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Invited Paper

Generation and applications of high-order harmonic radiation

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ABSTRACT

Coherent radiation in the extreme ultraviolet and soft X-ray spectral regions can be generated as high-order harmonics by focusing a high-power, short-pulse laser into a jet of free atoms or ions. This process is of basic physics interest, as a probe of the behavior of an atom strongly perturbed by an electromagnetic field. At the same time, the generated radiation has unique properties of short pulse length, high peak power and high spectral brightness which can be utilized in various applications. The perspectives for optimizing this process, both in efficiency, spectral range, and characteristics are discussed. Examples are presented, illustrating how this new source of coherent radiation can be used in different applications.

1. INTRODUCTION

Recent developments in laser technology and in particular the advent of table-top-terawatt lasers has made possible the efficient generation of coherent radiation in the extreme ultraviolet (XUV) and soft X-ray spectral regions as high-order harmonics. In the high-intensity (10^{13}- 10^{15} W/cm^2), low-frequency regime, the harmonic spectra are characterized by a broad plateau of nearly constant conversion efficiency, followed by an abrupt cutoff. Harmonics of very high orders, up to the 109th of 806 nm[1] and the 135th of 1053 nm[2] radiation using neon as the non-linear medium and up to the 143rd of 1053 nm radiation in helium[3] have been reported. Using rare gas ions as non-linear medium, harmonic orders up to the 37th of a KrF laser (249 nm) have also been observed recently[4]. All of these correspond to the generation of coherent radiation down to about 7 nm (170 eV).

This coherent XUV radiation has unique properties. It is well collimated and its spectral and temporal characteristics follow those of the pump laser. A short-pulse laser produces short-pulse harmonics (of duration shorter than that of the pump laser). A narrow-bandwidth laser (which implies that its pulse duration cannot be too short) produces narrow-bandwidth harmonics. The brightness of the harmonic radiation is several orders of magnitude higher than that achieved with more traditional XUV sources.

High-order harmonics now begin to be used as a source of radiation for various applications. In the present paper, presented at a meeting on *X-ray lasers and applications*, we will therefore discuss the current status of the high-order harmonic generation field from the point of view of a potential user. First we will describe the essential parts of an experimental setup. Then we continue by discussing the limits for the production of high-order harmonics, both in energy and in number of photons. Section 4 summarizes the main results concerning the characteristics of these harmonics, in particular in view of applications. In Section 5, we make a brief comparison between harmonic radiation and other "conventional" XUV radiation sources and, finally, in section 6 we present a few recent experiments demonstrating the applicability of high-order harmonics as an XUV source of radiation in atomic spectroscopy.

2. EXPERIMENTAL SETUP

The experimental setup can, in principle, be rather simple. Laser pulses from a short-pulse, linearly polarized, 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. This medium is usually in form of a jet of rare gas atoms emerging from a pulsed nozzle,

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but can also be, e.g., a plume of alkali-metal ions, produced by laser irradiation on a solid target. In either case, the laser interacts with free atoms or ions and therefore 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. To protect the spectrometer from optical damage by the intense laser beam, the beam must diverge to a large enough diameter before reaching the slit or grating. The slit, or if a slit-less arrangement is used, the grating, must therefore be placed at a relatively large distance from the interaction region. This leads either to low collection efficiency, or to the need of a large grating. Finally, the harmonics are detected, e.g. using an electron multiplier tube or a micro-channel-plate detector. Alternatively, the radiation can be sent into a second chamber where it is used for applications.

3. OPTIMIZATION OF HIGH-ORDER HARMONICS

3.1 Extension of the harmonic plateau

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. A major breakthrough in the understanding of this fact came in 1992, with the prediction of Krause *et al.*[5]. By performing time-dependent calculations for a number of atomic systems under various conditions of laser intensity and frequency, they showed that the width of the single-atom plateau varies as I_p+3U_p where I_p is the field-free ionization potential of the atom and U_p the ponderomotive energy. $U_p(\mathrm{eV}) = 9.33\cdot 10^{-14}\cdot I\cdot\lambda^2$ where I is the laser intensity (in W/cm^2) and λ the laser wavelength (in µm). The extent of the harmonic plateau obtained experimentally in rare gases[6,7] is found to be shorter than in the single-atom response. It varies with the intensity as approximately $I_p+\alpha\cdot U_p$ with $\alpha \approx 2\text{-}2.5$. Results obtained in Ne are shown in Fig. 1. This reduction in slope can be understood by considering the effect of propagation in the nonlinear medium, and more precisely by including the geometrical phase mismatch introduced by focusing[7]. These scaling laws are valid up to the intensity at which the atom becomes ionized (I_{sat}).

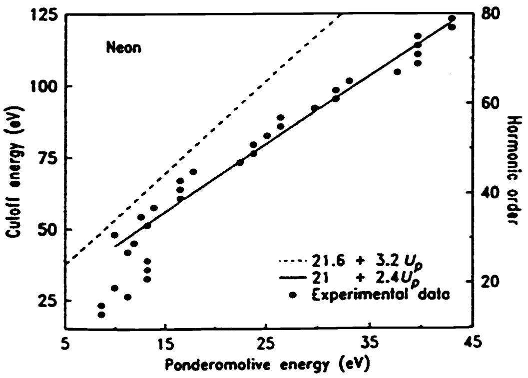

Fig. 1 Cutoff energy in neon as a function of the ponderomotive energy. The solid line is a linear best fit: 21+2.4 Up, the dashed line is the prediction of the single-atom response 21.6+3.2 Up. From Ref 6.

Some important observations can be made directly from this 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, in a given non-linear medium, 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 Up) require short pulse duration.

The most frequently used non-linear media are the rare gases. The ionization potentials 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).

3.1.1 Harmonic generation by ions

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 Ip+3Up formula, much higher harmonic orders should therefore be possible to generate in these ions. Coherent radiation in the biologically interesting "water window" (2.3 - 4.4 nm) might even be possible to generate as high-order harmonics of a laser field in the visible. The simplest way to create an ionic medium is to use the same laser pulse to ionize the neutral gas and to generate the harmonics. If the intensity is high enough, the ions are produced during the leading edge of the pulse, so most of the pulse propagates through an ionized medium. This leads to poorer phase matching, due to the free-electron dispersion and to defocusing of the fundamental beam which decrease the intensity in the medium. This severely reduces the macroscopic efficiency, in particular if long-wavelength lasers are used.

Harmonic generation from rare-gas ions has been observed using high-frequency lasers (KrF), for which the effect of the free electrons is of much less importance[8,4]. Experiments have also been performed in plasmas of alkali-metal ions, created by focusing a laser on a solid alkali-metal target, both with a high-frequency[9] and low-frequency[10] laser. The number of harmonics observed by Wahlström *et al.*[10] was far below that expected according to the single-atom cutoff law. This effect, however, was at least partly explained by ionization induced defocusing of the fundamental field, which reduced the effective intensity in the medium.

To summarize the situation regarding the spectral range reached by high-order harmonic generation, in contrast to the expectations of the single-atom cutoff law, experimentalists have not managed, so far, to reach energies much beyond the 170 eV obtained by Macklin *et al.*[1] three years ago. Propagation effects limit the photon energy, especially for ions excited by a low-frequency laser.

3.2 Optimization of the conversion efficiency

Focusing conditions have been shown to considerably affect the conversion efficiency[11]. Focusing as weak as possible should be used in order to optimize phase matching. The optimum conversion efficiency depends, of course, on the laser power available, since the intensity at the focus must be high enough to generate harmonics (to reach the plateau level).

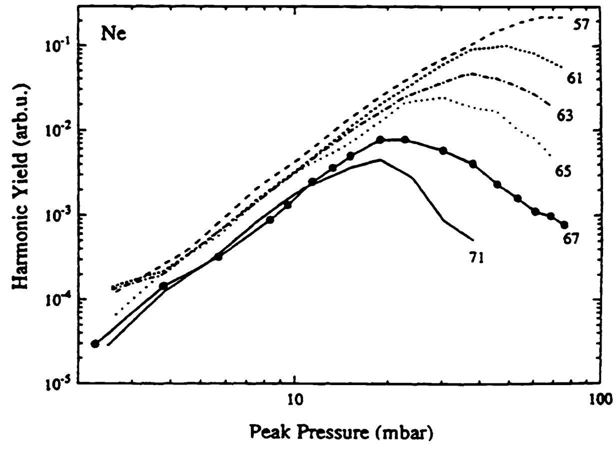

FIG 2 Number of photons at different harmonic frequencies (from the 57th, in the plateau to the 71st, last visible harmonic) as a function of atomic pressure. The laser used was a 150 fs Ti:S system. From Ref 12.

Another simple way to optimize the conversion efficiency is to increase the number of atoms in the interaction region. Harmonic generation is a coherent process, which is expected to vary as the square of the atomic density. However, the dispersion in the medium (due to atoms, ions and free electrons), which increases with the density, might alter this simple scaling. In a recent investigation by Altucci *et al.*[12], the number of photons produced was studied as a function of pressure for different process orders, laser intensities, etc. In general, the number of photons increased as the square of the atomic density over the range investigated (3-80 mbar). However, an interesting effect was observed in the cutoff region, which is shown in Fig 2. The harmonic yield goes through a maximum and then decreases. The pressure corresponding to this maximum decreases as the process order increases. Fig. 2 shows the evolution of several harmonics from the 57th, in the plateau region, to the 71st, the last visible harmonic in the cutoff. The intensity was estimated to be $6 \cdot 10^{14}$ W/cm^2.

This result implies that the cutoff region is significantly affected by the atomic density. The interpretation of these data shows that it may be understood by a defocusing of the fundamental beam. Increasing the atomic density increases the efficiency, but, at the same time, it leads to a reduction in the effective intensity in the medium, induced by defocusing, and, consequently, to a limitation of the plateau range.

Apart from these two parameters (geometry and atomic density), which affect the macroscopic conditions of the interaction, one can also change the parameters which affect the microscopic part. The efficiency of the conversion process is optimized by using high-frequency lasers and heavy rare gases, to the detriment, however, of the spectral range, as demonstrated by the cutoff law. In certain cases, where relatively low-order harmonics are wanted, it can therefore be useful to first frequency double the laser frequency in a non-linear crystal, before generating the harmonics in the gas.

3.3 Tunability

For a given frequency of the pump laser, the radiation consists of many different harmonics separated by twice the laser photon energy, (typically 1-3 eV), which can be selected in turn. Most applications demand, however, a finer tunability. Ti:sapphire lasers can, in principle, be tuned between about 720 and 900 nm, but not easily. For these chirped-pulse amplification systems, each change in wavelength requires, in general, a laser realignment. Dye lasers are easier to tune. By tuning the laser over a limited range and selecting different harmonic orders, the radiation may be continuously tuned over a large range in the XUV. Wave-mixing techniques, involving an intense fixed-frequency laser and a weaker tunable laser, such an Optical Parametric Amplifier (OPA), present alternative ways of obtaining continuous tunability of the harmonic radiation[13,14].

4. CHARACTERISTICS OF HIGH-ORDER HARMONICS

In this section, we summarize the results concerning the characterization of the harmonic emission.

4.1 Temporal and spectral properties of the harmonics

The temporal profiles of the harmonics have, so far, only been measured for low orders, generated from relatively long-pulse (picosecond) lasers[15,16], for obvious technical reasons. In general, from these measurements, as well as from numerical calculations[17,18], one can expect the harmonic pulse duration to be shorter than that of the incident laser, and larger than or equal to that given by the perturbative limit, i.e. $\tau_L/\sqrt{q}$, q being the process order and τ_L the laser pulse width. (Note that gratings used to separate the harmonics can introduce an unwanted temporal spreading.) The short-pulse aspect of the radiation might be the most interesting aspect of harmonics as a soft-X ray source. It opens the possibility to perform pump-probe experiments, with one pulse, the probe or the pump, in the XUV region. Numerous applications can be foreseen in atomic and molecular spectroscopy, solid-state physics, plasma physics, etc.

The spectral width of the harmonics of a short-pulse laser is quite large, owing to Heisenberg's principle. Typical relative widths of high-order harmonics, generated by sub-picosecond lasers, are[19] $\Delta\lambda/\lambda = 10^{-3}$. Moreover, 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 broadening and blueshift of the fundamental field and of the generated harmonic field [6].

For applications demanding better spectral resolution, one can use longer-pulse lasers (in the picosecond range). For example, the spectral width of the 13th harmonic of the tunable 80 ps dye laser used in Lund for radiative lifetime measurements[20], was measured to be 0.01 nm, which corresponds to $\Delta\lambda/\lambda = 3\cdot10^{-4}$.

4.2 Spatial properties of the harmonics

There has been no direct measurements of the spatial coherence reported yet. However, several measurements of the angular structures of the harmonic radiation have been reported[21,22,23]. 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, corresponding to a particular harmonic order is needed, one should therefore choose the intensity so that this harmonic order 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 should change the focusing conditions. By increasing the focal cross section, one can increase the total flux at the same time as the spatial profile is improved.

The understanding of the spatial coherence of the harmonics, which is very important in view of applications, has stimulated several theoretical studies[18,24,25]. All of them point out the influence of phase effects (phase of the dipole emission, dispersion due to free electrons, etc.) on the angular emission. Salières *et al.*[18] show that the spatial and spectral characteristics depend critically on the focusing conditions, more precisely on the position of the laser focus relative to the atomic medium.

5. COMPARISON WITH OTHER XUV SOURCES

To get an idea of when radiation generated as high-order harmonics might be the appropriate source for XUV radiation, comparisons with an undulator at a modern storage ring, and with X-ray lasers will be discussed.

5.1 Harmonics versus undulators

At 100 eV, harmonic radiation generated with the 150 fs, 150 mJ Ti:sapphire laser of the Lund High-Power Laser Facility[26], gives typically about 10^8 photons (~1 nJ.) per pulse at 10 Hz repetition rate. With the new undulator under construction at the 1.5 GeV synchrotron facility in Lund, MAX II, the corresponding pulse energy is about three orders of magnitude lower[27]. The repetition rate with the undulator, of course, is much higher; 500 MHz. The *average power* is therefore considerably higher with the undulator. The *peak power*, on the other hand, is about five orders of magnitude higher in the case of harmonic radiation. This is partly due to the higher number of photons per pulse, but also to the much shorter pulse duration; about 0.1 ps for the harmonics compared to about 60 ps from the undulator. The two sources therefore complement each other and are best suited for different applications.

This comparison was made at 100 eV, corresponding to about the 63rd harmonic. If the comparison would have been made instead 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.

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.

5.2 Harmonics versus X-ray lasers

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 duration 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, e.g., 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.

6. APPLICATIONS OF HIGH-ORDER HARMONICS

In a recent experiment by Balcou *et al.*[19], harmonic radiation in the 15 to 104 eV range was used to measure photo-ionization cross sections in various rare gases. They measured the photoionization cross-section from the threshold to about 43 eV in Xe, Kr and Ar and up to 104 eV in Ne using the successive harmonics of a 140 fs LiSAF laser. The rare gas used to produce the harmonic radiation was selected on the basis of the XUV spectral range desired. Argon gave a high yield of harmonics extending to the argon cutoff at the 29th harmonic (43 eV). Using neon, harmonics beyond the 81st (120 eV) could be produced, but with a lower conversion efficiency. 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, Haight and Peale[28] performed time-resolved photoemission studies on surfaces in a pump-probe experiment with picosecond resolution. 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. Up to the 19th (39 eV) of a 150 fs dye laser system was also used to do atomic core level spectroscopy[29].

In a more specific application, Larsson *et al.*[20] utilized the specific properties of the harmonics, namely, its relatively narrow bandwidth and short pulse duration, as well as the possibility of synchronization with a second laser pulse. They measured the short (subnanosecond) radiative lifetime of the excited 2p ^1P state in helium in a pump-probe configuration, using the 13th harmonic of a 80 ps tunable dye laser. The harmonic resonantly excited the atomic state, which was subsequently ionized by a laser pulse. The experimental setup is shown schematically in Fig 3. A distributed feedback dye laser oscillator was used to generate 80-ps pulses tunable between 715 and 900 nm. These pulses were amplified in two dye cells and in a Ti:S crystal up to an energy of 50 mJ[30]. Harmonics were generated in a pulsed jet of rare gas and separated by a normal-incidence spherical grating. The laser wavelength was tuned around 760 nm and the 13th harmonic (58 nm, 21 eV) was selected as the pump pulse. The excited atoms are subsequently ionized by the third harmonic (355 nm, 3.5 eV) of a fraction of the 80 ps Nd-YAG laser whose second harmonic was used to pump the dye laser (see Fig. 3). The two beams crossed at 45 degrees inside a time-of-flight spectrometer. A variable delay line placed in the beam path of the probe beam allowed the relative time between the two light pulses to be adjusted. The ions generated were separated in mass in the time-of-flight tube and detected by a microchannel plate (MCP).

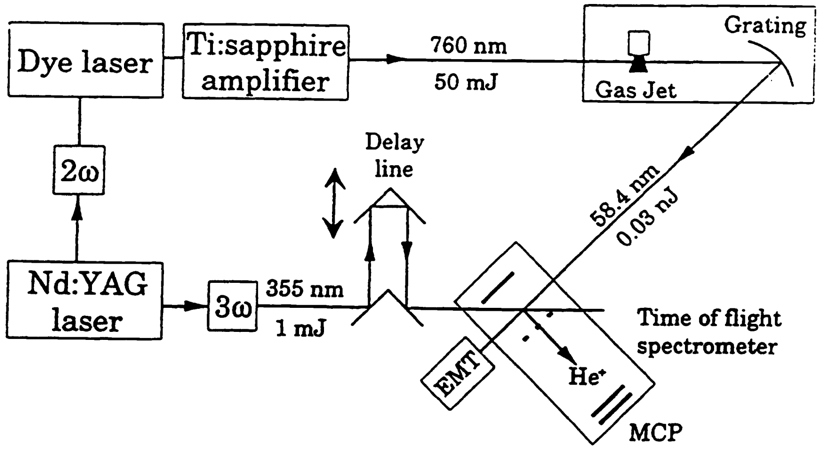

Fig. 3 Experimental setup for the pump-probe experiment. From Ref 20

This experiment demonstrates that the high-order harmonics can be used for two-color pump-probe experiments. It enables cascade-free studies of radiative properties of atomic and ionic systems in the XUV spectral range. Note that it is possible to go to much shorter wavelengths and shorter pulse durations than those used in the present experiment, by using a pump laser with a shorter pulse duration. This could be useful for measurements of very short radiative processes.

The very high peak power and the spatial coherence in the harmonic pulses should make it possible to focus the harmonics to intensities never before explored in the XUV. Intensities exceeding 10^{13} W/cm^2 at 100 eV should be possible within the near future. This development, of course, relies on the capabilities of modern X-ray optics. A reliable assessment, at this stage, of specific applications of this high-intensity XUV radiation is difficult to make. However, one need recognise only how high-intensity optical lasers have altered the scientific and technological landscape to appreciate the potential of such a capability at soft X-ray wavelengths. Nonlinear X-ray optical phenomenon can be used in the study of fundamental physical processes, such as nonlinear excitations and polarisation of inner-shells in atoms and molecules. Its potential for use in a number applications related to material science and plasma diagnostics might, however, turn out to be even greater. The long-term potential of this high-intensity source should be clear to anyone with more than a passing interest.

7. CONCLUSIONS

In summary, we have discussed the characteristics of the harmonic radiation: spectral range and conversion efficiency; spectral and spatial coherence properties. The spectral range available is typically from a few eV to more than 150 eV. The extension to energies above 200 eV, expected when using ions instead of neutral atoms, has not been demonstrated so far. Propagation effects in a plasma must be better understood in order to make some progress in this direction. The spectral and spatial properties of the harmonic light, which depend on the experimental conditions, have been reviewed. Finally, we have presented feasability experiments, illustrating some of the properties of the harmonic radiation. We believe that high-order harmonics of short-pulse lasers will become a useful source, complementary in its properties and applications to traditional XUV sources.

8. ACKNOWLEDGMENTS

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