# High-order harmonic generation

Wahlström, Claes-Göran

*Published in:*
The 4th International Colloquium: X-ray Lasers 1994

*DOI:*
10.1063/1.47918

1995

# High-Order Harmonic Generation

C.-G. Wahlström

*Department of Physics, Lund Institute of Technology*

*P.O. Box 118, S-221 00 Lund, Sweden*

**Abstract.** A brief overview of high-order harmonic generation is given. Special emphasis is made on the use of this technique as a source of coherent radiation in the extreme ultraviolet. The experimental techniques as well as some characteristics of the generated radiation are presented. In particular, the wavelength range that can be covered and the number of photons per pulse that can be generated will be discussed and compared with other sources of radiation in this wavelength range. Finally some applications of radiation generated as high order harmonics will be briefly mentioned.

## EXPERIMENTAL SETUP

High-order harmonic radiation is generated when an intense laser field interacts with free atoms, ions or plasma gradients. In this paper only interactions with free atoms and ions will be covered. The experimental setup can look like the one in Figure 1. Laser pulses from a short-pulse high-power laser are focused by a lens into a low density ($10^{17}$-$10^{18}$ cm$^{-3}$) non-linear medium inside a vacuum system. In Figure 1 the medium is a jet of a rare gas emerging from a pulsed nozzle, but can also be, e.g., a plume of alkali-metal ions, produced by laser irradiation on a rotating solid target. In either case, the focused high-power laser interacts with

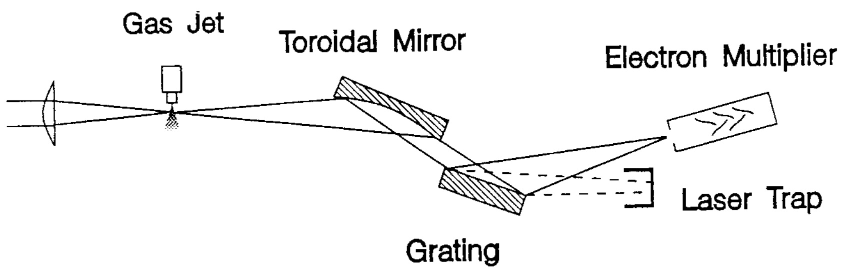


**FIGURE 1.** Example of experimental setup for high-order harmonic generation. From Ref [1].

 

free atoms or ions and only odd order harmonics are generated. The generated radiation and the laser beam propagate in the same direction after the interaction region and a VUV/XUV spectrometer is required to separate the laser radiation and the harmonics of different orders.

If a conventional spectrometer with an entrance slit is used, one must place the slit close to the interaction region to obtain a high collection efficiency. However, at this point the diameter of the diverging laser beam is still small, the intensity is very high, and there will be a severe risk for laser-plasma generation on the entrance slit. An alternative arrangement is to use the spectrometer without an entrance slit, placing the interaction region directly at the slit position (as in Fig. 1). With this arrangement, the grating is directly exposed to the diverging laser beam. To protect the grating from optical damage, the laser beam must diverge to a large enough diameter before reaching the grating. As will be discussed below, tight focusing, with a rapidly diverging laser beam after the interaction region, leads to much less efficient harmonic generation compared to loose focusing. The grating, therefore, must be placed at a relatively large distance from the interaction region. Finally, the harmonics can be detected, e.g. using an electron multiplier tube or a micro-channel-plate detector.

## HARMONIC SPECTRA

The yield of the harmonics, according to lowest-order perturbation theory, is expected to drop rapidly with increasing order. This is indeed observed for the lowest orders, but from the 7th or the 9th harmonic, a *plateau* is found where the photon yields of different harmonic orders are roughly the same. The plateau extends to increasingly higher harmonics as the peak intensity of the laser field is increased, but it always ends with a rather sharp cut-off. From recent experiments, harmonic orders as high as the 135th [1] and wavelengths as short as about 7 nm [2] have been reported. Both the formation and the extent of the plateau have been investigated by performing systematic studies of the intensity dependence of individual harmonics [3]. For the high order harmonics, the cut-off energy, i.e. the photon energy of the highest harmonic in the plateau, is found to increase linearly with laser intensity.

Numerical calculations, valid in the regime of tunneling ionization, have shown that the cutoff-energy in the single-atom response, is well approximated by the simple formula $I_p+3U_p$ [4]. $I_p$ is the ionization energy and $U_p$ is the ponderomotive energy, given by $Up(I) = 9.33 \cdot 10^{-14} \cdot I\lambda^2$, where the energy is expressed in eV, I, the intensity, in W/cm$^2$, and $\lambda$, the wavelength, in µm. (The maximum value of I in this expression is the saturation intensity for ionization,

$I_{sat}$.) In the experimental spectra, the cutoff energy is somewhat reduced due to intensity-dependent phasematching effects [5]. The constant of proportionality in the above expression is reduced from 3 to about 2, depending on the experimental conditions.

Some important observations can be made directly from the above expression for the cutoff energy. First, to generate harmonic radiation with very short wavelength (high cutoff energy), one should choose as non-linear medium atoms or ions with high ionization potential. Secondly, since the ponderomotive potential is proportional to the square of the laser wavelength, lasers in the near-infrared region can generate harmonic radiation with shorter wavelengths than, e.g. excimer lasers operating in the ultraviolet region. Finally, since $I_{sat}$ decreases with the duration of the laser pulses, very high harmonic orders (large $U_p$) require short pulse duration.

The most frequently used non-linear media for harmonic generation are the rare gases. The ionization potential as well as the saturation intensities for ionization for these gases decrease with atomic number. The harmonic conversion efficiency, on the other hand, increases with atomic number. For an application where radiation corresponding to a particular harmonic order is required, one should therefore choose as non-linear medium the heaviest rare gas possible for the given harmonic to be part of the plateau (which depend on the laser pulse duration). Some relevant parameters for the rare gases and estimates of the plateau extent in the single atom response ($I_p$+3$U_p$) and in a typical experiment ($I_p$+2$U_p$) are given in Table 1.

**TABLE 1.** Properties of the rare gases and estimated extents of the harmonic plateau cutoff when used as non-linear medium in high-order harmonic generation experiments.

| | $I_p$ (eV) | $I_{sat}$ ( 1ps, 1μm) ($10^{14}$ W/cm$^2$) | $I_p+3 \cdot U_p$ ($I=I_{sat}$) (eV) | $I_p+2 \cdot U_p$ ($I=I_{sat}$) (eV) |
|---|---|---|---|---|
| **He** | 25 | 7 | 230 | 160 |
| **Ne** | 22 | 5 | 170 | 120 |
| **Ar** | 16 | 1.5 | 60 | 45 |
| **Xe** | 12 | 0.7 | 25 | 21 |

The ionization energies and saturation intensities for the rare-gas ions or rare-gas-like alkali-metal ions are substantially higher than for the corresponding neutral atoms. According to the $I_p+3U_p$ formula, higher harmonic orders should therefore be possible to generate in these ions. A numerical calculation [6] for $He^+$, using a 527 nm laser at $5x10^{15}$ W/cm$^2$, predicts a cut-off energy well above 400 eV. This implies that coherent radiation in the biologically interesting "water window" (2.3 - 4.4 nm) might be possible to generate as high-order harmonics of a laser field in the visible. However, when producing ions through photo ionization, a significant electron densitiy is simultaneously produced. The dispersion of these free electrons severely reduce the macroscopic efficiency, in particular if long-wavelength lasers are used.

## PHASE MATCHING

The existence of a macroscopic field requires that there be proper phase matching between the induced and the driving fields. There are essentially two reasons for the phases not to be matched: Firstly, the dispersion in the non-linear medium makes waves with different frequencies travel at different speeds in the medium and therefore get out of phase with each other. (For frequency doubling, etc. in the visible or infrared spectral regions, this effect is circumvented by the use of birefringent crystals.) In most of the recent high-harmonic generation experiments, using gas jets, the atomic density was low so the dispersion was small. Ionization of the medium, however, creates free electrons that can introduce a substantial phase mismatch between the generated harmonics and the driving polarization.

The second reason for the phases not to be matched is connected to the unavoidable phase shift of $\pi$, occurring in a light wave in passing through a focus. Using a loosely focused laser beam, and a short non-linear medium, only a small fraction of this phase shift occurs in the medium. However, even if the shift inside the medium is only a fraction of the total phase shift $\pi$, the difference in phase between the driving field and the generated field will be $q$ times as large for a harmonic of order $q$. For sufficiently high harmonics, therefore, the phase lag will reach $\pi$, after a distance $L_{coh}$ (the coherence length) in the medium, resulting in destructive interference.

The focusing geometry has a drastic effect on the macroscopic efficiency. A useful quantity that characterizes the focus is the confocal parameter, b. For a Gaussian beam, b is equal to twice the distance on the propagation axis over which the beam section increases by a factor of two. It has been found that in the tight focusing limit, the measured number of photons is proportional to $b^3$ [7].

## CHARACTERISTICS OF THE RADIATION

There has been no direct measurements of the spatial coherence reported yet. However, measurements of the angular structures of the harmonic radiation have been reported [8]. Harmonic orders that are in the cut-off region show smooth, narrow angular profiles. Harmonics in the plateau, on the other hand, are often broader and can even exhibit ring structures. When preparing an experiment where XUV radiation with good focusability and corresponding to a particular harmonic order is needed, one should therefore choose the intensity so that the harmonic order in question is just at the end of the plateau or in the beginning of the cutoff region. Instead of decreasing the available laser energy, and thereby reducing the harmonic photon flux, one can change the focusing condition. By increasing the focal cross section (proportional to the confocal parameter, b), one can take advantage of the $b^3$ rule and increase the total flux at the same time as the spatial profile is improved.

Using peak intensities exceeding the saturation intensity for ionization leads to rapid ionization in the medium during the risetime of the laser pulse. The rapid temporal variation in the density of free electrons induces a time-dependent change in the refractive index, and consequently, a spectral blueshift of the fundamental field and of the generated harmonic field. The blueshift of the harmonic field is found to be due mainly to the shift of the fundamental. It is therefore approximately equal to the shift of the fundamental divided by the process order [3]. In Figure 2, the normalized spectral profiles of the 15th harmonic, obtained in xenon with a short-pulse laser are shown for a fixed peak intensity and different gas pressures.

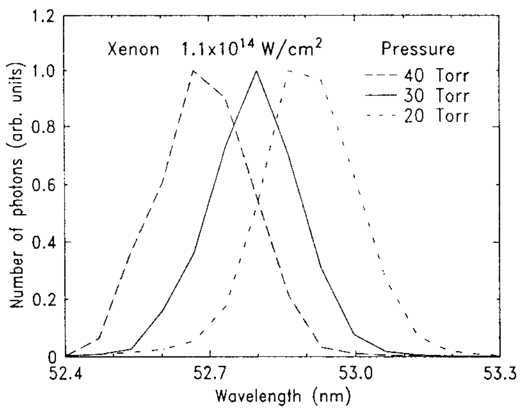


**FIGURE 2.** Normalized spectral profiles in Xe, obtained with a fixed peak intensity and different gas pressures. From Ref [3].

In order to get an idea of the characteristics of the radiation, generated as high order harmonics from a short-pulse laser, various parameters are presented in Table 2 together with the corresponding numbers for an undulator. The numbers for the harmonic radiation are estimates based on experiments performed with the 150 fs, 150 mJ Ti:sapphire laser of the Lund High-Power Laser Facility [9], operating around 800 nm. ($10^8$ photons per pulse, given in the table, corresponds to a pulse energy of about 1 nJ.) The undulator data are corresponding estimates for the new undulator under construction at the MAX II, 1.5 GeV synchrotron facility in Lund [10]. The comparison is made at 100 eV, corresponding to about the 63rd harmonic. If the comparison would be instead made at, say, 30 eV, the number of photons per pulse in the harmonic generation case would be at least two orders of magnitude higher since a heavier gas could be used. Making the comparison at much higher photon energies, on the other hand, would greatly favor the synchrotron/undulator source. (Above about 170 eV harmonic radiation has not yet been reported experimentally while synchrotron radiation with much higher photon energies are readily generated.) The first observations to be made from Table 2 is that the average power obtained from an undulator is considerably higher due to the very high repetition rate. The peak power, on the other hand, is several orders of magnitude higher in the case of harmonic generation. The two sources therefore complement each other and are best suited for different applications. Another, very important difference between these sources is the

**TABLE 2.** Estimated characteristics for radiation at 100 eV, generated as high-order harmonics of a 150 fs Ti:sapphire laser or in an undulator at a 1.5 GeV storage ring.

| | **Harmonics** | **Undulator** |
|---|---|---|
| | 150 mJ @ 1.5eV<br>150 fs, $\eta = 10^{-8}$ | Max II<br>200 mA<br>1.5 GeV |
| Photons /pulse | $10^8$ | $10^5$ |
| Repetition rate | 10 Hz | 500 Mhz |
| $P_{ave}$ | 15 nW | 1 mW |
| Pulse width | 0.1 ps | 20 ps |
| $P_{peak}$ | 15 kW | 0.1 W |
| Bandwidth | 0.01% | 0.1% |
| Coherence cycles | 3000 | 100 |

pulse duration. With a short-pulse laser, the harmonic pulse is short as well. (However, great care must be taken in order to maintain the short pulse duration when separating the harmonics of different orders from each other and from the fundamental radiation.) Many potential applications of high-order harmonic radiation depend on the ultra-short pulse duration. Experiments based on pump-probe technique for dynamic studies, e.g., molecular kinetics and surface studies, frequently require sub-picosecond resolution. These experiments cannot be performed with synchrotron radiation, but harmonic generation might be the ideal radiation source.

Comparing harmonic generation as a source of XUV radiation with existing x-ray lasers, the most striking differences are tunability, repetition rate and pulse energy. The wavelength of the harmonic radiation follows directly the laser wavelength, so a tunable high-power laser can be used to generate tunable radiation in the XUV. Even if the laser is tunable over only a limited wavelength range, the coverage in the XUV becomes complete by the possibility of choosing harmonics of successive orders. Ti-sapphire terawatt lasers with pulse durations in the 100 fs range, suitable for efficient high-order harmonic generation, can frequently be operated at repetition rates of 10 Hz or more. Most conventional x-ray laser schemes, on the other hand, require driving lasers with considerably higher pulse energies and correspondingly lower repetition rates. Therefore, x-ray lasers usually have considerably lower repetition rates than harmonic-generation sources and are best suited for applications requiring single-shot exposure. (X-ray lasers based on optical-field ionization might, one day, prove to be exceptions.) The pulse energies of conventional x-ray lasers, on the other hand, are in many cases several orders of magnitude higher than the most efficient harmonic generation source existing today. This might strongly favor x-ray lasers in applications such as x-ray holography. Some x-ray lasers have also been made to operate at shorter wavelengths than so far obtained with harmonic generation. X-ray lasers and harmonic generation sources therefore complement each other and might be found to be useful in completely different applications.

## APPLICATIONS

In a recent experiment harmonic radiation in the 10 - 120 eV range was used to measure photo-ionization cross sections in various rare gases [11]. This is one example of an experiment that could have been performed using synchrotron radiation. However, the experiment proved that "synchrotron experiments" can be brought into normal sized laboratories with table-top equipment. In another experiment time-resolved photoemission studies on surfaces was performed in a pump-probe experiment with picosecond resolution [12]. This experiment, in the VUV/XUV, could not have been done with any other conventional source due to the high temporal resolution required. This experiment thus illustrates how the harmonic generation source can open up new fields of investigations. The very high peak intensities expected in the XUV by focusing the harmonic radiation by suitable x-ray optics might open up the field of multi-photon processes and non-linear optics in the XUV.

## REFERENCES


1. L'Huillier, A. and Balcou Ph., *Phys. Rev. Lett.* **70**, 774 (1993).
2. Macklin J.J., Kmetec J.D. and Gordon III C.L., *Phys. Rev. Lett.* **70**, 766 (1993).
3. Wahlström C.-G., Larsson J., Persson A., Starczewski T., Svanberg S., Salières P., Balcou Ph. and L'Huillier A., *Phys. Rev. A* **48**, 4709 (1993).
4. Krause J.L., Schafer K.J., and Kulander K.C., *Phys. Rev. Lett.* **68**, 3535 (1992).
5. L'Huillier A., Lewenstein M, Salières P., Balcou Ph, Ivanov M. Yu., Larsson J. and Wahlström C.-G. *Phys. Rev. A* **48**, R3433 (1993).
6. Xu H., Tang X. and Lambropoulos P., *Phys. Rev. A* **46**, R2225 (1992).
7. Lompré L.-A., L'Huillier A., Monot P., Ferray M., Mainfray G. and Manus C., *J. Opt. Soc. Am.* **7**, 754 (1990).
   Balcou Ph., Cornaggia C., Gomes A. S. L., Lompré L.-A. and L'Huillier A., *J. Phys. B* **25**, 4467 (1992).
8. Tisch J.G.W., Smith R.A., Ciarrocca M., Muffett J.E., Marangos J.P. and Hutchinson M.H.R. *Phys. Rev A* **49**, 28 (1994).
   Salières P., Ditmire T., Budil K.S., Perry M.D. and L'Huillier A., *J.Phys B Letters*,
   Peatross J. and Meyerhofer D.D., to appear in *the Proceedings of the Int. Conf. on Multiphoton Processes*, Quebeck, Canada, July 1993.
9. Svanberg S., Larsson J., Persson A. and Wahlström C.-G., *Physica Scripta* **49**, 187 (1994).
10. Werin S. (1994), Private Communications.
11. Balcou Ph., Budil K.S., Ditmire T., Perry M.D., Salières P. and L'Huillier A., *Preprint* (1994).
12. Haight R. and Peale D.R., *Phys. Rev. Lett.* **70**, 3979 (1993).