Beam quality and conversion efficiencies of harmonics generated from overdense plasma

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Introduction

Coherent high order harmonic generation (HOHG) from overdense plasmas is expected to provide a unique source for producing intense, short wavelength, attosecond radiation from table top lasers. These attosecond pulses are generated by temporal truncation of the harmonic emission from relativistically oscillating plasma surfaces. Such an XUV/X-ray (1-100 nm) coherent light source is of great interest to many research fields. While promising to provide new insight into new research areas such as ultrafast biomedical imaging^[1], attosecond pulses may also be used to probe and explore the structural dynamics of atoms and molecules on unprecedented timescales. HOHG also has the prospect of seeding current generation X-ray free electron lasers^[2].

The advantage of generating high harmonics using solid targets in preference to gases is that the plasma medium can, in principle, permit the use of arbitrarily high laser intensities, and consequently the intensity and brightness of the attosecond pulses produced can be orders of magnitude greater.

However, the future and possible uses of this light source depends on the quality of the radiation that can be produced. This includes not only the conversion efficiency of the fundamental driving laser into attosecond pulses but also the beam quality/divergence of the emitted radiation.

Theory

The interaction of an intense ultrafast laser pulse with a short scale length ($<0.2\lambda_{I}$) overdense plasma will result in the generation of many harmonics of the incident laser pulse in the specular reflection. Recently it has been shown that the production of these harmonics depends strongly on the intensity of the interaction. At intensities $<10^{18}$ Wcm⁻² the harmonics are generated within the bulk plasma gradient via Coherent Wake Emission (CWE)^[3] and at intensities in the relativistic regime $(>10^{18} \text{ Wcm}^{-2})$ by a relativistically oscillating mirror-like critical density surface (ROM)^[4-6]. The CWE process can only support harmonic generation up to the maximum plasma frequency, corresponding to the maximum density of the solid target. However over the past several years the number of harmonics generated via ROM has steadily grown (>3000th)^[5] with increasing laser intensity and

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Figure 1. Spectrometer configurations. (A) Used to angularly select harmonic emission from the interaction. The on specular axis emission was apertured such that only onaxis emission could be observed on the CCD. Harmonic emission into larger angles was collected using a gold focussing optic. (B) Used for greater angular range.

should continue to do so with improving laser performance. Theoretical work by T. Baeva *et al.*^[6] has shown that the efficiency of the nth harmonic scales as $\sim n^{-8/3}$ in the relativistic limit 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).

Here we present a study of the beam quality and conversion efficiency of the harmonics generated by these two competing processes. It is hoped that the study of these properties will accelerate the optimisation of this light source.

Experiment

Using the Titanium sapphire Astra laser the angular divergence, beam quality and conversion efficiency of harmonics produced via CWE and ROM processes was studied. The high power laser pulse was focused onto a sub-nm rms surface roughness fused silica target at ~30°.

The Astra laser has a fundamental wavelength of 800 nm and delivers pulses of 800 mJ in ~50 fs. The intensity contrast (ratio of the peak of the pulse to pre-pulse level) of the Astra laser at <500 fs prior to the arrival of the main pulse was >10⁷:1. To improve this contrast and achieve the short scale length plasma required for HOHG a plasma mirror^[7] was inserted into the beam path improving the contrast to >10⁹:1.

The harmonics generated in the specular reflection were analysed using a Hitachi grazing incidence (4°) Flat field spectrometer. The spectrometer was set so that the source to detector distance was \sim 1.1 m. The spectrometer was used in two different configurations (figure 1) depending on experimental requirements.

Results

Spectrometer configuration A was used to characterise the angular distribution of CWE and ROM harmonics on a single shot basis, simultaneously distinguishing between on and off specular reflection axis emission. The length of the focussing mirror was \sim 20 cm, allowing the collection of harmonic radiation emitted into angles greater than \sim 35 mrad.

Figure 2 shows spectra recorded as the target was moved through focus. At approximately 200 µm out of focus (figure 2(a) – Intensity ~ 10^{17} Wcm⁻²) harmonic orders (14-19) were detected both along the specular axis and from the focussing optic, corresponding to harmonics with 1/e² divergence of approximately 35-40 mrad (2-3 times diffraction limited). These orders are unambiguously due to CWE ($< \omega_n$, sub-relativistic intensity). In tight focus (figure 2(b) – relativistic intensity $\sim 2 \times 10^{19}$ Wcm⁻²) many more orders were observed along the specular axis, extending to the 26th harmonic. However the higher order harmonics (20-26) are not collected by the gold focussing optic (off axis signal), and as such are beamed into a significantly narrower cone angle. These orders are unmistakably ROM ($\geq \omega_n$) and have divergence on the order of a diffraction limit. As the target was moved a further 200 µm through focus (figure 2(c) – Intensity ~10¹⁷ Wcm⁻²) leaving the relativistic regime the higher order ROM harmonics disappear.

This work demonstrates a clear distinction between the quality of the harmonics generated via the CWE and ROM mechanisms. While still exceptional (2-3 times diffraction limited at 40 nm) it is clear that the CWE process accumulates phase errors due to propagation through the bulk plasma. ROM orders on the other hand appear to maintain the high beam quality of the incident laser pulse, with near diffraction limited emission

Given this distinct difference in emission characteristics of CWE and ROM harmonics, the angular distribution of the harmonics produced via the two mechanisms was investigated in detail. The CCD detector was rotated through 90° (configuration B, figure 1) to increase the angular range of the flatfield spectrometer (figure 3). The signal recorded also provided data for an accurate measurement of conversion efficiencies.



Figure 2. Transition from the lower intensity (~10¹⁷ Wcm⁻²) CWE mechanism to the relativistic ROM (~2×10¹⁹ Wcm⁻²).

The angular divergence of the 17th, 18th and 19th CWE harmonics was measured to be 38 ± 6 mrad $1/e^2$ radius. approximately 2-3 times diffraction limited. Even at the upper limit these CWE harmonics are much narrower than any previous angular divergence data published. The relatively large error in angular distribution is due to the highly modulated harmonic signal of the CWE harmonics. This is in contrast to the near perfect Gaussian distribution of harmonics produced by ROM, due to the fact that the ROS harmonics are simply a reflection from a well defined surface, up-shifting the fundamental with some non-linearity as in frequency doubling. Whereas the CWE harmonics are produced at a range of positions throughout the plasma and suffer modulations as they are emitted due to interference from different density layers which is dependent on the evolving density gradient.

Moving the CCD detector to lower wavelengths ROM orders generated could be observed. A sharp cut off of the harmonic spectrum was observed at the 40th order even though detector efficiency was still increasing, corresponding to a roll over or cut off of the harmonic spectrum, which is in good agreement with the predicted $n_{max} \sim 8^{1/2} \gamma^3$. Observation of this theoretical cut off is a direct indication of the attosecond nature of the emission^[6].

Surprisingly, the angular distribution of all the ROM harmonics (20-40) was found to be uniform, approximately 19±3 mrad 1/e² radius. This is contrary to theory which suggests a narrowing of the harmonic divergence as order increases. The angular divergence of the nth harmonic order (γ_n) is expected to be given as $\gamma_n = \gamma_L/n$, where γ_L is the divergence of the incident laser beam. Further analysis suggests that the uniform divergence of the harmonics was due to generation from a critical density plasma curved due to a denting induced by the ponderomotive force of the laser.^[8]

The conversion efficiency of harmonics generated by the two competing processes (i.e. CWE and ROM) was calculated by angularly integrating the signals inferred from the divergence data and correcting for system



Figure 3. Comparison of the highly modulated CWE harmonics (a) with the perfect Gaussian distribution of the ROM harmonics (b).

transmission. The signal on chip was summed over the entire spectral width and background subtracted. This signal was then converted to relative signal in the entire harmonic beam by considering the geometrical limitations of the system. The overall transmission of the system was calculated by considering all of the limiting factors on the harmonic signal before detection.

From this a good estimate of the total transmission of the flatfield spectrometer was obtained, enabling an accurate measurement of the conversion efficiency into each harmonic. CWE harmonics were observed to have a maximum conversion efficiency of $2\pm0.5\times10^{-3}$ for the 17-19th orders, compared with $1.5\pm0.3\times10^{-5}$ for the first ROM harmonics observed (20-21).

Conclusion

A clear distinction between the intensity regimes in which CWE and ROM processes dominate has been presented. It has also been shown that the CWE harmonics are emitted with greater angular divergence than ROM harmonics. Initial results suggest it may also be possible to focus ROM harmonics from a curved critical plasma surface leading to near diffraction limited performance in the harmonic focus.^[8]

CWE harmonics are observed to have a conversion efficiency of $\sim 10^{-3}$, up to two orders of magnitude greater than the ROM mechanism ($\sim 10^{-5}$ @ 20th order). However, these more intense CWE harmonics can only be generated up to the maximum plasma frequency and do not have the high beam quality of the ROM.

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