High-Brightness Coherent Soft X-Ray Generation by High-Order Harmonics

Katsumi Midorikawa, Eiji Takahashi, and Yauso Nabekawa

Laser Technology Laboratory, RIKEN
Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan

Abstract. We investigate the energy scaling of high-order harmonics in Ar using a loosely focused beam under the phase-matched condition. By adjusting the argon gas density and the pump laser focusing condition, a total output harmonic energy as high as 0.7 \mu J is obtained in the spectral region of 34.8 to 25.8 nm (the corresponding order of the 23rd to 31st harmonic), while the 27th order harmonic (29.6 nm) energy attained is as high as 0.33 \mu J with almost perfect spatial profile. Both high efficiency and good spatial quality are achieved simultaneously.

INTRODUCTION

High-order harmonic generation (HHG)\(^1\), \(^2\) can produce ultrashort coherent extremely ultraviolet (XUV) pulses from a compact laser system. Compared to other XUV light sources, such as synchrotron orbit radiation (SOR), free electron lasers (FEL), X-ray lasers (XRL), and laser produced plasma, high-order harmonics (HH) have many advantages such as ultrashort pulse duration, high peak intensity and brightness, wide tunability, and low equipment cost. This emission source is useful for XUV nonlinear optics, attosecond physics, and XUV spectroscopy. Recently, several interesting applications using a HH have been demonstrated in solid-state and plasma physics\(^3\) as well as atomic and molecular spectroscopy. Also, some attempts were made in nonlinear physics to produce two-photon ionization of rare gases in the XUV.\(^4\), \(^5\) Moreover, Hentschel et al.\(^6\) recently demonstrated the measurement of attosecond pulse duration using a HH.

For further development of a variety of applications of HH, one of the most important issues is its energy scaling. The high-energy HH is expected to boost new physics in the soft X-ray region. For example, Bandrauk et al.\(^7\) reported the advantages of high intensity short wavelength radiations to Coulomb explosion imaging, and Ishikawa and Midorikawa\(^8\), \(^9\) pointed out the possibility of a nonlinear optical effect in the soft X-ray region with a high-energy harmonic pulse.

Experimental and theoretical optimization of conversion efficiency to increase an output energy of HH were reported with a few mJ pump laser energy.\(^10\), \(^11\) However, the energy scaling under such an optimized condition has not been studied experimentally, although we can utilize much larger pump energy. Furthermore, the conditions that give the maximum efficiency and output did not always generate high harmonics with good spatial quality in previous works.\(^12\)
Here, we report the energy scaling of HH in Ar under the optimized phase-matched condition. Our scaling method demonstrated a linear increase of harmonic energy with respect to the geometrical focusing area of the pump pulse, while keeping an almost perfect spatial profile of the harmonic output. Moreover, we performed the direct measurement of HH output energy using an XUV photodiode. The high output energy achieved in our scheme allows the direct energy measurement with a single shot.

ENERGY SCALING PROCEDURE

Most of the high-order harmonics generation (HHG) was performed by using a pulsed gas jet, where an intense laser pulse is focused. Phase-matching is dominated by a Gouy phase shift of the laser field and the medium’s dispersion as it passes through the focus. Ditmire et al.\(^{13}\) performed the direct measurement of the conversion efficiencies of HH using an X-ray charge coupled device (CCD) camera. Although they had generated individual harmonics with energies as high as 60 nJ at wavelengths as short as 20 nm, they had not commented on their beam quality.

Recently, by using a self-guided beam or a capillary waveguide in a static long gas-cell, conversion efficiencies were improved.\(^{10,14-18}\) In those cases, phase-matching was performed by balancing dispersions due to neutral atoms, free electrons, and focusing geometry. As pointed out by Constant et al.,\(^{11}\) when the medium length is comparable to or longer than the absorption length, it becomes important to consider the effect of the absorption term of the target medium. Therefore, the photon number, \(N_q\) of the qth harmonic on axis per unit of time and of area is given by

\[
N_q = N_0^2 d(q\omega_q) \exp \left[ \frac{4 (L_{\text{abs}} L_{\text{coh}})^2}{L_{\text{coh}}^2 + (2\pi L_{\text{abs}})^2} \right] \left[ 1 + \exp \left( \frac{L_{\text{med}}}{L_{\text{abs}}} \right) - 2 \cos \left( \frac{\pi L_{\text{med}}}{L_{\text{coh}}} \right) \exp \left( - \frac{L_{\text{med}}}{2 L_{\text{abs}}} \right) \right],
\]

where \(d(q\omega_q)\) is the atomic dipole moment induced by the laser field, and \(N_0\) and \(L_{\text{med}}\) are neutral gas density and medium length, respectively. Coherence length \(L_{\text{coh}}\) and absorption length \(L_{\text{abs}}\) correspond to \(\pi \Delta k\) and \(1/2\alpha\), respectively. Here, \(\alpha\) is the absorption coefficient for the qth harmonic. To estimate \(\Delta k\), one needs to consider the geometrical phase advance and the atomic dispersion for both fundamental and XUV light. As is found in Eq. (1), when the coherence length is comparable to the absorption length, \(N_q\) saturates as soon as the medium length becomes longer than the absorption length. Therefore, in order to increase the HH output, the coherence length should extend much longer than the absorption length. When the coherence length is much longer than both the absorption length and medium length, the net HH energy yield is proportional to the following relationship,\(^{13}\)

\[
N_q \propto A_{\text{spot}} (PL_{\text{med}})^2
\]

where \(A_{\text{spot}}\) is the spot area of the pump pulse at the focusing point, which corresponds to \(\pi \omega_p^2\), \(\omega_p\) corresponds to the spot size, and \(P\) is the target gas pressure.

As described above, the photon number of HH is proportional to the square of the
medium length and the target gas pressure. It is also proportional to the square of the dipole strength at the given harmonic order. The dipole strength is assumed to follow $d(q\alpha_0) \sim (1 - \eta) I_0^{5/2}$, where $\eta$ and $I_0$ correspond to the ionization probability and the laser intensity at the focus, respectively. The focused laser intensity is limited by the ionization threshold of the target gas medium, because the negative dispersion due to the free electrons breaks phase-matching and ionization decreases the target atomic gas density. In the nonionized condition of the target medium, the gas pressure must be adjusted to cancel the geometrical phase shift of the pump laser pulse.

To extend the interaction length between the laser pulse and the target medium, the Rayleigh length $z_0 = \pi\alpha_0^2/\lambda_0$ must be extended. Since a Gouy phase is given by $\Delta k_{\text{gouy}} = q/(z_0 + z^2/z_0^3)$, the increasing of the Rayleigh length results in the decreasing of the Gouy phase shift. Therefore, to satisfy the phase-matched condition, the target gas density must be decreased in inverse proportion to the medium length. In other words, the $PL_{\text{med}}$ product must be constant under the optimized phase-matched condition in neutral atoms. Consequently, as is derived from Eq. (2), to enhance the output energy of HH under the phase-matched condition, the spot size of the pump pulse at the focus should be increased. This leads to the increase of input laser energy to maintain a fixed pump intensity.

Designed harmonic energy scaling parameters are shown in Table 1. Optimized parameters for $f = 500$ mm pumping were obtained from the previous experiment. For newly designed parameters, the laser focusing condition and the pump energy are scaled up approximately 10-fold.

<table>
<thead>
<tr>
<th>Focusing Length [mm]</th>
<th>500</th>
<th>2500</th>
<th>5000</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pump Energy [mJ]</td>
<td>5</td>
<td>25</td>
<td>50</td>
</tr>
<tr>
<td>Sopt Size: $\alpha_0$ [\mu m]</td>
<td>60</td>
<td>130</td>
<td>200</td>
</tr>
<tr>
<td>Medium Length: $L_{\text{med}}$ [mm]</td>
<td>10</td>
<td>50</td>
<td>100</td>
</tr>
<tr>
<td>Gas Pressure: $P_{\text{Ar}}$ [Torr]</td>
<td>20</td>
<td>4</td>
<td>2</td>
</tr>
</tbody>
</table>

**EXPERIMENT**

Experimental studies were carried out with a 10 Hz Ti:sapphire laser system based on a chirped pulse amplification (CPA). This system produced an output of 200 mJ with a pulse width of 35 fs. The wavelength was centered at 800 nm. The pump pulse was loosely focused with a fused silica lens, and delivered into the target chamber through a CaF$_2$ window. We set the focus at the entrance pinhole of the interaction cell. The interaction cell had two pinholes on each end surface of the bellow arms. These pinholes isolated the vacuum and gas-filled regions. The interaction length was variable from 0 to 150 mm in the interaction cell. Argon gas was statically filled in the interaction cell. The generated harmonics illuminated a 120 \mu m (H) x 15 mm (V) slit of the spectrometer. A flat-field grating (1200 grooves/mm) relayed the image of the slit to a microchannel plate (MCP), while it was spectrally resolved along the horizontal axis. CCD camera detected two-dimensional fluorescence from a phosphor screen placed behind the MCP. Therefore, we measured the spectrally resolved far-field profiles of HH. The spectral response of the measurement equipment was
assumed to be flat, because the measured spectral region was sufficiently narrow. The absolute energy of HH was measured directly with an unbiased silicon XUV photodiode (XUV-100). This detector has a wide range of sensitivity from 200 nm to 0.07 nm (6 eV to 17.6 keV). The sensitivity of this photodiode follows the simple linear law $N_e = E_{ph}/3.63$ eV, where $N_e$ is the number of photoelectrons created by a single XUV photon of energy $E_{ph}$. The values of spectral sensitivity and quantum efficiency of the XUV photodiode were referred to in Ref. 21, and calibrated with 266 nm Q-switched YAG laser pulses in this experiment.

RESULTS AND DISCUSSION

The absolute output energy of HH was measured using an XUV photodiode. This photodiode was inserted in front of the spectrometer, and the output signal was recorded directly on an oscilloscope. A thin aluminum filter (0.2 μm) was placed between the sources and the detector to eliminate the pump pulse and pass only the 11th through 45th harmonics (17 to 70 eV). In practice, low order harmonics (<21st) are not phase-matched in our experimental condition and are strongly absorbed in argon gas. Therefore, the XUV photodiode detected only the 23rd to 31st order harmonic energies. Figure 1 shows the HH energy yield for the 10 cm interaction. The measured total energy of HH was ~0.7 μJ. The individual harmonic energies were estimated from the spectral distribution of HH. From the relative harmonic strength distribution, the 27th harmonic energy was estimated to be ~0.3 μJ. The error bars resulted from the fluctuation of the pump laser energy. The maximal output of 330 nJ was recorded with a pump energy of 20 mJ.

We also measured the pump spot area dependence of the 27th harmonic energy by changing the pump focusing length. Figure 2 shows the experimentally obtained energy yield of the 27th harmonic as a function of the spot area of the pump pulse at the focus. Note that the spot area is calculated from the focusing geometry in the vacuum. As is shown in Table 1, we initially planned to extend the input laser energy to ~50 mJ. However, since the frequency spectrum of the pump laser pulse was modulated by the self-phase modulation (SPM) at the entrance window (CaF2), we could use only 20 mJ in this experimental setup. When we increased the input energy above 20 mJ, we could obtain neither good spatial quality nor high energy within the observed spectral region. Therefore, the pump energy and the truncated diameter for
an f = 5000 mm pumping geometry were set at 20 mJ and 18 mm, respectively. The spot size of the pump pulse at the focus was measured to be approximately 200 μm, which was nearly Gaussian. The output energy of the 27th harmonic shows linear dependence on the spot size of the pump pulse. However, the input laser energy was not scaled up along the designed scaling parameters (see Table.1). For an f = 500 mm pumping, the energy and the diameter of the pump pulse were 5 mJ and 7 mm, respectively. In the middle range of our optimized scaling condition, we used an f = 2500 mm focusing lens. In this condition, the energy and the diameter of the pump pulse were 12 mJ and 13 mm, respectively, while the argon gas pressure and the medium length were 4 Torr and 5 cm, respectively. The $P_{Ar}L_{med}$ product was also kept constant for the f = 2500 mm pumping. When the interaction length was increased to 10 cm in the 1.8 Torr Ar gas, the output energy of the 27th harmonic was enhanced more than 10 times, compared with the 20 Torr, 1 cm interaction, while the pump energy was increased by only a factor of four. Therefore, the conversion efficiency was improved by a factor of two, and attained a value of 1.5 x10^5.

The evolution of HH intensities in the spectral region from the 23rd to 27th order harmonics measured as a function of medium length is shown in Fig.3. Based on Eq. (1), we estimated the coherence length $L_{coh}$ in the experiment. The absorption length $L_{abs}$ for 1.8 Torr argon gas was calculated to be 1.16 cm for the 23rd, 2.39 cm for the 25th, and
6.81 cm for the 27th harmonic from Ref. 24. The solid line shows theoretically fitted intensities for the 23rd, 25th and 27th harmonics. The coherence length was estimated to be ~15 cm by fitting the theoretical curves. As is pointed out by Constant et al., the optimizing conditions of medium, coherence and absorption lengths are given by $L_{med} > 3L_{abs}$, $L_{coh} > 5L_{abs}$. The 23rd and 25th order harmonics satisfied the optimized condition for this relation. Therefore, those orders were saturated under our experimental conditions. On the other hand, the 27th order harmonic did not satisfy the above conditions yet, because of low absorption.

Figure 4 shows the single-shot far-field spatial profiles of the 27th harmonic. The profile was integrated with respect to wavelength. The abscissa represents the emission angle, and the ordinate represents the normalized intensity. Emission angles (mrad) can be altered to the spatial profile of the harmonic by considering the distance between the harmonic source and the spectrometer slit. No data smoothing procedures were made for the experimental results described below. The thin line corresponds to the experimental result from a 20 Torr, 1 cm interaction under the optimized phase-matched condition. The output beam divergence was ~2 mrad (FWHM). The bold line corresponds to the 27th harmonic from a 1.8 Torr, 10 cm interaction. To extend the interaction length under the phase-matched condition, the pump pulse was loosely focused with an $f=5000$ mm planoconvex lens. As a result of phase-matching, the spatial quality of the 27th harmonic also showed a Gaussian-like profile, and the beam divergence decreased by a factor of three. The measured beam divergence was 0.7 mrad (FWHM).

The beam emittance of HH under the phase-matched condition was improved with the increase of the medium length. The beam divergence of HH is related to a noncollinear angle of HH. This type of phase-matching is called the Cerenkov phase matching. The Cerenkov phase matching is similar to the Gouy factor in that the phase of the pump pulse is independent of the gas density and the laser intensity. In the off-axis direction, the phase-mismatch is given by $\Delta k = k_q - qk_o - \Delta k_{\text{cere}}, \Delta k_{\text{cere}} = q\pi\theta \lambda_0$, where $\theta$ is the noncollinear angle of HH. By using Eq. (1), we performed the calculation of the 1D spatial distribution of the 27th harmonic including the Cerenkov phase matching. The dashed line in Fig. 4 shows a calculation result in the $f=5000$ mm experimental condition. In that calculation, the phase-mismatch factors were assumed to be zero, except for the Cerenkov factor $\Delta k_{\text{cere}}$. The spatial intensity distribution of the pump pulse at the focal plane can be described by $I(r) = I_0 \exp(-2r^2/w_0^2)$. The emitted photon number was defined to follow $I(r)^{1/3}$, and the
interaction laser intensity was assumed to be constant along the medium length. Calculated and experimental spatial distributions in the phase-matched condition of \( f = 5000 \text{ mm} \) are compared in Fig. 4. Good agreement in profile and divergence are clearly recognized.

Figure 5 shows beam divergence as a function of medium length under the optimized phase-matched condition. The solid line corresponds to the fitting curve for the experimental results, which was inversely proportional to the medium length with a power of 0.5. The beam divergence angle of HH under the phase-matched condition \( (k_2 \cdot q_k \sim 0) \) is given by \( \theta \propto 1/(L_{\text{med}})^{1/2} \). Therefore, the measured beam divergence of the 27th harmonic was proportional to \( 1/(L_{\text{med}})^{1/2} \). This result is in good agreement with the theoretical prediction including the Cerenkov phase matching. It is clearly demonstrated that to generate the low-emittance harmonic beam, the long medium length is more useful than a thin gas jet.

**CONCLUSION**

When the interaction length was increased to 10 cm in 1.8 Torr argon gas, the total output energy in the spectral region of 34.8 to 25.8 nm (the corresponding order of 23rd to 31st harmonics) attained was 0.7 \( \mu \text{J} \), and the maximal 27th harmonic energy was estimated to be 0.33 \( \mu \text{J} \). The output energy of the 27th harmonic (29.6 nm) was enhanced by more than 10-fold, compared with 20 Torr, 1 cm interaction, while the pump energy was increased by only a factor of four. Therefore, the conversion efficiency was improved by a factor of two, and attained a value of \( 1.5 \times 10^{-5} \). From the measured beam parameters for the 27th harmonic, the peak brightness of this coherent soft X-ray was estimated to be \( 8 \times 10^{26} \) photon/mm\(^2\)/mrad\(^2\)/s, assuming a beam diameter of 60 \( \mu \text{m} \) at the exit of the gas cell. This is the maximum brightness achieved to date, compared to the values produced by various coherent soft X-ray sources.\(^{22, 23}\)

**REFERENCES**

20. United Detector Technologies Inc..