Experimental observation of attosecond control over relativistic electron bunches with two-colour fields

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1. Influence of the Coherent Wake Emission mechanism

Apart from the Relativistically Oscillating Mirror model and Coherent Synchrotron Emission, another important high harmonic generation mechanism that is relevant for the parameters used in the present experiment is Coherent Wake Emission (CWE)[1]. This process occurs via plasma oscillations triggered in the wake of the dense electron bunches as they are driven back into the surface plasma density gradient. These oscillations are able to couple into electromagnetic modes with a frequency matching that of the local plasma frequency within this density gradient. As a result, radiation generated via CWE is limited to frequencies below the maximum plasma frequency of the target. The process is very linear and can dominate at moderate intensities (≈10^{18}Wcm^{-2}) for harmonic orders below the cut-off.

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As well as its characteristic frequency cutoff, another key difference is the scale length dependence of CWE. CWE favours very short scale lengths with the efficiency dropping rapidly as it is increased as opposed to the ROM mechanism which reaches an optimum before falling off more slowly for longer scale lengths[2]. The exact optimum depends on the interaction intensity but is typically on the order of λ/10 and is lower for CWE. In the current experiment, the optimal prepulse timing for the ROM orders was set by scanning a small time range around 1.5ps earlier than the main pulse based on previous observations[2]. The optimal timing was found to be 1.64±0.15ps.

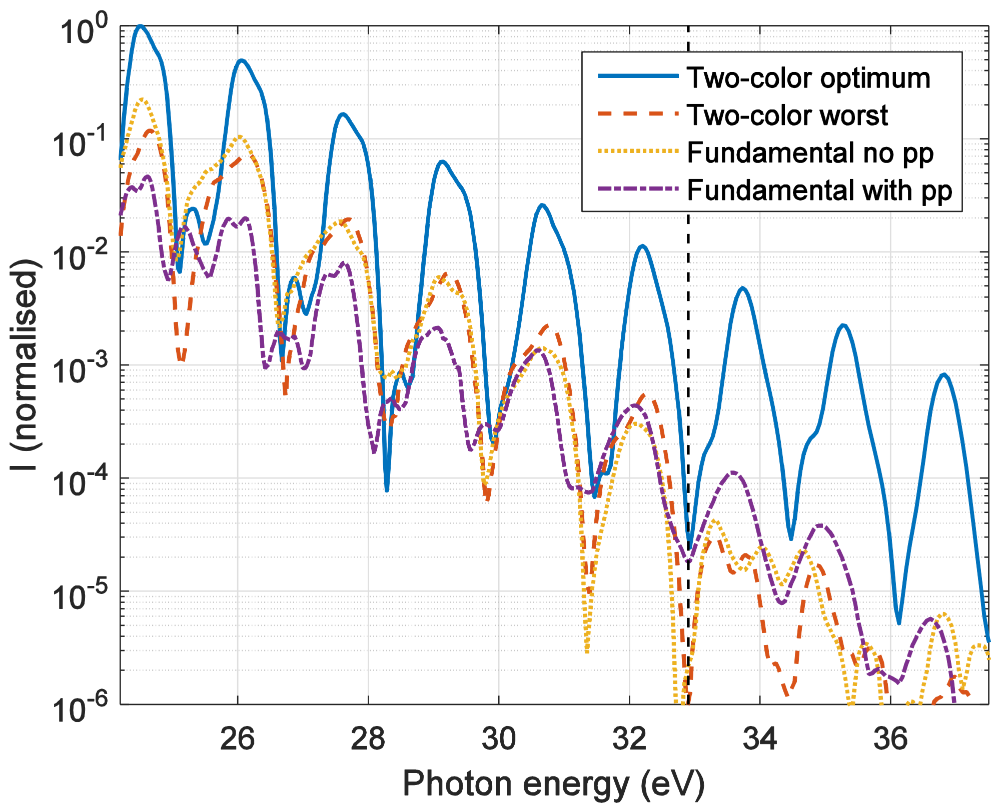

Figure S1 – CWE contribution – Comparison of spectra taken with a two-colour field and the fundamental laser only without any prepulse (the optimum for the two-colour field) (blue solid – two-colour with optimum phase, red dashed – two-colour at π phase from optimum, yellow dotted – fundamental) and with the fundamental only and a prepulse ≈1.64±0.15ps early (optimum for the ROM signal for the fundamental only) (purple dot-dashed). The black dashed line indicates the cutoff for the CWE orders.

In figure S1 a number of spectra are shown that illustrate the level of contribution made by the CWE process for the current study. With only the fundamental pulse and no prepulse present there is clear harmonic signal up to the 21st harmonic, beyond which there are no discernible harmonics. This distinct cut-off at the peak plasma frequency present of the glass targets is characteristic of the CWE process which dominates under these conditions (low intensity, steep gradient). We note that the observed spectrum is similar in shape and intensity to that observed when using the two-colour field with the incorrect phase. Once the prepulse is introduced to the fundamental, harmonics orders beyond the cutoff appear whilst those below are weakened as the interaction conditions change to favour the ROM process. The smooth transition from the low orders to those above the 21st in this case suggests that the CWE contribution is no longer significant (as this would create a sharp change in the signal around the cutoff) and is consistent with previous observations[2].

More importantly, it can be seen that at the optimum phase for the two-colour field the harmonic signal far exceeds that of the other cases and the smooth trend of the harmonic peaks suggest there is negligible contribution from the CWE process. In calculating the efficiency in the two-colour case (which was found to be $\approx 1 \times 10^{-3}$) the harmonic signal found with the fundamental only (and no prepulse) was subtracted to ensure that only the additional signal originating from the two-colour source (assumed to be from the FWHM of the smaller 2nd harmonic focal spot) was used.

2. Second Harmonic Polarisation Angle

The mechanism has been described in the main text as a sub-cycle modification of the driving electric field thus it is expected that this effect will be most prevalent when the 2nd harmonic beam is polarised in the same direction. Experimentally, it was observed that orthogonal polarisation did not produce any enhancement or any clear phase dependence. In this section we present simulations showing the full trend of the harmonic yield with both 2nd harmonic phase and polarisation angle.

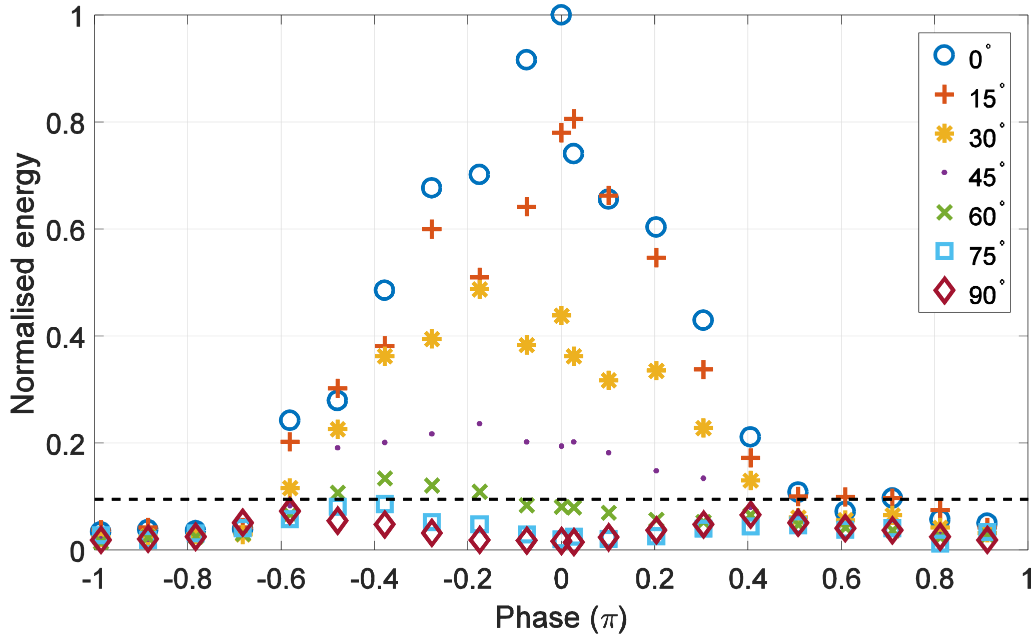

Figure S2 – Effect of second harmonic polarisation – Total harmonic yields from PIC simulations, integrated between the 22nd and 27th harmonics for the same simulation parameters as described in the methods section of the main text. Here, the angle of polarisation of the second harmonic is indicated in the legend where 0° means parallel to the fundamental. The black dotted line indicates the case with no 2nd harmonic beam and a slightly higher fundamental peak a_0 of 1.3 to account for the extra unconverted energy.

In figure S2 we see that, as expected, parallel polarisation produces the strongest enhancement. There is a gradual decrease as the polarisation is rotated and the 2nd harmonic field component in the direction of the fundamental polarisation is reduced. Interestingly, for angles above 60° a clear drop in the harmonic yield is seen and the phase dependence changes shape to have two peaks located close to relative phases of ±0.5π. This is likely due to the **v**×**B** force exerted in the forward direction by the 2nd harmonic magnetic field which oscillates at twice the frequency of the electric field force and is significantly weaker at the current intensities. The weakness of this effect may be the reason it was not observed in the current experiment. In any case, these simulations confirm that parallel polarisation is optimal for efficient harmonic generation on solid targets with two-colour fields, in contrast to similar methods using gaseous targets where orthogonal polarisation was also found to be efficient[3-5].

3. Energy ratio and scale length

In figure S3, the experimentally observed phase dependence for additional scale lengths and 2nd harmonic energy ratios is plotted. In figure S3a, the yield for the full observed spectral range is shown, including contributions from CWE orders, whilst figure S3b only shows clear ROM harmonics. Additionally, datasets were obtained for 5% energy conversion and no prepulse and 10% energy conversion with prepulse but no clear phase dependence or enhancement was observed within the experimental error.

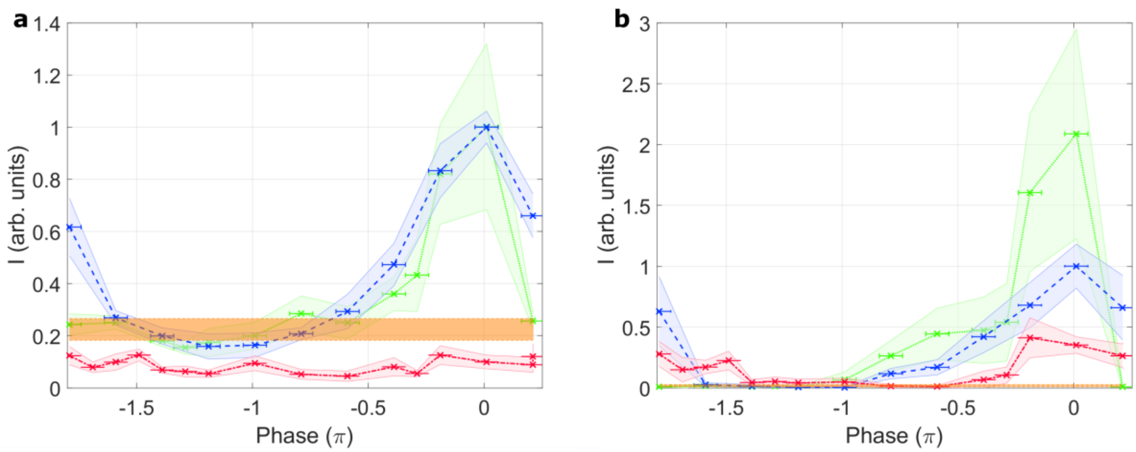

Figure S3 – Experimental data for different scale lengths and energy ratios – Total measured harmonic yield summed between (a) the 16th and 27th harmonic and (b) the 22nd and 27th harmonic. (Green dotted) is for the case of 20% conversion into the 2nd harmonic and for no added prepulse, (blue dashed) is for 10% conversion and no added prepulse and (red dot-dashed) is for 20% conversion and a prepulse at 1.64±0.15ps. All points have been normalised to the 10% conversion case optimum to be comparable to the data of figure 4 of the main text. The shaded regions correspond to the standard deviation of the data. The shaded orange region indicates the harmonic yield for the same spectral ranges for the fundamental beam only both without (a) and with (b) prepulse.

The key finding from these datasets is that very short scale lengths appeared to be more ideal for optimisation of the harmonic efficiency for the current parameter range of study, however intermediate prepulse positions were not investigated. A possible reason is that, for an exponential density gradient, the critical density for the 2nd harmonic laser will be a factor of 4 higher than for the fundamental and hence will be closer to the initial target surface than that for fundamental, thus there may be a less efficient interaction between the two fields. At higher intensities, such as those studied in reference [6], the pressure of the laser itself will be sufficient to steepen the density gradient which may lead to better mixing of the two frequencies at the generation point.

Additionally, we also see that, for the higher orders, a larger 2nd harmonic energy conversion leads to a stronger yield, albeit with much less stable signal. In figure S4 the effect of the energy ratio is examined using PIC simulations. Here the total energy is assumed constant and the intensity of the 2nd harmonic has been calculated by accounting for a focal spot area which is three times smaller than the fundamental (as measured for the current experimental conditions – in the ideal case this would be a factor of four). As a result, the 2nd harmonic intensity increases very rapidly as the energy ratio is increased. Combining this with the improved matching between the target density and the driving frequency, as discussed in reference [6], we see, as expected, a clear increase in the harmonic yield for larger ratios. The scanned conversion efficiencies range up to 90% but it should be stressed that for pulses on the order of 30fs and lower, efficiencies on the order of 10% to 20% are realistic experimentally.

Additionally, the optimum phase position is seen to shift as the 2nd harmonic becomes dominant. For low conversions, the peak yield is seen for a relative phase of 0 but as the energy ratio increases, this shifts to higher phases which are more consistent with the findings of the simulations of reference [6].

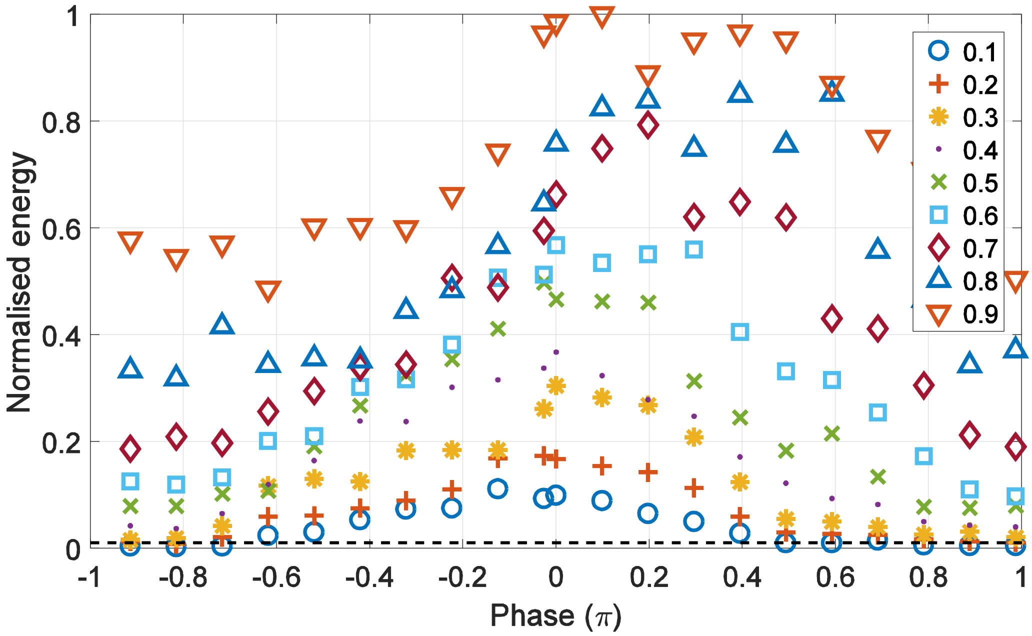

Figure S4 – Simulations of the effect of the second harmonic energy ratio – Total harmonic yields from PIC simulations, integrated between the 22nd and 27th harmonics for the same simulation parameters as described in the methods section of the main text. Here the legend indicates the proportion of energy converted to the second harmonic. The second harmonic intensity at each point has been calculated by also considering its improved focusability. The black line indicates the case for all energy in the fundamental and a peak a_0 of 1.3.

References

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