Angular emission and polarization dependence of harmonics from laser–solid interactions

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Abstract. Laser plasma interaction experiments have been performed to characterize high order harmonic emission up to the 18th order using high rep rate mJ level laser pulses at relativistic intensities. The experiments were compared to two- and three-dimensional particle-in-cell simulations. The harmonic divergence was found to be less than 4° (full-width at half-maximum) at highest intensity and increased as the laser was defocused (i.e. as the intensity was reduced). The polarization dependence on the harmonic generation efficiency and divergence was also measured. Circular polarization was found to cause a deflection in the angle of emission of the harmonics—an effect which may be beneficial in the use of such harmonics for efficient isolated attosecond pulse production.

The generation of short wavelength coherent radiation is motivated by a host of potential applications. In studies of ultrafast dynamics, higher frequencies are useful for synthesizing shorter duration (attosecond) pulses and consequently for making measurements with high temporal resolution. In imaging, shorter wavelengths can provide higher spatial resolution.

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Figure 1. Diagram of the high-harmonic experiment. OAP: gold-coated, \( f/1.4 \), off axis paraboloidal mirror; FS: fused silica target; DG: iridium-coated, 1200 grooves \( \text{mm}^{-1} \) diffraction grating; AF: 650 nm aluminum filter; MCP: phosphor-backed microchannel plate; L: zoom lens.

and enhanced image contrast. High order harmonic conversion of the laser frequency is one potential method for producing such coherent radiation. High-order harmonic production using gas targets has been investigated extensively, but the fundamental physics of gas harmonic generation makes scaling of the energy of attosecond pulses formed in this way difficult. In contrast, through the use of much higher intensity laser interactions with solid targets and subsequent harmonic generation, such limits can potentially be avoided so that bright and coherent x-ray sources may be produced with very high conversion efficiency [1–3].

Two mechanisms have been proposed to explain recent harmonic measurements from laser solid interactions: coherent wake emission (CWE) [4] and the relativistic oscillating mirror model (ROM) [5–9]. These mechanisms have been associated with harmonic emission in particular regimes of laser intensity and plasma density scale-length. Roughly, CWE is predicted to occur at lower laser intensities (\( a_0 < 1 \)) and shorter plasma density scale-lengths (\( L \ll \lambda/2 \)), while ROM dominates for higher laser intensities (\( a_0 > 1 \)) and longer scale-lengths (\( L \gg \lambda/2 \)). Here, \( a_0 = eA/m_ec \) is the normalized vector potential of the focused laser field. In CWE, electrons accelerated into the overdense plasma form plasma wakes that match the momentum of harmonics leaving the plasma and can therefore coherently amplify optical waves. In the case of ROM, harmonic emission results from Doppler shifting of the incident radiation reflected from the oscillating critical surface driven by the laser.

This paper describes a series of experiments to investigate the laser polarization dependence and the effect of focusing conditions on the divergence of high order harmonics using mJ laser pulses from a kHz repetition rate laser system. From these experiments it was concluded that the polarization gating technique [10–12] is a very promising method for generation of isolated attosecond pulses from high harmonic generation (HHG) emission using solid targets. The experiments took place using laser intensities and density scale-lengths which were in a transition regime between the CWE and ROM generation mechanisms.

A diagram of the experiment is shown in figure 1. Wavefront corrected, 50 fs, 2 mJ pulses were focused onto a bulk target by an \( f/1.4 \) off-axis paraboloidal mirror. The experimental conditions were unique in that very tight focusing was used in conjunction with adaptive optics in order to achieve relativistic peak intensities [13]. The measured focal spot size was 1.7 \( \mu \text{m} \) (full-width at half-maximum (FWHM)), resulting in a peak intensity of \( \sim 2 \times 10^{18} \text{ W cm}^{-2} \).
For most experiments, the target material was polished fused silica. The laser beam was incident horizontally at $35^\circ$ to target normal. The vertically oriented target was held by a motorized stage that enabled precise positioning about focus and raster-scanning to expose fresh surface for each shot. A shot separation distance of $100 \mu m$ was found to be sufficient for reliable operation. The specularly reflected beam was collected by an iridium coated, 1200 grooves $\text{mm}^{-1}$, spherical grating (Princeton Instruments) which dispersed the spectrum onto a microchannel plate (MCP). The grating incidence angle was either $5^\circ$ or $6^\circ$ depending on the observed spectral range.

The diffraction grating had reflectivity nulls at 72, 39, 27 nm, etc. The null at 72 nm was clearly visible in the spectrum between harmonics 10 and 11. Harmonics up to 18th order were detectable, but the grating efficiency limited detection below these wavelengths. A small amount of higher frequency emission was typically visible in the data. This emission had no observable harmonic structure because of the spectrometer resolution—but had a similar divergence as the measured harmonics. This suggests that such radiation was not atomic ‘line-emission’ but may have been due to harmonic emission beyond the 20th order. Harmonic spectra were recorded with different driving pulse energies and the efficiency was fit to a power law scaling of $I^{0.64\pm0.05}$. This scaling falls between the $I^{0.4}$ of CWE harmonics previously observed by Quéré [4] and the $I^2$ scaling from simulations of relativistic harmonics by von der Linde [5]. It should be noted that as the intensity in our experiments was increased for the main pulse, the intensity should similarly increase for the prepulse indicating that the experimental conditions will also change during an ‘intensity’ scan (i.e. the density scalelength during the ‘main’ interaction will increase). This implies that our observed harmonics should scale more slowly with respect to increasing intensity than for ‘clean’ pulses.

Because the diffraction grating was used off-normal, astigmatism separated the vertical and horizontal foci. To achieve the highest spectral resolution, the MCP was positioned at the horizontal focus. Consequently the vertical displacement on the MCP could be mapped directly to angular emission from the source. Indeed when poor focusing conditions generated highly divergent harmonics, the boundaries of the grating became visible in the acquired images, providing a simple method of calibration. The height of the grating was 40 mm, so that at a distance of 14.6 cm the full vertical collection angle of the grating was $15.3^\circ$. The divergence for harmonics 13–16, those with sufficient signal to accurately measure their divergence was measured to be $4\pm1^\circ$ (FWHM) with a small but clear trend toward increasing divergence for higher order harmonics.

The effect of driving polarization was also measured. Zero-order mica wave plates were used to generate s- and circular polarization. For s-polarization a half-wave plate was placed between the deformable mirror and the vacuum chamber. A reference shot was taken by rotating the wave plate so as to leave the horizontal polarization unaffected. The wave plate was then rotated $45^\circ$ to produce vertical polarization (s-polarization). Typical data from both situations is shown in figure 2. The reference shot shows a similar harmonic structure as data with no wave plate. The UV emission of the s-polarized shot was approximately 30 times weaker and showed no harmonic structure.

For circular polarization a quarter wave plate was placed after the deformable mirror. As before, a reference shot was taken. The behavior of circular polarization was quite distinct from that of linear polarization. Rather than eliminating the harmonic signal, circular polarization changed the angular distribution of the harmonic emission. For left handed circular polarization the harmonic spectrum was observed to be deflected vertically by approximately $3^\circ$, while for right-handed polarization the spectrum was deflected by a similar amount in the opposite direction.
Figure 2. Comparison of p and s polarization driven harmonics. s-polarization (lower figure) produces weak UV emission with no resolvable harmonic structure compared to p-polarization (upper figures).

Figure 3. Harmonic spectra for left and right circular polarization (a), (c) compared to p-polarization (b). One of the axes aligned with the horizontal beam polarization.

direction (figure 3). The intensity of the harmonics was also reduced by more than a factor of 3 although it should be noted that changing the polarization also has the effect of reducing the peak intensity somewhat [14]. The observed deflection of the harmonic beam also greatly reduced the on-axis signal, there was no change in the number of harmonic orders measurable.
Finally the effect of the laser spot-size/beam profile was investigated experimentally. The Rayleigh range for the \( f/1.4 \) focus in these experiments is approximately 2 \( \mu \text{m} \)—which places a significant demand with regard to proper positioning of the target, especially when operating at the target rotation speeds necessary for high repetition rate operation. Figure 4 shows several high-harmonic spectra taken at various positions of the target. While the harmonics remain detectable as much as 10 \( \mu \text{m} \) from the optimum position, a shift of only 2 \( \mu \text{m} \) leads to a noticeable drop in intensity. In addition, as the target moved, the shape and divergence of the harmonics are observed to change.

In these experiments the laser intensity during the interaction can be varied by working at a different point along the beam focus. Correspondingly, the observed drop in harmonic signal level is expected to be due largely to the drop in intensity, and the increasing harmonic divergence angle perhaps initially results from a reduction in the ponderomotive force and consequently a decrease in the ‘denting’ of the reflecting surface. This phenomenon has been shown to be a cause of the narrow divergence of the HHG beams at very high intensity [15, 16]. The increasing divergence angles are inconsistent with the simple argument that the harmonics should diverge with angles \( \theta_n = \theta/n \) [15, 16]. However for these tight focusing experimental conditions, the intensity of the laser during the interaction rapidly becomes less as the target is moved away from best focus. Indeed at a position 10 \( \mu \text{m} \) away from best focus the intensity is about 70 times less than at the peak (i.e. \( \sim 3 \times 10^{16} \text{ W cm}^{-2} \)) and the density scalelength of the interaction is also significantly less. It is consequently clear that for these lower intensities the CWE regime can be accessed—and in which the divergence of the measured harmonics should increase significantly as observed.

The lack of harmonic production for s-polarization is a signature of the CWE generation mechanism however it should be noted that we have also subsequently performed experiments using a laser pulse with a similar focusing geometry focused to three orders of magnitude higher intensity which is clearly in the ROM regime [17]. This experiment also showed almost no HHG emission for incident s-polarized laser interactions which suggests that in this tightly focusing regime neither CWE nor ROM mechanisms are able to generate harmonics for incident
s-polarized light [17]. The efficiency of the grating is well approximated by scalar diffraction, and the near normal incidence eliminates any other polarization effects that might occur. The only other components in the detection system (the aluminum filter and MCP) are used near normal incidence and are largely polarization independent.

The harmonic production in these experiments is likely in a transition regime between CWE and ROM. At lower intensities (when the scalelength is also short) the harmonics are clearly produced by a CWE mechanism. However at the highest intensity used ($a_0 \sim 1$) the divergence of the harmonic emission becomes much less and the scalelength increases so it is likely that there should be a significant contribution from ROM. In addition, at the highest intensity there are experimental evidence of harmonic-like (i.e. narrow divergence) emission greater than 20th order which could only be due to ROM. Previously the two mechanisms have been distinguished mainly by observation of a CWE harmonic cutoff at the maximum plasma frequency [7]. Fully ionized fused silica has an electron density of $n_e = 6.6 \times 10^{23} \text{cm}^{-3}$. This corresponds to a plasma frequency of $\omega_{pe} = (4\pi e^2 n_e / m_e)^{1/2} = 4.6 \times 10^{16} \text{rad s}^{-1}$ or a wavelength of 41 nm, approximately the 20th harmonic of 800 nm. Unfortunately, this occurs very near a null in the efficiency of the diffraction grating in our experiments, making it unclear whether the loss of harmonics at that wavelength during the intensity scaling is gradual or abrupt.

In addition while the scaling of conversion efficiency with intensity suggests that an important mechanism is CWE, the scaling matched a higher power law exponent than observed at lower intensities where CWE is expected to dominate. This is again evidence that the dominant harmonic generation mechanism is in a transition region between CWE to ROM at the highest intensities employed in these experiments ($2 \times 10^{18} \text{W cm}^{-2}$) and that the increasing density scalelength is a critical factor in determining the nature of the interaction at best focus.

A series of two-dimensional (2D) simulations using the particle-in-cell (PIC) code OSIRIS [18] using the experimental parameters and with a front surface plasma density scalelength of $\lambda/4$ were performed. This scalelength was similar to previous simulations performed to match previous measurements of hot electron emission from similar experiment on this laser system [19, 20]. A 2D spatial Fourier transform of the reflected field is shown in figure 5(a). The HHG emission in these simulations was clearly due to ROM and no emission from a CWE mechanism in the overdense plasma could be discerned. The FWHM divergence angle is 3.6°, which agrees well with the measured divergence. An efficiency of harmonics 13–19 scaling as $n^{-5.3}$ was obtained, where $n$ is the order number of the observed harmonic similar to the experiment, although the absolute efficiency is about an order of magnitude higher. Absolute efficiency can be affected by a number of factors. Target surface roughness and wavefront distortion of the driving beam can cause UV harmonic energy to be scattered outside the collection angle. Some energy at the fundamental frequency can also be scattered outside of the main focus by wavefront errors and roughness of the paraboloidal mirror. Finally, the calculated calibration of the MCP is imprecise and its gain could be lower than specifications. The total conversion efficiency into all measurable harmonics transmitted by the aluminum filter was $9 \times 10^{-7}$. This value is comparable to those reported from recent gas harmonic experiments [21]. However, chirped pulse amplification (CPA) systems operating at kHz rep rates can now produce sub-30 fs pulses with 10–20 mJ of energy. Focusing such pulses to the diffraction limit may yield intensities in excess of $10^{19} \text{W cm}^{-2}$ where the conversion efficiency for solid target harmonics should be even higher.
Figure 5. (a) Spatial Fourier transform of the reflected beam from 2D PIC simulation. (b) Angular emission profile of the third harmonic from 3D simulation showing the shift in angular emission from the left and right circularly polarized pulses (blue and green) with respect to a linearly polarized pulse with the same time averaged intensity (red).

To reproduce the circular polarization effects it was necessary to perform three-dimensional (3D) PIC simulations of the interaction, however computational constraints greatly limit the resolution so that only the first few harmonics can be resolved. To clearly show the effect it was also necessary to increase the laser $a_0$ to 2, since numerical dispersion tends to limit propagation at a small angle to the grid. The $s$-polarized component of the circular polarization leads to a distortion of the ‘mirror’ surface reflecting light both up and down in 2D, with a $\pi$ phase shift between the two. However, in 3D the $p$-polarized component results in an oscillating mirror that steepens the field at a particular $2\pi$ phase, which results in harmonics in only one of the vertical directions, depending on the handedness of the polarization. Figure 5 shows the integral over $\phi$ and $k$ of a full 3D spatial Fourier transform over the third harmonic, leaving just the angular distribution in $\theta$. When the polarization is varied from left-hand circular to right-hand circular, the harmonic intensity peak shifts in $\theta$ and the intensity is diminished. The polarization dependence leads one to consider the possibility of generating isolated attosecond pulses using
long (~20–30 fs) driving pulses and polarization techniques demonstrated in gas harmonics. By combining two delayed, counter-rotating circular polarizations a long pulse with only a few (or even a single) linearly polarized cycles can be created [22]. Because circular polarization does not produce harmonic emission in gases, the few cycles of linear polarization are the only source along the laser propagation direction. The same type of experiment may be possible with solid target harmonics and has been studied numerically [23]. While circular polarization did not eliminate the harmonic signal in the experiments presented here, it did change its angular emission. It is possible that by collecting only the central 4° of the emission, the signal generated by one cycle of linear polarization (in the form of a sub-femtosecond pulse) could be separated from the signal generated by the other cycles of circular polarization.

It should be noted that the CWE mechanism might also be able to generate deflected harmonic emission due to phase differences of Brunel accelerated electron bunches generated by for circular polarized light at very short scalelengths [10]. This however may be less interesting for isolated attosecond pulse generation because of the slower scaling with intensity when compared to ROM.

In conclusion, harmonics of a 800 nm high rep rate laser up to the 18th order have been observed from solid targets. An energy conversion efficiency of $2 \times 10^{-7}$ per harmonic was measured. This is comparable to current efficiencies achieved in gas harmonic experiments, but has the advantage of being scalable to much higher intensities, where the conversion efficiency should also increase. Despite using a very short focal length required to achieve the necessary intensity, the harmonics were emitted with low divergences of approximately 4° (FWHM) making the capture and manipulation of the harmonic beam easier. The low pulse energy in the experiment and the lack of complex contrast enhancement mechanisms make this harmonic source very accessible and operable at high repetition rates. Similar systems operating at multi-kilohertz repetition rates should be capable of producing comparable harmonics with higher brightness.

The effect of polarization on the divergence of the harmonics has also been measured for the first time. Modification of the angular emission for circular polarization suggests that a polarization gating technique like that used in gas harmonics may also be used on solid targets to generate single sub-femtosecond pulses using long (>20 fs) driving pulses. These experiments have shown that a wealth of data stands to be collected now that such easy access to solid target harmonics is available.

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