Extreme ultraviolet radiation with coherence time greater than 1 s

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Many atomic and molecular systems of fundamental interest possess resonance frequencies in the extreme ultraviolet (XUV) where laser technology is limited and radiation sources have traditionally lacked long-term phase coherence. Recent breakthroughs in XUV frequency comb technology have demonstrated spectroscopy with unprecedented resolution at the megahertz level, but even higher resolutions are desired for future applications in precision measurement. By characterizing heterodyne beats between two XUV comb sources, we demonstrate the capability for sub-hertz spectral resolution. This corresponds to coherence times >1 s at photon energies up to 20 eV, more than six orders of magnitude longer than previously reported. This work establishes the ability of creating highly phase-stable radiation in the XUV with performance rivalling that of visible light. Furthermore, by direct sampling of the phase of the XUV light originating from high-harmonic generation, we demonstrate precise measurements of attosecond strong-field physics.

XUV comb generation and XUV interferometer

In the experiment, we pumped two independent femtosecond enhancement cavities20,21,23 (fsECs) with a high-power Yb:fibre frequency comb outputting 120 fs pulses at 154 MHz with an average power of 80 W centred at 1,070 nm (ref. 24). To pump two fsECs the frequency comb was split into two by an AOM so that the two resulting combs had a carrier envelope offset frequency detuning of $\Delta f_{\text{CEO}} \equiv 1$ MHz. Each fsEC (XUV1 and XUV2, Fig. 1a) was actively stabilized using the Pound–Drever–Hall technique with piezo-electric transducers on cavity mirrors as actuators. The carrier envelope offset was not actively stabilized, but its passive stability was sufficient to maintain good power enhancement. Each fsEC typically operated with an average power of $\sim 4.5$ kW and a peak intensity of $4 \times 10^{13}$ W cm$^{-2}$. Xenon gas for HHG was injected at the cavity foci via quartz nozzles with $\sim 150 \mu$m apertures backed by $\sim 1$ atm of pressure. Harmonics were coupled out of the fsECs in a co-linear fashion using 330-μm-thick intracavity sapphire plates set at the Brewster angle for the fundamental radiation to limit intracavity loss. The finesse of the fsECs was intentionally kept low to mitigate the nonlinearities of the Brewster plate and the plasma22,25,26, but enhancement factors of $\sim 200$ were still obtained. The two XUV beams were combined (more on this later) and detected using either an electron multiplier or a photomultiplier tube (PMT) with a phosphor screen. A schematic of the optical layout is shown in Fig. 1a. Because the pump frequency combs were offset by $\Delta f_{\text{CEO}}$, the resulting XUV frequency comb had a relative detuning of $q \times \Delta f_{\text{CEO}}$ and heterodyne beatnotes could be
observed at these frequencies, so a series of beatnotes appeared in the radiofrequency output of the detector, effectively mapping the XUV spectrum of the harmonics to the radiofrequency domain (Fig. 1c,e).

Splitting the NIR frequency comb beam at the beginning of the interferometer with the AOM is straightforward, but recombination of the XUV beams at the end of the interferometer is not. Because there are no transparent materials in the XUV, there are no standard beamsplitters available for recombination. Instead, we rely on a wavefront division scheme. A silicon wafer with a 100 µm pyramidal aperture produced by KOH etching was used as the beam combiner (Fig. 1d). The beam from XUV2 was focused through the aperture, but the reflected beam from XUV1 is much larger than the aperture. All the optics for the XUV light, including the beam combiner, were coated with boron carbide (B₄C) to enhance their reflectivity in the XUV. In the near field, there was no overlap between the two beams, but in the far field the beams interfered with a circular, ‘bulls-eye’ fringe pattern (Fig. 1b). The fringe pattern could be easily observed by setting Δf_{ceo} = 0 and adjusting the relative path length of the interferometer so that pulses from XUV1 and XUV2 overlapped in time at the detector. To observe beat signals at finite Δf_{ceo}, the central portion of the fringe was selected with an aperture before detection. A key feature of this beam combination scheme is that the fringe period scales weakly as \( l \sqrt{\lambda} \), so the interferometer can work well over a broad range of wavelengths, allowing us to simultaneously observe beats at the fundamental, the 17th harmonic, and all the harmonics in between.

Because fsECs XUV1 and XUV2 were both pumped by a common NIR laser, the noise in the radiofrequency beatnotes was immune to the common-mode frequency noise of the Yb:fibre laser. Thus, the apparatus directly measured noise from the HHG process or the cavity-plasma dynamics. However, because the interferometer was not actively stabilized, there were small amounts of relative noise induced by vibrations in the optics, giving the two sources a non-zero relative linewidth that is technical in origin.

Phase noise and long coherence time
Here, we show that HHG shares many common features with classical frequency multiplication of radiofrequency signals. In frequency multiplication, the power spectral density (PSD) of the

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**Figure 1 | Schematic of experiment and harmonic spectra.**

(Left) **a.** A high-powered frequency comb is split with an AOM to create an f_{ceo} shift of 1 MHz. The split beams are then coupled into independent femtosecond enhancement cavities (XUV1, XUV2) to reach intensities suitable for HHG. The harmonic light is coupled out with a Brewster plate and is steered to the beam combiner (shown in d) with XUV optics.  

**b.** Transmitted, reflected and combined beam profiles. The central interference fringe is selected with a spatial filter before detection.  

**c.** Schematic of the harmonic spectrum. Each harmonic order has a frequency comb structure with teeth spaced by the laser repetition rate.  

**d.** Measured radiofrequency beatnotes of the 3rd to 17th harmonics. The beatnotes have been mixed down to lower frequencies from \( q \times 1 \) MHz for clarity. The beatnotes are shown on a 30 Hz resolution bandwidth.
phase noise $S_\delta(f)$ increases quadratically with harmonic order\textsuperscript{28}. Even if the multiplication process is noiseless, any noise around the carrier will still increase with harmonic generation, thus setting a fundamental limit on how phase noise transforms under frequency multiplication and thus the achievable coherence level in the XUV (see Methods). The linewidth of a carrier can be related to $S_\delta(f)$ by the approximation\textsuperscript{29}

$$1 \text{ rad}^2 \approx \int_{f_{3\text{dB}}}^{\infty} S_\delta(f) \, df$$ (1)

The limit of the integral $f_{3\text{dB}}$ is the point where approximately half of the power is in the carrier and half in the noise. Therefore, the 3 dB linewidth of the carrier will increase quadratically with harmonic order. If one is not careful to have a low-noise carrier before multiplication, the carrier will start to disappear, leading to the known phenomenon of carrier collapse\textsuperscript{30}. Figure 2 shows the full-width at half-maximum (FWHM) of each harmonic comb tooth plotted versus harmonic order. A simple fit to FWHM = $aq^2$, where $a$ is the scaling parameter, highlights the quadratic dependence of the linewidth. The linewidth dependence on harmonic order was also independent of intracavity power. This analysis shows that there is a fundamental scaling of phase noise from the generating laser to the resulting XUV light. For example, assuming the best available c.w. laser with a linewidth of 30 mHz (ref. 4) and that the fundamental comb will faithfully follow the phase of the c.w. laser\textsuperscript{31}, the linewidth at the 17th harmonic would be 8.7 Hz. Fortunately for HHG, we do not observe any additional linewidth-broadening mechanisms other than the unavoidable classical frequency multiplication scaling.

We show that the previously observed linewidth originated from the differential path noise and not HHG physics, so higher levels of coherence can be observed. The differential path length noise in the interferometer can be removed by phase-stabilizing the two optical paths. A small amount of pump light leaks through the XUV optics to the detection plane, which is then picked off and detected on a photodetector to provide an error signal sensitive to interferometer fluctuations. The error signal is filtered and used to apply feedback correction to the AOM frequency, thus removing the relative noise (see Methods). With the interferometer phase locked, we can probe any noise processes intrinsic to intracavity HHG. A dramatic change in the linewidth of the XUV beatnotes is observed with a stable interferometer. The unstabilized beatnote of the pump laser and the beatnote at the 17th harmonic are shown in Fig. 3a,c and the corresponding stabilized case in Fig. 3b,d with a 250 mHz resolution bandwidth. Similar resolution limited beats were observed on the 3rd–19th harmonic. The coherence of the pump lasers is faithfully transferred to the harmonics as seen by the resolution-limited beatnote of 62.5 mHz in Fig. 3f. This is equivalent to a coherence time of 16 s or a stable phase relation maintained over $\approx 1 \times 10^9$ consecutive pulses. This is a nearly seven orders of magnitude larger coherence time than ever reported in the XUV\textsuperscript{16,18,32}. This establishes that our XUV comb system is extremely phase-coherent and has the capability to support sub-hertz coherences in the XUV. Furthermore, by also observing coherent beats from harmonics generated from xenon gas in one cavity and krypton in the other (Fig. 3e), we demonstrate that the common-mode noise rejection of this measurement scheme is not conditioned on the two XUV comb systems being identical.
Application to attosecond strong-field physics

The heterodyne beatnotes in the XUV provide unique and unprecedented access to the phase of XUV radiation. We can use this to probe attosecond physics and measure the intensity-dependent dipole phase. This technique is a fundamentally different method for probing this phenomenon than used in previous realizations and does not rely on extensive spatial and spectral filtering to observe interference between multiple quantum pathways. Furthermore, this technique does not rely on referencing the short to long trajectory contributions, or vice versa. It is complementary to RABBITT and related methods based on photoelectron spectroscopy, which seek to determine the time delay (or equivalently phase shift) between adjacent harmonic orders at a given intensity. In this experiment, we isolate individual harmonics and precisely measure their phase shift as a function of intensity.

These phase shifts can be directly linked to the temporal dynamics of the electron in the intense laser field. By using amplitude modulation (AM) on one arm of the interferometer, we can measure the amount of induced phase modulation (PM) on the XUV light (see Methods). For this measurement, the peak intensity of the modulated cavity was $3.4 \times 10^{13} \text{ W cm}^{-2}$, with a 15% AM depth. The intensity-dependent dipole phase can be expressed as

$$\phi = -\alpha_j \frac{U_p(I)}{\omega} = -\alpha_j \frac{I}{4\omega^3}$$

where $I$ is the laser intensity, $U_p(I)$ the pondermotive energy, $\omega$ the laser frequency, $\phi$ the phase of the emitted XUV light and $\alpha_j$ is the phase coefficient. As a result of our on-axis spatial filtering (needed to observe a high-contrast beatnote) we are primarily sensitive to the

Figure 3 | Demonstration of sub-hertz coherence in the XUV.

a, c. Unstabilized beatnotes of the pump laser and the 17th harmonic. b, d. Stabilized beatnotes of the pump laser and the 17th harmonic. e, f. Comparison of the two sources when one is injected with krypton and the other with xenon. A 1 Hz resolution bandwidth limited signal is achieved. f. The resolution of the 17th harmonic can be improved further to a 62.5 mHz resolution bandwidth showing a coherence time of >16 s. RBW, resolution bandwidth.
short trajectory. The schematic of the measurement is shown in Fig. 4a. The measurement requires a two-step demodulation process to extract the amount of PM induced on the XUV light from the AM applied to the pump laser. Great care was taken to ensure that the amount of PM on the XUV light was not induced by parasitic PM on pump laser (see Methods). The phase of the XUV light depends on both the intensity of the light and the particular quantum path the electron traverses (Fig. 4b). The result of the measurement is shown in Fig. 4c, where the $\alpha$ of equation (2) is expressed in atomic units according to the convention of Yost and co-authors. The shaded purple region corresponds to values predicted by the standard semi-classical model (SCM). The grey shaded region represents the range of values predicted by different approximations to below-threshold harmonics.

Below threshold, short-trajectory harmonics do not originate from tunnel ionization as in the above-threshold case and are much more sensitive to the atomic potential and ionization dynamics. Our measurement is able to discriminate between contributions of the standard SCM and a model that includes over-the-barrier (OTB) ionization to confirm that the below-threshold harmonics mostly originate from OTB ionization. Furthermore, our measurement for an above-threshold harmonic (15th and 17th) agrees well with the predictions of the SCM and the below-threshold with the predictions of Yost and co-authors and the theoretical framework of Hostetter and co-authors. Further exploration of intensity-dependent phases in atoms and molecules with comparison to calculations is the subject of future work. Our phase measurement technique was able to resolve phase shifts with uncertainties at the $10^{-2}$ rad level, which corresponds to a time uncertainty of $\sim 6$ as. In contrast to typical experiments that utilize direct attosecond timing resolution, we measure the attosecond electron dynamics imprinted on the phase of the emitted XUV light originating from HHG. With system improvements, it is feasible to extend this into the $<1$ as regime, rivalling the highest achievable temporal resolution of attosecond electron dynamics. Thus, our apparatus provides direct, unambiguous access to the phase of XUV radiation and will prove to be a valuable tool for attosecond science and the dynamics of atoms and molecules in intense laser fields.

Summary and future outlook

We have demonstrated that HHG is extremely phase-coherent. We have identified the primary noise requirements on the pump laser and shown that it is possible to support coherence times greater than 1 s in the XUV. We have also developed an interferometer capable of operating from 1,070 nm to 56 nm, but with different optics and a new beam combiner extension to even shorter wavelengths is possible. Such an apparatus will be a vital tool for future work in dual-comb spectroscopy, Fourier transform spectroscopy, high-resolution molecular spectroscopy, attosecond electron dynamics in intense laser fields and HHG spectroscopy. We have successfully probed physics at attosecond timescales using the tools of frequency metrology to measure the intensity-dependent dipole phase. Future work will require improved output coupling and power-scaling schemes to extend the high level of phase coherence to shorter wavelengths, possibly enabling spectroscopy of a nuclear isomer transition where highly phase-stable light will be needed for excitation.

Methods

**AOM and beat detection.** To create a small frequency offset between the two XUV sources we relied on an AOM to frequency-shift the pump laser such that the relative detuning was 1 MHz. Because there are no available AOMs at this frequency, we drove the AOM such that $1 \text{ MHz} = f_{\text{rep}} - f_{\text{det}}$. Two beats at frequencies less than $f_{\text{rep}}$, one at $f_{\text{det}}$ and the conjugate at $f_{\text{rep}} - f_{\text{det}}$ were therefore observed. The conjugate beatnote is sensitive to any noise in $f_{\text{rep}}$. To remove this, we derived $f_{\text{det}}$ by phase-locking a voltage-controlled oscillator to $f_{\text{rep}}$ detuned by 1 MHz such that $f_{\text{det}} = f_{\text{rep}} - 1$ MHz. This removed the dependence of $f_{\text{rep}}$ from the conjugate beatnote and put it in $f_{\text{det}}$. Accordingly, the low-frequency beatnote can be detected noise-free where we have detectors of adequate bandwidth.

**Phase noise.** A signal oscillating at frequency $\omega$ with PM can be expressed as

$$A = A_0 e^{-\beta (\text{phase}) + \omega t}$$

where $\beta$ is the modulation depth and $\text{phase}$ is the modulation frequency. When $\beta$ is small, we can express the power in the first-order modulation sideband (SB) relative...
to the carrier (C) by $P_{IR}/P_1 = (I_v(\beta)/I_v(\beta_0))^2 \approx I_v(\beta)^2$, where $I_v$ are Bessel functions. We can extend this to the case of general PM and not at a particular discrete tone by $P_{IR}/P_1 \approx (1/2)\Delta f_{dc}$. We can define the phase noise power spectral density as the noise around a carrier within a bandwidth (BW) as

$$S(f) = \frac{1}{2} \Delta f_{dc}^2 \frac{BW}{(20 \log_{10}(f))} \, (4)$$

By integrating $S_{dc}(f)$, one arrives at equation (1) and gets the approximate linewidth of a signal. The $f_{dc}$ point is the cutoff of the integral where there is approximately half the power in the carrier and half in the noise.

### Phase-stable interferometer

The XUV light is very sensitive to path length fluctuations and any phase fluctuations in the driving laser. It is therefore imperative to keep the interferometer stable to generate the highest levels of coherence in the XUV. The passive stability is sufficient for sub-kilohertz levels of coherence. To stabilize the interferometer, we used the small amount of pump laser light that co-propagates with the XUV light. Because the pump light diverges more than the XUV light, it was simple to separate it out before the detection plane. A small amount of light was picked off and sent to a photodetector where a 1 MHz beatnote was used to measure the phase fluctuations in the interferometer caused by mechanical noise on the mirrors. The beatnote was mixed with a radiofrequency synthesizer to generate an error signal. The error signal was filtered and the correction signal was applied to the 1 MHz reference frequency for the AOM.

Because the fluctuations of the interferometer were small, the phase-lock loop that sets the AOM frequency could easily compensate for the small, necessary corrections.

### Measurement of intensity-dependent dipole phase

The intensity-dependent dipole phase is expressed in equation (2). By trying to measure the intensity-dependent phase that results from HHG, we are effectively measuring the AM–PM coupling, with the AM being on the pump laser and the PM being on the XUV light. We can mathematically describe a beatnote signal as

$$S(t) = [1 + A \sin(\Omega t + \phi_m)] \cos(\omega t + P \sin(\Omega t + \phi_m)) \, (5)$$

where $\omega$ is the frequency of the beatnote, $\Omega$ is the frequency of the applied modulation and $\phi_m$ is its phase, $A$ is the AM depth and $P$ is the PM depth. Each beatnote is characterized by its own values for $A$ and $P$. To avoid confusion, the subscripts will refer to which signal it represents. For example, $A_{IR}$ is for the AM of the pump laser and $P_{IR}$ is the PM of the $q$th harmonic. Our task is to determine the values for $A$ and $P$ at the pump and the harmonics. By taking the ratio of $P_{IR}$ to $A_{IR}$ and using proper units, we can extract the intensity-dependent phase coefficient $\alpha$. We need to simultaneously extract the relevant parameters of equation (5) at both the harmonic of interest and the IR. To do this, we use a two-step demodulation process. By taking equation (5) and mixing it with a stable radiofrequency signal ($\Omega_0 t$) at a frequency of $\omega$ and relative phase offset $\phi$, we obtain

$$S_1(t) \approx V_1 \cos(\omega t + \phi) = S_1(t) \, (6)$$

$$S_1(t) \approx V_1 \cos(\phi) - AP \sin(\phi) + A \cos(\phi) \sin(\Omega t + \phi_m) \approx V_1 \cos(\phi) - AP \sin(\phi) \, (7)$$

We have ignored terms at $2\omega$. We can further low pass the signals at $\Omega$ and obtain a ‘dc’ signal

$$S_1(t) \rightarrow V_1 \cos(\phi) = S_1(t) \, (8)$$

Equation (8) will be one of our primary signals. Note that the phase is set by the phase of $\Omega_0$. This also assumes that the phase of the XUV beatnote is stable. This is true when we phase-stabilize our interferometer.

The signal $S_1(t)$ contains terms that oscillate at the applied modulation frequency $\Omega$. We can demodulate our signal once more using a lock-in amplifier at the correct phase $\phi_m$ and ignore terms at $2\Omega t$ to obtain

$$S_2(t) \approx V_2 \sin(\Omega t + \phi_m) = S_{1,dc} \approx V_2 \cos(\phi) - P \sin(\phi) \, (9)$$

where $S_{1,dc}$ is our second signal. With equation (8), equation (9) and some independently measured parameters (for example, the existing AM–AM coupling), we can extract our parameters of interest. To measure the $A_{IR}$–$A_{IR}$ coupling, we can use our beatnote signals. Our XUV beatnotes are directly proportional to the amount of XUV power in each beam. The amount of beatnote power can also be easily measured on a radiofrequency spectrum analyser. By changing the amount of power in one of the enhancement cavities and observing how the beatnote power changes, we can determine how much the XUV power must have changed for a given laser intensity change. $A_{IR}$ can be determined by

$$\Delta f_{dc} = 20 \log_{10}(1/\sqrt{2})$$

where $\Delta f_{dc}$ is the measured beatnote power. By applying AM to the pump laser on one arm of the interferometer, we can control $A_{IR}$ very well. It is also easily measured with a photodetector. By varying the phase of $\Omega_0$, we can measure $S_{1,dc}$ and $S_{1,IR}$ (equations (8) and (9)) simultaneously. With the modulation ($\Omega_0$) turned off, $S_{1,dc}$ tells us the phase of the beat. With the modulation on, the relative phase between $S_{1,dc}$ and $S_{1,IR}$ can tell us the ratio $A/P$. Because $A$ can be measured independently, we can extract the amount of PM, $P$. This procedure needs to be done with the infrared signal and the XUV signal simultaneously to prevent any systematic errors.

The results of the measurements are shown in Fig. 4c. Each data point is the average of approximately ten runs of data where equations (8) and (9) were measured while varying the phase $\phi_0$ from 0 to $2\pi$. The measured values for $P_{IR}$ were corrected for any parasitic $P_{IR}$ using $P_{IR} \rightarrow P_{IR} - q \times P_{IR}$. This was verified by measuring the effect with gas present and absent. This can easily corrupt the intensity-dependent phase measurement and necessitates the correction described previously. Furthermore, any cavity oscillation due to bistability can render the signals too noisy. Therefore, by careful measurement of the PM on the NIR laser and the XUV beatnote, we can extract the intensity-dependent phase originating from HHG and not other macroscopic effects.

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Author contributions
C.B., T.K.A., A.C., D.C.Y. and J.Y. contributed to the design and planning of the experiment. C.B., I.H. and F.L. acquired the data. C.B., T.K.A., I.H., F.L. and J.Y. analysed the data. All authors discussed the results and contributed to the writing of the final manuscript.

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Competing financial interests
The authors declare no competing financial interests.