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Energy-yield and conversion-efficiency measurements of high-order harmonic radiation

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Experimental measurements of the absolute energy yields and conversion efficiencies of high-order harmonic radiation in the spectral range of 31 to 17 nm are presented. We examine energy yields as a function of a number of parameters including drive laser wavelength, target atom, focal geometry, and peak laser intensity, and we have generated individual harmonics with energies as high as 60 nJ at wavelengths as short as 20 nm. Under optimum conditions, we find that conversion efficiencies of as high as 10^{-7} into each harmonic are possible.

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The generation of high-order harmonic radiation by an intense laser pulse is now a well documented phenomenon [1,2] and is one of the principal new sources of bright XUV radiation being developed for a variety of applications that rely on the interaction of short-wavelength radiation with matter. To date, wavelengths as short as 7 nm have been generated by harmonic generation [3,4]. When compared to other XUV sources such as synchrotrons, FELs (free-electron lasers), and x-ray lasers, harmonic generation exhibits many advantages such as short pulse duration (40 fs–100 ps), high peak brightness, and tunability. Harmonic generation has been extensively studied by a number of groups using drive lasers with a range of wavelengths and pulse widths [3–8] and much work has been done to understand the short-wavelength limit and related scaling formulas.

In spite of these early studies, the work of characterizing and optimizing high-order harmonic generation as a general purpose XUV source is incomplete. A number of groups have examined the angular divergence and spatial coherence of the harmonics [9–11]. Previous experiments have also characterized the linewidths of the harmonics [12,13]. The utilization of high-order harmonics in applications has been recently demonstrated by Balcou *et al.*, who have used photon energies in excess of 100 eV in photoionization spectroscopy [14]. Though there have been preliminary studies [7], a complete measurement of the photon yields achievable with high-order harmonics has not yet been undertaken. In this Rapid Communication we report on measurements of the absolute energy yields and conversion efficiencies of harmonics generated in the 31–17-nm wavelength range under a variety of conditions.

These experiments were performed using a Nd:glass laser that produced 650-fs pulses at 1052.7 nm with energies up to 8 J, and pulses of its second harmonic at 526 nm with energies up to 4 J [15]. The laser was focused into the plume of a pulsed, supersonic nozzle, gas jet. This jet produces localized atomic densities from 10^{18} to 2×10^{19} atoms/cm^3 and exhibits a linear density dependence with gas jet backing pressure, verified by backward stimulated Raman scattering measurements [16]. The interaction length through the gas jet is 0.8 mm. Our measurements were conducted with an atom density of 2×10^{19} atoms/cm$^3\pm30\%$. The harmonic radiation is sampled by an astigmatic compensated, grazing incidence, XUV spectrometer. A calibrated aluminum foil filter prevents scattered laser light from reaching the detector.

The harmonics are detected with an absolutely calibrated x-ray charge-coupled device (CCD) detector (Princeton Instruments). This detector uses a thermoelectrically cooled Tektronix 1024B back illuminated CCD chip [17]. The quantum efficiency of the chip (which is lowered in the spectral region of interest by a thin layer of SiO_2 on the surface of the chip of approximately 5–10 nm thick) was calibrated at the Brookhaven National Synchrotron Light Source. The quantum efficiency ranges from 0.7 for wavelengths between 20 and 10 nm and drops to below 0.2 at 30 nm. The response of this camera drops very quickly with wavelengths longer than 31 nm. The correlation factor between accumulated charge and detected counts on the camera was measured by exposing the CCD chip to single-photon hits of $K\alpha$ emission from tin at a photon energy of 25 keV.

The throughput of the XUV spectrometer was measured by comparing signals of harmonics on the x-ray CCD produced under identical conditions with and without the spectrometer in the system. Data taken with the spectrometer yielded the relative energy in each harmonic while data taken without the spectrometer gave the total integrated harmonic yield per laser shot. Two calibrated aluminum filters (860 nm total thickness) were used to pass the harmonics and completely block all laser light for data taken without the spectrometer. Comparison of these shots with harmonics shots taken with the spectrometer at the same laser intensity yielded the spectrometer throughput of those harmonics between 30 nm and the aluminum L edge at 17 nm. This measurement was repeated for a range of laser intensities and for four different laser focal configurations, $f/25$, $f/35$, $f/50$, and $f/70$ focusing.

Figure 1 shows the typical energy yields of harmonics produced in neon for drive wavelengths of 1053 and 526 nm. The error bars result from uncertainties in the spectrometer throughput measurement and in the CCD chip quantum efficiency. These shots were taken with the confocal parameter b held constant at a value of 2.5 cm (which is much longer than the gas jet interaction length of 0.8 mm) and a constant peak intensity of 1×10^{15} W/cm^2. This intensity is above the ionization saturation intensity for neon (which is $\approx4.5\times10^{14}$ W/cm^2) [18]. The harmonics of the 526-nm light were produced with 50 mJ focused with an $f/50$ geometry to

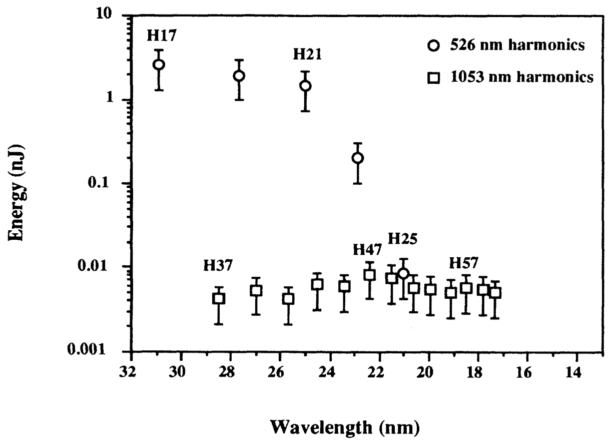

FIG. 1. Energy yields of harmonics generated in neon by 526-nm light (circles) and harmonics generated by 1053-nm light (squares). The peak intensity was 1×10^{15} W/cm^2 and the confocal parameter was 2.5 cm.

a spot size of 140 μm ($1/e^2$ diameter), and the 1053-nm harmonics were produced with 90 mJ focused with an $f/35$ geometry to a spot size of 180 μm ($1/e^2$ diameter). The actual plateau of the 1053-nm harmonics extends well beyond the L-edge cutoff of the aluminum filter used for these measurements; we have observed harmonics out to the 121st in neon in previous experiments under similar conditions [4]. The cutoff of the 526-nm harmonics, however, occurs at a much longer wavelength, 22.9 nm. This is expected on the basis of the prediction that the high-order harmonic cutoff occurs at $\sim I_p+3.17U_p$ [19]. I_p is the atom ionization potential, and U_p is the electron quiver energy and is given by $U_p=9.33\times10^{-14}I(\mathrm{W/cm^2})\lambda_0^2(\mu\mathrm{m})$. This expression favors the longer-wavelength driver for generating shorter harmonic wavelengths.

The harmonics in the 1053-nm plateau exhibit energies of between 6 and 8 pJ per harmonic, corresponding to a conversion efficiency of $\sim10^{-10}$. The yields of the harmonics in the plateau of the 526-nm harmonics (the 17th through the 21st) have energies of 1–2 nJ. This dramatic difference in the conversion efficiency between the two colors has been reported previously [20], and can be explained on the basis of two factors: a single-atom response and a phase-matching term. Quantum-mechanical calculations of Krause, Schafer, and Kulander show that the single-atom response favors the shorter-wavelength driver [19]. This can be easily understood if one considers the quasiclassical model of Corkum [21]. In this model the electron oscillates in a classical trajectory about the atom in the laser field. Whenever the electron returns to the atom during its oscillations it can emit a harmonic photon. Since the oscillation amplitude scales as λ_0^2, the electron spends more time in the vicinity of the atom for the shorter-wavelength driver and, therefore, has a larger probability of emitting a harmonic photon per unit time.

The energy yield is also enhanced for the shorter-wavelength by phase matching. It can be shown that if the qth harmonic varies with an effective nonlinear order p, the number of harmonic photons emitted can be written as [2]

$$N_q=\frac{\pi^2b^3}{4\hbar}\,\tau_p n_0^2|d(q\omega)|^2|F_q|^2, \tag{1}$$

where b is the laser confocal parameter, τ_p is the integral of the pth power of the laser temporal envelope, n_0 is the atom density, and $d(q\omega)$ is the atomic dipole moment induced by the laser, representing the atomic contribution to the harmonic yield. $|F_q|^2$ is the phase-matching factor. In the weak focusing limit (i.e., when the confocal parameter is much longer than the gas jet) for a Gaussian beam focused through a uniform gas jet of length l, the phase-matching factor for a square laser pulse can be written as [2]

$$|F_q|^2=\frac{q}{p}\frac{4l^2}{b^2}\left|\frac{\sin(\Delta kl/2+ql/b-ql/pb)}{(\Delta kl/2+ql/b-ql/pb)}\right|^2, \tag{2}$$

where Δk is the phase mismatch defined by $\Delta k=k_q-qk_0$. When $I>I_{\mathrm{sat}}$, the phase matching is dominated by free electrons. In this case the phase mismatch is given by

$$\Delta k_e\cong\frac{2e^2n_e\lambda_q}{c^2m_e}\left(1-\frac{\lambda_0^2}{\lambda_q^2}\right). \tag{3}$$

If $\lambda_0^2/\lambda_q^2\gg1$, then the phase-matching factor becomes

$$|F_q|^2\approx\frac{q}{p}\frac{4l^2}{b^2}\left|\frac{\sin[(2e^2n_e\lambda_q/c^2m_e)\lambda_0^2/\lambda_q^2]}{[(2e^2n_e\lambda_q/c^2m_e)\lambda_0^2/\lambda_q^2]}\right|^2,$$

which means that the net harmonic yield scales like

$$N_q\sim\frac{bn_0^2l^2}{\lambda_0^4}. \tag{4}$$

This expression also favors the shorter-wavelength drive. Our measurements were taken with a peak intensity that is well above the ionization saturation intensity, so we expect significant ionization throughout the entire focal volume. Relation (4) largely explains the difference in our measured harmonic yields for the two drive wavelengths.

The energy yields of harmonics produced in neon and helium generated by 250 mJ of 526-nm light focused with an $f/50$ geometry to an intensity of 6×10^{15} W/cm^2 are compared in Fig. 2. This intensity is well above the saturation intensity of both gases (I_{sat} for He is 7×10^{14} W/cm^2) [19]. The cutoff in He extends beyond that of Ne, as predicted by the cutoff law of Krause, Schafer, and Kulander, because of the greater ionization potential of helium. The energy yields of harmonics in the plateau in both species, however, are approximately the same. Both gases exhibit harmonic yields of between 6 and 8 nJ in the plateau region. The yields drop to a few hundred pJ for the harmonics in the cutoff, which occurs at the 25th in neon and at the 29th in helium.

Though earlier results found harmonic yields greater in neon than in helium [7], we find the conversion yields for both gases to be roughly the same. This can be understood when one considers that our measurements were taken with a peak intensity well above I_{sat} for both gases, so we expect ionization throughout the focal volume. Though neon is more polarizable than helium, implying higher photon yields, the helium survives to a higher intensity than does the neon during the pulse. Because of the high nonlinearity of the

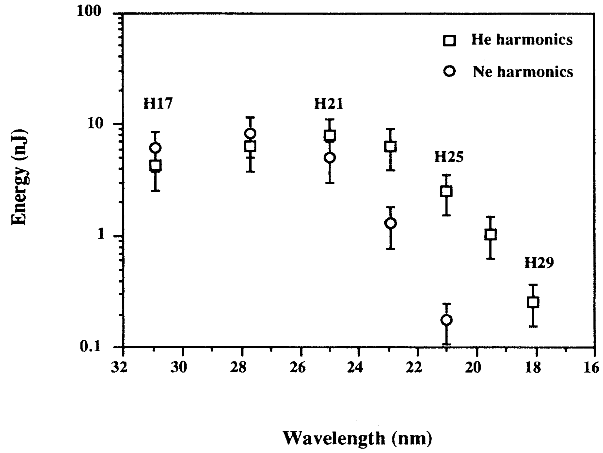

FIG. 2. Energy yields of harmonics produced by 526-nm light in neon (circles) and in helium (squares). The peak intensity was 6×10^{15} W/cm^2 and the focal geometry was $f/50$ ($b=2.5$ cm).

process, the majority of the harmonic photons are produced at an intensity near I_{sat}; consequent ionization prohibits harmonic generation at high intensities later in the pulse. The relative yields of the neon, N_q^{Ne}, and the helium, N_q^{He}, are therefore roughly

$$\frac{N_q^{Ne}}{N_q^{He}} \sim \frac{|\chi_{Ne}(q\omega)|^2(I_{sat}^{Ne})^p}{|\chi_{He}(q\omega)|^2(I_{sat}^{He})^p}. \quad (5)$$

Data at intensities below I_{sat} indicate that $|\chi_{Ne}(q\omega)|/|\chi_{He}(q\omega)|\approx 3$ for harmonics of 526-nm laser light [6]. These measurements also indicate that the harmonics in the plateau tend to vary with an effective order p of between 5 and 7. This implies that N_q^{Ne}/N_q^{He} is of the order of $3^2x(4.5\times10^{14}/7.0\times10^{14})^6\approx0.6$ for our conditions, consistent with our results. Thus, high conversion (>10 nJ) is possible in both noble gases with the added advantage that shorter wavelengths are attainable in helium.

Equation (4) also implies that higher yields are attainable by changing the focal geometry to increase the confocal parameter b. Figure 3(a) shows energy yields of harmonics of 526 nm produced in helium at a constant intensity of 6×10^{15} W/cm^2 for three different f-stop configurations. The harmonics were generated with 130 mJ at $f/25$, 280 mJ at $f/50$, and 460 mJ at $f/70$. The location of the cutoff is the same for all three focus speeds. The harmonic yield in the plateau, however, varies from roughly 1 nJ for the $f/25$ harmonics, to between 6 and 8 nJ for harmonics generated at $f/50$, and increases to better that 15 nJ per harmonic for those generated with $f/70$ focusing. The yield of the 21st harmonic (at 25 nm) is plotted in Fig. 3(b) as a function of the confocal parameter for an intensity of 6×10^{15} W/cm^2. The yield is linear with b, confirming the scaling predicted by Eq. (4). The conversion efficiency, however, is constant with confocal parameter for any given intensity. Though the harmonic energy yield increases linearly with b, the confocal parameter is proportional to the focal spot area, so a commensurate increase in laser energy is required to maintain a given peak intensity.

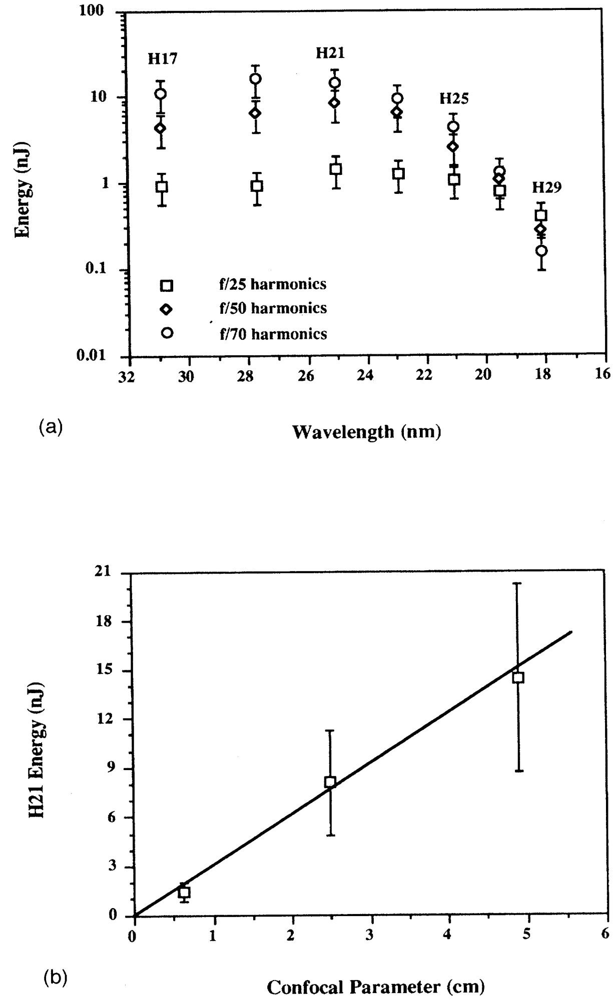

FIG. 3. (a) Energy yields of harmonics produced by 526-nm light in helium at a peak intensity of 6×10^{15} W/cm^2 for three different focal configurations: $f/25$ (squares), $f/50$ (diamonds), and $f/70$ (circles). (b) Energy yield of the 21st harmonic as a function of confocal parameter. The line shows the linear dependence of the yield with b.

We find that the best harmonic yields and conversion efficiencies are achieved with intensities well above the saturation intensity. Figure 4(a) shows the yields of the 21st harmonic of 526-nm light generated in helium as a function of peak laser intensity for $f/50$ focusing. The corresponding conversion efficiencies are shown in Fig. 4(b). For this focusing geometry we measured harmonic energies near 60 nJ at a peak intensity of 1.1×10^{16} W/cm^2 with a drive energy of 590 mJ. This corresponds to a conversion efficiency of $>10^{-7}$ from the laser into the harmonic at 25 nm. Similar yields ($\sim$40 nJ) were attainable at a lower intensity with $f/70$ focusing, giving conversion efficiencies that are comparable to those generated at $f/50$. Harmonics generated in helium are also capable of generating harmonic yields of nanojoules at wavelengths below 20 nm. The yields [Fig. 4(a)] and conversion efficiencies [Fig. 4(b)] of the 27th harmonic (at 19.5 nm) are shown for $f/50$ focusing. We mea-

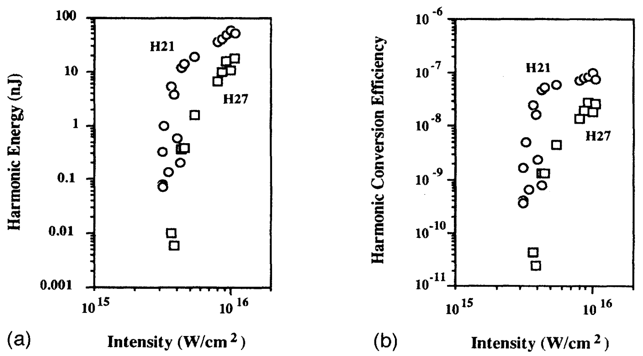

FIG. 4. (a) Energy yields of the 21st harmonic (at 25.1 nm) and the 27th harmonic (at 19.5 nm) as a function of intensity. Harmonics were produced by 526-nm light in helium with $f/50$ focusing. (b) Harmonic conversion efficiencies of the 21st and 27th harmonics under the same conditions.

sured harmonic energies of over 15 nJ at this wavelength at an intensity of 1.5×10^{16} W/cm^2. This corresponds to a conversion efficiency of 3×10^{-8}.

The saturation in the harmonic yields at the highest intensities is due to the depletion of the ground state and the creation of free electrons by photoionization. When the peak intensity is roughly ten times the ionization saturation intensity, the entire focal volume undergoes nearly complete ionization by the laser pulse. This effect manifests itself in the far-field profile of the harmonics as well. At a peak intensity around ten times I_{sat} we see ionization-induced refraction of the laser beam. We also observe severe breakup of the harmonics profile at this intensity attributable to the refraction of the fundamental beam. This degradation of the harmonic beam profile above an intensity of 10^{16} W/cm^2 essentially precludes the generation of harmonics at higher intensities for any applications.

In conclusion we have measured the energy yields of harmonics produced in the soft-x-ray spectral range of 31–17 nm. We have measured energy yields of up to 60 nJ in the 31–23-nm spectral range corresponding to a conversion efficiency of 10^{-7} for harmonics generated in helium by 650-fs, 526-nm laser pulses. We have also measured harmonics with energies in excess of 15 nJ for wavelengths below 20 nm. The harmonic yields in this wavelength range can be optimized by using a short-wavelength drive laser at focused intensities well above the ionization saturation intensity. We have measured three-orders-of-magnitude enhancement in the harmonic yields by using the second harmonic of our Nd:glass laser at 526 nm over harmonics generated by the laser fundamental at 1053 nm. At very high intensities we find that yields in the plateau are roughly similar for He and Ne, though shorter wavelengths are attainable with helium because of its higher ionization potential. The harmonic yields for a given intensity can also be increased by decreasing the speed of the focus, thereby increasing the confocal parameter. We find that the optimum condition for the generation of harmonics at wavelengths below 30 nm with a subpicosecond 1-μm laser is with the second harmonic focused to intensities of around 10^{16} W/cm^2.

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