High photon flux table-top coherent extreme-ultraviolet source

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High harmonic generation (HHG) enables extreme-ultraviolet radiation with table-top set-ups1. Its exceptional properties, such as coherence and sub-femtosecond pulse durations, have led to a diversity of applications1. Some of these require a high photon flux and megahertz repetition rates, for example, to avoid space charge effects in photoelectron spectroscopy2–4. To date, this has only been achieved with enhancement cavities5. Here, we establish a novel route towards powerful HHG sources. By achieving phase-matched HHG of a megahertz fibre laser we generate a broad plateau (25 eV–40 eV) of strong harmonics, each containing more than $1 \times 10^{11}$ photons s$^{-1}$, which constitutes an increase by more than one order of magnitude in that wavelength range. The strongest harmonic (H25, 30 eV) has an average power of 143 µW ($3 \times 10^{10}$ photons s$^{-1}$). This concept will greatly advance and facilitate applications in photoelectron or coincidence spectroscopy, coherent diffractive imaging or (multidimensional) surface science2.

In the late 1980s, the first experiments on high harmonic generation (HHG) driven by intense pulsed lasers were performed6,7,8. The process rapidly attracted a great deal of attention because of its non-perturbative behaviour9,10, its coherence11,12, the potential for sub-femtosecond pulse trains13 or isolated attosecond pulses14, and its table-top set-up12. Today, significant efforts to further advance this still growing field are leading to an ever increasing demand for novel laser sources. The established laser technology for HHG is based on Ti:sapphire chirped pulse amplifiers, which deliver multi-millijoule, ultrashort (25 fs) pulses with 130 eV of a megahertz chirped pulse amplifier (FCPA), resulting in a conversion efficiency of more than 10$^{-4}$ into a single harmonic at 30 eV. This demonstrates a new class of coherent extreme-ultraviolet sources where the average power in the 25–40 eV range is increased by more than one order of magnitude over previous systems6–8.

HHG can be understood with a simple three-step model that describes the response of a single atom to a strong laser field16. The single-atom response to a certain intensity, that is, the dipole amplitude $A_q$, can be obtained quantum-mechanically19 or modelled by empirical scaling laws20. The overall yield obtained in HHG, however, critically depends on macroscopic effects (phase matching), that is, the coherent build-up along the generation medium. The dipole amplitude $A_q$ significantly increases with intensity, but at the same time the increased ionization fraction strongly influences the phase matching21. It has to be noted that the use of ultrashort laser pulses reduces the impact of ionization and allows the use of higher intensities1, therefore increasing $A_q$. Ultimately, the signal build-up is limited by linear absorption of the harmonics in the generation medium itself22.

For the present experiments we used an FCPA with subsequent nonlinear compression (see Methods). The system delivers laser pulses with 130–150 µJ of energy, a duration of ~30 fs (Fig. 1a) and variable repetition rates23. These pulses were focused to a focal spot diameter of 90 µm (1/e$^2$ intensity, Fig. 1b) inside a vacuum chamber containing gas jets (Fig. 1c) of different diameter. The generated harmonics and the remaining infrared light pass two SiO$_2$ surfaces (see Methods), reducing the average power of the driving laser to protect the following two aluminium filters (1 µm thickness) against damage. Subsequently, the harmonics are analysed with a flat-field grating-based spectrometer (see Methods). Optimization of the experimental parameters was carried out for xenon gas, as it has the lowest ionization potential and highest $A_q$ of all noble gases. The intensity in the focal spot was reduced gradually from $1.5 \times 10^{14}$ W cm$^{-2}$ until blueshifting of the harmonics with increasing pressure disappeared at ~9 $\times 10^{13}$ W cm$^{-2}$. Figure 2a presents the signals of the 23rd, 25th and 27th harmonics, obtained at this intensity level, with respect to various nozzle thicknesses.

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Figure 1 | Experimental set-up of the high harmonic generation experiments. 

a. The pulses of a fibre chirped pulse amplifier (CC-FCPA) are post-compressed (PC) (see Methods). Autocorrelation traces of the 340 fs (black) fibre laser and the 30 fs (red) compressed pulses are shown. The data is normalized (norm.). 

b. Intensity profile of the focal spot (diameter of 90 µm; 1/e² intensity). 

c. Experimental set-up after the nonlinear compression stage, comprising a vacuum chamber used for the experiments on high harmonic generation. Pulses are focused (b) into a gaseous target (krypton, xenon) provided by simple cylindrical opening nozzles of various sizes (see main text). The generated harmonics co-propagate with the infrared beam and impinge on a chicane of two SiO₂ substrates under a 75° angle of incidence. Two aluminium filters (each of thickness 1 µm), isolate the harmonics, which are then sent into a flat-field grating spectrometer or onto a photodiode (PD) (see Methods).

Figure 2 | Optimization and phase matching in high harmonic generation. 

a. Spatially and spectrally integrated signals (in arbitrary units (a.u.)) of three high harmonics (H23, H25 and H27) generated in xenon are shown with respect to nozzle opening size. 

b. Spectrum (blue curve) of the high harmonics transmitted through a 200 nm aluminium filter and a 200 nm zirconium filter, and respective percentages of the overall signal, as used for photodiode measurements (see main text and Methods). Inset: harmonic signal (sum over H21–H29) as a function of repetition rate. 

c. Signals of harmonics H23–H27 (solid lines) generated in a 1 mm xenon gas jet, recorded with respect to pressure. The results of a simulation (dashed lines, Supplementary Section II) are shown for an I = 8 × 10¹⁵ W cm⁻², 30 fs pulse. 

d. Signals of harmonics H23–H27 (solid lines) generated in a 600 µm krypton gas jet, recorded with respect to pressure. The results of a simulation (dashed lines, Supplementary Section II) are also shown for an I = 9.7 × 10¹⁵ W cm⁻², 30 fs pulse.
openings. Clearly, the signal increases up to the maximum opening diameter of 1 mm, where it starts to saturate. This behaviour is very similar for all observed harmonics (H19–H31), leading to broad plateau of strong harmonics. The absolute signal of the harmonics is obtained either with the known detection efficiency or with a photodiode (see Methods). The latter method was used to measure the average power of three harmonics (Fig. 2b) at 50 kHz, yielding values of 10.7 µW (H19), 13.3 µW (H21) and 15.5 µW (H23), which are in good agreement with our estimates (based on the known detection efficiency) of 8 µW (H19), 10 µW (H21) and 13 µW (H23), respectively. Because of this consistency between the two techniques, the photon flux for the remaining harmonics and for higher repetition rates was obtained using detection efficiencies. We also studied the phase-matching behaviour by varying the backing pressure of the gas jet and, simultaneously, recording the signal of the respective harmonics. The signal growth of the harmonics was also in good agreement with a numerical model20,22 (Fig. 2c; see Supplementary Section I). According to a calculation with the Ammosov–Delone–Krainov (ADK) ionization model, the ionization fraction at the peak of the pulse is 24% and higher than the critical ionization level21. The experiment was therefore performed in a transient phase-matching regime, where the coherence length $\lambda_{\text{coh}} = n \Delta k$ is larger than the medium length over a short time interval at the rising edge of the pulse where the harmonics are generated and phase matching is achieved. The absorption lengths at optimal pressure (~60 mbar in the interaction region, Fig. 2c) are 590 µm (H23), 881 µm (H25), 1.1 mm (H27) and 2.4 mm (H31), which means that the lower-order harmonics are close to the absorption limit, and a longer interaction length would be required for higher orders22 (Supplementary Section III). More importantly, the repetition rate of the laser system can be increased to 0.6 MHz (80 W of average power) with very similar pulse parameters (intensity, pulse duration and focal spot size), which is corroborated by the almost linear increase of the harmonic signal (Fig. 2b, inset). The spatial profiles and spatially integrated spectra for the experiments at 0.6 MHz are shown in Fig. 3a. The gas jet was positioned slightly behind the focus (~150 µm), which results in phase matching for the short trajectories, as indicated by the excellent spatial profiles of the low diverging harmonics13. A similar optimization was carried out for krypton gas targets (Fig. 2d), resulting in the use of a 600 µm nozzle placed ~180 µm behind the focus. The intensity of $\sim 9 \times 10^{13}$ W cm$^{-2}$ (600 kHz) is similar, but the ionization is significantly reduced to <3% at the pulse peak. Consequently, phase matching can be achieved over an increased time window around the pulse peak, which results in spectrally narrower harmonics (Fig. 3b). In this case the coherence length is ~1 mm and the absorption lengths for an optimal pressure of 110 mbar are 162 µm (H23), 221 µm (H25), 286 µm (H27) and 730 µm (H33), which means, again, that the lowest harmonic orders are generated absorption-limited (Supplementary Section III). The obtained average power of the harmonics generated in xenon and krypton are shown in the lower panels of Fig. 3. Strong harmonics with more than 30 µW per harmonic, that is, more than $1 \times 10^{12}$ photons s$^{-1}$, are obtained over a broad plateau extending from 25 eV to 40 eV. The highest average power is obtained for H25 (30 eV) with 143 µW, which corresponds to $3 \times 10^{13}$ photons s$^{-1}$ and a conversion efficiency of $1.8 \times 10^{-9}$. Experiments performed at a repetition rate of 300 kHz...
and a different spectrometer configuration show that microwatt-level harmonics are generated up to H39 (47 eV). The achieved photon flux is within one order of magnitude of typical free-electron-laser operation, although these facilities can offer up to a few $10^{11}$ photons s$^{-1}$ with high peak intensity. In conclusion, we have demonstrated the most powerful source of coherent extreme-ultraviolet radiation (25–40 eV) enabled with HHG to date. In combination with megahertz-level repetition rates, this will greatly advance applications in various fields, in particular quantum (information) and nonlinear surface science, coincidence detection experiments, photoelectron emission spectroscopy (PES) and microscopy (PEEM), (time-resolved) coherent diffractive imaging (CDI), among others. Due to the excellent scaling properties of coherent combination, the availability of kilowatt-average-power femtosecond lasers will further increase the HHG signal by another order of magnitude. Moreover, a second nonlinear compression stage can be implemented for achieving, potentially, sub-10 fs pulses with high average power and repetition rate. In combination with carrier-envelope phase stabilization this could also enable high-photon-flux megahertz isolated attosecond pulses. Accordingly, the presented results are a major milestone towards new applications of coherent extreme-ultraviolet radiation in science and technology.

Methods

Fibre laser with nonlinear compression. The front end is an FCMA system that incorporates coherent combination (CC) of pulses from up to four main amplifier channels. In the experiments presented here, the system was operated with a pulse energy of 270 μJ, a compressed pulse duration of ~340 fs and a repetition rate between 50 kHz and 0.6 MHz, which corresponds to an average power between 14 W and 163 W. Laser pulses were sent to the nonlinear compression set-up as described in ref. 25, and coupled into the 1.1-m-long hollow-core fibre (inner diameter of 250 μm). After vacuum, the tube was filled with 4 bar of krypton gas to enable spectral broadening. Subsequently, the pulses were compressed in time by a chirped mirror compressor with a group delay dispersion of -1,600 fs. After propagation through this set-up, the pulses were 29 fs short (Fig. 1a) with an energy of 130–150 μJ. At the highest repetition rate of 600 kHz, the average power used for HHG was >80 W.

HHG and characterization of extreme-ultraviolet radiation. The nonlinear compressed pulses were focused with an f = 300 mm lens (f̸= 350 focusing) onto a gaseous target inside a vacuum chamber. Simple cylindrical nozzles with different opening diameters (see main text) provided the gas targets. The generated harmonics and fundamental infrared laser co-propagated and impinged on a chicane of two SiO$_2$ substrates, which were used with an angle of incidence of 4°. We used a one-dimensional model to determine the availability of kilowatt-average-power femtosecond lasers will further increase the HHG signal by another order of magnitude. Moreover, a second nonlinear compression stage can be implemented for achieving, potentially, sub-10 fs pulses with high average power and repetition rate. In combination with carrier-envelope phase stabilization this could also enable high-photon-flux megahertz isolated attosecond pulses. Accordingly, the presented results are a major milestone towards new applications of coherent extreme-ultraviolet radiation in science and technology.

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Author contributions
J.L., S.H., J.R. and M.K. conceived the experiment. The experiments were planned and performed by S.H., J.R., A.K., A.H. and M.K. Data were analysed by S.H. with support from J.R. and M.K. All authors discussed and contributed to interpretation of the results. J.L. and A.T. supervised the project and acquired funding. The idea for and design of the anti-reflection-coated SiO₂ substrates originate from O.P. and V.P., who also fabricated the samples used in this experiment. All authors contributed to writing the manuscript.

Competing financial interests
The authors declare no competing financial interests.

Additional information
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