ mm the variation of the harmonic emission intensity is dominated by reabsorption, with an exponential decrease with interaction length, as indicated by the exponential fit (solid line) to the data. With an effective useable photon number for the fundamental laser of $\sim 2.5 \times 10^{15}$ photons/pulse, a total HHG photon number of $\sim 1 \times 10^{9}$ photons/pulse at around 30 nm is obtained which corresponds to an energy conversion efficiency of $\sim 10^{-5}$. The high conversion efficiency and the quadratic scaling of the HHG intensity with interaction length confirm that the generation process is phase matched.

The dispersion can be controlled by changing the angle of the axicon cone which offers the possibility of choosing the cone angle to compensate for the phase mismatch between the pump pulse and the harmonics because the contribution of the geometric term has the same sign as that of the plasma dispersion term. In a waveguide the dispersion Δk is proportional to $1/a^2$, where a is the diameter of the waveguide. For example, for $a = 100$ μm, $\Delta k \approx 36$ m^{-1}, while for an axicon with a base angle of 2.8°, $\Delta k \approx 2337$ m^{-1}.[13,14] The BG beam is characterized by a derivation angle γ, which depends on the parameters of the axicon and the confocal parameter of the Gaussian beam. The variation of the beam size at the axicon through a different distance between the lens and the axicon can be the same as the change of the angle of the axicon cone. This leads to a different contribution of the geometric term in the phase-mismatch expression and therefore a different harmonic intensity can be obtained as shown in Fig. 1. The use of axicon focusing can be treated as the same as a hollow core fiber in which the coupling efficiency is much higher. When phase matching is established the limit for generation of harmonics for a given laser intensity is determined by reabsorption of medium.[8,19] The balance of the phase matching and the absorption through the variation of the interaction length and gas pressure allows control of the generation of a few harmonics in the absorption edge of the medium.

For an ideal Bessel beam, the central spot radius can be extremely narrow without being subjected to the diffraction limit[20] which can also reduce the influence of the atomic phase.[21] The reduction of the diffraction limit of the Bessel–Gaussian beam through the axicon plus lens system provides better spatial coherence which is reflected in the very good Gaussian beam profile as shown in the inset of Fig. 2, in which the harmonic beam is more divergent when generated with a Gaussian beam. The better beam profile can lead to a small-size well-collimated harmonic beam, and consequently to high brightness or high photon fluxes. Using a Young's double slit, a very high fringe visibility (>0.98) of the interference pattern is obtained which indicates a high spatial coherence of emission.

In conclusion, we have been able to improve the generation of just a few harmonic orders in a semi-infinite Ar gas cell with a combination of a lens and axicon which provide a Bessel–Gaussian beam. Improved macroscopic phase-matching is achieved for harmonics in the cutoff region compared to a Gaussian beam. Given the better control of the spectrum and the coherence we find that such a source is more suitable for multiple-wavelength coherent diffractive imaging,[3] and we anticipate a significant improvement of the spatial resolution and a significant reduction in exposure time.

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