Bright multi-keV harmonic generation from relativistically oscillating plasma surfaces

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Introduction

High order harmonic generation (HOHG) from intense laser - solid density interactions has emerged as a promising route to the generation of attosecond pulses extending to keV energies [1-3]. With efficiency of the nth harmonic order, η_n, scaling as ~ $n^{-2.5}$–n^{-3} in the relativistic limit [4] up to a maximum harmonic order $n_{max} \sim 8^{1/2}\gamma^3$ (where γ is the maximum relativistic Lorentz factor of the oscillating plasma surface), the potential for a bright solution to attosecond science is a distinct possibility. For an attosecond pulse with a fixed fractional bandwidth at a given central frequency $n_{cf}\omega_{Laser}$ the energy in the pulse scales as $\eta_{att} \sim n_{cf}^{-1.5}$ (Eqn. 1) [2], where n_{cf} is the harmonic order of the carrier frequency and ω_{Laser} the laser frequency.

The unique properties of such a source has lead to the investigation of its potential for use in many exciting applications. 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 and the study of new regimes of high field physics.

Here we show, for the first time, HOHG extending to multi-keV energies and the first experimental evidence for a high frequency roll-over of relativistic limit conversion efficiency scaling [3]. This result corresponds to the most extreme nonlinear optical process observed to date in the laboratory (harmonic order $n > 3200$).

HOHG essentially results from an oscillatory extension to Einstein's prediction for the frequency up-shift 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 $\Delta t' = \Delta t/4\gamma_{max}^2$. 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, causing the electrons to oscillate in phase effectively constituting a relativistic mirror oscillating at the laser frequency ω_{Laser}.

The most recent theoretical development in the field, based on similarity theory [4], identifies the sharp spikes in the temporal variation of the Lorenz 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 $\eta_n \sim n^{-Prel}$ (Eqn. 2), with Prel = 8/3 [4]. Another important result of this theory is the prediction of the highest harmonic where the Eqn. 1 still applies up to an order $n_{RO} \sim 8^{1/2}\gamma_{max}^3$, beyond which the conversion efficiency decreases more rapidly or 'rolls over' (where $\gamma_{max}=(1+3.6\times10^{-19}I\lambda^2)^{1/2}$ corresponds to the maximum surface velocity, I (Wcm^{-2}) is the peak intensity and λ (µm) the wavelength of the laser).

Experiment

The Vulcan Petawatt laser at the Rutherford Appleton Laboratory is ideally suited for generating such harmonic radiation as it can readily reach the desired peak intensities of ~10^{21}Wcm^{-2}, delivering up to ~600 J on target in ~500 fs. However, with an intrinsic prepulse pedestal that extends for ~5 ns at a contrast of 10^7:1, it is unsuitable for the formation of the short plasma scalelength, L_s, ~ 0.1λ required for efficient harmonic generation, where λ is the laser wavelength. As a result the contrast of the incident pulse was increased to >10^{10}:1 at ~10 ps from the peak of the pulse by the use of a double plasma mirror setup to achieve the required sharp plasma-vacuum interface.

The laser focus size was routinely monitored by imaging the size of the emission region at $3\omega_{Laser}$ to ensure high intensity both with and without plasma mirrors. The x-ray signal was observed using a broadband crystal spectrometer consisting of mica crystal in a von Hamos geometry and a Fujifilm 'Image Plate' detector (BAS with 9 µm CH protective layer). The point spread function of the spectrometer was experimentally determined and the resolution ($\lambda/\Delta\lambda$) of the spectrometer determined to be ~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.

Results

Harmonic spectra from smooth CH targets (few nm surface roughness) and under high contrast conditions, in the ~ 9Å - 3.3Å spectral region, are presented in Figure 1.

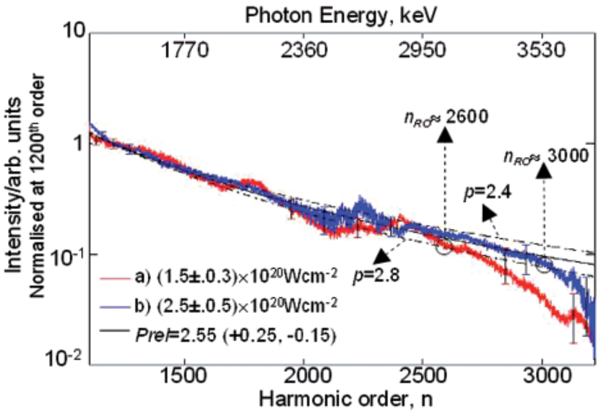

Figure 1. The relative intensity of the harmonic spectra for two intensities: a) $(1.5\pm0.3)\times10^{20}$ Wcm^{-2} (red trace) and b) $(2.5\pm0.5)\times10^{20}$ Wcm^{-2} (blue trace). Spectra are integrated along the spatial dimension and normalised at the 1200th harmonic (~1.4 keV, 8.8 Å). The lines 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 keV – 3 keV and b) in the range 1.2 keV – 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 (red ends for red trace, 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 the limited resolution of the crystal spectrometer used.

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 (Eqn 2) in the relativistic limit up to ~2600th (~3 keV) order for an incident intensity of $(1.5\pm0.3)\times10^{20}$ Wcm^{-2}, and up to ~3000th order (~3.5 keV) for $(2.5\pm0.5)\times10^{20}$ Wcm^{-2}. At the highest intensity the observed spectra extend to ~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[5], 850th order[6]).

The relative signal strengths shown in Figure 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 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.

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 characterised by a departure from the scaling given by Eqn. 2. The exact position of this roll-over 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_{RO} ~2600 and ~3000 for incident intensities of 1.5 and 2.5×10^{20} Wcm^{-2} respectively.

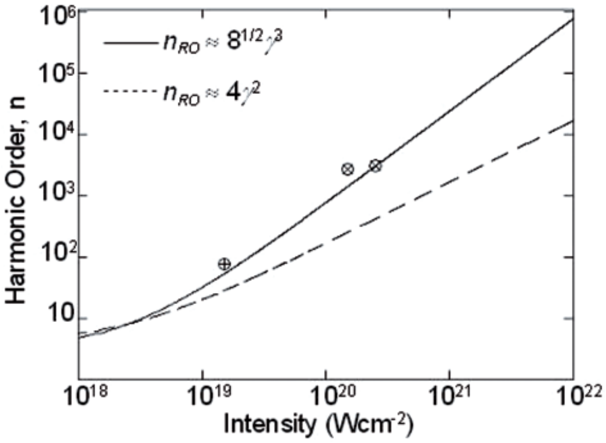

Figure 2. Comparison of the $8^{1/2}\gamma^3$ (solid line) and $4\gamma^2$ (dashed line) scaling for the maximum relativistic limit harmonic order, n_{RO}, with experimentally observed data. The $8^{1/2}\gamma^3$ scaling is clearly a better fit to the highest observed harmonic from previous experiments, ⊕[5], and the two values of n_{RO} presented in this paper, ⊗. The rapid scaling of n_{RO} with intensity allows the extension of atto- and zeptosecond pulse generation to sub-Angstrom wavelengths with realistic lasers.

For our experimental conditions we estimate γ_{max} =10…13 which would correspond to a value of n_{RO} ~500 based on the roll over at n_{RO} ~$4\gamma_{max}{}^2$ derived from the moving mirror model. From the results presented in Figure 1, it is clear that the onset of this roll-over regime begins at much higher orders with n_{RO} >3000 for $(2.5\pm.5)\times10^{20}$ Wcm^{-2}. These measurements are however in good agreement with most recent theory[4], which predicts $n_{RO} \approx 8^{1/2}\gamma_{max}{}^3$, 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 Figure 2 the dependence of n_{RO} is also consistent with the highest observed harmonics from previous measurements[5].

Based on the results presented in this letter and current theory, it should in principle be possible to generate 10 keV zeptosecond pulses[1] with conversion efficiency of $>10^{-7}$ using intensities ~7×10^{20} Wcm^{-2} – a level readily achievable with the current generation of lasers. The experimentally observed signal is consistent with harmonic energies of ~17 μJ at 1200th (1.4 keV) and ~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 nm surface roughness) CH targets, the HOHG signal >1 keV is observed to be emitted into a cone angle ~4° full width half max with a low-intensity halo of ~13° (Figure 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. 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

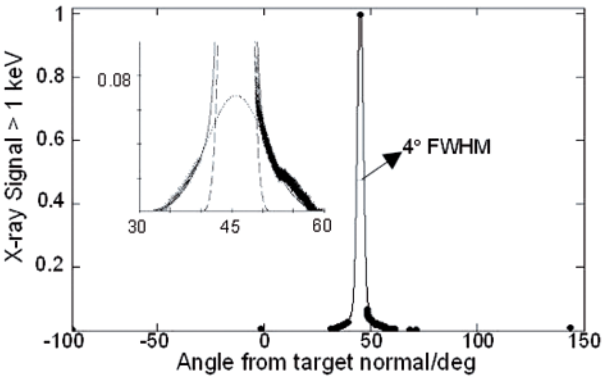

Figure 3. Angular distribution of > 1 keV x-ray signal under high contrast conditions. The signal is emitted into a narrow cone peaked in the specular direction at 45° (the laser incidence angle is minus 45°). The inset shows a 13° full width half max Gaussian fit to the scattered x-ray halo (black dotted) and the 4° fit to the strongly peaked HOHG signal (black dashed). The full width half max (FWHM) 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[5].

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

By contrast, for gold foils with micron scale surface roughness, $3\omega_{Laser}$ emission was observed both in specular and at 90° to specular, indicating that the angular distribution is indeed strongly correlated to the shape/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 to 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.

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 rollover in efficiency scaling corresponding to $n_{RO} \approx 8^{1/2}\gamma_{\max}^{3}$. These results are consistent with the most recent analytical theory[4].

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