Bright Multi-keV Harmonic Generation from Relativistically Oscillating Plasma Surfaces

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(Received 12 January 2007; published 23 August 2007)

The first evidence of x-ray harmonic radiation extending to 3.3 Å, 3.8 keV (order $n > 3200$) from petawatt class laser-solid interactions is presented, exhibiting relativistic limit efficiency scaling ($\eta \sim n^{-2.5}$–n^{-3}) at multi-keV energies. This scaling holds up to a maximum order, $n_{\mathrm{RO}} \sim 8^{1/2}\gamma^3$, where γ is the relativistic Lorentz factor, above which the first evidence of an intensity dependent efficiency rollover is observed. The coherent nature of the generated harmonics is demonstrated by the highly directional beamed emission, which for photon energy $h\nu > 1$ keV is found to be into a cone angle $\sim 4°$, significantly less than that of the incident laser cone (20°).

DOI: 10.1103/PhysRevLett.99.085001 PACS numbers: 52.59.Ye, 42.65.Ky, 52.38.Ph, 52.50.Jm

Coherent high order harmonic x-ray generation (HOHG) has the potential to open up the world of physical processes on an attosecond time scale [1–3]. The key to this is converting high-power optical laser pulses into broad, phase-locked harmonic spectra extending to multi-keV photon energies—which can be achieved, with unprecedented efficiency and brightness, by reflection off relativistically oscillating plasmas [2,3]. Of particular note is the implication this has for the production of high brightness attosecond pulses [3]. For an attosecond pulse with a fixed fractional bandwidth at a given central frequency $n_{\mathrm{cf}}\omega_{\mathrm{laser}}$ the energy in the pulse scales as [3]

$$\eta_{\mathrm{att}} \sim n_{\mathrm{cf}}^{-1.5}, \tag{1}$$

where n_{cf} is the harmonic order of the carrier frequency and ω_{laser} the laser frequency.

The unique properties of such a source have lead to the investigation of its potential for use in many exciting applications [1,3,4]. The availability of bright attosecond x-ray pulses will allow the probing of the dynamics and properties of atoms and molecules on temporal scales shorter than that of the period of atomic vibrations, i.e., attosecond resolution of bound-free electronic transitions (e.g., from the $4p$ state of krypton) [5,6].

Recently, HOHG pulse production has been cited as a possible route to achieving the huge intensities required for probing the nonlinear quantum electrodynamical properties of the vacuum, providing a significant intensity boost for existing or imminently anticipated laser technology and highlighting the enormous potential of HOHG [4]. These predictions rely on the fact that the focused harmonic radiation can in principle have a substantially *higher* intensity $I_{\max}$ than that of the laser I used to generate them, scaling as $I_{\max} \sim I n_{\mathrm{cf}}^{+1.5}$. This is the result of the slow decay of the conversion efficiency for pulse generation ($\sim n_{\mathrm{cf}}^{-1.5}$), coupled with the increased focusability ($\sim n_{\mathrm{cf}}^{2}$) and temporal compression of the reflected energy ($\sim n_{\mathrm{cf}}$). For example, an incident intensity of $\sim 10^{22}$ W cm^{-2} could be refocused to $> 10^{29}$ W cm^{-2} corresponding to the critical Schwinger limit [7] electric field of $\sim 10^{16}$ V cm^{-1} for electron-positron pair production from the vacuum [8].

In this Letter we show, for the first time, HOHG extending to multi-keV energies and the first experimental evidence for a high frequency rollover of relativistic limit conversion efficiency scaling [2]. This result corresponds to the most extreme nonlinear optical process observed to date in the laboratory (harmonic order $n > 3200$).

HOHG [9–12] essentially results from an oscillatory extension to Einstein's prediction for the frequency upshift of light reflected off a perfect mirror moving at relativistic velocities. In this theory a pulse of duration Δt at frequency ω_0 is shifted to a frequency of $\omega' = 4\gamma_{\max}^2\omega_0$ and compressed to a pulse duration of $\Delta t' = \Delta t/4\gamma_{\max}^2$ (where $\gamma_{\max}$ is the maximum relativistic Lorentz factor of the oscillating plasma surface) [13]. When an intense laser pulse interacts with a near discontinuous plasma-vacuum boundary the electric field of the laser can efficiently couple to the plasma surface [14], causing the electrons to oscillate in phase [1–3,10–12,15–17], effectively constituting a relativistic mirror oscillating at the laser frequency ω_{laser}.

As the position of this mirror surface is a temporal function of the incident optical laser cycle, the phase of the reflected light wave is modulated such that it is no longer sinusoidal and, as can be understood from Fourier theory, contains many high order harmonics of the fundamental frequency. For lower intensities the expected harmonic spectrum is very well described by the moving mirror model [11]. However, the moving mirror model does not have any predictive power as to the ultimate efficiency that might be expected in the relativistic limit ($a0 \gg 1$) and the ultimate extent of the harmonic spectrum.

The most recent theoretical development in the field, based on similarity theory [16], identifies the sharp spikes in the temporal variation of the Lorentz factor γ as the key to the production of the highest harmonics. From this theory of "γ spikes" the conversion efficiency in the relativistic limit for the nth harmonic is predicted to scale as

$$\eta(n) \sim n^{-\mathrm{Prel}}, \tag{2}$$

with Prel $= 8/3$ [16]. Another important result of this theory is the prediction of the highest harmonic where Eq. (1) still applies up to an order $n_{\mathrm{RO}} \sim 8^{1/2}\gamma_{\max}^3$, beyond which the conversion efficiency decreases more rapidly or "rolls over" [where $\gamma_{\max} = (1 + 3.6 \times 10^{-19} I\lambda^2)^{1/2}$ corresponds to the maximum surface velocity, I (W cm^{-2}) is the peak intensity, and λ (μm) the wavelength of the laser]. This is in contrast to the moving mirror model, where one would anticipate a cutoff or at least a rapid decrease in efficiency for harmonic orders $n > 4\gamma_{\max}^2$ [15]. The physical origin of this new prediction derives directly from the γ spikes. Since the emission of high harmonic orders only takes place for large values of $\gamma_{\max}$ a sharp temporal localization of the emitted harmonics results. The temporal duration of the γ spikes reduces for increasing intensity as $T_{\mathrm{spike}} \sim T_0/\gamma_{\max}$ (with $T_0 = 2\pi/\omega_0$). The pulses of duration T_{spike} are up-shifted and compressed by the factor of $4\gamma_{\max}^2$—familiar from the relativistic mirror. As a result the harmonics are emitted in short temporal bursts with $T_{\mathrm{burst}} \sim T_{\mathrm{spike}}/\gamma_{\max}^2 \sim T_0/\gamma_{\max}^3$. The fact that the harmonics extend to substantially higher frequencies than would be expected from the moving mirror model alone is therefore a direct result of the attosecond temporal bunching of the emitted harmonics—and consequently an observation of the $n_{\mathrm{RO}} \sim 8^{1/2}\gamma_{\max}^3$ is evidence for the attosecond temporal bunching of the harmonic emission [note that in Eq. (1) the conversion efficiency into a single attosecond pulse $\eta_{\mathrm{att}} \sim n_{\mathrm{cf}}^{-2.5}\Delta n \sim n_{\mathrm{cf}}^{-1.5}$ takes the increased bandwidth with increasing n_{cf} into account [3]]. In effect, the surprising new result of the theory of relativistic spikes is that the high energy cutoff and the ultimate slope of the spectrum are governed by the temporal compression and truncation of the electromagnetic pulse rather than the maximum up-shift expected from a relativistic mirror moving at constant γ.

Experiments were performed at the Vulcan petawatt laser at the Rutherford Appleton Laboratories [18] which reaches peak intensities of $\sim 10^{21}$ W cm^{-2} (up to 600 J on target in $\sim$500 fs). However, with an intrinsic prepulse pedestal that extends for $\sim$5 ns at a contrast of 10^7:1, it is unsuitable for the formation of the short plasma scale length $L_s \sim 0.1\lambda$ required for efficient harmonic generation [14], where λ is the laser wavelength. As a result the contrast of the incident pulse was increased to $>10^{10}$:1 at $\sim$10 ps from the peak of the pulse by the use of a double plasma mirror setup [19–21] to achieve the required sharp plasma-vacuum interface.

The laser focus size was routinely monitored by imaging the size of the emission region at $3\omega_{\mathrm{laser}}$ to ensure high intensity both with and without plasma mirrors [22]. The x-ray signal was observed using a broadband crystal spectrometer consisting of mica crystal in a von Hamos geometry [23] and a Fujifilm "Image Plate" detector (BAS with 9 μm CH protective layer) [24]. The point spread function of the spectrometer was experimentally determined and the resolution ($\lambda/\Delta\lambda$) of the spectrometer determined to be $\sim$200 (limited by the crystal bending inaccuracies). Both detectors were placed in the specular direction for oblique (45°) incidence and were not moved when the target was rotated to normal (0°) incidence. Angular distribution measurements were performed using pieces of Image Plate giving data over a 2π range. Differential filtering (5 μm Mg, 5 μm and 15 μm Al) was used for signal discrimination. The observed signal levels were consistent only with x-ray emission. The possibility of low energy electrons significantly contributing to the signal was eliminated by the inclusion of deflecting magnets.

With recent experimental observations [19] confirming HOHG efficiency scaling in the relativistic limit to <4 nm, the essential question remains as to whether this efficiency scaling does indeed extend to the very high harmonic orders required to produce intense attosecond pulses at angstrom wavelengths.

Harmonic spectra from smooth CH targets (few nanometer surface roughness) and under high contrast conditions, in the $\sim$9–3.3 Å spectral region, are presented in Fig. 1. These demonstrate for the first time that harmonics with keV photon energies can indeed be generated using this technique. The observed spectrum is consistent with the predicted efficiency scaling [Eq. (2)] in the relativistic limit [16,19] up to $\sim$2600th ($\sim$3 keV) order for an incident intensity of $(1.5 \pm 0.3) \times 10^{20}$ W cm^{-2}, and up to $\sim$3000th order ($\sim$3.5 keV) for $(2.5 \pm 0.5) \times 10^{20}$ W cm^{-2}. At the highest intensity the observed spectra extend to $\sim$3.8 keV, or the 3200th order of 1055 nm fundamental (3.3 Å) before dropping to the noise floor of the detector. This is the first ever observation of >1000th order from any harmonic generation process and is substantially higher than the previous best results from solid targets (75th order [25], 850th order [19]).

The relative signal strengths shown in Fig. 1 were obtained by correcting the detected signal for the transmission function of the spectrometer (mica crystal reflectivity, Al filter, CH protective layer, and image plate response). The mica crystal reflectivity curve is given by Henke *et al.* [26] and the relative strength and resolution of the Bragg diffraction orders is given by Holzer *et al.* [27]. The largest uncertainty in correcting the signal is due to the uncertainty in the spectral shape at the highest photon energies which affects the accuracy with which the spectrum can be determined at longer wavelengths due to contributions from higher Bragg order reflections.

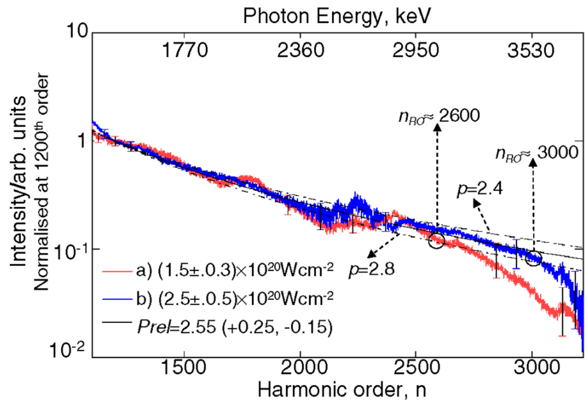

FIG. 1 (color online). The relative intensity of the harmonic spectra for two intensities: (a) $(1.5 \pm 0.3) \times 10^{20}$ W cm^{-2} [gray (red) trace] and (b) $(2.5 \pm 0.5) \times 10^{20}$ W cm^{-2} [black (blue) trace]. Spectra are integrated along the spatial dimension and normalized at the 1200th harmonic ($\sim$ 1.4 keV, 8.8 Å). The lines are fits to the data such that $I(n)/I(1200) = n^{-p}/1200^{-p}$, where p is the fitting parameter. The best fit (solid line) is for a value of Prel = 2.55 for (a) in the range 1.2–3 keV and (b) in the range 1.2–3.5 keV, and is consistent with that expected for harmonic generation in the relativistic limit. The dashed lines represent $p = 2.4$ and $p = 2.8$ scaling, as labeled. Error bars [gray (red) ends for red trace, black (blue) ends for blue trace)] represent the uncertainty in the relative signal strength arising from the detector and filter transmission, taking into account the individual uncertainty in each of the relevant quantities. The absence of strong modulation in the spectra is due to the limited resolution of the crystal spectrometer used.

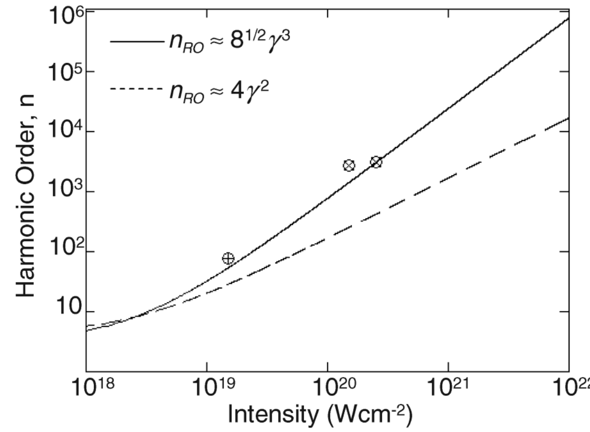

FIG. 2. Comparison of the $8^{1/2}\gamma^3$ (solid line) and $4\gamma^2$ (dashed line) scaling for the maximum relativistic limit harmonic order n_{RO} with experimentally observed data. The $8^{1/2}\gamma^3$ scaling is clearly a better fit to the highest observed harmonic from previous experiments (⊕) [26], and the two values of n_{RO} presented in this Letter (⊗). The rapid scaling of n_{RO} with intensity allows the extension of attosecond and zeptosecond pulse generation to subangstrom wavelengths with realistic lasers.

The relativistic exponent derived from the data is Prel = 2.55(+0.25, −0.15), demonstrating the slow decay to high orders that is essential for efficient attosecond production. At the highest orders the efficiency scaling is characterized by a departure from the scaling given by Eq. (2). The exact position of this rollover is found to be intensity dependent and is defined here as the point where the fitting parameter $p > 2.8$ relative to $n = 1200$. Using this definition we obtain $n_{\mathrm{RO}} \sim 2600$ and $\sim$3000 for incident intensities of 1.5 and 2.5×10^{20} W cm^{-2}, respectively.

For our experimental conditions we estimate $\gamma_{\max} = 10, \ldots, 13$ which would correspond to a value of $n_{\mathrm{RO}} \sim 500$ based on the rollover at $n_{\mathrm{RO}} \sim 4\gamma_{\max}^2$ derived from the moving mirror model. From the results presented in Fig. 1, it is clear that the onset of this rollover regime begins at much higher orders with $n_{\mathrm{RO}} > 3000$ for $(2.5 \pm 0.5) \times 10^{20}$ W cm^{-2}. These measurements are, however, in good agreement with most recent theory [16], which predicts $n_{\mathrm{RO}} \approx 8^{1/2}\gamma_{\max}^2$, based on interpreting the x-ray harmonic generation process as being due to the sharp spikes in the relativistic γ factor of the plasma surface described above. As shown in Fig. 2 the dependence of n_{RO} is also consistent with the highest observed harmonics from previous measurements [25].

As stated above the observation of $n_{\mathrm{RO}} \approx 8^{1/2}\gamma_{\max}^3$ is consistent only with the emission of attosecond radiation bursts (a pulse train under the conditions of this experiment). Therefore our measurements imply conversion efficiencies as high as $\sim 10^{-5}$ ($h\nu > 1$ keV) and $\sim 10^{-2}$ ($h\nu > 20$ eV) into an attosecond pulse train. Based on the results presented in this Letter and current theory, it should in principle be possible to generate 10 keV zeptosecond pulses [2] with conversion efficiency of $>10^{-7}$ using intensities $\sim 7 \times 10^{20}$ W cm^{-2}—a level readily achievable with the current generation of lasers. The experimentally observed signal is consistent with harmonic energies of $\sim$17 μJ at 1200th (1.4 keV) and $\sim$5 μJ at 2600th (3.1 keV) in 1% bandwidth, respectively. This is comparable to the state of the art at even the largest facilities for coherent x-ray generation at such energies.

For high pulse contrast and smooth (few nanometer surface roughness) CH targets, the HOHG signal >1 keV is observed to be emitted into a cone angle $\sim$4° full width at half maximum (FWHM) with a low-intensity halo of $\sim$13° (Fig. 3). This 4° cone is consistent with harmonics being reflected from a surface with a small amount of curvature induced by the laser ponderomotive pressure [28]. Assuming that the beam divergence is primarily determined by surface curvature, it is possible that the observed beam corresponds to a near diffraction limited, highly focusable beam. Using shorter pulses would substantially reduce the surface denting for a given intensity and is consequently expected to lead to reduced beam divergence. The low-intensity halo is likely due to surface roughness. These observations are indicative of strong smoothing of the initial interaction surface due to plasma expansion and/or Lorentz contraction. The total energy in the angularly integrated signal is estimated to be consistent with that expected from the power-law efficiency scaling [Eq. (1)] to within experimental uncertainties, which are

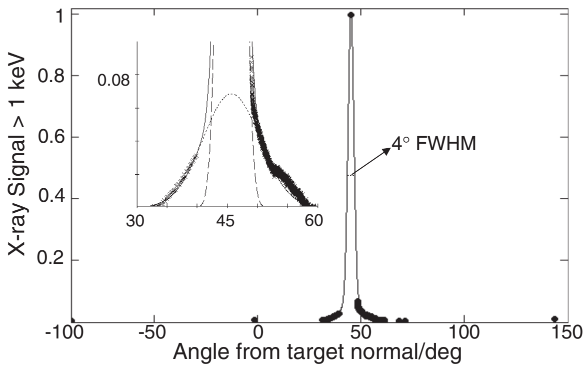

FIG. 3. Angular distribution of >1 keV x-ray signal under high contrast conditions. The signal is emitted into a narrow cone peaked in the specular direction at 45° (the laser incidence angle is −45°). The inset shows a 13° full width at half maximum (FWHM) Gaussian fit to the scattered x-ray halo (black dotted line) and the 4° fit to the strongly peaked HOHG signal (black dashed line). The full width at half maximum of the summed double Gaussian fit to the signal is ~4° (solid black line), which is considerably less than the $f/3$ laser cone angle. The specular emission of the harmonics is clear indication of the coherent nature of the process. It also demonstrates the absence of significant laser induced surface modulation present on previous, lower contrast experiments [26].

estimated to be approximately a factor of 2–3. These are dominated by the angular distribution (with further contributions from filters, crystal, and the detector). Ultimately the reflection of subnanometer wavelengths into a narrow cone demonstrates clearly the coherent nature of HOHG. Consistent with the result for keV harmonics, the $3\omega_{\text{laser}}$ radiation was only observed with the collection mirror positioned in the specular direction for nanometer-roughness targets.

By contrast, for gold foils with micron scale surface roughness, $3\omega_{\text{laser}}$ emission was observed both in specular and at 90° to specular, indicating that the angular distribution is indeed strongly correlated to the shape or roughness of the target surface. No x-ray harmonic signal was recorded for such rough targets. It is assumed that the wide angular distribution due to scattering reduced the signal level to below the noise floor of the x-ray detector.

Without contrast enhancing plasma mirrors and for comparable intensities, the x-ray spectra were observed to be dramatically different from those obtained with plasma mirrors. Under these low contrast conditions the x-ray signal is found to be consistent with that from bremsstrahlung due to hot electrons, resulting in a peaked spectrum—approximately Planckian in shape—which shifts to higher energies for higher intensities.

Based on the scalings for efficiency and n_{RO} typical photon numbers [from Eq. (1)] expected per attosecond/zeptosecond pulse [3] for the given central energies are $\sim 7 \times 10^{12}$ photons/pulse at 1.5 keV, $\sim 2 \times 10^{12}$ photons/pulse at 2.5 keV, and $\sim 8 \times 10^{11}$ photons/pulse at 3.5 keV. In this context, these photon numbers appear suitable for the proposed high resolution (10 s of angstrom) imaging of biomolecules using ultrashort x-ray pulses [4]. This technique has recently been verified using intense radiation from the FLASH soft-x-ray free-electron laser at 32 nm for the diffraction limited imaging of a complex nanostructured object [29]. Based on this, and the results presented here, the prospects for future femtosecond, attosecond, and zeptosecond keV x-ray pulses looks very bright indeed.

In conclusion we have demonstrated harmonic efficiency scaling in the relativistic limit for $h\nu > 1$ keV and presented the first evidence of an intensity dependent roll-over in efficiency scaling corresponding to $n_{\text{RO}} \approx 8^{1/2}\gamma_{\text{max}}^3$. These results are consistent with the most recent analytical theory [16].

The authors would like to acknowledge helpful discussions with Teodora Baeva and the excellent support from the VULCAN staff. This work was supported by EPSRC . M. Z. acknowledges support from the Royal Society.

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[1] A. Pukhov, Nature Phys. **2**, 439 (2006).
[2] Gordienko *et al.*, Phys. Rev. Lett. **93**, 115002 (2004).
[3] G. D. Tsakiris *et al.*, New J. Phys. **8**, 19 (2006).
[4] R. Neutze *et al.*, Nature (London) **406**, 752 (2000).
[5] M. Hentschel *et al.*, Nature (London) **414**, 509 (2001).
[6] C. Pellegrini and J. Stöhr, Nucl. Instrum. Methods Phys. Res., Sect. A **500**, 33 (2003).
[7] J. Schwinger, Phys. Rev. **82**, 664 (1951).
[8] W. Heisenberg and H. Euler, Z. Phys. **98**, 714 (1936).
[9] R. L. Carman, D. W. Forslund, and J. M. Kindel, Phys. Rev. Lett. **46**, 29 (1981).
[10] S. V. Bulanov, N. M. Naumova, and F. Pegoraro, Phys. Plasmas **1**, 745 (1994).
[11] R. Lichters, J. Meyer-ter-Vehn, and A. Pukhov, Phys. Plasmas **3**, 3425 (1996); R. Lichters and J. Meyer-ter-Vehn, *Multiphoton Processes 1996*, Institute of Physics Conference Series Vol. 154 (IOP, London, 1997), p. 221.
[12] P. Gibbon, Phys. Rev. Lett. **76**, 50 (1996).
[13] A. Einstein, Ann. Phys. (Leipzig) **17**, 891 (1905).
[14] M. Zepf *et al.*, Phys. Rev. E **58**, R5253 (1998).
[15] S. Gordienko *et al.*, Phys. Rev. Lett. **94**, 103903 (2005).
[16] T. Baeva, S. Gordienko, and A. Pukhov, Phys. Rev. E **74**, 046404 (2006).
[17] L. Plaja *et al.*, J. Opt. Soc. Am. B **15**, 1904 (1998).
[18] C. N. Danson *et al.*, Nucl. Fusion **44**, S239 (2004).
[19] B. Dromey *et al.*, Nature Phys. **2**, 456 (2006).
[20] B. Dromey *et al.*, Rev. Sci. Instrum. **75**, 645 (2004).
[21] G. Doumy *et al.*, Phys. Rev. E **69**, 026402 (2004).
[22] B. Dromey (to be published).
[23] L. Von Hamos, Z. Kristallogr. **101**, 17 (1939).
[24] S. G. Gales and C. D. Bentley, Rev. Sci. Instrum. **75**, 4001 (2004).
[25] P. Norreys *et al.*, Phys. Rev. Lett. **76**, 1832 (1996).
[26] B. L. Henke *et al.*, At. Data Nucl. Data Tables **54**, 181 (1993).
[27] G. Holzer *et al.*, Phys. Scr. **57**, 301 (1998).
[28] S. C. Wilks *et al.*, Phys. Rev. Lett. **69**, 1383 (1992).
[29] H. Chapman *et al.*, Nature Phys. **2**, 839 (2006).