Spatio-temporal characterization of attosecond pulses from plasma mirrors

Ludovic Chopineau¹, Adrien Denoeud¹, Adrien Leblanc², Elkana Porat³⁴, Philippe Martin¹, Henri Vincenti⁵ and Fabien Quéré⁶

Reaching light intensities above $10^{25} \text{ W cm}^{-2}$ and up to the Schwinger limit of order $10^{29} \text{ W cm}^{-2}$ would enable the testing of fundamental predictions of quantum electrodynamics. A promising—yet challenging—approach to achieve such extreme fields consists in reflecting a high-power femtosecond laser pulse off a curved relativistic mirror. This enhances the intensity of the reflected beam by simultaneously compressing it in time down to the attosecond range, and focusing it to submicrometre focal spots. Here we show that such curved relativistic mirrors can be produced when an ultra-intense laser pulse ionizes a solid target and creates a dense plasma that specularly reflects the incident light. This is evidenced by measuring the temporal and spatial effects induced on the reflected beam by this so-called plasma mirror. The all-optical measurement technique demonstrated here will be instrumental for the use of relativistic plasma mirrors with the upcoming generation of petawatt lasers that recently reached intensities of $5 \times 10^{22} \text{ W cm}^{-2}$, and therefore constitutes a viable experimental path to the Schwinger limit.

Quantum field theory predicts that even the most perfect vacuum has a complex structure, characterized by a jostling of so-called virtual particles. Probing the nonlinear optical properties of vacuum resulting from the coupling of light with these virtual particles would enable unprecedented tests of these predictions²⁷−¹. The typical amplitude of the electromagnetic fields required to do so corresponds to the onset of the Sauter–Schwinger effect²⁷−¹, where charged particle–antiparticle pairs are predicted to spontaneously appear from a vacuum in which a sufficiently strong electric field is applied. For electron–positron pairs, this is expected for field amplitudes exceeding $E_0 = m_e^2 c^3 / e h = 1.32 \times 10^{19} \text{ V m}^{-1}$, corresponding to electromagnetic waves with intensity $I_0 \geq e_0 s_0 E_0^2 / 2 \approx 4.7 \times 10^{22} \text{ W cm}^{-2}$. Because these values are so high, this so-called Schwinger limit has never been reached or even approached in experiments. The concentration of light energy allowed by ultrashort lasers⁶ has raised the hope of approaching such intensities by focusing near-visible laser light⁴⁰. However, the critical value $I_0$ still far exceeds the present record laser intensity of $\approx 5 \times 10^{21} \text{ W cm}^{-2}$ that was recently achieved with a tightly focused petawatt femtosecond laser⁴¹. Further increasing the laser pulse energy appears to be a technologically hopeless path to close this gap of about seven orders of magnitude. A more realistic approach would consist in considerably increasing the concentration of this light energy. This requires a conversion to electromagnetic waves of shorter wavelengths, which can be more tightly focused in space and compressed in time.

A promising path to implement such a frequency conversion for high-power laser pulses is the concept of the curved relativistic mirror²²−²³. Upon reflection on a mirror moving at $v \leq c$, an ultra-intense laser pulse is compressed in time and downconverted in wavelength by a factor $\sim 4\gamma^2$ (where $\gamma = 1/\sqrt{1 - v^2/c^2}$ is the mirror’s Lorentz factor) due to the Doppler effect. For large $\gamma$, the reflected pulse can be compressed to the attosecond time range, and can now converge to a submicrometre focal spot, thus boosting the intensity of the initial laser by orders of magnitude. Although appealing, the major and so far unsolved challenge of this approach is the experimental generation of such curved relativistic mirrors.

Different implementations to achieve this goal have been proposed²²−²⁵ and debated²⁷. Experimentally, substantial advances have already been made in the last 15 years by using so-called plasma mirrors. These are dense plasmas produced at the surface of initially solid targets, ionized by intense femtosecond laser pulses²⁶−²⁷. They have the ability to specularly reflect high-power ultrashort laser pulses, just like ordinary mirrors do for ordinary light. At intensities exceeding $10^{18} \text{ W cm}^{-2}$, the laser-driven oscillation of the plasma surface becomes relativistic and thus induces a periodic Doppler effect on the reflected beam²⁷−²⁹. This results in the generation of a train of attosecond light pulses—with one pulse every laser period—associated in the frequency domain to a comb of harmonics of the laser frequency. In addition to these temporal effects, the surface of plasma mirrors can become strongly curved under the effect of the radiation pressure exerted by the incident laser field, which can provide a way to focus these attosecond pulses immediately after their generation. Implemented with the emerging generation of petawatt lasers, this scheme appears to be one of the few viable paths towards the Schwinger limit²⁸.

Pursuing this path requires the accurate measurement of the spatio-temporal properties of the reflected beam, down to the attosecond scale in time and nanometric scale in space, to be able to both assess and optimize the properties of the relativistic mirror. Despite the development of attosecond metrology in the last 20 years²⁸−³⁰, attosecond pulses generated from plasma mirrors have never been accurately characterized in time due to the challenge of implementing such advanced techniques in the extremely harsh conditions of laser–plasma experiments³⁵−³⁷. In this Article, we report the spatio-temporal characterization of attosecond pulses generated from relativistic plasma mirrors with a 100-TW-class femtosecond laser, using an all-optical technique. We thus get direct evidence for the effects that compress the light energy of the reflected beam in time and space. The measurement method that we demonstrate

¹Université Paris-Saclay, CEA, CNRS, LIDYL, Gif-sur-Yvette, France. ²Laboratoire d’Optique Appliquée, ENSTA-ParisTech, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France. ³The School of Physics and Astronomy, Tel Aviv University, Tel Aviv, Israel. ⁴Applied Physics Department, Soreq Nuclear Research Center, Yavne, Israel. ⁵E-mail: henri.vincenti@cea.fr; fabien.quere@cea.fr
Fig. 1 | Principle of dynamical ptychography. a–d. An example of dynamical ptychography for a case where the diffracting object (light grey area in a and c) is a sinusoidal reflective surface with a spatial period $D$. The ptychographic measurement consists in measuring the diffraction pattern of the illuminating beam (Gaussian profiles in a and c) as a function of the propagation angle $\theta$ (top insets in a and c) while scanning the position offset $x_0$ of this oscillating surface with respect to the beam position (from left to right in a and c). In dynamical ptychography, the probe results from the superposition of multiple frequencies $\omega$ represented by the differently coloured Gaussian profiles. Ptychographic traces $S(\theta, x_0, \omega)$ are measured for several of these frequencies, and such traces are displayed in b and d for two frequencies, $\omega_1$ and $\omega_2$. In this example, the object is assumed to drift with time $t$ at constant velocity $v$. The temporal information on the probe pulse is encoded in the frequency dependence of the phase of the oscillating ptychographic traces, as illustrated here for the case of an optimally compressed probe (a,b) and a chirped probe (c,d). The black dashed lines in b and d are guides for the eye.

Principle of dynamical ptychography

The measurement method implemented in our experiment is an extension of a powerful lensless imaging technique known as ptychography. This technique consists in illuminating a microscopic object, described by a transmission or reflection function $O(x-x_0)$, with a focused beam of coherent light of wavevector $k_0$, described by a field $E(x)$. The angular diffraction pattern $S(\theta, x_0) \propto |\int dx O(x-x_0)E(x)e^{i\theta k_0 x}|^2$ produced as the beam diverges away from the object is measured along the propagation angle $\theta$ as a function of the position $x_0$ of the object with respect to the beam. As an example, we consider the simple case of a sinusoidal surface as an object (Fig. 1a). This object is illuminated by a probe beam whose size at focus is smaller than the surface oscillation period $D$. The resulting ptychographic trace $S(\theta, x_0)$ is displayed in Fig. 1b. As the position $x_0$ is scanned, the peaks and dips of the surface are alternatively probed, and both the angular width and direction of the diffracted beam oscillate. The power of ptychography lies in the fact that iterative phase-retrieval algorithms can be applied to such a trace to reconstruct the spatial structures $O(x)$ of the object and $E(x)$ of the illuminating beam, in amplitude and phase. Ptychography is thus an advanced spatial measurement method for both microscopic objects and coherent beams of light or particles.

We now consider an illuminating probe consisting of an ultrashort pulse of light, and aim to add temporal resolution to ptychography to obtain both spatial and temporal information for this beam. We show that this is possible by using an object $O(x,t)$ that evolves in time in a known manner. The measurement then consists in spectrally resolving the diffraction pattern of the probe beam on this evolving object as the offset position $x_0$ is scanned. This provides a three-dimensional (3D) dataset $S(\theta, x_0, \omega)$, where $\omega$ is the frequency within the spectral width of the probe pulse. To illustrate this measurement scheme, we now assume that the object of Fig. 1 drifts in time with a known and constant velocity, $v$ (Fig. 1).

An ultrashort pulse of light is formed by the superposition of multiple frequencies. For a given spectrum, the shortest pulse is formed if all frequencies are perfectly synchronized in time (that is, the group delay is constant). In such a case, all these frequencies will probe the evolving object at the same instant (Fig. 1a). The multiple ptychographic traces measured at different frequencies $\omega$ would then be observed to oscillate in phase (Fig. 1b).

We now consider a probe pulse with the same spectrum, but with the frequencies arriving at different times (here, the arrival time of frequency $\omega$ refers to the group delay at this frequency, that is, it corresponds to the temporal position of the pulse formed by the superposition of frequencies within a small interval centred at $\omega$). This corresponds to a chirped pulse whose duration is longer than the minimum allowed by the pulse spectral bandwidth. Different frequencies $\omega$ within the pulsed beam will now probe the object at different instants of its motion, and will be diffracted differently (Fig. 1c). The oscillations of the multiple ptychographic traces measured at different frequencies would then be observed to be dephased (Fig. 1d), that is, to present a maximal deflection for different values $x_0(\omega)$ of $x_0$. If the object velocity $v$ is known, this dephasing directly encodes the chirp of the probe pulse through the following straightforward relationship:

$$\tau(\omega) = x_0(\omega)/v$$

where $\tau(\omega)$ is the group delay of frequency $\omega$ within the probe spectrum. Combined with the spatial information provided by each ptychographic trace, we can thus get access to the complete spatio-temporal structure of the illuminating beam.
This measurement scheme, which we call dynamical ptychography, is very general and can in principle be applied to very different types of object and probe, over a broad range of timescales, to determine the temporal properties of the probe if the evolution of the object is known, or vice versa.

**Application to attosecond pulses from plasma mirrors**

We can apply this general measurement scheme to determine the spatio-temporal structure of attosecond pulses generated from plasma mirrors. This calls for a diffracting object evolving very quickly in time—that is, typically moving by a fraction of its spatial period on an attosecond timescale. We fulfill this condition by using a transient optical grating as the diffracting structure: this consists of an evolving spatial interference pattern, applied on the ultra-intense laser beam that drives the interaction with the plasma mirror and generates the attosecond pulses. The spatio-temporal structure of this driving field is imprinted on the dynamics of the plasma mirror and hence on the generated attosecond pulses. This acts as the evolving diffracting object for the dynamical ptychographic measurement of this light source (Supplementary Section 2).

Such a fast transient optical grating can be generated by perturbing the main driving laser field of frequency $\omega_0$, with a much weaker secondary beam of frequency $2\omega_0$, arriving at a small angle $\theta_p$ with the main beam (Fig. 2a). At any given time $t$, the crossing of these two fields generates a sinusoidal spatial interference pattern—exactly as the object of Fig. 1. Because the two waves have different frequencies, the relative phase of the two beams changes as time evolves, and the resulting interference pattern therefore drifts spatially along the plasma mirror surface—again, like in the example of Fig. 1. The spatio-temporal structure of this transient optical grating can be easily calculated analytically (Supplementary Section 2), and is displayed in the inset of Fig. 2a. Its temporal period is that of the main driving field ($T_L=2.7 fs$). The methods of attosecond metrology developed in the last 20 years have shown that such a modulation at petahertz frequency indeed enables temporal measurements with attosecond resolution\(^{24,25}\).

A ptychographic measurement requires scanning the position $x_p$ of the diffracting structure with respect to that of the harmonic source, imposed by the spatial profile of the main driving beam. This can be achieved with high accuracy by changing the relative delay $\delta t$ between the two beams by small fractions of the laser optical period. At any given time $t$, this shifts the spatial interference pattern formed by their superposition, while the position of the harmonic source remains fixed. To ensure the required interferometric delay stability, we generate the perturbing $2\omega_0$ beam from a fraction of the main beam, thanks to an all-solid in-line optical set-up (Supplementary Section 1), and vary its delay with attosecond accuracy by tiny rotations of a glass plate used in transmission. Using an angularly resolved spectrometer (Fig. 2a), we then measure in a single delay scan the ptychographic trace of each individual harmonic in the spectrum of the attosecond pulse train.

We have performed such dynamical ptychographic measurements in different interaction regimes (Supplementary Section 1)\(^{19,21}\), from laser intensities exceeding $10^{19}$ W cm\(^{-2}\), where...
the Doppler effect described in the introduction is the main source of harmonic generation (relativistic oscillating mirror regime, ROM), down to much lower intensities of \( \sim 10^{11} \text{ W cm}^{-2} \), where attosecond pulses are produced by collective plasma oscillations, periodically triggered into the dense plasma by fast electron bunches (coherent wake emission regime, CWE)\(^{32}\). The comparison of the results obtained in these two regimes will provide stringent tests of the measurement method, as explained below.

We note that different measurement schemes based on the perturbation of a laser field by its second harmonic have been demonstrated in the last decade\(^ {33-35} \) for the characterization of attosecond pulses generated in gases or solids at laser intensities of \( \sim 10^{14} \text{ W cm}^{-2} \). In particular, the method implemented in ref. \(^ {34} \) for the spatio-temporal characterization of isolated attosecond pulses from gases is similar to the one used here. The justification of these schemes, so far, has been based on a quantum picture. The present work provides a different perspective, which suggests that dynamical ptychography can equally apply to some other systems where quantum effects do not play a role, such as plasmas exposed to laser intensities \( 10^{15} \) times higher than in all previous attosecond measurements.

**Spatio-temporal field reconstructions**

Figure 2b,d (top) displays two experimental ptychographic traces obtained for harmonic 9 in the CWE and ROM interaction regimes, respectively. These were processed by a ptychographic phase-retrieval algorithm (Supplementary Section 3), which converges to the reconstructions displayed below the measured data.

The first information obtained from the reconstruction of a single ptychographic trace is the spatial structure of the harmonic beam in the plane of the plasma mirror—along one spatial axis only, although an extension of the method to the two transverse spatial dimensions is, in principle, possible. The amplitude and phase profiles of harmonic 9 retrieved in the two interaction regimes are displayed in Fig. 2c,e. These spatial properties and, more particularly, the phase profile carry rich information on the physics of the harmonic generation process\(^ {36,37} \) and are consistent with analytical models\(^ {38,39} \) (black dotted lines, Fig. 2c,e) as well as previous spatial-only measurements\(^ {40,41} \).

In both cases, the observed curvature of the spatial wavefront is due to the spatial variation of the laser intensity across the focus. In the CWE regime, the intensity dependence of the electron dynamics at the plasma surface results in a diverging beam\(^ {31,36} \). The opposite curvature observed in the ROM regime is that of a converging harmonic beam, and has a different physical origin. At the ultra-high laser intensities involved in this regime, the radiation pressure exerted by the incident field on the plasma mirror surface reaches the gigabar range and leads to a recession of this surface typically by a few tens of nanometres\(^ {37,38} \). Because of the varying laser intensity, this recession is stronger at the centre of the focal spot than on the edges, and the plasma mirror surface thus becomes dented, acquiring a parabolic shape. This is precisely the type of curved relativistic mirror that is needed to boost the reflected light intensity.

The intensity boost that will be achieved at the plasma mirror focus is not only determined by the spatial properties of individual harmonics (obtained in Fig. 2). It also critically depends on the attosecond temporal compression of the reflected field. This is precisely the second type of information provided by dynamical ptychography. For each individual harmonic of frequency \( \omega_n = n \omega_L \), the ptychographic algorithm provides a reconstruction of the diffracting object. According to the previous sections, the emission time \( \tau(\omega_n) \) of this harmonic is encoded in the measured position offset of this object, following equation (1). We now experimentally validate this key feature of the measurement method.
The optical gratings retrieved from the two ptychographic traces of Fig. 2 are displayed in Fig. 3a. Although these two measurements have been performed in different laser-plasma interaction conditions, the moving grating used for dynamical ptychography remained the same in the two cases (Supplementary Section 1). As expected, it is sinusoidal in space, with a spatial period $D = 11.5\,\mu m$ determined by the angle between the main and perturbing beams. More importantly, we observe that the overall position of the object retrieved for the ninth harmonic differs by $\Delta x_n = 7.5\,\mu m$ in the two regimes. This position shift should be related to the different times of emission (within the laser optical cycle) of the attosecond pulses produced in these two measurements carried out in different regimes. Converted in time using equation (1) with $v = 14.3c$, this corresponds to a delay of $\Delta t_n = 1.8\,fs$ between these attosecond pulses (Fig. 3c).

Such a difference is indeed what is expected physically\textsuperscript{[12]}. In the ROM regime, attosecond pulses are emitted in the part of the optical cycle where plasma surface electrons are pulled outward by the incident laser field (Fig. 3d, bottom). Later in the laser optical cycle, these surface electrons are pushed back into the plasma, where they trigger the emission of CWE attosecond pulses (Fig. 3d, top). The measured delay quantitatively matches the one observed in particle-in-cell (PIC) simulations of the laser-plasma interaction ($\Delta t_n = 1.75\,fs$, Fig. 3d): this provides a striking validation of the temporal sensitivity of the measurement method.

We can now use the position offset $x_n(\omega_n)$ of the multiple objects retrieved from the ptychographic traces measured for different harmonics, to determine the group delay $\tau(\omega_n)$ of the associated attosecond pulses (Supplementary Section 3.2). These results are plotted in Fig. 3b for both the CWE and ROM regimes. In the CWE regime, we observe that the harmonics are emitted at different times, with a total delay of $\sim 150\,as$ between the emission of harmonics 9 and 14, leading to pulses slightly longer than the Fourier-transform limited duration (Fig. 3c, top). This varying group delay results from the fact that higher harmonics are emitted from denser parts of the plasma, which are located deeper into the target\textsuperscript{[32]}: they are therefore emitted later in the laser optical cycle. By contrast, all harmonics are found to be synchronized in the ROM regime, leading to attosecond pulses with the minimum duration allowed by the spectral bandwidth of the beam (Fig. 3c, bottom). This is because all harmonics are emitted at the same time when the plasma mirror moves outward at relativistic velocity (Fig. 3d, bottom). These results constitute an accurate temporal measurement of attosecond pulses generated from plasma mirrors. Although we do not access the group delay for all harmonic orders generated in this experiment due to the detection constraints of the spectrometer, the measured range is sufficient to validate a key prediction of simulations, that is, the fact that harmonics should be in phase in the relativistic regime.

Combining the spatial and temporal information obtained from our ptychographic measurements, we can reconstruct the spatio-temporal field of the attosecond pulses formed by the superposition of harmonics 9 to 14. This field is displayed in Fig. 4a for the ROM regime. Owing to the spatial curvature of the wavefronts imprinted by the curved plasma mirror surface, this optimally compressed attosecond pulse will get focused $\sim 150\,\mu m$ away from this surface. The combination of these temporal and spatial compressions leads to an intensity gain compared to the incident laser field. Determining the effective gain requires considering the field produced by the entire harmonic spectrum, and knowing the conversion efficiency of the laser energy to these harmonics. To this end, we use a PIC simulation (Supplementary Section 4) validated by our experimental results (Fig. 4a, side and bottom), and estimate that the intensity gain induced by the plasma mirror can reach a value of up to 8 in the physical conditions of our experiments.

**A path to the Schwinger limit**

The excellent spatio-temporal compression measured in our experiment suggests that considerably higher gains should be possible using incident laser fields with peak intensities of the order of $10^{24}\,W\,cm^{-2}$, as are now available from the emerging generation of petawatt lasers. At these much higher intensities, the involved physical processes—temporal compression and tight focusing—qualitatively remain the same as the ones observed in our experiment, but their effect can be made much stronger\textsuperscript{[16]}. As illustrated by the simulation results of Fig. 4b, shorter attosecond pulses ($\sim 100\,as$), associated with broader harmonic spectra, are generated with much higher conversion efficiencies, reaching 40% for the cumulation of all harmonic orders $\geq 2$ in the physical conditions of Fig. 4b. Owing to the larger size of the harmonic source, converging harmonic beams with larger numerical apertures are produced, which are thus focused to tighter focal spots. In the case of Fig. 4b, a $300\,nm$ focal spot is produced $60\,\mu m$ away from the plasma mirror surface, resulting in a total intensity gain of $10^8$ that brings the light intensity into the $10^{25}\,W\,cm^{-2}$ range. Different approaches can be used to further enhance the focusing by these highly controllable plasmas\textsuperscript{[22-49]} and thus bring the Schwinger limit within reach\textsuperscript{[9]}; such as creating the plasma mirror on a micro-fabricated curved target, using optically shaped plasmas\textsuperscript{[43,45]} or tailoring the laser wavefronts to control the harmonic beam divergence\textsuperscript{[52,56]}.

In all of these cases, the advanced measurement method described here will equally apply, enabling the accurate characterization and optimization of the spatio-temporal effects induced by relativistic plasma mirrors on the reflected field. All key experimental...
concepts and tools are available to pursue this challenging yet realistic path to the Schwinger limit based on plasma mirrors, which our results set as a new type of attosecond light source of high spatio-temporal quality. These results are also highly relevant for the field of ultrafast science, confirming experimentally that plasma mirrors can provide attosecond pulses for time-resolved experiments on electronic dynamics in matter.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-021-01253-9.

Received: 4 June 2020; Accepted: 26 April 2021; Published online: 3 June 2021

References

1. Marklund, M. & Shukla, P. K. Nonlinear collective effects in photon–photon and photon–plasma interactions. Rev. Mod. Phys. 78, 591–640 (2006).

2. Di Piazza, A., Müller, C., Hatae, S. & Keitel, C. H. Extremely high-intensity laser interactions with fundamental quantum systems. Rev. Mod. Phys. 84, 1177–1228 (2012).

3. Mourou, G. A., Tajima, T. & Bulanov, S. V. Optics in the relativistic regime. Rev. Mod. Phys. 78, 309–371 (2006).

4. Sauter, F. Über das Verhalten eines Elektrons im homogenen elektrischen Feld nach der Relativistischen Theorie Diracs. Z. Phys. 98, 714–732 (1936).

5. Schwingel, S. & Euler, H. Folgerungen aus der Diracschen Theorie des positrons. Z. Phys. 98, 749–764 (1935).

6. Heisenberg, W. & Euler, H. Folgerungen aus der Diracschen Theorie des positrons. Z. Phys. 98, 714–732 (1936).

7. Ringwald, A. Pair production from vacuum at the focus of an X-ray free electron laser. Phys. Lett. B 510, 107–116 (2001).

8. Strickland, D. & Mourou, G. Compression of amplified chirped optical pulses. Opt. Commun. 55, 447–449 (1985).

9. Bahk, S.-W. et al. Generation and characterization of the highest laser intensities (10^24 W/cm^2). Opt. Lett. 29, 2837–2839 (2004).

10. Gerstner, E. Extreme light. Nature 446, 16–18 (2007).

11. Yoon, J. W. et al. Achieving the laser intensity of 5.5×10^22 W/cm^2 with a wavefront-corrected multi-PW laser. Opt. Express 27, 20412–20420 (2019).

12. Landecker, K. Possibility of frequency multiplexing and wave amplification by means of some relativistic effects. Phys. Rev. 86, 852–855 (1952).

13. Bulanov, S. V., Esirkepov, T. & Tajima, T. Light intensification towards the Schwinger limit. Phys. Rev. Lett. 91, 085001 (2003).

14. Gordienko, S., Pukhov, A., Shirshov, O. & Baeva, T. Coherent focusing of high harmonics: a new way towards the extreme intensities. Phys. Rev. Lett. 104, 095003 (2005).

15. Gornakov, A. A., Korzhimanov, A. V., Kim, A. V., Marklund, M. & Sergeev, A. M. Ultrarelativistic nanoplasmonics as a route towards extremely-intensity attosecond pulses. Phys. Rev. E 84, 046403 (2011).

16. Vincenti, H. Achieving extreme light intensities using optically curved relativistic plasma mirrors. Phys. Rev. Lett. 123, 105001 (2019).

17. Solodov, A., Malkin, V. & Fisch, N. Limits for light intensification by reflection from relativistic plasma mirrors. Phys. Plasmas 13, 093102 (2006).

18. Kapteyn, H. C., Murnane, M. M., Zozou, A. & Falcone, R. W. Prepulse energy suppression for high-energy ultrashort pulses using self-induced plasma scattering. Opt. Lett. 16, 490–492 (1991).

19. Thaury, C. et al. Plasma mirrors for ultrahigh-intensity optics. Nat. Phys. 3, 424–429 (2007).

20. Lichters, R., Meyer-ter Vehn, J. & Pukhov, A. Short-pulse laser harmonics from oscillating plasma surfaces driven at relativistic intensity. Phys. Plasmas 3, 3425–3437 (1996).

21. Thaury, C. & Quéré, F. High-order harmonic and attosecond pulse generation on plasma mirrors: basic mechanisms. J. Phys. B 43, 213001 (2010).

22. Baeva, T., Gordenko, S. & Pukhov, A. Theory of high-order harmonic generation in relativistic laser interaction with overdense plasma. Phys. Rev. E 74, 046404 (2006).

23. Dromey, B. et al. High harmonic generation in the relativistic limit. Nat. Phys. 2, 456–459 (2006).

24. Krausz, F. & Ivanov, M. Attosecond physics. Rev. Mod. Phys. 81, 163–234 (2009).

25. Quéré, F., Mairesse, Y. & Itatani, J. Temporal characterization of attosecond XUV fields. J. Mod. Opt. 52, 339–360 (2005).

26. Orfano, L. et al. Attosecond pulse metrology. APL Photon. 4, 080901 (2019).

27. Nomura, Y. et al. Attosecond phase locking of harmonics emitted from laser-produced plasmas. Nat. Phys. 5, 124–128 (2009).

28. Quéré, F. Attosecond plasma optics. Nat. Phys. 5, 93–94 (2009).

29. Rodenburg, J. M. Psychography and related diffusive imaging methods. Adv. Imaging Electron Phys. 150, 87–184 (2008).

30. Thibault, P. et al. High-resolution scanning X-ray diffraction microscopy. Science 321, 379–382 (2008).

31. Kahaly, S. et al. Direct observation of density-gradient effects in harmonic generation from plasma mirrors. Phys. Rev. Lett. 110, 175001 (2013).

32. Quéré, F. et al. Coherent wake emission of high-order harmonics from overdense plasmas. Phys. Rev. Lett. 96, 125004 (2006).

33. Dudovich, N. et al. Measuring and controlling the birth of attosecond XUV pulses. Nat. Phys. 2, 781–786 (2006).

34. Kim, K. T. et al. Manipulation of quantum paths for space–time characterization of attosecond pulses. Nat. Phys. 9, 159–163 (2013).

35. Vampa, G. et al. Linking high harmonics from gases and solids. Nature 522, 462–464 (2015).

36. Quéré, F. et al. Phase properties of laser high-order harmonics generated on plasma mirrors. Phys. Rev. Lett. 100, 095004 (2008).

37. Dromey, B. et al. Diffraction-limited performance and focusing of high harmonics from relativistic plasmas. Nat. Phys. 5, 146–152 (2009).

38. Vincenti, H. et al. Optical properties of relativistic plasma mirrors. Nat. Commun. 5, 3403 (2014).

39. Malvache, A., Borot, A., Quéré, F. & Lopez-Martens, R. Coherent wake emission spectroscopy as a probe of steep plasma density profiles. Phys. Rev. E 87, 035101 (2013).

40. Leblanc, A., Monchoçé, S., Bourassou-Bouchet, C., Kahaly, S. & Quéré, F. Psychographic measurements of ultrahigh-intensity laser–plasma interactions. Nat. Phys. 12, 301–305 (2016).

41. Leblanc, A. et al. Spatial properties of high-order harmonic beams from plasma mirrors: a psychophysical study. Phys. Rev. Lett. 119, 155001 (2017).

42. Thaury, C. et al. Coherent dynamics of plasma mirrors. Nat. Phys. 4, 631–634 (2008).

43. Wheeler, J. A. et al. Attosecond lighthouses from plasma mirrors. Nat. Photon. 6, 829–833 (2012).

44. Monchoçé, S. et al. Optically controlled solid-density transient plasma gratings. Phys. Rev. Lett. 112, 145008 (2014).

45. Leblanc, A. et al. Plasma holograms for ultrahigh-intensity optics. Nat. Phys. 13, 440–443 (2017).

46. Denouël, A., Chopineau, L., Leblanc, A. & Quéré, F. Interaction of ultraintense laser vortices with plasma mirrors. Phys. Rev. Lett. 118, 033902 (2017).

47. Yeung, M. et al. Experimental observation of attosecond control over relativistic electron bunches with two-colour fields. Nat. Photon. 11, 32–35 (2017).

48. Gao, J. et al. Divergence control of relativistic harmonics by an optically shaped plasma surface. Phys. Rev. E 101, 033202 (2020).

49. Quéré, F. & Vincenti, H. Reflecting petawatt lasers off relativistic plasma mirrors: a realistic path to the Schwinger limit. High Power Laser Sci. Eng. 9, e6 (2021).

50. Quintard, L. et al. Optics-less focusing of XUV high-order harmonics. Science 365, eaau7175 (2019).

51. Maroju, P. et al. Attosecond pulse shaping using a seeded free-electron laser. Nature 578, 386–391 (2020).
Data availability
The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Acknowledgements
We thank F. Réau, C. Pothier and D. Garzella for operating the UHI100 laser. The research received financial support from the European Research Council, LASERLAB-EUROPE and CREMLINplus (grants 694596, 871124 and 871072, European Union Horizon 2020 Research and Innovation Programme), from Investissements d’Avenir LabEx PALM (ANR-10-LABX-0038-PALM) and from Agence Nationale de la Recherche (ANR-18-ERC2-0002). An award of computer time was provided by the INCITE programme (project ‘PlasmInSilico’). This research used resources of the Argonne Leadership Computing Facility, which is a DOE Office of Science User Facility supported under contract DE-AC02-06CH11357. We also acknowledge the financial support of the Cross-Disciplinary Program on Numerical Simulation of CEA (Commissariat à l’Energie Atomique et aux énergies alternatives).

Author contributions
F.Q. conceived the experiment, and F.Q. and A.D. conceived the experimental set-up. A.D. and L.C. performed the experiment with the help of E.P. The data analysis was carried out by L.C. with the help of A.L. H.V. performed all numerical simulations. F.Q. was in charge of the manuscript, to which all authors contributed.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-021-01253-9.

Correspondence and requests for materials should be addressed to H.V. or F.Q.

Peer review information Nature Physics thanks Zhengming Sheng and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

Reprints and permissions information is available at www.nature.com/reprints.