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Optimised optical waveforms drive efficient water window attosecond pulse generation

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The ability to precisely tailor optical waveforms provides a powerful means of controlling strong-field interactions, including high-harmonic generation (HHG) for attosecond pulse production. Although numerous tailored waveforms have been proposed to extend the harmonic cut-off or enhance conversion efficiency, achieving such improvements in the soft X-ray region remains challenging because plasma formed in the medium can strongly distort the driving field during propagation. Moreover, isolated attosecond pulse (IAP) generation demands sub-cycle waveform shaping to enhance a single attosecond burst while suppressing contributions from neighbouring optical cycles. Here, we demonstrate order-of-magnitude enhancements in IAP generation efficiency up to ~ 450 eV, by driving HHG in helium with tailored fields spanning the $0.7 - 2.2$ μm range, surpassing a 1.5-cycle driver centred at 1.8 μm . Direct waveform measurement and simulations allow us to track the propagation of optimised optical transients through the nonlinear medium and to disentangle the intertwined microscopic and macroscopic contributions that govern soft X-ray HHG. These results establish sub-cycle waveform engineering as a robust strategy for optimising soft X-ray IAP generation and provide an experimentally validated framework for next-generation attosecond sources.

1 Main

Attosecond pulses in the soft X-ray (SXR) regime (above ~ 120 eV) are unique tools for tracing coherent charge dynamics on the native timescale of electrons¹⁻³. Interrogating core-level electrons provides element- and site-specificity, enabling clear differentiation of contributions from distinct atomic sites in transient spectral signatures. This synergy of attosecond resolution and local chemical sensitivity enables unprecedented access to time-evolving electronic correlations. Of particular interest is the “water window” (WW) region, spanning from the carbon to the oxygen K-edge, covering as well the L-edges of few transition metals, while remaining transparent to water. This makes WW attosecond pulses ideal for probing real-time electron motion in aqueous environments^{4,5}, offering unprecedented mechanistic insight into photochemical reactions in molecular systems^{6,7}.

The generation of isolated attosecond pulses (IAPs) within the WW range is necessary to fulfill this potential. Pioneering time-resolved experiments have been performed using both free-electron lasers (FELs)⁸⁻¹⁰ and high-harmonic generation (HHG) sources¹¹⁻¹⁴. FELs can deliver gigawatt-scale IAPs^{15,16}, but the absence of sub-femtosecond synchronization with external optical lasers and limited facility access underscore the value of table-top alternatives. HHG remains the most accessible technique, providing attosecond pulses intrinsically phase-locked to optical pulses. It has demonstrated kiloelectronvolt spectral reach and ultrabroad bandwidths¹⁷ that enable simultaneous probing of multiple elemental core-level transitions^{18,19}. Yet, its inherently low efficiency has effectively capped femtosecond time-resolved experiments at ~ 500 eV^{5,20}, and continues to hinder more photon-hungry experiments even within this range.

48 The low efficiency of attosecond pulse generation stems from both microscopic and macroscopic
 49 factors that worsen at longer driving wavelengths, necessary to reach higher photon energies. Mi-
 50 croscopically, the high-harmonic cutoff scales with $\propto \lambda^2$ ^{21,22}, requiring shortwave-infrared drivers
 51 (typically $\geq 1.5 \mu\text{m}$)²³ to reach the WW. According to the three-step model²⁴ -tunnel ionization,
 52 acceleration in the laser field, and recombination to the parent ion- longer wavelengths prolong
 53 electron excursion, increasing wavepacket diffusion, and reducing recombination probability. This
 54 results in a steep efficiency penalty^{25,26} scaling as $\propto \lambda^{-6}$. Macroscopically, phase-matching im-
 55 poses strict bounds on allowable plasma fractions (scaling as $\propto \lambda^{-2}$)²⁷, which curtail driving peak
 56 intensities and reduce the number of emitters. In parallel, conventional IAP gating techniques²⁸
 57 discard nearly all but a single optical cycle of the driving field, wasting much of the energy that
 58 would otherwise boost the photon yield and instead increases plasma-induced dephasing.
 59 To counteract these efficiency losses, strategies have been developed that tackle both single-atom
 60 and macroscopic scales. On the single-atom scale, sub-cycle waveform tailoring via controlled
 61 superposition of multicolour pulses enables fine control over each step of the HHG process^{29–35}.
 62 At fixed laser fluence, this control can allow the synthesis of waveforms that maximise recollision
 63 energy while minimising excursion time³⁶, thereby reducing wavepacket spreading and extend-
 64 ing the harmonic cutoff. Waveforms can also be tailored to selectively launch short trajectories³⁷,
 65 whose naturally short excursion times translate into smaller dipole phases and hence easier phase-
 66 matching³⁸. Steering ionisation into these trajectories makes more efficient use of the driving
 67 energy and can lead to order-of-magnitude gains in the macroscopic HHG output³⁹. On the macro-
 68 scopic scale, HHG is typically optimised by balancing neutral and plasma dispersion, Gouy phase,

69 and dipole phase²⁷. With longer wavelengths, achieving this balance usually requires higher pres-
 70 sures (often ≥ 1 bar)^{40–42}, due to the λ^2 -scaling of the phase-matching pressure⁴³, making macro-
 71 scopic effects increasingly significant. Losses from conventional IAP gating approaches can be
 72 circumvented using single- or sub-cycle drivers (i.e., shorter than the centre oscillation period),
 73 which naturally confines HHG to the most prominent optical cycle^{34, 44–47}.
 74 Up to ~ 200 eV, multicolour tailored drivers, predominantly of multi-cycle duration, have achieved
 75 order-of-magnitude gains in HHG efficiency via mechanisms including ionization boosting⁴⁸, pref-
 76 erential enhancement of short-excursion trajectories⁴⁹, and more generally the reshaping of elec-
 77 tron trajectories that suppresses wavepacket spreading^{50–53}. Applying these capabilities to IAP
 78 generation with few-cycle (or shorter) drivers, however, creates new challenges. Temporal gating
 79 then demands pronounced cycle-to-cycle variations in amplitude and shape of the driving field
 80 to avoid additional ionisation events, or at least prevent them from contributing to recombining
 81 trajectories. This raises the question of whether waveform engineering can robustly deliver IAP
 82 efficiency gains above ~ 200 eV, extending well into the WW.
 83 Here, we demonstrate that tailoring 1.7-octave-spanning waveforms centred at $1.4 \mu\text{m}$ and capa-
 84 ble of reaching durations as short as 0.6 cycles⁴⁵, can significantly enhance IAP generation over
 85 the $\sim 200 - 450$ eV spectral range. By fully characterizing hundreds of driving fields directly at
 86 the HHG medium⁵⁴ together with the corresponding SXR spectra and flux, we identify waveforms
 87 that yield over an order-of-magnitude efficiency improvement compared to an optimised 1.5-cycle,
 88 $1.8 \mu\text{m}$ driver. Experimental results are in good agreement with simulations under diverse genera-
 89 tion conditions.

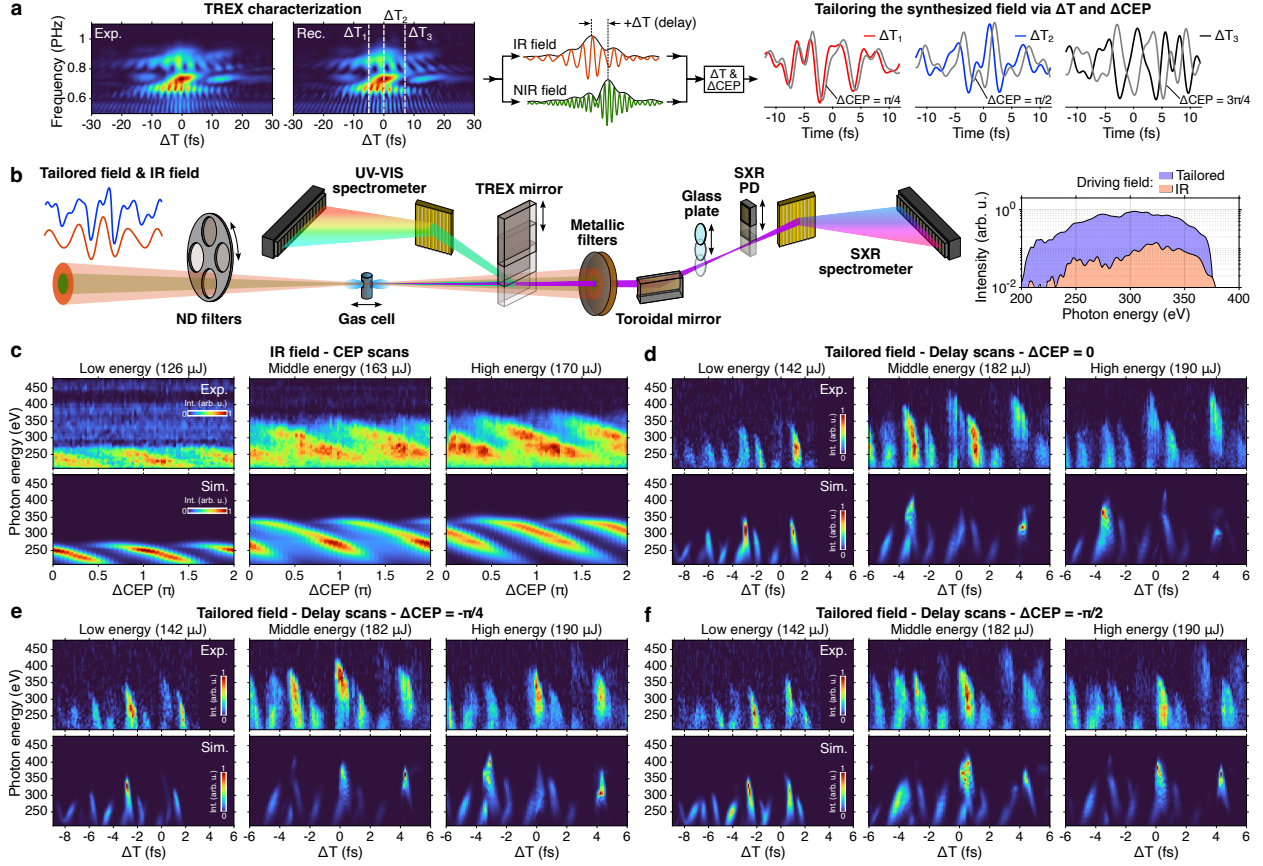


Figure 1: Experiment: waveform characterization and HHG scans. (a) Synthesised waveforms are generated by coherently combining two CEP-stable pulses centred at 0.8 and 1.8 μm with parallel polarisations, referred to as near-infrared (NIR) and infrared (IR) pulses, with FWHM durations of 7 fs (2.5 cycles) and 9 fs (1.5 cycles), respectively. Tailoring is achieved by adjusting the common CEP and relative delay ΔT between the pulses (positive ΔT : NIR arrives after IR; $\Delta T = 0$: overlapping pulse envelopes). The electric fields, including CEP, calibrated ΔT , and amplitude ratio, are retrieved using TREX⁵⁴. (b) Experimental setup. The driving waveforms, attenuated by neutral-density (ND) filters are focused into a ~ 1 mm helium cell. Ultraviolet and visible emission from the third-order response is recorded as a function of ΔT using the TREX mirror and UV–VIS spectrometer at 1 bar backing pressure. HHG is generated at 6–8 bar and analysed with an SXR spectrometer after blocking light below ~ 200 eV using Al and Cu filters; a glass plate for background subtraction and an SXR photodiode (PD) provide photon-flux calibration. (c) Measured IR-driven HHG spectra as a function of CEP, together with macroscopic simulations. (d–f) Tailored-field-driven HHG spectra as a function of CEP and ΔT , together with macroscopic simulations. Each scan is repeated for three input-energy values (low, middle, high). Simulations exclude long trajectories and incorporate the experimental CEP r.m.s fluctuation (300 mrad) and the beamline transfer function (see Methods).

2 Results

The experiment is summarised in Fig. 1a–b, with further details given in the Methods and Supplementary Information. The macroscopic conditions -namely the gas pressure and target position- were first optimised to maximise HHG emission above ~ 200 eV using the IR field alone, while rapidly sweeping its carrier-envelope phase (CEP) to average out CEP-sensitive effects. This optimisation was then repeated for low-, middle-, and high-energy input energy values of the single IR pulse (126, 163 and 170 μJ , respectively), corresponding to increasing peak intensities.

With the macroscopic conditions established for each energy value, the driving waveforms were scanned while recording the corresponding HHG spectra. For the IR field, this simply consisted of varying the CEP (Fig. 1c). For the synthesised fields, obtained by coherently combining the NIR and IR pulses, waveform scans span a two-parameter space defined by the relative delay ΔT and the CEP of the synthesised waveform. Experimentally, this was realised by finely sampling ΔT (~ 100 as steps) at a fixed CEP, followed by repeated delay scans after shifting the CEP of both pulses by a fixed amount ($\pi/4$ and $\pi/2$), as shown in Fig. 1d–f. Such procedure was also repeated for low-, middle- and high-input energies of the synthesised pulse (142, 182 and 190 μJ , respectively), allowing to study propagation effects at different plasma densities.

To directly link each measured SXR spectrum to its corresponding driving field, all synthesised waveforms, together with the IR field, were fully characterised *in situ* at the HHG target using the TREX technique⁵⁴ (see Fig. 1a). This approach provides the complete NIR and IR electric fields, including their CEPs, relative delay (ΔT), and amplitude ratio.

We now define the IAP conversion efficiency, η_{iap} , as the fraction of incident optical energy con-

111 verted into an IAP emission above ~ 200 eV:

$$\eta_{\text{iap}} = \frac{E_{\text{sxr}}^{\text{IAP}}}{E_{\text{opt}}}, \quad (1)$$

112 where E_{opt} is the energy of the driving field (at the gas cell input) and $E_{\text{sxr}}^{\text{IAP}}$ is the energy of the
113 IAP (at the HHG source), where the latter was obtained from photodiode and CCD measurements
114 corrected for the beamline transfer function (see Methods). Our analysis focuses on IAPs, exclud-
115 ing cases in which the emission consists of multiple bursts. For this study, a pulse is considered
116 isolated if its peak intensity exceeds that of any secondary pulses by at least a factor of ten.
117 To access the temporal domain of the HHG emission, on-axis macroscopic HHG simulations were
118 implemented that use the *in situ*-measured driving waveforms as input. For the optical field, the
119 propagation model includes neutral dispersion, the time-dependent plasma response, as well as
120 intensity and phase variations arising from focusing, whereas for the SXR beam neutral dispersion
121 and absorption are considered (see Methods). By neglecting transverse propagation, this on-axis
122 approach enables simulations across the full experimental parameter space while preserving the
123 essential dynamics of the SXR generation. The simulations shown in Fig. 1c–f were performed
124 following the experimental procedure. For each input energy, the optimal gas pressure and target
125 position were first optimised with the CEP-averaged IR field. With these parameters fixed, CEP
126 scans were then carried out for the IR field, and delay scans were performed for the combined
127 (IR+NIR) fields for different CEP shifts. The close agreement between experiment and theory
128 highlights both the exceptional fidelity of the waveform characterization as well as the accuracy
129 of the simulations. This consistency allows us to confidently assign each measured emission to
130 its driving waveform and identify which waveforms generate IAPs, excluding from the efficiency

131 analysis cases associated with multiple HHG bursts.

132 With this validation in place, we aim to address three questions: (i) Are there specific waveforms
133 that yield higher η_{iap} than a CEP-optimised 1.5-cycle pulse centred at 1.8 μm ? (ii) If so, which
134 waveforms enable such enhancement, and (iii) how do the single-atom and macroscopic responses
135 contribute?

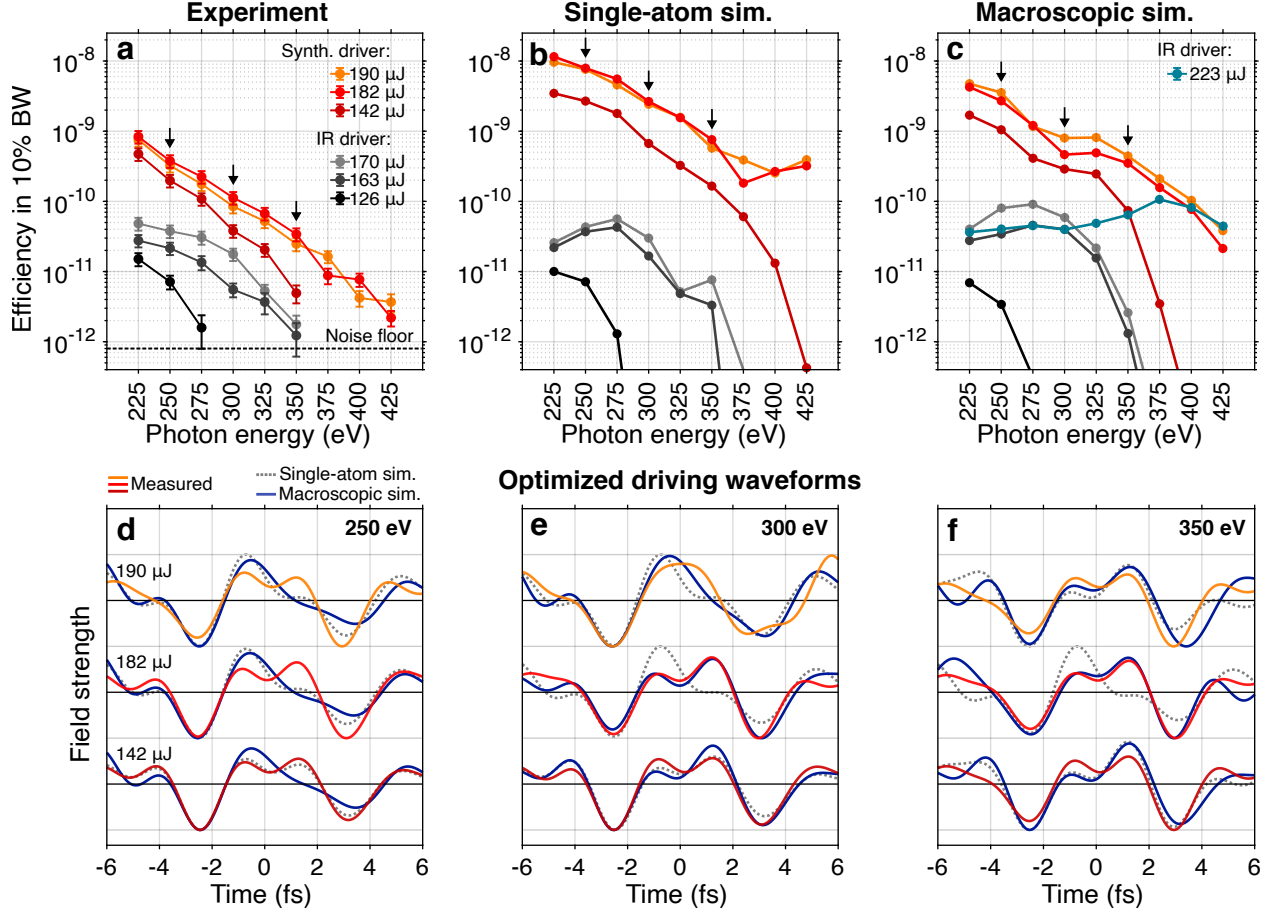


Figure 2: IAP conversion efficiencies in helium and driving waveforms. (a) Highest measured IAP efficiencies ($\eta_{\text{iap}}/10\% \text{ BW}$) from low to high input-energy values across different photon energies, driven by the IR pulse (grey scale) and tailored waveforms (red scale)- Corresponding efficiencies (and photon flux values) without IAP filtering are provided in the Supplementary Information Fig. 31. For the IR pulse, points correspond to optimised CEP; for tailored waveforms, to optimised synthesis settings defined by ΔT and a CEP offset ($0, \pi/4$ or $\pi/2$). (b,c) Simulated maximum conversion efficiencies from single-atom and macroscopic response models, respectively. For tailored fields, the CEP optimisation spans a finer grid over $[0, \pi)$. Single-atom simulations use the electric field at the gas-cell centre. Colour coding in (a-c) denotes the experimental/simulated input energy, and simulated efficiencies are rescaled to the experimental range for direct comparison of relative enhancements. An additional simulation at 223 μJ predicts the maximum efficiency attainable with the IR pulse. (d-f). Measured tailored waveforms (solid lines, colour-coded as in (a-c)) yielding the highest efficiencies at 250, 300, and 350 eV, respectively, together with the corresponding single-atom (dashed grey) and macroscopic (solid blue) simulations. Electric fields are normalised to their peak amplitude and time-shifted so that their first field maximum at ~ 2.5 fs coincides.

The first question is addressed in Fig. 2a–c, which shows the IAP efficiency η_{iap} (Eq. 1) across successive spectral bands centred between 225 and 425 eV, comparing optimised IR and synthesised waveforms at different input energies. For the IR field, each data point corresponds to the maximum η_{iap} extracted from the measured CEP scan, reflecting optimisation of the CEP for the given spectral band. For the synthesised fields, each value represents the highest efficiency obtained from the set of measured delay scans acquired at the few applied CEP offsets, and is therefore associated with a specific ΔT and CEP setting. In the simulations, the optimisation is refined by sampling the CEP on a much finer grid ($< 0.1\pi$ step size) than in the experiment. In both measured and simulated data, η_{iap} is calculated from the IAP energy ($E_{\text{SXR}}^{\text{IAP}}$), obtained by integrating the HHG spectra after excluding cases that, in the simulation, do not qualify as isolated emissions. The same experiment and analysis were also carried out in neon, where cutoff extension was observed (see Supplementary Information 1.6).

Figure 2a shows that the measured $\eta_{\text{iap}}/10\%\text{BW}$ for the IR driver spans values between 10^{-12} and 10^{-10} , depending on photon energy. By contrast, the synthesised waveforms yield efficiencies ranging from 10^{-11} to 10^{-9} —a more than tenfold improvement on average across the 225–350 eV range, with the highest gain obtained at the middle energy value. Note that the gas target position and pressure for the synthesised fields were not re-optimised as they were for the IR driver; hence, even higher efficiencies could be possible upon further macroscopic optimisation. Single-atom and macroscopic simulations (Figs. 2b–c) reproduce these enhancement trends qualitatively. The simulations also provide access to IR pulse energies beyond those experimentally available (blue solid line in Fig. 2c). In particular, the macroscopic simulations suggest that increasing the IR pulse en-

ergy to 223 μJ reduces $\eta_{\text{iap}}/10\%\text{BW}$ in the region up to 300 eV, while at higher photon energies it could still be increased, although the efficiency remains below 10^{-10} and generally lower than that achieved with synthesised waveforms. Taken together, these results indicate that specific waveform configurations can lead to enhanced efficiency, and allow to associate the observed increase primarily with waveform tailoring itself, rather than arising solely from increased peak intensity or phase-matching conditions.

We next examine the second question about which specific waveforms drive the efficiency boost. Figures 2d–f show the measured waveforms maximising the efficiency at photon energies of 250, 300 and 350 eV, together with the corresponding predictions from single-atom and macroscopic simulations. Most measured waveforms share a similar shape across all photon energies and input energy values, with a first peak around -2.5 fs, followed by an M-shaped half-cycle of opposite sign spanning -1.5 to +2 fs, and a secondary peak near +3 fs. This temporal structure closely resembles those reported in previous works^{37,53}. Despite their overall similarity, subtle variations emerge as the input energy is increased. For instance, the amplitude of the first peak decreases with increasing input energy in favour of the secondary peak. In addition, the symmetry of the M-shaped half-cycle changes, with variations in the relative amplitudes of its two lobes. At the low input energy, the predicted driving waveforms for the 300 eV and 350 eV cases agree well with the measured waveforms at both the single-atom and macroscopic levels. As the input energy increases to the middle energy value, the single-atom results begin to diverge from the measurements, whereas the macroscopic simulations remain consistent with the experiment. At the highest energy value, and for waveforms optimised around 250 eV, discrepancies between experiment and

macroscopic simulations are more apparent. In the latter case, the fields from macroscopic simulations lack the characteristic M-shaped half-cycle and instead exhibit a quasi-sinusoidal profile that produces higher ionization. Such deviations may arise from the on-axis nature of the model, which cannot capture volume-averaging effects and plasma defocusing^{55,56}, which become increasingly relevant at high intensities.

3 Discussion

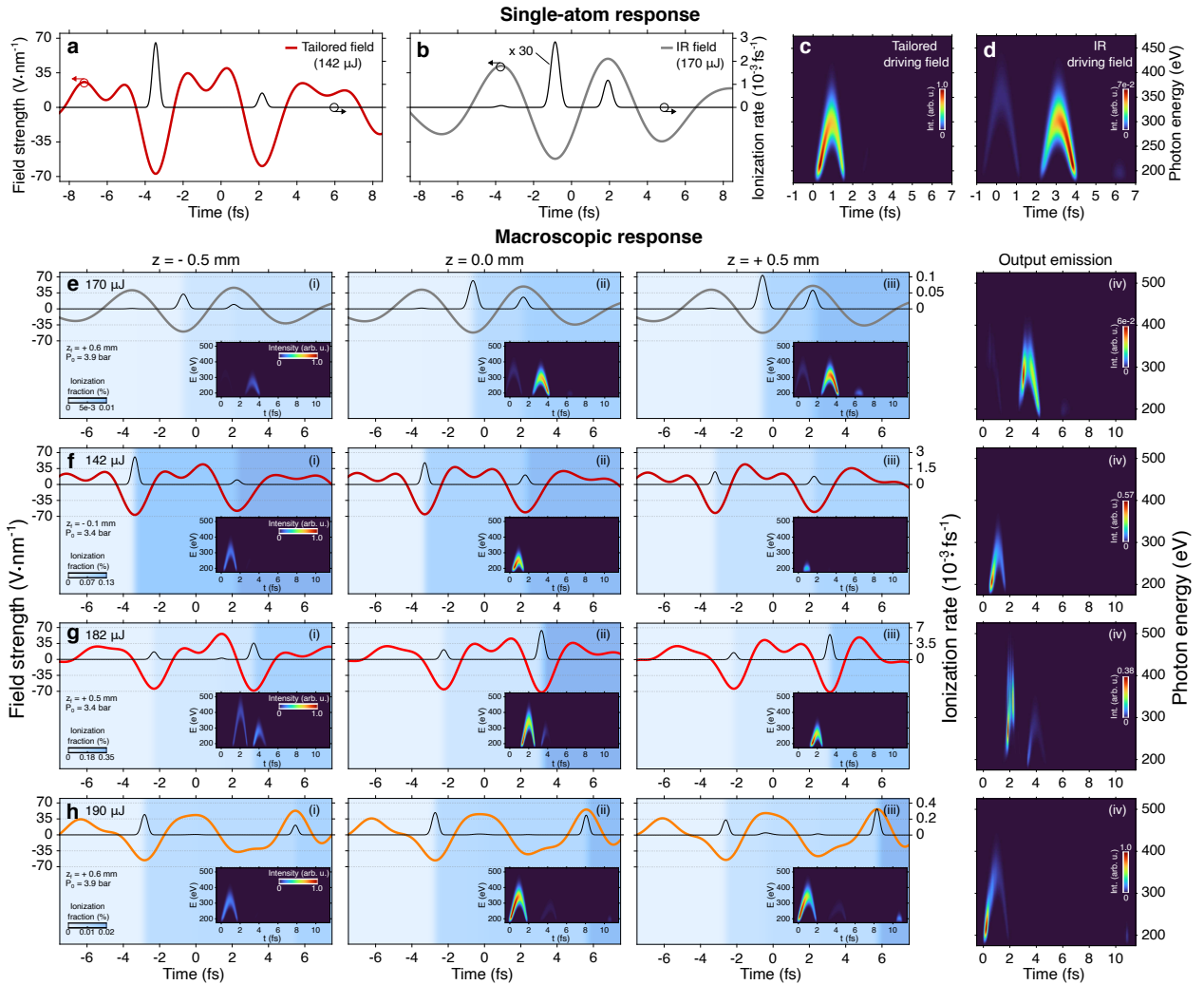


Figure 3: HHG driven by optimised driving fields. (a) Measured optimised tailored field at low input energy (left axis) with the computed ionisation rate (right axis). (b) Same as (a) for the IR driver at high input energy; the ionisation rate is scaled by $\times 30$. (c,d) Time–frequency profiles of the SAR driven by the fields in (a,b). (e–h) Macroscopic propagation dynamics for the measured IR driver (e) and the optimised tailored fields at low, medium and high input energies (f–h), shown at (i) $z = -0.5$ mm, (ii) $z = 0$ mm and (iii) $z = +0.5$ mm and displayed as in (a,b), with the ionisation fraction shown as a background colormap. The ~ 1 mm helium medium is centred at $z = 0$ mm; the simulated focus position (z_f) and pressure (P_0) are indicated for each case. Insets show the corresponding local single-atom time–frequency profiles, normalised to the maximum yield in each row (see Methods). (iv) Macroscopic SXR emission at the medium exit for the cases in (e–h). All analysed waveforms were optimised for emission at $300 \text{ eV} \pm 5\%$, and time–frequency profiles are multiplied by a Tukey window to suppress emission below $\sim 200 \text{ eV}$.

The third question, on how the single-atom response (SAR) and macroscopic response contribute to the enhanced efficiency, is addressed in Fig. 3, illustrating the corresponding mechanisms for waveforms maximising emission around 300 eV. We begin by considering the SAR. To isolate this contribution, we focus on the low-energy tailored field, for which the single-atom, macroscopic and experimental HHG scans show close agreement (see Supplementary Information Fig. 13). Figures 3a–b compare the optimised measured low-energy tailored field with the high-energy IR driving field, together with the corresponding time–frequency maps shown (c–d) for the maximised emission. Despite having lower input energy, the tailored waveform reaches a higher field strength ($\sim 70 \text{ V}\cdot\text{nm}^{-1}$) than the IR driver ($\sim 50 \text{ V}\cdot\text{nm}^{-1}$). Both waveforms generate isolated emissions with cutoffs beyond 300 eV, yet the IR-driven signal intensity is merely $\sim 7\%$ of that produced by the tailored field. This disparity stems primarily from the underlying ionisation dynamics. The tailored field induces two ionisation events: a dominant event peaking at -3.5 fs, launching electrons at a rate of $\sim 3 \times 10^{-3} \text{ fs}^{-1}$, and a secondary event at $+2.1$ fs with a rate below 10^{-3} fs^{-1} . Electrons from the primary event recombine within the 0–2 fs window –partially over-

lapping with the M-shaped half-cycle— to produce the single broadband burst, while the secondary event launches trajectories returning between 6—8 fs but with energies well below 200 eV. The IR driver instead generates three ionisation events. The first, at -3.7 fs, is negligible and results in a barely discernible emission in the -1 to $+1$ fs window, while another at $+2$ fs produces a contribution that remains largely confined below 200 eV. Notably, the dominant event at -1 fs—which produces the brightest HHG contribution in the 2–4 fs window, is $\sim 30\times$ weaker than the strongest ionisation event of the tailored field. This ionisation advantage^{48,57} is further reinforced by favouring short-trajectory contributions, which naturally experience reduced quantum diffusion. In the tailored case, the strongest field crest has a minimal temporal separation from the onset of short trajectories (~ 54 as), facilitating their generation³⁷. For the IR field, however, the corresponding separation is much larger (~ 280 as), leading to an emission dominated by long trajectories.

We now concentrate on the macroscopic scale. Figures 3e–f illustrate the propagation of the optimised high-energy IR and tailored waveforms (spanning low to high input energies) through the simulated ~ 1 mm helium medium (see Methods) at selected positions, alongside the resulting macroscopic HHG outputs. To highlight the impact of field reshaping on the SXR emission, the time-dependent ionisation fraction and SAR are overlaid, revealing the sub-cycle plasma influence and how the evolving field drives the local emission at each position.

In the IR case (Fig. 3e), the low plasma level ($\leq 0.01\%$) has a negligible effect on the waveform, and HHG is therefore governed by the evolution of the focused field in an effectively neutral gas. A dominant SAR emission is generated throughout the medium, consistently exhibiting spectral content around 300 eV. As the pulse approaches the focus at $z_f = +0.6$ mm, the rising field

235 strength causes the ionisation rate to nearly double between the medium entrance ($z = -0.5$ mm)
 236 and the centre, before rising further towards the exit ($z = +0.5$ mm), with the corresponding SAR
 237 yield increasing accordingly. This growth towards the exit compensates for the falling gas density.
 238 As a result, the macroscopic output is primarily shaped by emissions from the second half of the
 239 medium. The only notable difference is that the macroscopic emission shows a stronger short-
 240 trajectory character, owing to a more favourable phase-matching^{55,58}.

241 In the low-energy tailored field case (Fig. 3f), the first ionising half-cycle produces an ionisation
 242 fraction of $\sim 0.11\%$ near the medium entrance. As the pulse propagates, the resulting plasma-
 243 induced blueshift becomes the principal mechanism distorting the subsequent M-shaped half-cycle.
 244 It strengthens the left lobe at the expense of the right one while progressively reducing its area be-
 245 neath, thereby lowering the final electron acceleration and recombination energy. This makes the
 246 local SAR cutoff to drop rapidly from ~ 350 eV at the entrance to below 300 eV within the first
 247 half of the medium, barely reaching 250 eV near the exit. Consequently, the macroscopic yield
 248 around 300 eV is primarily generated near the entrance; nevertheless, it ultimately delivers a higher
 249 efficiency than the IR case, as shown in Fig. 2a.

250 At the middle-energy value (Fig.3g), the measured waveform near the medium entrance shows dif-
 251 ferences compared to the low-energy case. The first peak, at -2.3 fs, is no longer the strongest and
 252 the excess field energy is shifted to the following negative ionising crest at $+3.2$ fs. This causes
 253 the main half-cycle to ionise slightly less ($\sim 0.07\%$) than in the low-energy case. Such change in
 254 shape when moving from low- to mid-energy arises because the low-energy waveform (Fig. 3e),
 255 if driven at higher intensity, would undergo stronger plasma dispersion, leading to a more severe

256 distortion of the M-shaped segment responsible for the brightest emission. By shifting the excess
 257 ionisation to the later peak, this segment propagates with less distortion, enabling the coherent
 258 growth of 300-eV photons over most of the medium, yielding a higher flux than in the low-energy
 259 case. Note that the increased "M" asymmetry triggers a secondary ionisation at +1.4 fs, generating
 260 a 300 eV emission in the 3–5 fs window, which is rapidly suppressed by the action of plasma on the
 261 optical field. In addition, the pronounced ionisation at +3.2 fs inhibits the build-up of additional
 262 bursts, since the subsequent cycles quickly reshape during propagation. Together, these plasma-
 263 induced effects result in the macroscopic output effectively manifesting as an isolated emission.
 264 Contrary to expectation, in the high-energy optimal waveform (Fig. 3h), the early peak at -2.8 fs
 265 is considerably weaker than in the low energy case, ionising only $\sim 0.01\%$ of the atoms. This re-
 266 duced ionisation lowers the local single-atom yield, but, in turn, it allows the following flat-top-like
 267 half-cycle to sustain a high amplitude over a longer propagation distance than in previous cases.
 268 This permits the SAR to reach the target 300 eV photon energy through almost the full medium
 269 length, ultimately delivering an efficiency that nearly matches the mid-energy case. Additional
 270 ionisation events occur; however, the resulting SAR contributions are either too faint or reside be-
 271 low ~ 200 eV, ensuring the generation of a single and broadband burst at the macroscopic level.
 272 A common feature across all optimised (NIR + IR) waveforms is that they consistently favour
 273 short-trajectory emission throughout propagation. Moreover, the trajectories launched in the first
 274 event always recombine before a second strong ionisation event occurs, so their coherent growth
 275 is unaffected by the subsequent plasma formation. This contrasts with the IR driver, which favours
 276 long trajectories and whose dominant emission overlaps with a later ionisation event, so the asso-

277 ciated plasma growth can perturb its coherent build-up.

278 The previous analysis of the most efficient waveforms suggests that, within the ionisation model

279 and macroscopic conditions considered here, the driving cycle must limit plasma to below 0.1%

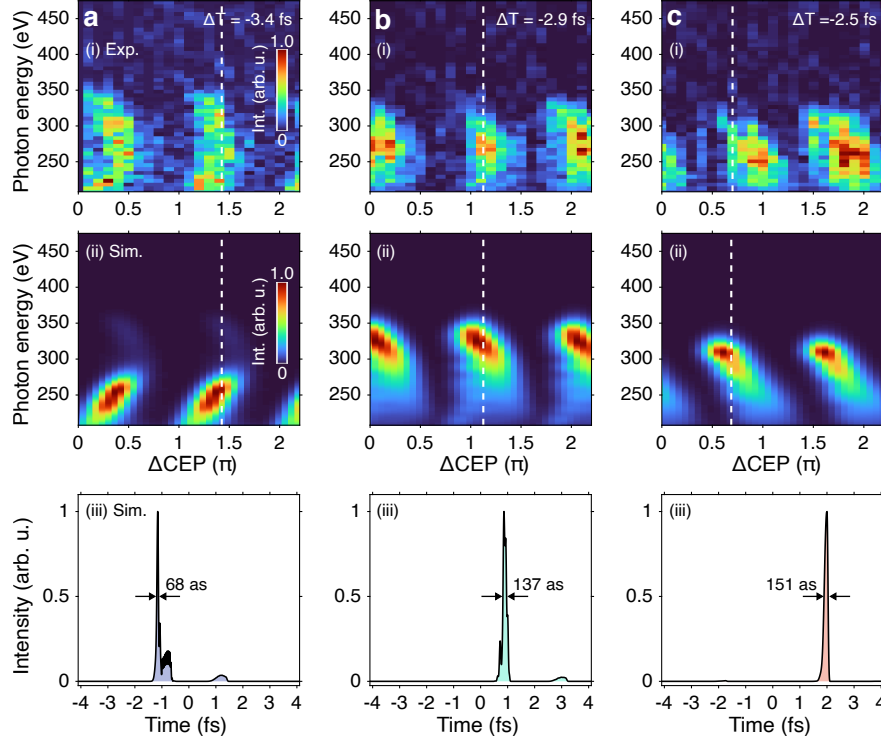
280 to ensure the return half-cycle propagates with minimal distortion. The amplitude and temporal

281 extent of this half-cycle must be such that electrons are accelerated as much as possible to return in

282 the minimum possible time without triggering additional ionisation events. Such a condition can

283 be achieved with an M-shaped or flat-top half-cycle, where its amplitude is $\approx 80\%$ of the preceding

284 ionising peak, and its duration is extended relative to that same peak.



285

286 **Figure 4: HHG dependency on CEP and IAP durations.** CEP dependency of the HHG driven by tailored
 287 waveforms obtained at $\Delta T = -3.4$ fs (a), -2.9 fs (b) and -2.5 fs (c). Each row shows (i) measured
 288 spectra, (ii) macroscopic simulations and (iii) the corresponding instantaneous intensity envelope at selected
 289 CEP values (white dashed lines in (i - ii)). The CEP scans were performed using the same waveforms and
 290 experimental conditions as the delay-CEP scans in Fig. 1d-f. Measured and simulated spectra are shown at
 291 the CCD plane, whereas the intensity envelopes are evaluated after the toroidal mirror (i.e., without grating
 292 and CCD transfer functions), corresponding to a virtual target. Simulations include far-field propagation,
 293 with long trajectories removed owing to their larger divergence (see Supplementary Information 2.3).

295

296 Finally, to verify that the enhanced emission corresponds to IAPs, we recorded HHG spectra
 297 driven by the tailored waveforms while scanning the CEP, which is an established indirect diagnos-
 298 tic for assessing IAP generation^{40,59-61}. Figure 4 shows measured (i) and simulated (ii) CEP scans,
 299 together with the corresponding simulated intensity profiles (iii) as they would appear after the
 toroidal mirror (i.e., at a virtual sample position). All scans were acquired using the same driving

300 waveforms and macroscopic conditions as in the low-energy delay scans (Fig. 1c–e.) Three delays
 301 are examined, -3.4 fs, -2.9 fs, and -2.5 fs, with the last one corresponding to the tailored wave-
 302 form of Fig. 3a,f. CEP scans display the expected π -periodic pattern and exhibit good agreement
 303 between experiment and simulation. The simulations further predict that the generated IAPs are
 304 below 200 as in duration, with one reaching ~ 68 as.

305 The delay window from -3.4 fs to -2.5 fs captures the evolution from a spectrum containing a
 306 main burst with a weak satellite to a clean isolated emission as ΔT is tuned. At $\Delta T = -3.4$ fs and
 307 -2.9 fs (Fig. 4a–b), the CEP scans reveal contributions from two consecutive half-cycles⁶². One
 308 branch reaches higher photon energies and shows a decreasing cutoff with CEP, while the other lies
 309 at lower energies with an increasing cutoff. The former originates from trajectories recombining
 310 in the M-shaped half-cycle, and the latter from trajectories recombining in an adjacent half-cycle.
 311 Owing to the ~ 2 -octave bandwidth of the tailored waveforms, pronounced amplitude and shape
 312 variations occur among the M-shaped half-cycle and its neighbours, such that the associated cut-
 313 offs no longer spectrally overlap, producing a characteristic “boomerang” shape. A clean isolated
 314 emission appears only at $\Delta T = -2.5$ fs (Fig. 4c), where the CEP scan shows a single cutoff that
 315 red-shifts monotonically with CEP and does not show any detectable contribution from adjacent
 316 half-cycles.

317 This phase-dependent behaviour provides a practical way to diagnose whether more than one half-
 318 cycle contributes to the emission. Delay scans in Fig. 1d–f display similar “boomerang” features in
 319 regions where two half-cycles contribute, as well as intervals in which no such structure appears,
 320 suggesting that the emission originates from a single half-cycle and is therefore isolated.

In conclusion, our results establish waveform synthesis as a viable and robust route for producing SXR IAPs with enhanced efficiency. The directly measured optimal waveforms closely match the predictions of earlier theoretical studies, and we show that the advantages provided at the microscopic level can be maintained macroscopically, even without optimising the phase-matching conditions. Supported by macroscopic simulations in quantitative agreement with experimental data, this work elucidates how propagation effects influence the isolation of single attosecond SXR emissions, establishing a framework for future waveform optimization that accounts for macroscopic propagation. Accurately including propagation effects on tailored non-sinusoidal waveforms is essential for designing the next generation of high-flux water-window attosecond sources, as they ultimately determine the achievable conversion efficiency.

4 Methods

Laser source The driving fields in this study are delivered by a two-channel parametric waveform synthesiser^{34,45}, which coherently combines two CEP-stable, few-cycle pulses with parallel p -polarisation. The NIR pulse is centred at 0.8 μm and has a FWHM duration of 7 fs, while the IR pulse is centred at 1.8 μm and has a 9 fs FWHM duration. The two pulses are collinearly combined (by a dichroic mirror) so that they both focus in the same position, with similar beam waists of ~ 40 μm for the NIR pulse and ~ 45 μm for the IR pulse. The intensity ratio at focus is $I_{\text{NIR}}/I_{\text{IR}} \sim 0.4$. The synthesis yields waveforms centred at 1.4 μm , with durations reaching down to 2.7 fs FWHM (0.6 cycles). Sub-cycle field shaping is achieved by tuning the relative delay (ΔT) between the two pulses and applying a common CEP shift.

HHG setup and measurement

SXR HHG is driven by either the synthesised waveforms or the IR pulse, both focused by a spherical mirror ($f = 400$ mm) into a ~ 1 -mm-long, gas cell filled with helium. This focusing yields Rayleigh lengths of $z_R \sim 2$ mm and 4 mm, for the IR and NIR beams, respectively. The input energy was adjusted using reflective neutral-density (ND) filters (ND0.0–ND0.3), where ND0.0 corresponds to an uncoated 2 mm UV-grade fused-silica plate to preserve pulse compression at the gas target. The transmitted energy decreases relative to ND0.0, with transmission values of approximately 96% (ND0.1), 75% (ND0.2), and 46% (ND0.3). The resulting HHG propagates through 150-nm-thick Al and 100-nm-thick Cu foils, that suppress photon energies below ~ 200 eV. The SXR beam is focused by a gold-coated toroidal mirror ($f = 500$ mm) at the entrance of a grating spectrometer (McPherson 251 MX) equipped with a CCD detector (Newton 940). The CCD detector features an array of 2048×512 pixels and was maintained at -50°C during operation. Each spectrum was recorded with an exposure time of 2 s, and the recorded spectra were binned into 128 pixels along the dispersion axis. A calibrated photodiode downstream of the toroidal mirror is used to measure the absolute SXR photon flux. The SXR photodiode (Opto Diode Corp. SXUV100, 10×10 mm² active area) was connected to an operational amplifier (IRD Inc. AXUB100HYB1V), converting the photocurrent to a voltage with a 10 G Ω resistor. The photon flux was then calculated from the responsivity curve provided by the manufacturer. Phase-matching was optimised for the IR driver by scanning the gas-cell position along the laser focus and adjusting the helium backing pressure (up to 10 bar); the corresponding optimal parameters can be found in the Supplementary Information Tab. 2. Relative-delay scans were acquired in ~ 100 as steps, and CEP scans in increments of $\sim 0.03\pi$.

Waveform characterization Waveform characterization is performed *in situ* using the TREX method⁵⁴, with helium as the nonlinear medium. To minimize propagation effects, the most attenuating neutral-density filter (ND0.3) is used and the gas-cell backing pressure is reduced to ~ 1 bar. The resulting UV–VIS emission is extracted from the chamber with a kick-out mirror (see Fig. 1). Phase-stable ΔT scans provide third-order cross-correlation signals, recorded by a spectrometer and processed through an iterative retrieval algorithm.

Efficiency measurement The measured spectra were intensity-calibrated using a SXR photodiode (see Supplementary Information 1.7). The SXR photodiode (Opto Diode Corp. SXUV100, 10×10 mm² active area) was connected to an operational amplifier (IRD Inc., AXUB100HYB1V) converting the photocurrent to a voltage with a 10 G Ω resistor. From the calibration, the energy within a 10% bandwidth was obtained at the HHG source, while the total driving-field energy was measured directly before the gas cell using a standard energy meter (Coherent J-10MT). These quantities were then used to calculate the conversion efficiency. The conversion from CCD counts to absolute energy at the HHG source accounts for the detector sensitivity, the CCD quantum efficiency, the number of electrons generated per photon at a bandgap energy of 3.65 eV, the grating diffraction efficiency, the reflection coefficient of the gold surface for *p*-polarised light, and the transmission of the metal filters. The CCD and grating parameters were taken from manufacturer specifications, while the gold reflectivity and filter transmissions were obtained from the Henke optical constants database⁶³.

On-axis HHG macroscopic model

An on-axis model, closely following Refs.^{55,64}, was used to calculate the macroscopic harmonic signal (see Supplementary Information). The propagation of the optical driving fields in the nonlinear medium was first simulated by solving

$$\begin{aligned} \frac{\partial \tilde{E}}{\partial z}(\omega, z) = & i \left[k(\omega) - k_n(\omega, z) \right] \tilde{E}(\omega, z) \\ & + \frac{i}{2\omega c} \mathcal{F} \left\{ \frac{\partial J_{\text{abs}}}{\partial t}(t, z) + \omega_p^2(t, z) \tilde{E}(t, z) \right\} \\ & - \frac{i}{2k} \nabla_{\perp}^2 \tilde{E}(\omega, r, z) \Big|_{r=0} . \end{aligned} \quad (2)$$

Here $\tilde{E}(\omega, z)$ denotes the on-axis optical electric field in the frequency domain, with $k(\omega)$ and $k_n(\omega, z)$ the vacuum and medium wavevectors, respectively. The first term describes linear propagation in the neutral gas medium. The second term accounts for absorption due to ionisation through the current density J_{abs} , as well as plasma dispersion and the associated blueshift via the plasma frequency ω_p . Tong–Lin ionisation rates were used throughout⁶⁵, with justification and comparison to alternative models provided in the Supplementary Information 2.5. The final term represents the on-axis contribution of transverse diffraction and focusing, evaluated at $r = 0$, accounting for the intensity variation and Gouy phase shift.

With the propagated optical field known at each position z , the SAR is evaluated at each step using the open-source code *HHGmax*⁶⁶, which computes the Lewenstein integral. The macroscopic build-up of the harmonic field is then obtained by integrating the nonlinear polarisation at each position along the medium,

$$\tilde{E}_h(\omega, z = L_{\text{med}}) = -\frac{i\omega}{2c\epsilon_0} \int_0^{L_{\text{med}}} \tilde{P}_{\text{NL}}(\omega, z) e^{-i[k_n(\omega, z) - k(\omega) - i\frac{\alpha(\omega, z)}{2}]} (L_{\text{med}} - z) dz, \quad (3)$$

where L_{med} denotes the medium length and absorption is incorporated through the coefficient $\alpha(\omega, z)$. The macroscopic polarisation is given by,

$$\tilde{P}_{\text{NL}}(\omega, z) = \rho(z) \mathcal{F}\{[1 - \eta_e(t, z)] d_h(t, z)\}, \quad (4)$$

derived from the single-atom dipole moment $d_h(t, z)$ and weighted by the neutral atom density, with $\rho(z)$ the gas density profile and $\eta_e(t, z)$ the ionisation fraction. The gas density distribution along the propagation axis was modelled as an asymmetric $\cos^2$ profile, featuring a flat 0.9 mm central region and smoothly tapered wings over a total length of 1.2 mm, with a slight asymmetry reproducing the experimental gas-cell geometry. The propagation step size was set to 2–4 μm . The influence of the pressure-profile geometry on the simulated scans is discussed in the Supplementary Information Fig. 18. The propagation of the optical waveforms was also computed with the 2D open source code Luna.jl⁶⁷ for comparison with the 1D code (see Supplementary Information 2.4).

Long trajectories are not expected to reach the detector, located few meters away, due to their larger divergence. To account for this, the far-field projection of the macroscopically simulated emission is restricted to short trajectories. These are filtered out by retaining only the positive-chirp component of the dipole phase for each atom during the HHG build-up.

411 The peak intensities corresponding to the measured input energies were estimated from the unat-
 412 tenuated IR driving beam, corresponding to the high-input energy value. Using the pulse energy
 413 (170 μJ), beam waist ($\sim 45 \mu\text{m}$), and the time-integral of the reconstructed IR intensity envelope
 414 ($\sim 13 \text{ fs}$), the peak intensity was estimated to be $400 \text{ TW}/\text{cm}^2$. Comparison with the measured
 415 HHG cutoff indicated that this value overestimates the effective intensity by about 5%, likely ow-
 416 ing to residual astigmatism or partial energy loss at the gas-cell entrance. The peak intensity used
 417 in the simulations was therefore set to $380 \text{ TW}/\text{cm}^2$, defining the high-input energy value. The
 418 middle- and low-input energy values were obtained by scaling this value according to the trans-
 419 mission of the ND filters, yielding peak intensities of $365 \text{ TW}/\text{cm}^2$ and $281 \text{ TW}/\text{cm}^2$, respectively.
 420 For the synthesised (NIR+IR) fields, the peak intensities were determined using the NIR-to-IR in-
 421 tensity ratio extracted from the TREX measurements.

422 Each simulated spectrum was computed within a 140 fs temporal window to fully encompass the
 423 synthesised field. A 2 as time step ensured sufficient spectral coverage to capture the highest cutoff
 424 energies. The delay (ΔT) grids closely matched the experimental ones (with 100 as steps), while
 425 the CEP grid spanned from -0.25π to $+0.75\pi$ in 0.0625π increments, thus covering the three CEP
 426 shifts and accounting for the CEP noise. The 300 mrad r.m.s. CEP noise of the driving laser was
 427 modelled by averaging each spectrum over the CEP axis using a Gaussian weighting centred on
 428 the nominal CEP. Time–frequency analyses of the HHG emission were performed using a con-
 429 tinuous wavelet transform (CWT) implemented in the MATLAB Wavelet Toolbox^{68,69}. For SAR
 430 simulations, the analysis was applied to the dipole acceleration $\ddot{d}_h(t, z)$, and the squared modulus
 431 of the resulting wavelet coefficients was used to obtain the corresponding time-frequency intensity

432 profile. For the macroscopic emission, the same wavelet analysis was applied to the harmonic
433 electric field at the exit of the medium, obtained from Eq. (3).

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Author contributions G.M.R. conceived the experiment, that was performed by M.A.S.T., F.S., M.K., R.E.M. and G.M.R.. F.S. extended the beamline for water window HHG. M.A.S.T., M.K. and R.E.M. and developed the data-acquisition systems. M.A.S.T., M.K. and F.S. carried out the data analysis. M.A.S.T. and R.Q.G. developed the HHG simulation framework and performed simulations. G.M.R. and F.X.K. supervised the project. M.A.S.T., G.M.R. and R.Q.G. wrote the manuscript with input from all authors.

Data and code availability The experimental data and computer codes used in this paper are available from the corresponding author upon request.

Competing Interests The authors declare that they have no competing financial interests.

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