# Optimizing High Harmonic Generation in Absorbing Gases: Model and Experiment

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We study high-order harmonic generation in the general case of absorbing and dispersive atomic gas media. For ultrashort laser pulses, the harmonic conversion efficiency tends to a limit mainly imposed by the harmonic reabsorption in the gas. This limit, independent on the gas density, is the same for both the case of a loosely focused beam or a beam guided in a gas-filled hollow-core fiber. Under optimum conditions, we measured the highest conversion efficiency to date ($4 \times 10^{-5}$) for the 15th harmonic generated in xenon using a 40 fs, 1.5 mJ, 800 nm pulse at a 1 kHz repetition rate. [S0031-9007(99)08502-6]

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The emission of high-order harmonics (HH) occurring when an intense linearly polarized laser field interacts with an atomic gas is highly nonperturbative, and the single atom response is currently well understood [1,2]. The coherence and short duration of HH make it an attractive extreme ultraviolet (XUV) source in atomic, molecular, plasma, and solid state physics, and major efforts are currently devoted to increase both its efficiency and the photon flux. Here, we show that, even if high harmonic generation (HHG) is strongly influenced by collective effects such as propagation and phase matching, the maximum efficiency is mainly imposed by absorption of the harmonics by the emitting gas in the short pulse loose focus regime.

With tight focusing and long pulses, the efficiency of HHG is limited by the geometrical phase shift [3] experienced by the fundamental beam (Gouy shift) and/or by defocusing [4] due to the large density of free electrons. Loose focusing reduces the effect of the Gouy shift. Ultrashort pulses minimize ionization and increase the atomic response as required to optimize HHG [5,6], while guiding the fundamental beam in hollow-core fibers [7] should further reduce the defocusing. When defocusing and geometrical phase mismatch are minimized, we show that dispersion and reabsorption become important. For a loosely focused Gaussian beam, the Gouy shift, $\varphi(z) = -\arctan(2z/b)$ ($b$ is the confocal parameter and $z$ is the propagation coordinate) can be compensated, at focus, by the atomic dispersion at low pressures: With $b = 5$ cm and $\lambda = 800$ nm, the positive dispersion induced by $\sim 18$ mbar of argon (or 8 mbar of xenon) cancels the Gouy shift (equivalent to a negative dispersion) for the fundamental. When the laser beam is guided in the fundamental ($EH_{11}$) mode of a hollow-core fiber [8–10] with an $a = 100$ $\mu$m bore radius, these pressures also compensate for the geometrical phase shift [7], $\varphi(z) = -0.46\lambda z/a^2$. However, at this pressure, the absorption length ($L_{\mathrm{abs}} = 1/\sigma\rho$, where $\rho$ is the gas density and $\sigma$ is the ionization cross section [11]) is only $\sim 600$ $\mu$m for the 15th harmonic (H15) generated in argon and limits the useful medium lengths. Although typical ionization cross sections are smaller at shorter wavelengths, the higher densities used in jets to generate higher order harmonics can also limit the HHG efficiency. For instance, the absorption length is $L_{\mathrm{abs}} = 1$ mm for H45 generated in 60 mbar of neon.

In this work, we perform an analysis of the time dependent factors that control the efficiency of HHG including atomic response, phase matching conditions, and absorption of the atomic medium. We derive general conditions for optimum conversion efficiency. An experimental test allowed us to obtain $\sim 1.5 \times 10^{10}$ photons per shot for H15 generated in Xe and more than $10^9$ photons per shot for every odd harmonic between the 11th and 23rd generated in argon with a 1.5 mJ, 40 fs fundamental pulse at a repetition rate of 1 KHz.

Although off-axis emission and the transverse variation of the beam intensity can be important (as discussed later), one can derive the main features of HHG with a one-dimensional (1D) model. For the $q$th harmonic, the number $N_{\mathrm{out}}$ of photons emitted on axis (i.e., $k_q$, wave vector of the harmonic, is collinear to $k_0$, wave vector of the fundamental) per unit of time and of area is

$$\frac{\omega_q}{4c\varepsilon_0\hbar}\left|\left[\int_0^{L_{\mathrm{med}}}\rho A_q(z)\exp\left(-\frac{L_{\mathrm{med}}-z}{2L_{\mathrm{abs}}}\right)\exp[i\varphi_q(z)]\,dz\right]\right|^2,$$

where $A_q(z)$ (units: C $\cdot$ m) is the amplitude of the atomic response at the harmonic frequency ($\omega_q$), $\varphi_q(z)$ is its phase at the exit of the medium, $c = 3 \times 10^8$ m/s, $\varepsilon_0$ is the vacuum permittivity, and $\hbar$ is the Planck's constant. Assuming that $A_q$ is independent of $z$ (loose focus or optical guiding) and that $\rho$ is constant, $N_{\mathrm{out}}$ is proportional to

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$$\rho^2 A_q^2 \frac{4L_{\rm abs}^2}{1 + 4\pi^2(L_{\rm abs}^2/L_{\rm coh}^2)} \left[ 1 + \exp\left(-\frac{L_{\rm med}}{L_{\rm abs}}\right) - 2\cos\left(\frac{\pi L_{\rm med}}{L_{\rm coh}}\right) \exp\left(-\frac{L_{\rm med}}{2L_{\rm abs}}\right)\right], \tag{1}$$

where $L_{\rm coh} = \pi/\Delta k$ is the coherence length ($\Delta k = k_q - qk_0$). To estimate $\Delta k$, one needs to consider the geometrical phase advance and the atomic dispersion for both the fundamental and the XUV light [8]. The evolution of $N_{\rm out}$ as a function of the medium length is plotted on Fig. 1 for several coherence lengths. Even when the coherence length is infinite, the HH emission saturates as soon as the medium length is longer than a few $L_{\rm abs}$, since harmonics emitted beyond that are reabsorbed. As $L_{\rm coh}$ decreases, the efficiency saturates at smaller values. For $L_{\rm coh} < L_{\rm abs}$, the efficiency is more than 10 times smaller than the asymptotic value corresponding to perfect phase matching. The optimizing conditions,

$$L_{\rm med} > 3L_{\rm abs}\,, \tag{2}$$

$$L_{\rm coh} > 5L_{\rm abs}\,, \tag{3}$$

ensure that the macroscopic response is more than half that maximum response. Another important result from this simple analysis is that the asymptotic value is independent of the density and increases as $|A_q/\sigma|^2$. Optimizing HHG requires therefore to simultaneously maximize $A_q/\sigma$ and to fulfill conditions (2) and (3). Those optimizing conditions are time dependent and strongly influenced by ionization.

In the regime where high harmonics are generated, ionization can be calculated with a reasonable accuracy from the Ammosov-Delone-Krainov model [12]. To simulate the ground state depletion, the atomic response was estimated as $A_q(t) = [1 - \Gamma_i(t)]d_q(I)$, where $\Gamma_i(t)$ is the ionization probability, and $d_q(I)$ is the intensity dependent dipole obtained quantum mechanically [1] and averaged over quantum interferences [13]. The negative dispersion due to the free electrons was added to the atomic dispersion. The intensity-dependent refractive index, negligible here, was ignored.

Integrating Eq. (1) over the pulse duration gives the number of photons emitted on axis for the $q$th harmonic. The number of H15 photons as a function of the xenon pressure is plotted in Fig. 2 for several peak intensities, $I_{\rm max}$, of a 40-fs-long Gaussian pulse (FWHM) guided in the $EH_{11}$ mode of a 4-cm-long, 200-$\mu$m bore diameter, hollow-core fiber. If ionization is negligible ($I_{\rm max} \leq 5 \times 10^{13}$ W/cm$^2$), the emission peaks around the optimum pressure of 6 mbar where phase matching is achieved for H15. As $I_{\rm max}$ changes from $5 \times 10^{13}$ to $10^{14}$ W/cm$^2$, the maximum number of emitted photons does not change significantly because of a balance between collective effects and atomic response. Indeed, for low intensities, at which ionization is negligible, conditions (2) and (3) can be fulfilled (for adequate pressure and fiber length) during all of the pulse duration while the atomic response remains relatively low. In contrast, for higher peak intensities, where ionization occurs, the atomic response is higher but condition (3) may be achieved only transiently. Note that, provided that the initial dispersion is positive, phase matching is then automatically transiently achieved when the negative dispersion introduced by the photoelectrons compensates the initial positive dispersion.

The situation of a thin jet can be derived from that of a fiber at high pressure. Indeed, fulfilling condition (2) in a thin jet requires a high gas pressure. The gas dispersion is then larger than the geometrical dispersion, as is the case of high pressures in the fiber. The maximum number of photons that can be obtained with a thin jet is therefore close to the asymptotic value obtained with

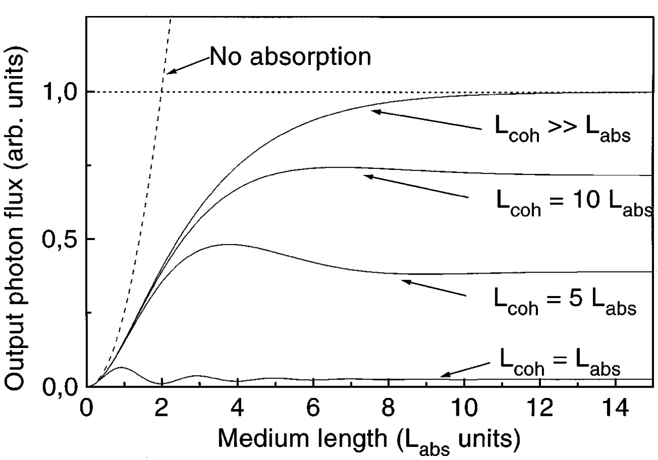


FIG. 1. Number of on-axis emitted photons (arbitrary units) as a function of the medium length (in units of absorption length). The dotted line corresponds to a zero absorption case.

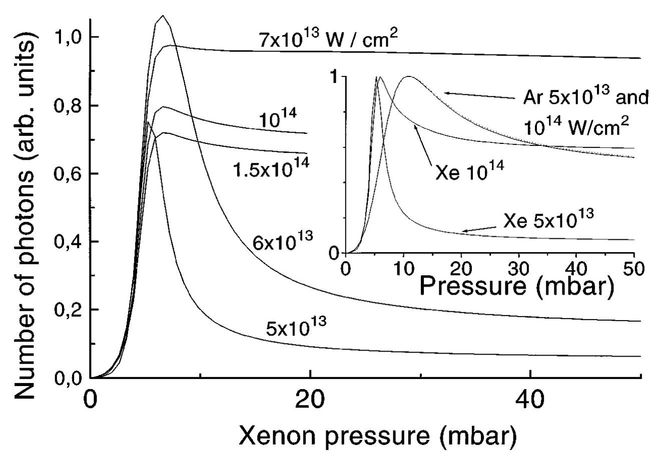


FIG. 2. Calculated number of H15 photons emitted on axis in xenon as a function of the gas pressure for several peak intensities of a 40-fs-long pulse propagating in the fiber. The inset shows the same (normalized) evolution after integration on the spatial profile of the fundamental for $I_{\rm max} = 5 \times 10^{13}$ and $10^{14}$ W/cm$^2$ both in xenon and argon.

a fiber. Figure 2 shows that, under optimum conditions, one should expect similar efficiencies for a jet or a fiber when ionization is significant. If ionization could be maintained at a low level, gas-filled hollow-core fibers could result in higher efficiencies than gas jets. However, the ionization yield is unfortunately not a free parameter, since observing a given harmonic order requires using a sufficiently high intensity.

To account for the radial distribution of peak intensities for the fundamental beam, one needs to integrate the number of emitted harmonic photons over the transverse coordinate. For the on-axis emission of H15 in a 4-cm-long fiber, we used a fundamental Gaussian profile with $w_0 = 80$ $\mu$m. This integration led to a pressure dependence (inset of Fig. 2) similar to the 1D case. The maximum number of H15 photons is, again, similar in a jet or in a fiber when ionization is significant. For a given beam profile, this number increases with $I_{\max}$. However, if one considers that, for a given energy, larger beam diameters can be used with lower intensities, the total number of emitted photons does not change significantly with $I_{\max}$ provided that the studied harmonic is well within the plateau. In addition to the 1D model, this 3D analysis shows that the optimum beam diameter should maximize the beam section where the intensity is high enough to generate the required harmonic.

The model can be extended to account for the off-axis HH ($k_q$ noncollinear to $k_0$) emission which can be significant especially when the total dispersion is negative since off-axis phase matching (annular harmonic beam in the far field) [14] can then be obtained. All of our conclusions concerning absorption, phase matching, and atomic response are still valid in this case. Off-axis emission will therefore evolve with the pressure similarly to on-axis emission and similar results are expected.

To test this model, we performed an experiment with a Ti:Sapphire laser system [15] that delivers 1.5 mJ, 40 fs pulses at a repetition rate of 1 KHz. The beam was focused with a 1-m focal length lens to a 250-$\mu$m diameter spot ($w_0 = 125$ $\mu$m) inside a vacuum chamber leading to an estimated peak intensity of $1.5 \times 10^{14}$ W/cm$^2$. Harmonics were generated either in a gas jet or in a gas-filled hollow-core fiber. The jet was collimated by a continuously opened simple cylindrical nozzle (800 $\mu$m wide). The jet position was adjusted to optimize the harmonic production. The entrance of the (4-cm-long) fiber was placed at the focus of the beam in order to maximize the coupling. The beam waist was too large for an optimum coupling in the $EH_{11}$ mode of the fiber, and we estimate that half of the energy was coupled in this mode. A hole drilled in the wall of the fibers (at half length) allowed us to inject rare gases in the fiber core. The resulting gas density decreases from the center of the fiber to the tips. However, our model requires only that the gas density is constant over $\sim 3L_{\text{abs}}$ in the end part of the medium (where the emitted harmonics can exit the medium without being reabsorbed). In argon at 12 mbar (calculated optimum pressure), the absorption length is 885 $\mu$m for H15 and $\rho$ can be considered constant. In xenon, at 6 mbar, the absorption length is 1.8 mm and $\rho$ may change over $3L_{\text{abs}}$. This case is therefore an intermediate case between a thin jet (strong density gradient) and the constant density case [16]. This setup ensures that HH can be generated up to the very end of the fiber and prevents reabsorption that occurs when a fiber is placed in a cell [8,9].

The harmonic photons were detected by photoionizing argon inside a time-of-flight electron spectrometer [17] in which argon was leaked at a known pressure. The known detection efficiency of the system allowed us to estimate the number of photons that crossed the sensitive region (3-mm long, 300-$\mu$m diameter) which was located 70 cm away from the jet/fiber. The harmonic beam is likely larger than the sensitive zone, and the number of emitted photons inferred from the number of photoelectrons is likely underestimated. This detection scheme allowed us to sample mainly the center of the far field harmonic beam (on-axis emission).

We observed all odd harmonics from the 11th (lower limit imposed by our detection system) to the 19th in xenon and the 27th in argon with both the jet and the fiber.

When the jet was used, we could increase the xenon backing pressure enough to reach the peak efficiency for the emission of H15 and we obtained up to $8 \times 10^9$ photons per shot (Fig. 3). The maximum backing pressure (500 mbar) was imposed by the pumping capacity of our system (1000 l/s turbo pump). With argon, we did not increase the pressure enough to reach the peak efficiency and obtained up to $7 \times 10^7$ photons per shot for H15 at 120 mbar.

When the jet was backfilled with xenon, the HHG efficiency was not limited by defocusing. If defocusing were the main limitation, the highest harmonics would be suppressed first as the pressure increases [3] since the

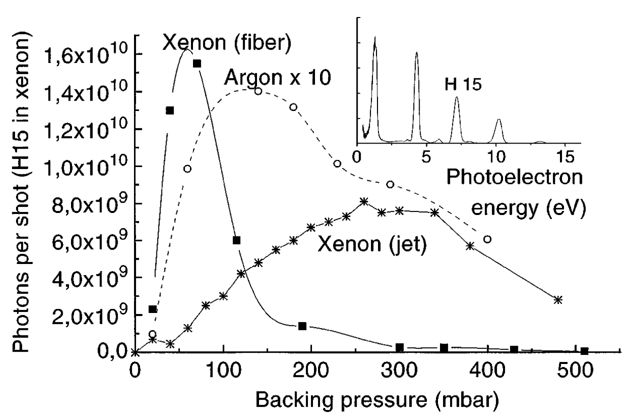


FIG. 3. Number of detected photons per shot for H15 generated in xenon in a 4-cm-long fiber (square) and in an 800-$\mu$m jet (cross). The number of H15 photons emitted with the fiber filled with argon (circle) was multiplied by 10 to use the same scale. The inset shows a harmonic spectrum generated in a xenon filled fiber.

cutoff is very sensitive to intensity. On the contrary, we observed that the optimum pressure increased with the harmonic order: For H13, the signal decreased by more than a factor 2 between the optimum pressure of 260 and 380 mbar, while the signal due to the 17th harmonic peaked at 380 mbar and the signal due to H19 was still increasing with the pressure of 480 mbar. This can be understood within our model since, for these harmonics, the absorption cross section in xenon decreases as the harmonic order increases: Hence, the higher the order the higher the pressure needed to fulfill $L_{\mathrm{med}} > 3L_{\mathrm{abs}}$.

When high harmonics were generated in a 4-cm-long, 200-$\mu$m wide gas-filled hollow-core fiber, the gas flow was much smaller than with a jet, and we could reach the maximum HHG efficiency both with xenon and argon. Figure 3 shows the number of detected photons for H15 as a function of the backing pressure. The number of photons peaks at a maximum value of $1.5 \times 10^{10}$ photons per shot in xenon. This corresponds to an energy conversion efficiency of $4 \times 10^{-5}$ which is the highest reported to date [8,18]. In argon, the maximum numbers of photons were higher than $10^9$ photons per shot (per harmonic) for all odd harmonics between H13 and H21. Within the accuracy of our pressure measurement, the optimum backing pressures were the same for all of the observed harmonics.

The optimum backing pressures that we measured were 50 mbar for xenon and 120 mbar for argon (Fig. 3). Based on gas flow measurement, the corresponding pressures at the tip of the fiber were estimated to 2.5 and 6 mbar. These values are in reasonable agreement with the optimum values predicted by our model (6 and 12 mbar, respectively).

Although we obtained a similar maximum H15 photon number with the fiber or with the jet under optimum conditions as predicted by our model for $I_{\max} = 1.5 \times 10^{14}\ \mathrm{W/cm^2}$, we did not observe the expected saturation of the emission for very high xenon pressures in the fiber. However, the shape of the calculated curves (Fig. 2, inset) is in qualitative agreement with the experimental curves (Fig. 3) for $I_{\max} \sim (5\text{–}6) \times 10^{13}\ \mathrm{W/cm^2}$ in the xenon filled fiber. These peak intensities correspond to the estimated peak intensity coupled in the $EH_{11}$ mode of the fiber ($\sim 7 \times 10^{13}\ \mathrm{W/cm^2}$). For argon, the measured pressure dependence of the emission of H15 (Fig. 3) is much broader than for xenon in agreement with our model (Fig. 2, inset), and the calculated shape (Fig. 2, inset) agrees qualitatively with our results.

In conclusion, based on a simple model, we have established the conditions (2) and (3) for optimal harmonic generation in absorbing gases. We showed that, under strong ionization, phase matching is automatically achieved (albeit transiently), provided that the gas pressure is high enough, but that the maximum achievable efficiency is limited by reabsorption. It follows that the efficiency is similar, under optimal conditions, with a jet in the loose focus regime or with a fiber, as confirmed by experiment, when defocusing can be neglected. We showed that, with our 40 fs pulse, defocusing is indeed not the limiting parameter for the moderate order harmonics that we observed. Large conversion efficiencies were obtained with both systems.

However, hollow-core fibers should lead to higher efficiencies than a jet in the cases where the absorption is low (Cooper minimum [8], zero of Fano profile) since the optimum conditions are easier to fulfill. For instance, gas-filled hollow-core fibers promise to increase the harmonic flux in the water window [6] by several orders of magnitude.

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*Note added in proof.*—For HHG in fibers, see also Ref. [19].

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