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Generation of High Harmonics of Coherent Radiation in the Extreme Ultraviolet Spectral Range in the Low-Intensity Regime

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Abstract. Generation of high harmonics is the only available way to obtain coherent extreme ultraviolet (EUV) and soft X-ray radiation in laboratory conditions. We present a system for high harmonics generation and study of its parameters. Capillary waveguide filled with Ar gas is used to convert the pumping laser radiation from a Ti:Sapphire mode-locked femtosecond laser system. In the capillary the laser interacts with the gas and generates high harmonics of orders from 13 to 31 with good spatial coherence. In the relatively low-intensity regime ($I \sim 10^{14}$ W/cm^2) we determined the optimum pressure for the generation of high harmonics with our system to be 36 mbar of Ar gas.

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1 Introduction

In the recent decades, the great interest in the generation of higher-order harmonic frequencies (HHG - higher-order harmonic generation) is due to the wide range of their possible applications in science and technology. A major contribution to their generation was the appearance of lasers capable of generating ultrashort pulses (with pulse durations of the order of tens of femtoseconds or less) with terawatt peak output powers. The amplitude of the electric field of pulses with such intensity reaches values exceeding the 10^9 V/cm range. The strength of these electric fields is larger than that of the Coulomb field retaining the electrons of the outer layers of atoms. Therefore, the laser field is strong enough to overcome the Coulomb potential and to cause ionization. Eventually, the above-threshold excitation process [1] can end up by recombination of high-energy electrons with ion cores leading to the generation of high harmonic frequencies [2,3].

Generation of harmonic frequencies was first observed in the University of Michigan in 1961 [4]. Ruby laser ensuring 105 V/cm strength of the electric field of the laser pulses at a wavelength $\lambda = 694.2$ nm was focused on anisotropic material (quartz) and generated the second harmonic frequency with a wavelength of 347.1 nm. In subsequent experiments on the generation of harmonic frequencies usually birefringent crystals are used as nonlinear media. To convert the frequency effectively, the electromagnetic waves corresponding to the input signal and the generated harmonic radiation must be synchronized in phase having the same phase velocities while passing through the nonlinear medium. Main method of achieving such phase synchronization is to use of birefringent materials and pump beams with carefully selected orientation of their polarizations, thereby ensuring that the fundamental and the harmonic waves propagate at the same speed in the optical medium.

Despite the undoubted advantages that the crystals have, their use is limited by their absorption of the generated harmonics with wavelengths of the order of 200 nm or less. For generation of high harmonic frequencies, which have much shorter wavelengths, the use of crystalline nonlinear media is impossible not only because they are not transparent to wavelengths in the deep ultraviolet but also because they cannot withstand the very high intensity of the pump radiation needed in high harmonic generation. In contrast to the solid materials gases allow the use of radiation with a high intensity and many of them are transparent to the extreme ultraviolet (EUV) and soft X-ray (XUV) range. The major problem with gases as nonlinear media is the inability to use the established methods for synchronization of the phase, since the gases are isotropic. In these early days of nonlinear optics it is worth mentioning the first generation of 5th, 7th and 9th harmonic of a Nd:YAG laser in alkali metal vapors by researchers of our Department [5-7].

The first generation of high harmonic frequency was achieved in 1987 [8]. The used laser emitted pulses at a wavelength of 248 nm with a peak intensity of $10^{15} - 10^{16}$ W/cm^2. Neon gas was used and 17-th harmonic frequency is generated in this work. Since then many attempts have been made in the field of generation of high harmonic frequencies by lasers with different wavelengths, pulse durations and intensities, and using various gases. The highly nonlinear nature of the ionization/excitation/recombination process [1-3] allows the generation of harmonic frequencies with wavelengths shorter than 3nm. One advantage of the generation of high harmonics is that the generated radiation is coherent, with polarization similar to that of the input radiation [9] and with a smaller divergence [10]. The low divergence of the high harmonic is particularly important for application in EUV lithography, ellipsometry and reflectometry [11], just to mention a few. Despite the huge number of possibilities for the use of high harmonics, the actual application is severely limited by their relatively low pulsed power.

The ultimate aim of our project is the development of a table-top system for generation of ultrashort XUV pulses. High harmonic generation (HHG) is now a widely accepted method for laboratory-based generation of coherent EUV and

XUV light. A XUV generation system based on HHG would provide a relatively cheap and compact source of coherent, short pulse XUV radiation. HHG produces pulses that are highly coherent, with a pulse length in the femtosecond or attosecond timescales thus opening the way for future precise time-resolved measurements. The scientific uses of such a system are widespread in a range of fields across the physical and life sciences.

2 Physical Background of the HHG Process

In a nonlinear-optical process, a coherent beam/pulse of light passes through a material, and some light is converted to a different frequency while retaining the coherent properties of the original beam/pulse. Microscopically, electrons in the material act as driven oscillators, responding to the electric field of an intense laser. In conventional nonlinear optics, these electrons remain bound to the atoms but are driven strongly enough that the potential binding an electron to its atomic core no longer appears to be a purely parabolic ("harmonic-oscillator") potential. As a consequence, the motion of the electrons themselves becomes anharmonic. The dipole moment of the atom thus re-radiates electromagnetic radiation, not only at the driving laser frequency, but also at higher harmonics of the driving laser, that correspond to the Fourier spectrum of the electron motion. High harmonic generation (HHG) takes this concept to its extreme. By further increasing the driving laser intensity, at some point the laser field is strong enough to ionize this electron. Once ionized, it will begin to oscillate in response to the field. Oscillating free electrons themselves do not emit harmonics in the nonrelativistic limit; however, some of these electrons can recollide with their parent ions after being driven for a fraction of a single optical cycle, and can recombine [1-4]. If this happens, the electron must give-up its kinetic energy at the time of the recollision, as well as the binding energy of the electron. It does this by emitting an energetic photon [8-13].

High-harmonic generation is observed in the interaction of high-power femtosecond laser wave with matter. By focusing such a laser beam/pulse to intensities of the order of $10^{13} - 10^{16}$ W/cm^2 into a gas jet [8,10] or into a gas-filled waveguide [11,12], the beams that emerge from the focal region contain not only the original laser frequency, but also a tightly-collimated beam of light at much higher frequencies, i.e. at gradually shorter wavelengths. The photon energies correspond approximately to odd-order harmonics of the original laser, i.e. to photon energies corresponding to $(2n + 1)h\nu$, where n is an integer. Photon energies in the range of tens to hundreds of eV can be generated. The gas used in HHG experiments is usually a noble gas because a high ionization potential is desirable. Argon, Neon and Helium are all commonly used [12,13]. High-harmonic generation is now an accepted method for laboratory based generation of coherent femtosecond EUV and soft X-ray light.

3 Experimental Setup and Results

The generation of high harmonics in our system (see Figure 1) is performed by a gas-filled capillary waveguide. Use of the capillary waveguide gives an extended interaction length between the laser and the gas at high intensity, and adds an extra means to control the generation process. In our experimental setup the used laser source was a commercial Ti:Sapphire mode-locked femtosecond system (femtosecond oscillator Ti-Light and a two-stage amplifier Integra-C UPS, Quantronix). It generates nearly chirp-free transform-limited pulses with temporal duration of about 47 fs (FWHM), emitted at a 1 kHz repetition rate, with an average output power of about 3.4 W at a central wavelength of 800 nm. The output of a Ti:Sapphire pulse amplifier system is focused with a lens of a long focal length ($f = 50$ cm) through a thin fused silica window into a vacuum chamber held at low pressure (less than 10^{-5} mbar). The high harmonic generator, which produces coherent radiation in the EUV and XUV ranges of the spectrum, is a module from KMLabs Inc. (model XUUS - eXtreme Ultraviolet Ultrafast Source). The HHG process occurs in a hollow quartz fiber, 55 mm long and with an internal diameter of 150μm. At both ends the capillary tube has holes to ensure the circulation of the noble gas (Argon in our case). The whole volume of converter box [14], except the working volume in the capillary tube, is evacuated. This is necessary, because of the large absorption of the generated radiation with $\lambda < 100$ nm in the air. The gas dispensing system consists of a module for fine adjustment of the pressure and of the flow of the working

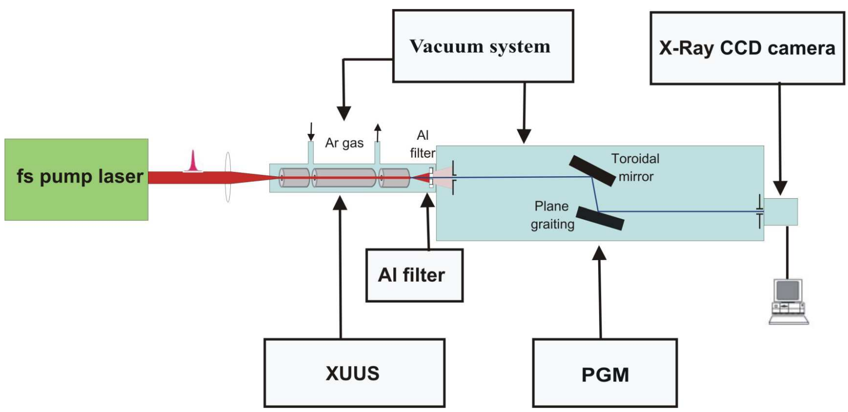

Figure 1. Experimental set-up for phase-matched EUV high-harmonics generation in a capillary waveguide. Fs pump laser – Ti:Sapphire mode-locked femtosecond system (47 fs/1 kHz/3.4 mJ/800 nm). The vacuum system ensures volume evacuation down to $p \sim 10^{-5}$ mbar. X-Ray CCD camera – Newton DO920-BN-9HC, 1024×255 pixels., (Andor Technology). XUUS – eXtreme Ultraviolet Ultrafast Source (KMLabs Inc.). PGM – plane grating vacuum monochromator with 1 m focal length (Horiba Scientific).

gas. To achieve HHG it is necessary to ensure the flow of clean gas through capillary tube with adjustable pressure in the range from 5 mbar to 300 mbar. The Argon gas we used was of a purity of 99.9995%. The capillary waveguide is placed near the focus of the laser beam. In the capillary the laser beam/pulse interacts with the gas and, eventually, generates high harmonics. In this regime, bright emission over a comb of harmonics of orders 13 to 31 was obtained, with perfect spatial coherence.

The high harmonics radiation has wavelengths in-between 26 nm to 66 nm, which actually falls in the so called "extreme vacuum ultraviolet" (EUV) range. That forced us to assemble a specific setup for signal registration and optimization. To control the low pressure gas flow through the hollow fiber, we used a precise electronic controllable valve. At the output of the XUUS converter, besides the high harmonics of the generated EUV radiation, there is also significantly stronger pump radiation. Passing to the registration system this radiation is able to cause serious disturbances in the measurement of the generated short-wave radiation. To overcome this problem, we used special ultra-thin metal foils with thicknesses from 50 nm to 500 nm (Lebow Company). These filters have good transmittance in the 10–100 nm range and a very good attenuation for the pump radiation with a wavelength of about 800 nm. The filters are placed in a special vacuum revolver with 3 positions, allowing selecting different combinations of filters. The technology of preparation of these filters is mechanically thinning of aluminum foil, which leads to the presence of pinholes in the area of the filter located in a random manner. To eliminate their influence, we put together a combination of at least two filters. The spectral distribution of the high harmonics was measured with a vacuum monochromator PGM-PGS 1000 with 1 m focal length (Horiba Scientific). This monochromator is especially designed for analyzing Extreme UV (EUV) to Far UV (FUV) radiation as a monochromator (slit-slit) or as a spectrograph (slit-CCD port). We used the spectrograph mode (slit-CCD). At the exit we placed special CCD camera, sensitive in the EUV spectral range (see Figure 1). In the particular measurement reported here we used specialized CCD camera NEWTON DO920-BN-9HC X-Ray Camera (ANDOR Technology) with a high sensitivity in the EUV part of the spectrum. The camera has 1024×255 pixels and is placed in a special housing that allows the sensor to be cooled down to -60°C in order to reduce the noise and keeping the possibility of prolonged exposure.

The spectral distribution of the pump radiation is shown in Figure 2.

One important parameter for the correct operation of the system is the length of the generated pump laser pulses τ_p. The pulse durations are monitored continuously during the measurements by a frequency-resolved optical gating device GRENOUILLE (Swamp Optics). The results presented here refer to $\tau_p = 40$ fs.

In order to achieve high brightness of the harmonic emission, the phases of the fundamental beam and the desired harmonic order need to be phase-matched, or

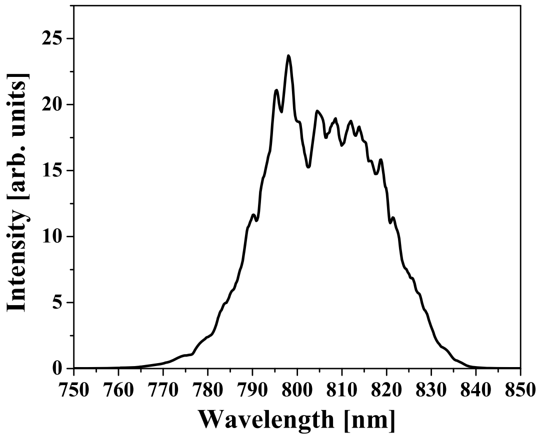

Figure 2. Spectral distribution of the femtosecond pump radiation.

the phase-mismatch needs to be minimized. This can be achieved by optimizing the pressure of the working gas Ar [14,15]. We performed series of measurements by changing the working pressure for achieving the optimal phase matching (see Figure 3). From the recorded spectra we determined the optimum pressure of Ar for the generation of high harmonics in our system to be 36 mbar.

The full spectrum of the generated higher harmonics at a pressure of 23 mbar of Ar gas is shown in Figure 4. It consists of spectral lines corresponding to the 13th – 31st harmonics of the pump radiation with a central wavelength of 800 nm.

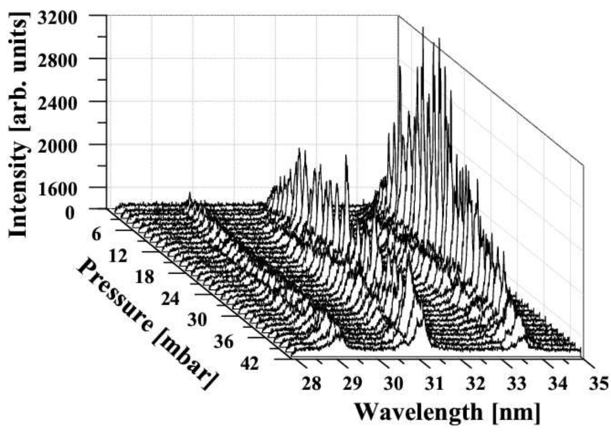

Figure 3. Relative intensities of the 25th, 27th, and 29th harmonic of the Ti:Sapphire laser vs. Ar gas pressure.

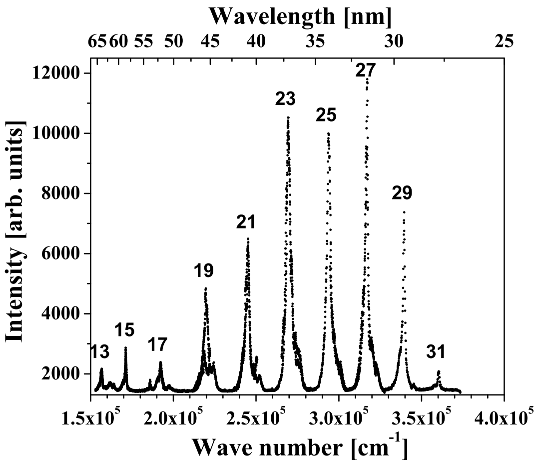

Figure 4. Spectrum of the HHG in Ar with pressure 23 mbar with identified orders of the high harmonics.

In the case of HHG the light-matter interactions are depending on the magnitude of the electric field in the interaction region relative to the ionization potential of the atoms [16,17]. One relevant quantity is the ponderomotive energy. It describes the average oscillation energy that is acquired by a free electron in the radiation field of the laser pulse. The ponderomotive energy U_p is given by the following relation

$$U_p = 9.34 \times 10^{-20} \times (\lambda\,[\text{nm}])^2 \times I\,[\text{W/cm}^2]\,. \tag{1}$$

U_p can be used as a criterion for determining whether the optical E-field is strong in comparison to the characteristic ionisation energy E_{Ar}. In the case of $E_{\text{Ar}} > U_p$ this is a low intensity regime and the electric field is strong enough to induce a perturbative non-linearity in the matter, but not strong enough to cause significant ionisation of atoms. In the opposite case, or when $E_{\text{Ar}} < U_p$, this is the high intensity regime. The electric field is sufficiently strong to provide a high probability of ionisation in the target material. With our parameters of the laser - pulse energy $E_p = 3.5$ mJ, pulse duration $\tau_p = 40$ fs, and focal diameter of the beam in the interaction zone $d = 0.1$ mm – we estimated $U_p = 6.6$ eV. The ionization potential of Ar is $E_{\text{Ar}} = 15.6$ eV. So, $E_{\text{Ar}} > U_p$, and we are in the so called low intensity regime of HHG ($I \sim 10^{14}$ W/cm^2).

In this low intensity regime (3.5 mJ, 40 fs) there is very little ionization, and phase matching occurs primarily as a balance of the dispersion of the neutral atoms and the waveguide dispersion. Figure 5 shows the dependence of the signal of some of the harmonics (23rd, 25th and 27th) on the pressure of the Argon gas in the capillary section. In the scheme of generation that we use, a key role to achieve optimal phase matching plays the pressure of the working

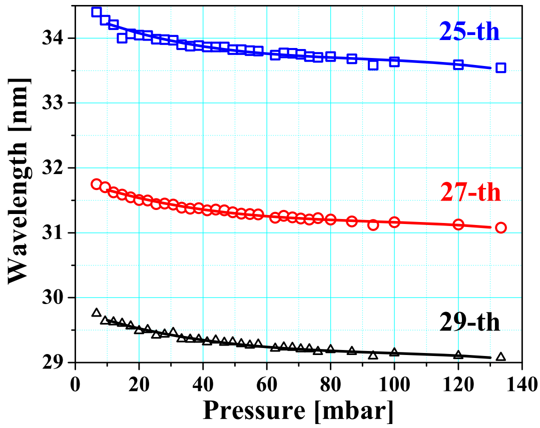

Figure 5. Wavelength tuning of the 25th, 27th, and 29th harmonics vs. Ar gas pressure.

gas. When changing this pressure the conditions for phase matching are also changed resulting in a tuning in the frequency of the generated harmonics. In Figure 5 we demonstrate this behavior for three of the generated harmonics.

This wavelength tuning is accompanied by changes in the relative strength of the harmonic signals (see Figure 6) from which we deduce an optimal Ar gas pressure of 36 mbar in our arrangement and for our particular laser beam/pulse parameters.

For further optimization of the phase matching and of the EUV HHG efficiency we are developing a folded $4-f$ system for pulse shaping [18] involving a two-dimensional liquid-crystal spatial light modulator (HoloEye GmbH) driven by

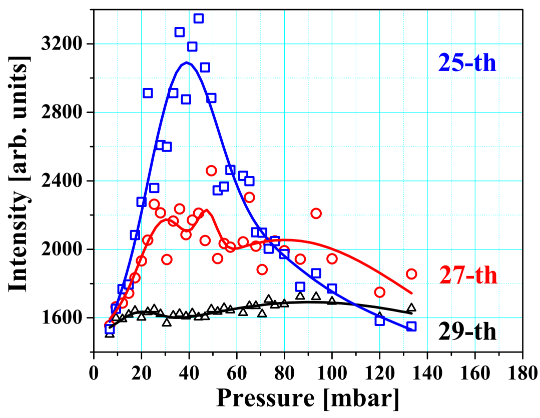

Figure 6. Relative intensities of the 25th, 27th, and 29th harmonics vs. Ar gas pressure.

a suitable genetic algorithm. This module should allow to pre-chirp the femtosecond pulses from the oscillator and to optimize the EUV signal of a particular pre-selected high-harmonic signal. This system will also allow to study the HHG process with structured (singular) femtosecond laser beams [19,20].

4 Conclusion

In this work we presented first results for high harmonic generation in the extreme ultraviolet spectral range down to 26 nm by a newly developed femtosecond-pulse-pumped setup in 55 mm long Ar-filled hollow capillary. In the relatively low intensity regime ($I \sim 10^{14}$ W/cm^2) we observe clear tuning of the HHG signal vs. Argon gas pressure, which is due to the changing phase-matching conditions. Further measurements in other noble gases (e.g. Neon) will provide the possibility to penetrate deeper in the XUV spectral region. Planned steps in optimizing the efficiency in the HHG with this setup are discussed.

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