Maximizing the brilliance of high-order harmonics in a gas jet

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New Journal of Physics 11 (2009) 023016 (12pp)

Received 29 July 2008
Published 9 February 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/2/023016

Abstract. We have measured the wave front of high-order harmonics generated in a gas jet with a Hartmann sensor. With this setup we have investigated the influence of typical adjustable parameters such as gas pressure and focal position as well as the spatial laser beam profile on the conversion efficiency and the extreme ultraviolet (XUV) beam profile. Independent of the control parameter we always observed the highest conversion efficiency in connection with the best beam profile, i.e. the highest flux corresponds also to the highest brilliance. Furthermore, we have shown that aberrations of the fundamental laser beam are not simply imprinted onto the XUV beam.

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High-order harmonic generation serves as a table-top source of XUV radiation with unprecedented temporal and spatial properties [1] and references therein; [2]. As has been known since the early 1990s, harmonic radiation is the coherent macroscopic buildup of a microscopic quantum-mechanical process [3]. This has been described semi-classically in the three-step model, which assumes the ionization of a single valence electron from an atom, its nearly free propagation in vacuum and its recombination with the parent ion [4, 5]. Typically, harmonic spectra extend over a plateau where all harmonics have approximately the same intensity and reach a cutoff energy that depends on the fundamental laser intensity, the wavelength and the ionization potential of the interaction medium. In the last few years, a lot of effort has been put into the control of the temporal properties—mainly the pulse duration—in the extreme ultraviolet (XUV) regime with the successful demonstration of pump–probe spectroscopy with attosecond pulses [6, 7]. In these experiments the temporal coherence of high harmonics is utilized. However, the spatial coherence opens the way to interferometry in the XUV [8] or high-resolution diffraction experiments [9] to name just two possibilities. Nevertheless, all these experiments rely on XUV sources with the highest possible brilliance. Most of the previous works on the optimization of harmonic sources have concentrated on the photon flux, the temporal evolution or the spectral shape [1]. Only very recently, there were first attempts to optimize the wave front of high-order harmonics [10].

For a given set of laser parameters there are several ways to optimize the high harmonic yield. First of all, the gas pressure plays an important role in all experiments. One should keep in mind that the harmonic intensity scales quadratically with the number of atoms, i.e. with the pressure in the interaction region [11]. Additionally, reabsorption in the medium and phase-matching through the index of refraction are also pressure dependent. Assuming a Gaussian beam shape, it can be easily shown that phase-matching can only be fulfilled in a limited spatial region [3]. So, varying the pressure influences not only the number of generated XUV photons but also the beam profile [12]. The position of the gas jet relative to the focus is yet another macroscopic parameter with a large microscopic impact [1]. As has been shown previously, it is even possible to suppress the harmonic contribution of the longer or shorter quantum path by choosing the position carefully [13]. One can see that, although experimentally easily accessible, these two parameters have a significant impact on the XUV beam profile in the far field. So the question arises whether a maximum photon flux corresponds to the highest brilliance. The brilliance or spectral brightness is defined as

$$B = \frac{\Delta P}{\Delta \Omega \Delta A(\Delta \omega/\omega)} = \frac{N_{\text{photons}}}{\Delta t \Delta \Omega \Delta A(\Delta \omega/\omega)}.$$  

For a pulse the peak brilliance depends on the number of photons per pulse ($N$) during the pulse duration ($\Delta t$) that are emitted from a source area of the size $\Delta A$ into the solid angle $\Delta \Omega$ with a given relative spectrum of $\Delta \omega/\omega$. Radiation emitted in a beam-like wave with a perfect spherical wave front is called a diffraction-limited beam. For diffraction-limited beams the product of source area and solid angle is a minimum by definition [14]. For a given spot size such a beam has the highest brilliance and all wave front distortions reduce the brilliance.

In the present work, we investigated the influence of the gas pressure in a gas-filled tube and its relative position with respect to the focal point on the harmonics generation. Besides recording the photon yield, we also estimated the brilliance after measuring the wave front
of the harmonics with a Hartmann sensor for each setting [15]–[18]. We found a coincidence between the maximal photon flux and the wave front with the lowest aberrations.

2. Experimental setup

The high harmonics are generated with ultrashort laser pulses with an energy of 500 µJ, a center wavelength of about 800 nm and a pulse duration of approximately 40 fs at a repetition rate of 1 kHz. Although the duration of the pulse is not intrinsically important for the wave front of the generated harmonics, a shorter laser pulse yields a higher cutoff energy and a stronger ionization rate [19]. Therefore, the measurements conducted here refer only to many-cycle pulses but the results should be transferable for few-cycle pulses as well.

We focused the laser pulses with a fused silica glass lens ($f = 300$ mm, $f/# = 15$) into a sealed nickel tube with a diameter of about 2 mm inside a vacuum chamber to drill a hole of approximately 40–60 µm in diameter (figure 1). Therefore the interaction of laser pulses with a peak intensity of $2 \times 10^{15}$ W cm$^{-2}$ is limited to approximately the tube diameter of 2 mm. After the initial ablation process, we injected Ar into the nickel tube with an adjustable backing pressure between 0 and 300 mbar limited by the maximum pumping rate. The background pressure in this vacuum chamber ranges from $5 \times 10^{-3}$ mbar up to $8 \times 10^{-2}$ mbar depending on the Ar backing pressure. At background pressures much higher than 0.1 mbar, reabsorption in both the interaction medium and the chamber becomes significant and reduces the measured signal to very low levels preventing the extraction of meaningful data.

The fundamental beam is blocked by a 200 nm Al foil placed about 20 cm behind the gas target. The XUV photons are detected with a back-illuminated x-ray CCD camera (Andor DO420BN). Between the filter and the camera we inserted either a transmission mask for wave front measurements or a reflection grating in grazing incidence for recording the spectrum.

A typically measured spectrum is displayed in figure 2. No special care was taken to record the spectrum with a high resolution. However, knowing the spectrum is important for retrieving the precise phase information from the evaluated wave fronts. As one can see, we detect a typical ‘window’ of the plateau region of our harmonics where the long wavelength cut-off is given by
Figure 2. Typical high-order harmonic spectrum with a gas pressure of about 100 mbar Ar gas and a pulse energy of 500 µJ at a pulse duration of 30 fs detected behind an Al filter.

The reabsorption of Ar at about 30 nm and the short wavelength cut-off is determined by the onset of the absorption from the Al L-edge at about 18 nm. These two absorption edges together with pressure dependent phase-matching effects determine the observed spectral shape [2]. During wave front measurements, the spectrum was not recorded. In previous studies, the spectral shape remained roughly the same for the whole accessible pressure range. Therefore, it can be safely assumed that all wave front measurements were conducted with a similar spectrum.

The wave front is characterized by a Hartmann sensor similar to that in [18]. This sensor consists of an array of holes, allowing the XUV photons to pass only through the holes and cast a shadow on the detector (CCD chip). The local tilt of the wave front across the mask can then be calculated from the position of the illuminated spots on the sensor. By sampling an array of holes, all these local tilts can be measured and the whole wave front reconstructed. In our wave front analysis experiment, we placed a custom-made mask 50 mm in front of the CCD camera. The mask is an array of 50 × 50 holes, each 40 µm in diameter and spaced 140 µm apart, laser-drilled into a molybdenum plate. The pixel size of the camera is 26 × 26 µm² and thereby defines the lower limit of the hole size of the mask, since it is necessary to illuminate at least 4 pixels to compute the center position precisely. Larger holes or separations are also disadvantageous because they decrease the spatial resolution due to a given harmonic beam diameter of 1 mm (foot-to-foot width). Thus, the above described hole array is a compromise between the resolution of the complete harmonic beam and the pixel size of the XUV camera. Diffraction is not a problem because the XUV wavelength is typically three orders of magnitude smaller than the diameter of the holes. The typical integration time for measuring the wave front is 1 s. Therefore all our measurements represent a wave front averaged over 1000 shots. A slightly increased or decreased exposure time did not affect the evaluated wave front substantially. Simple wave front aberrations can be evaluated almost in real time, which allows implementation of this method in closed-loop setups or other time-sensitive procedures.

For a reliable evaluation of the wave front, knowledge of the exact position of each hole is necessary, because the information on the local tilt of the wave front is extracted from the spot.
pattern. Each beam with a different wave front casts a different shadow on the CCD, and by comparing the deviations, it is possible to reconstruct the wave front bit by bit. For calibration we put the hole array in front of the CCD and illuminated it with an expanded He–Ne laser beam, ensuring a flat wave front across the whole mask. Although diffraction is quite strong and only a very small fraction of the laser beam energy reaches the detector, it is possible to define center positions for all holes assuming a plane wave. Furthermore, by using a weighting algorithm, it is possible to determine the center position for the reference and measured beams with subpixel resolution.

A very convenient way to describe the wave front of a Gaussian beam profile quantitatively is the use of Zernike polynomials, which construct a 2D orthogonal basis system on the detector plane [14]. The wave front amplitude \( W(x, y) \) can be written as

\[
W(x, y) = \sum_{i=0}^{\infty} c_i Z_i(x, y).
\]

The Zernike polynomials \( Z_i \) are selected because their coefficients \( c_1, c_2, c_3, \ldots \) correspond to specific aberrations of the wave front. In this description, the wave front amplitude \( W(x, y) \) can be given in nm or \( \lambda \). For example, for a plane wave, the wave front amplitude is zero or constant over the whole beam. Thus, it is possible to analyze the measured wave fronts and determine exactly which aberrations contribute to the final wave front distortion. In practice, the reconstructed higher order coefficients are not very reliable and easily affected by experimental noise so that we restricted ourselves to the use of the lower order polynomials such as divergence \( (Z_4) \), astigmatism \( (Z_3, Z_5) \) and partly trefoil \( (Z_6, Z_9) \) and comatic aberrations \( (Z_7, Z_8) \).

3. Experimental results

3.1. Optimizing the gas pressure

After setting up the harmonic sources and calibrating the wave front sensor, we optimized the harmonic source. In a first experiment we varied the backing pressure for the gas tube for a fixed focal position of \( z = 1.5 \text{ mm} \) (see section 3.2 for further information) and measured the temporally integrated photon flux with the CCD. The results are shown in figure 3. The red curve represents the number of spatially integrated counts on the x-ray CCD camera as a function of backing pressure. The black curve gives the peak intensity, meaning the highest counts at one pixel, which is usually near the optical axis. One can see a characteristic trend where almost no harmonics are generated at very low pressure. Then, for pressures from about 25 mbar to about 100 mbar, the XUV radiation increases roughly with the square of the pressure. At about 100–120 mbar, the signal reaches its maximum before declining again. At the maximum, we have a peak brilliance of about \( 2 \times 10^{18} \text{ photons s}^{-1} \text{ sr}^{-1} \text{ mm}^{-2} \) per 0.1\% BW. First of all, it is not trivial that both curves have their maxima at roughly the same pressure, which gives the first evidence that the optimal pressure in terms of conversion efficiency also corresponds to the highest brilliance.

To underline this statement we measured the beam profiles with the Hartmann sensor. In figure 4, we show the beam profile reconstructed from the measured intensity distribution and local tilt of the wave front for two different gas backing pressures. For 120 mbar, the photon flux reaches its maximum and the beam profile can be well fitted to a Gaussian-like distribution. But also for a backing pressure of 220 mbar, where the photon flux is almost one order of magnitude
Figure 3. Plot of the peak intensity (black squares, solid line) and integrated intensity (red triangles, dashed line) versus gas pressure at $z = 1.5$ mm. The divergence (blue circles, dotted line) has its minimum in the same pressure range where the intensities peak (110–130 mbar). The lines here are just a guide for the eye.

smaller, no obvious degradation of the beam profile is visible. For a more quantitative statement, we calculated the coefficients for a few low-order Zernike polynomials. In figures 4(c) and (d), we plot the terms representing the astigmatism and, in figures 4(e) and (f), we plot the astigmatism together with the high-order terms.

However, the high-order distortion cannot explain the different behavior of the peak intensity and the integrated intensity for pressures higher than 200 mbar. An obvious explanation is a divergence as a function of the gas pressure. In figure 3, we have also plotted the corresponding Zernike coefficient as a function of pressure. The divergence (blue curve) has a distinct minimum in the region of 110–140 mbar corresponding to the maximum signal. Together, this leads to the conclusion that—as stated above—the best pressure for the overall harmonics generation provides also the least divergent beam in the XUV regime.

For applications requiring a tight focus, a good beam quality is indispensable [20, 21]. The additional divergence can be easily compensated by adjusting the lens, but higher order aberrations like astigmatism cannot be compensated for in such an easy way. Thus, we also checked whether astigmatism depends on the gas pressure. The astigmatism of the harmonic beam dependent on the gas pressure reveals some interesting behavior, as is shown in figure 5. The black curve (45° astigmatism) refers to a different divergence of the laser beam along the two diagonal axes, resulting in a focal spot at one $z$-position that looks like a ‘forward slash’ and, at a second $z$-position, a second focus that looks like a ‘backslash’.

One can see that this aberration remains nearly constant and shows only little ‘quadratic’ behavior when the pressure is changed. The red curve (90° astigmatism) describes the same ‘two-focal-points’ phenomenon but this time the foci look like vertical and horizontal bars. Of course, in both cases the beam profiles at the focal spots do not form an exact bar but more an elliptic structure. However, it is important to notice that this second aberration depends much more on the gas pressure and its minimum coincides with the maximum intensity (compare with figure 3).
3.2. Optimizing the z-position

The second macroscopically adjustable parameter is the position of the focal spot with respect to the nickel tube. In our setup, we can move the focusing lens along the z-direction and thus place the focal spot in arbitrary distances to the gas medium. The z-position in these measurements refers to the relative scale of the micrometer reading of the translation stage holding the focusing lens. The focal spot and the gas-filled nickel tube coincide at a z-position of 0.0 mm. It is hard to set this up exactly because of the self-focusing effect in air and no easy way to directly measure it in vacuum. Thus, this number is an estimate given with the naked eye and has only a limited accuracy. During our measurements the gas medium was always behind the focus. Increasing numbers for the z-position mean moving the focus in front of the nickel tube further away. The Rayleigh length of the focal point in our setup is about 1.5 mm. We performed our measurements mostly within the Rayleigh range but were also able to record data outside of it.

When we look at the same observables (peak intensity, overall counts and divergence) as before in figure 3, we see a similar behavior with some peculiarities in figure 6. First, peak intensity and integrated counts are very much alike and differ only in magnitude.

Figure 4. Experimental examples of the spatial beam profile (a, b), wave front astigmatism (c, d) and higher order wave front (e, f) at two different gas pressures (a, c, e: 120 mbar; b, d, f: 220 mbar).
Figure 5. Comparison of the two contributions to the astigmatic aberration, the ‘diagonal’ 45° (black squares, solid line) and the ‘orthogonal’ 90° (red triangles, dashed line) part. While the 45° astigmatism remains nearly unchanged over the pressure scan, the 90° aberration has its minimum in the same pressure regime as the intensity (see figure 3). Again, the lines serve just as a guide for the eye.

The dotted blue curve shows the divergence and has a minimum at approximately \( z = 1.0 \text{ mm} \). This quantity does not change dramatically near the maximum counts, but at small and large distances between focal spot and gas tube, the divergence grows (which is in good agreement with [12]), while the harmonic intensity decreases. Interestingly, the count rates fall steeper when moving the focal spot toward the gas tube and do not decrease as fast when the lens is moved away from the jet. This can be explained by the importance of phase-matching: when approaching the gas jet, very little harmonics can be generated efficiently and the optimal phase-matching conditions require a focus in front of the interaction medium. Another reason why we can safely assume that the lack of phase-matching is the main reason for the steeper drop closer to the gas is the still acceptable divergence. If the lower count rate were caused by a distorted wave front, this would have been detected, which is not the case.

The astigmatism as a function of \( z \)-position as shown in figure 7 cannot be easily interpreted. The 90° astigmatism exhibits a clear plateau between 1.0 and 2.0 mm; in this region the aberration is comparatively small. Further away from the gas jet, the astigmatism increases, while at shorter distances, the astigmatism is not quite as pronounced. Unfortunately, we were not able to analyze the beam profile when the focus is exactly at and behind the gas jet, because of the low signal. Increasing the integration time was not possible because of a decreasing signal-to-noise ratio. With an improved laser system and experimental setup and a tighter focusing lens, we expect to be able to characterize the wave front in the whole range in the near future. In such a measurement series, we could not only study the influence of the \( z \)-position on the wave front in more detail but also evaluate the influence of the short and long trajectories on the beam profile.
Figure 6. Plot of the peak intensity (black squares, solid line) and integrated intensity (red triangles, dashed line) versus z-position (at $z = 0.0$ mm, the focus lies in the middle of the gas medium; positive $z$ values mean the focus is in front of the nickel tube). The blue dotted graph (circles) again gives the divergence of the beam during the position scan. Similar to figure 4, the divergence is lowest when the intensity of the harmonics is high.

3.3. Aberration corrections with phase masks

Finally, we studied the origin of the wave front distortions. For these experiments, we put a controllable phase mask (Hamamatsu PPM-SLM X8267) in the fundamental beam in front of the focusing lens. A similar setup has already been used to optimize the high harmonic yield. By applying a genetic algorithm, the signal could be enhanced in a free space focusing geometry [22], as well as in a capillary-based setup [23]. For the free focusing geometry it has been shown that a correction of laser beam aberrations increases the detected photon flux, but the harmonic beam profile has not been recorded. By exciting different spatial modes in a capillary, not only has the high harmonic yield been enhanced, but also the brilliance has been substantially increased. These experiments provide clear evidence that spatial shaping can improve parameters such as intensity and divergence.

We were not so much interested in an optimization of the flux as in the possibility of controlling the harmonic wave front by spatial beam shaping. The fundamental laser beam without the phase modulator has been carefully characterized and we found an $M^2$ of 1.22 and 1.15 for the horizontal and vertical directions, respectively. The excellent beam quality and the long focal length ($f/# = 15$) high-quality lens ensure an almost perfect Gaussian beam profile in the focal spot. Under optimized generation conditions, we measured a nearly perfect harmonic beam, as demonstrated in the measurements described above. Then we applied a phase modulation corresponding to astigmatism, tilt, divergence, trefoil or comatic aberration. We defined the phase pattern via Zernike polynomials with very large modulation depth. For such strong aberrations the detected signal decreased significantly. This can be explained by the
Figure 7. The two components of astigmatism (45° astigmatism: black squares, solid line; 90° astigmatism: red triangles, dashed line) are plotted versus focal position change. Both components change slightly with the focal position. Near the focal position for maximum intensity (compare figure 6), the astigmatism clearly shows a plateau where the aberrations remain constant.

very poor focus quality. Only in a very few cases did an applied wave front distortion improve the harmonic intensity. Here we assume that an unknown existing aberration was effectively compensated. If one of the above-mentioned aberrations was introduced by the SLM, we were not able to ‘recognize’ the applied wave front distortion by the Hartmann sensor. For example, applying an additional astigmatism onto the fundamental beam, we observed a lot more high-order aberrations on the harmonic beam, but no clear evidence for an additional astigmatism. This dependence differs from the results reported by Gautier et al [10] that astigmatism of the harmonics is influenced by the astigmatism of the fundamental laser. This difference might be due to the different target design, and further studies on this topic are necessary. From these observations, we conclude that there is a nontrivial correlation between fundamental and harmonic wave fronts. We consider it likely that easily accessible experimental parameters such as gas pressure or focal position have a larger and more evident impact on the harmonic wave front than spatial phase shaping. This does not mean that spatially shaping the wave front would not be a way to improve the XUV output, but it requires a feedback algorithm [24].

4. Conclusion

We investigated the dependence of harmonic output, brilliance and wave front on external parameters such as gas pressure and focus position. Doing so, we could demonstrate that adjusting these parameters to an optimal overall count rate automatically also delivers the best brilliance as well as the lowest aberrations. This statement is underlined by the quantitative analysis of the harmonics wave front in terms of Zernike polynomials. The divergence and
astigmatism show a clear dependence on the two adjustable parameters and reveal the best output when they are tuned for maximum harmonic photon counts.

Furthermore, we applied spatial phase masks containing pure aberrations (e.g. astigmatism and divergence) and analyzed the resulting XUV beam using Zernike polynomials and wave front reconstruction. We could see that the impact of artificially applied phase masks on the harmonics wave front is nontrivial and could not be easily identified with our evaluation technique. However, changing the macroscopic parameters revealed a strong influence on the wave front behavior and gave plausible results in terms of physical effects.

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