PHYSICAL REVIEW A, **66**, 021802(R) (2002)

Generation of highly coherent submicrojoule soft x rays by high-order harmonics

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(Received 26 October 2001; revised manuscript 22 March 2002; published 26 August 2002)

We demonstrated the energy scaling of high-order harmonics using a self-guided beam under the phase-matched condition. By adjusting the argon gas density and pump laser focusing condition, a total output harmonic energy as high as 0.7 μJ was 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 was as high as 0.3 μJ.

DOI: 10.1103/PhysRevA.66.021802 PACS number(s): 42.65.Ky, 32.80.Rm

For the development of a variety of applications of high-order harmonics (HH) [1,2], one of the most important issues is energy scaling. Ditmire *et al.* [4] performed the direct measurement of output energy and conversion efficiencies of HH using an x-ray charge-coupled device (CCD) camera. They examined the energy yields as a function of a number of parameters, including the pump laser wavelength, the target atoms, the focal geometry, and the peak laser intensity. They realized the generation of the maximum harmonics energy as high as 60 nJ at a wavelength as short as 20 nm. But the conversion efficiencies attained were 10^{-7}.

To improve the conversion efficiency of HH, the phase-matching between the harmonic field and the atomic dipole becomes essential. High-order harmonics generation (HHG) is usually 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 medium's dispersion as it passes through the focus. Salières *et al.* [3] demonstrated phase matching by balancing the Gouy phase and the dipole phase shift, which was achieved experimentally by controlling the relative position between the laser focus point and the gas jet. The obtained harmonic profile was improved to be smooth and nearly Gaussian.

Recently, by using a self-guided beam or a capillary waveguide in a static long gas cell, conversion efficiencies were improved [5–10]. The best conversion efficiency with phase matching was 4×10^{-5} [7], which was achieved using a Xe gas. In those cases, the phase-matching was performed by balancing dispersions due to neutral atoms, free electrons, and focusing geometry. Moreover, as is pointed out by Constant *et al.* [7], when the medium length is comparable to or longer than the absorption length, it becomes important to consider the effect of absorption. Consequently, the photon number N_q of the qth harmonic on the axis per unit of time and of area is given by [7]

$$N_q \sim N_0^2 |d(q\omega_0)|^2 \frac{4(L_{\mathrm{abs}}L_{\mathrm{coh}})^2}{L_{\mathrm{coh}}^2+(2\pi L_{\mathrm{abs}})^2}\left[1+\exp\left(-\frac{L_{\mathrm{med}}}{L_{\mathrm{abs}}}\right)-2\cos\left(\frac{\pi L_{\mathrm{med}}}{L_{\mathrm{coh}}}\right)\exp\left(-\frac{L_{\mathrm{med}}}{2L_{\mathrm{abs}}}\right)\right], \quad (1)$$

where $d(q\omega_0)$ is the atomic dipole moment induced by the laser pulse, N_0 is the neutral gas density, ω_0 is the frequency of the laser pulse, and L_{med} is the medium length. Coherence length L_{coh} and absorption length L_{abs} correspond to $\pi/\Delta k$ and $1/2\alpha$, respectively. Here, Δk is the total phase mismatch between the harmonic field and the atomic dipole, and α is the absorption coefficient for the given harmonics. To estimate Δk, one needs to consider the geometrical phase advance and the atomic dispersion for both the fundamental and the XUV light [5]. Equation (1) indicates that 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 $S_{\mathrm{spot}}(PL_{\mathrm{med}})^2$ [4], where S_{spot} is the spot area of the pump pulse at the focus point, which corresponds to πw_0^2, w_0 corresponds to the spot size, and P is the target gas pressure.

Our research group has reported the phase-matching technique of HHG using a hollow fiber and a self-guided beam as in previous publications [8]. In the phase-matched HHG, the output energy was optimized [10] with a perfect Gaussian profile having the visibility of 0.73 [11] at the 29th harmonic (27.6 nm) in Ar. In this study, as the next step, we investigated the energy scaling of the phase-matched HH using a loosely focused beam. Moreover, we performed the direct measurement of HH output energy using an XUV photodiode.

First, let us explain our energy scaling procedure under the phase-matched condition. The efficiency and the spatial quality of HH depend on the target gas density, the medium length, and the pump laser focusing condition. The photon number of HH is proportional to the square of the medium length, 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 $d(q\omega_0)\sim(1$

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TABLE I. Designed harmonic energy scaling condition.

Focusing length (mm)	500	5000
Pump energy (mJ)	5	50
Spot radius w_0 (μm)	$\sim$60	$\sim$200
Medium length L_{med} (cm)	1	10
Target gas pressure P_{Ar} (Torr)	20	2

$-\eta)I_0^5$ [12], where η 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 $z_0=\pi w_0^2/\lambda_0$ must be extended. Since a Gouy phase is given by $\Delta k_{\mathrm{gouy}}=q/(z_0+z^2/z_0)$, 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_{med} product must be constant under the optimized phase-matched condition in neutral atoms. Consequently, 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 I. Optimized parameters for the $f=500$ mm pumping were obtained from the previous experiment [10]. For newly designed parameters, the laser focusing condition and the pump energy are scaled up by approximately tenfold. Therefore, we can expect that the output HH energy is enhanced by approximately tenfold.

The 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 μm $(H)\times 25$ mm (V) slit of the spectrometer. A flat-field grating (1200 grooves/mm) relayed the image of the slit to a microchaneel plate (MCP), while it was spectrally resolved along the horizontal axis. A CCD camera detected two-dimensional fluorescence from a phosphor screen placed behind the MCP. Therefore, we measured spectrally resolved far-field profiles of high-order harmonics. The absolute energy of HH was measured directly with an unbiased silicon XUV photodiode (XUV-100) [13]. 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 $N_e=E_{\mathrm{ph}}/3.63$ 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 Ref. [14], and calibrated with 266 nm Q-switched yttrium aluminum garnet (YAG) laser pulses in this experiment.

Figure 1(a) shows the far-field spatial profiles of the 27th harmonic. The dashed line corresponds to one from a 20 Torr, 1 cm interaction under the optimized phase-matched condition. In this case, the energy and diameter of the pump pulse were 5 mJ and 7 mm, respectively. The pump pulse was focused with an $f=500$ mm planoconvex lens. The output beam divergence was $\sim$2 mrad [full width at half maximum (FWHM)]. The solid 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 is shown in Table I, we initially planned to extend the input laser energy to $\sim$50 mJ. However, since the frequency spectrum of the pump laser pulse was modulated by the self-phase-modulation (SPM) at the entrance window (CaF_2), we could use only $\sim$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 were set at 20 mJ and 18 mm, respectively. As a result of phase matching, the spatial quality of the 27th harmonic also showed a Gaussian-

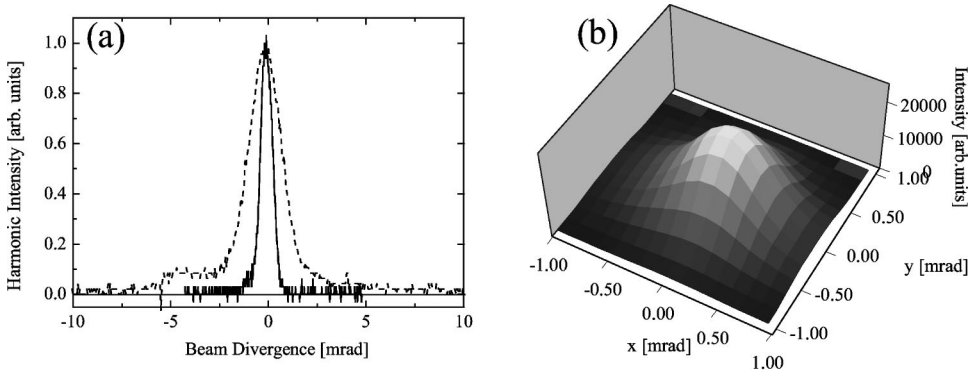

FIG. 1. The measured spatial profile of the 27th harmonic. (a) Normalized 1D distribution of the 27th harmonic. The solid line and dashed line correspond to $P_{\mathrm{Ar}}=1.8$ Torr, 10 cm medium length, and $P_{\mathrm{Ar}}=30$ Torr, 1 cm medium length, respectively. (b) 2D spatial distribution of the 27th harmonic in the optimized condition.

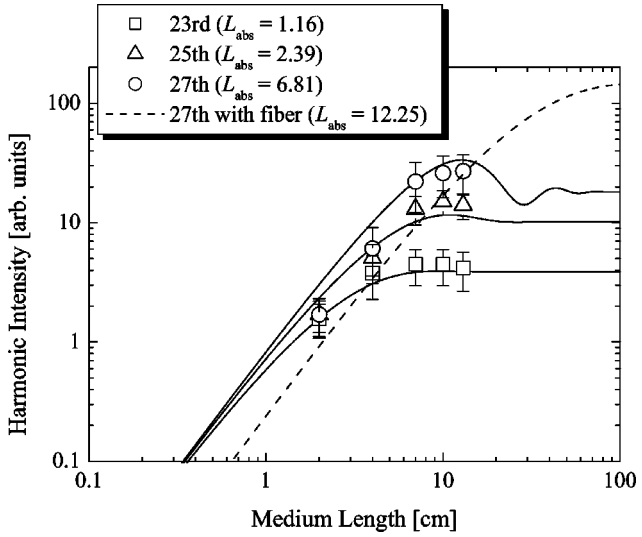

FIG. 2. The emitted photon number of the harmonics on axis in argon as a function of the medium length. The solid line corresponds to the calculated photon number for $L_{coh} \sim 15$ cm with 1.8 Torr argon in free space. The dashed line corresponds to the calculated photon number of 27th for $L_{coh} \sim 1000$ cm with 1 Torr argon in a hollow fiber.

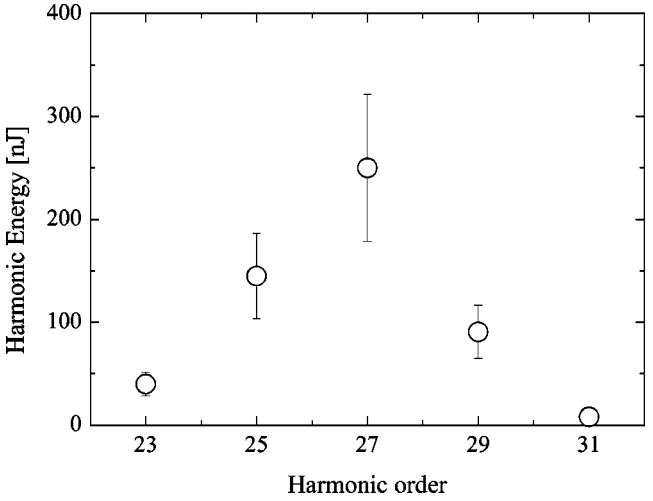

FIG. 3. Experimentally obtained energy yield per shot of the 27th harmonic for a 10 cm long medium length with 1.8 Torr argon gas.

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 proportional to 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 [9]. In the off-axis direction, the phase mismatch is given by $\Delta k = k_q - qk_0 - \Delta k_{cere}, \Delta k_{cere} = q\pi\theta^2/\lambda_0$, where θ is the noncollinear angle of HH. The beam divergence angle of HH under the phase-matched condition ($k_q - qk_0 \sim 0$) is given by $\theta \propto 1/\sqrt{L_{med}}$. Therefore, the measured beam divergence of the 27th harmonic was proportional to $1/\sqrt{L_{med}}$.

Figure 1(b) shows the two-dimensional (2D) spatial profile of the 27th harmonic with the 10 cm medium length, 1.8 Torr of argon. The 2D spatial profile was measured by moving the spectrometer along the perpendicular direction of the entrance slit in 500 μm intervals. Thus, we can reconstruct the 2D spatial profile of harmonics. The measured 2D spatial profile of the 27th harmonic shows perfect beam quality.

Under the optimized condition of 10 cm medium length, the evolution of HH intensities in the spectral region from the 23rd- to 27th-order harmonics measured as a function of the medium length are shown in Fig. 2. Based on Eq. (1), we estimated the coherent 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. [15]. The solid line shows theoretically fitted intensities for the 23rd, 25th, and 27th harmonics. The coherent length was estimated to be ~ 15 cm by fitting the theoretical curves. As is pointed out by Constant *et al.* [7], the optimizing conditions of medium, coherence, and absorption length 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.

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 3 shows the HH energy yield for 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 result from the energy fluctuation of the pump laser energy.

We also measured the pump spot area dependence of the 27th harmonic energy by changing pump focusing length. In the middle range of our energy scaling condition, we used an $f = 2500$ mm focusing lens. In this condition, an energy and a 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. The maximum output energy of the 27th harmonic was measured to be ~ 150 nJ with a good beam quality. The output energy of the 27th harmonic revealed the linear dependence of the spot size of the pump pulse. However, the input laser energy was not scaled up along the designed scaling parameters (see Table I). When the interaction length was increased to 10 cm in 1.8 Torr argon gas, the output energy of the 27th harmonic was enhanced more than 10 times, compared with 20 Torr, 1 cm interaction, while the pump energy was in-

creased by only a factor of four. Therefore, the conversion efficiency was improved by a factor of two, and attained a value of 1.5×10^{-5}. This might be explained by considering the propagation of the pump pulses. The effective interaction intensity of the pump pulses will be fixed by ionization-induced defocusing.

From the experimental results, the coherent length was estimated to be less than ~15 cm with an $f=5000$ mm lens and 1.8 Torr argon gas. Since the Gouy phase in the free space changes along the pump laser propagation in the medium, the estimated coherence length is an averaged value over the interaction length. To attain the perfect phase matching, a useful method is to use a capillary tube. The phase mismatch of the pump pulse for a fundamental traveling wave in the lowest mode of the capillary tube is given by $qu_{11}^2\lambda_0/(4\pi a^2)$. Here, u_{11} and a_0 correspond to the first zero of the Bessel function J_0 [16] and the radius of the capillary tube, respectively. Therefore, the amount of phase mismatch does not change in the capillary tube. The dashed line in Fig. 2 shows the calculated photon number of the 27th harmonic from 1 Torr argon gas as a function of the capillary length in the perfect phase-matched condition. Because the geometrical phase shift of a pump laser pulse in the capillary tube becomes smaller than free space, the optimized argon gas pressure becomes ~1 Torr ($L_{\mathrm{abs}}=12.25$ cm). In our experiment, the spot size of the pump pulse was measured to be ~190 μm. With a $a=300$ μm capillary tube, the coupling efficiency was estimated to be $>95\%$. The intensity decay of the laser pulse and the ionization of neutral argon gas were neglected in this calculation. With a longer than 40-cm capillary tube, we can expect to generate 1 μJ in the 27th harmonic under the phase-matched condition.

In summary, 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 the 23rd to 31st harmonics) attained was ~0.7 μJ, and the 27th harmonic energy was estimated to be ~0.3 μJ. The output energy of the 27th harmonic (29.6 nm) was enhanced more than 10 times, 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 1.5×10^{-5}. From the measured beam parameters for the 27th harmonic, the peak brightness of this coherent soft x ray can be estimated to be $\sim10^{26}$ photon/mm^2 mrad^2 s, assuming a beam diameter of 60 μm 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 [17,18]. Finally, the possibility of the generation of 1 μJ energy in the 27th harmonic was revealed by performing energy scaling.

This work was supported by the Special Postdoctal Researchers Program of RIKEN.

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