

Laser-driven proton acceleration beyond 100 MeV by radiation pressure and Coulomb repulsion in a conduction-restricted plasma

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An ultrahigh-intensity femtosecond laser can establish a longitudinal electric field stronger than 10^{13} Vm^{-1} within a plasma, accelerating particles potentially to GeV over a sub-millimetre distance. Laser-accelerated protons with high brightness and picosecond duration are highly desired for applications including proton imaging and flash radiotherapy, while a major limitation is the relatively low proton energy achieved yet, primarily due to the lack of a controllable acceleration structure. Here, we report the generation of protons with a cutoff energy exceeding 110 MeV, achieved by irradiating a multi-petawatt femtosecond laser on a conduction-restricted nanometre polymer foil with a finite lateral size. The enduring obstacles in achieving ultrahigh laser contrast and excellent laser pointing accuracy were successfully overcome, allowing the effective utilization of size-reduced nanometre foils. A long acceleration structure could be maintained in such a quasi-isolated foil since the conduction of cold electrons was restricted and a strong Coulomb field was established by carbon ions. Our achievement paves the road to enhance proton energy further, well meeting the requirements for applications, through a controllable acceleration process using well-designed nano- or micro-structured targets.

Plasma without a breakdown threshold can support an acceleration gradient six orders of magnitude higher than that in conventional accelerators¹. When a driving laser propagates inside a gas-density plasma, its wakefield can form a well-controlled bubble structure, resulting in the successful acceleration of electrons to around 10 GeV on a decimetre scale^{2,3}. For the acceleration of protons with a much larger mass-to-charge ratio, solid-density plasmas are usually utilized as they can support a stronger electric field^{4,5}. The accelerated protons

with ultralow transverse emittance and picosecond duration have been applied for the production of high-energy-density matter⁶, measurement of transient electromagnetic field⁷, and flash radiotherapy of cancer at an ultrahigh dose rate⁸. However, limitations, including the lack of a long acceleration structure⁹, deflection of protons by the self-generated magnetic field¹⁰, and dephasing of the fastest protons during acceleration¹¹, lead to a proton energy below 100 MeV in experiments^{12–14} except one very recent work reporting the

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generation of 150-MeV protons¹⁵. Improving the energy of laser-accelerated protons is thus essential for applications, especially for the instantaneous radiography of dense objects and clinical therapy of deep-seated tumours¹⁶.

To increase the proton energy, a series of efforts has been made in the past two decades¹. Among them, the utilization of structured targets has been intensively studied, and several effective designs have been demonstrated. A straightforward solution is modifying the target front surface to enhance the laser-plasma coupling, and different coating structures, including surface grating¹⁷, carbon nanotube foam¹⁸, and arrays of microwire¹⁹ or microtube²⁰, have been attempted. Another method is modulating the acceleration field in the target rear surface by adopting hydrogen-rich micro-dots²¹, hemispherical pits²², or helical coils²³. Besides, a number of experiments have employed mass-limited targets to improve the proton quality, which can be roughly categorized into two schemes. The first utilizes isolated high-repetition targets, like microspheres^{24–26}, and cryogenic hydrogen jets²⁷, where protons are accelerated mainly via the mechanism of collisionless shock acceleration²⁴ or Coulomb explosion^{25,26}. The second scheme uses foils with a reduced transverse size to enhance the target normal sheath acceleration (TNSA) of protons, since the laterally spreading electrons would be reflected around the foil edges^{28,29} and reaccelerated to higher energies if a sub-picosecond driving laser^{10,30–34} was employed. Several experiments have also tried a femtosecond driving laser with a relatively low power of less than 200 TW, demonstrating a robust enhancement of proton energy^{35–37}.

The major challenge in further improving the proton energy by irradiating a petawatt femtosecond laser on structured targets is to preserve the nano- or micro-structures before the arrival of the main pulse. High temporal contrast of laser is a prerequisite for such experiments since the intensity of prepulse should be suppressed below the damage threshold of the targets, which is relatively low due to the structures³⁸. Another difficulty is to maintain an excellent laser pointing accuracy within a few microns²⁸, which is crucial to precisely align the focal spot on the structures of targets. Moreover, since the peak intensity of a multi-petawatt laser can be orders of magnitude higher than that of terawatt pulses, proton acceleration may occur via a novel and complex mechanism, and a proper understanding of the acceleration process is critical for the optimization of laser parameters and the design of target structures.

In this article, we demonstrate an enhanced acceleration of protons from a structured nanometre-thick polymer foil driven by a multi-petawatt femtosecond laser. The temporal contrast and pointing stability of the petawatt laser were well controlled, and the transverse shape of the polymer foils was tailored with a 100-kHz, μ J femtosecond laser. From a 100-nm-thick, 30- μ m-wide foil, 110-MeV protons were successfully measured by several detection methods and unambiguously confirmed by the CR-39 plate in a stack detector. With the help of two- and three-dimensional (3D) particle-in-cell (PIC) simulations, we revealed that such energetic protons were accelerated by a hybrid mechanism of radiation pressure and Coulomb repulsion, the latter of which was enhanced in laser-tailored foils by suppressing the conduction of cold electrons. Our experimental result, thus, clearly demonstrated the enhancement of proton energy by employing conduction-restricted nanometre targets. Furthermore, our work indicates that the current technology is ready to support the desired interaction of a petawatt femtosecond driving laser with structured foils, and further improving the proton quality can be realized by employing well-designed targets with controllable acceleration structures.

Results

We utilized the CoReLS 4-PW Ti:Sapphire laser with 32-J energy and 24-fs duration on the target as the driver of protons. To obtain an ultraclean laser pulse, techniques including the cross-polarized wave, optical parametric chirped-pulse amplification, and double plasma mirror were employed, achieving a temporal contrast higher than 10^{12} up to 2 ps before the main pulse³⁹. The pointing stability of the focal spot was also improved to a standard deviation of several microns (Supplementary Fig. 3) by reducing the vibration of optical components and aligning the whole surface of plasma mirrors. For the targets, we chose 100-nm-thick polymer (F8BT) foils with relatively high mechanical and optical damage thresholds, whose size and shape were tailored by ablating the needless region using a 100-kHz, μ J femtosecond laser to restrict the conduction of surface current. As displayed in Fig. 1, electrons and protons, accelerated from a conduction-restricted foil irradiated by a petawatt laser, were detected online by an electron spectrometer and two Thomson parabola spectrometers (TPSSs), respectively. After the optimization of the laser and target parameters according to the results of TPSSs, a stacked detector was inserted to

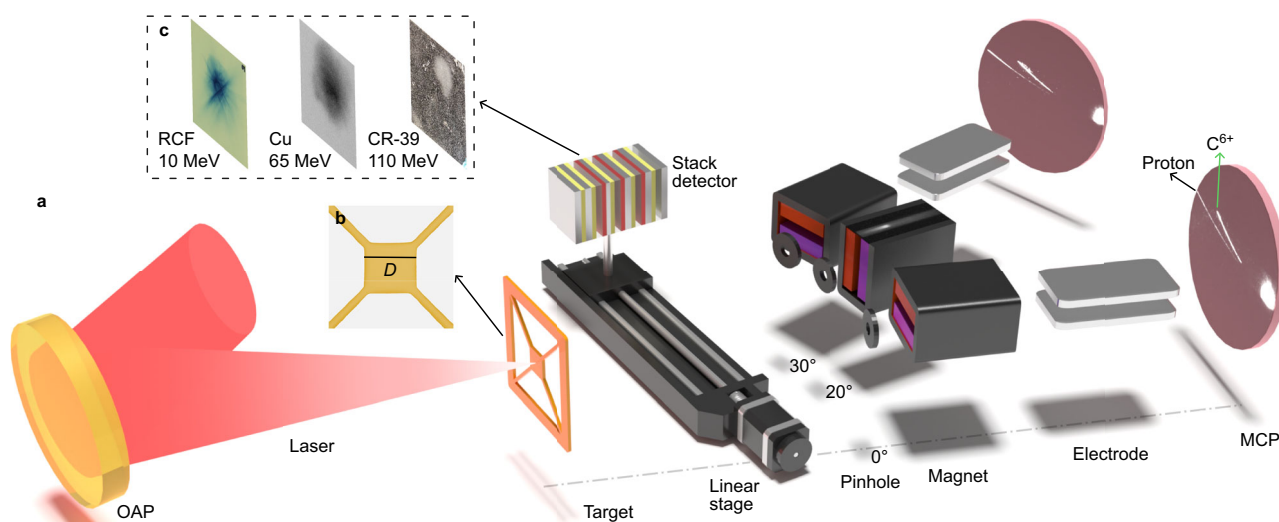


Fig. 1 | Layout of the proton acceleration experiment. **a** An ultrahigh-contrast, multi-petawatt laser irradiated on the centre of a conduction-restricted nanometre foil at an incident angle of 20° to accelerate protons within tens of microns. The accelerated protons at 0° and 30° to the laser axis were measured online by two Thomson parabola spectrometers. An electron spectrometer was installed in the normal

direction of the target rear surface (20°). **b** Microscope photo of a conduction-restricted square foil with a transverse width D of 80 μ m. **c** For the selective shot, one stack detector, composed of aluminium, copper, and CR-39 plates as well as radiochromic films (RCFs), was inserted to measure the spatial profile and spectrum of the protons simultaneously. The insets are results from a 30- μ m-wide foil.

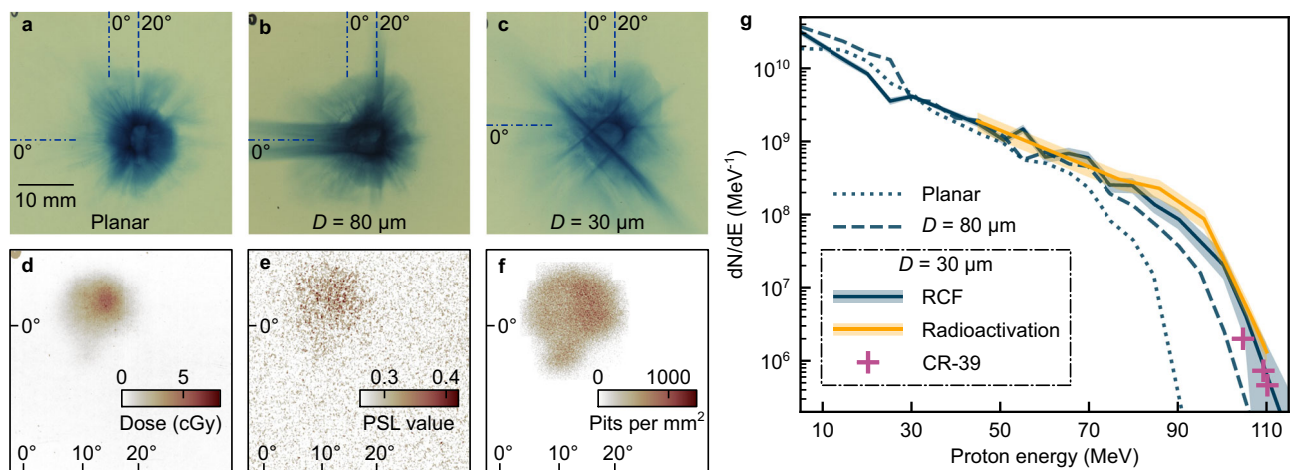


Fig. 2 | Spatial profile and spectrum of accelerated protons. a–c Raw data of the first (Bragg peak of 10-MeV protons) RCF using 100-nm-thick foil targets: planar (a), $D = 80 \mu\text{m}$ (b), and $D = 30 \mu\text{m}$ (c). D is the transverse width of a foil, and 0° is the laser propagation direction. **d–f** Signals of protons with an energy around 110 MeV measured by RCF, radioactivation, and CR-39, respectively, corresponding to the layer numbers 55, 53 and 56 in Supplementary Table 1. A $D = 30 \mu\text{m}$ foil was utilized,

and the electron signals in the RCF were negligible in this laser shot. **g** Unfolded proton spectra of the three targets obtained from the RCF measurement. For $D = 30 \mu\text{m}$, the results from radioactivation and CR-39 are also shown together. Both the front and rear surfaces of the last CR-39 plate were analysed to deduce the spectral intensities at approximately 110 MeV. The shaded regions indicate the uncertainties induced from the calibrations of RCF and imaging plate (IP).

simultaneously measure the spatial profile and spectrum of the protons⁴⁰. Details of the laser manipulation, target fabrication, and data analyses are given in Methods.

Generation of over-110 MeV protons

An enhancement of the proton cutoff energy was investigated by employing conduction-restricted foils with the structure shown in Fig. 1b. The energy-dependent spatial profiles of the protons were obtained directly from the RCFs in a single-shot stack measurement (Supplementary Fig. 13). Different from the case of planar targets, patterns caused by protons with large divergence angles were observed on the first RCF when the conduction-restricted foils were used (Fig. 2a–c), consistent with previous works³⁶. These patterns, induced by protons accelerated from the edges (Fig. 2b) or supporting stalks (Fig. 2c) of the conduction-restricted foils, imply that the laser contrast in our experiment has been improved enough to ensure the survival of the foil structure before the arrival of the main pulse. The asymmetric profiles of these patterns indicate that the laser focal spot was not perfectly aligned on the foil centre, due to the shot-to-shot fluctuation of laser pointing. Despite this, the acceleration of over-110 MeV protons from a 30- μm -wide foil was realized, as depicted in Fig. 2d–f. The proton profiles measured by the RCF, radioactivation, and CR-39 plate agreed well with each other, manifesting the emission of high-energy protons at angles between the laser axis and the target normal direction. It should be noted that the result from CR-39 can provide a faultless proton cutoff energy, since RCF is also sensitive to electrons, and gamma-rays can introduce noises on the radioactivation measurement⁴⁰. For instance, one may measure signals in the RCF with a Bragg-peak energy of 120 MeV, which are induced by electrons in the laser direction according to the CR-39 result (Supplementary Fig. 19).

Apart from the spatial profiles, the spectra of the protons were also obtained from the stack results by a differential evolution algorithm (details in Methods). The proton spectra generated from the RCF and the radioactivation measurements were consistent and confirmed by the CR-39 data. A decrease in spectral intensity with proton energy was observed for all the spectra in Fig. 2g, and the cutoff energies for 80- μm - and 30- μm -wide foils were 105 and 112 MeV, respectively, 17% and 24% higher than that of 90 MeV for planar targets. The number of protons with energies higher than 70 MeV from the 30- μm -wide foil reached 5×10^9 , five times greater than that from the planar target.

While the total number of $>5 \text{ MeV}$ protons was 4×10^{11} for both cases, showing no enhancement, which is different from the previous results using terawatt lasers^{33,36}. This is because our petawatt laser pulse is strong enough to accelerate all the protons around the focal spot: 4×10^{11} corresponds to the total number of protons resided in the target with a radius of $4.8 \mu\text{m}$, larger than the waist width of the focal spot. For the same reason, the energy conversion efficiency from the laser to protons only slightly increased from 3.6% (planar) to 3.7% ($D = 30 \mu\text{m}$).

The enhanced proton acceleration was also verified by the multi-shot results of the two TPSs (Fig. 3a, b). The data from the TPSs were consistent with those from the RCFs in the stack detector, showing a higher proton cutoff energy E_{cut} for the conduction-restricted foils. For the 30- μm -wide foils, the averaged E_{cut} at 0° was 100 MeV, enhanced by 19% compared to the value of 84 MeV from the planar foils. A larger E_{cut} was measured at 0° rather than 30° for the majority of the shots, since the most energetic protons were emitted at around 10° with respect to the laser axis (Fig. 2f). A robust modulation of the electron spectra, showing a peak at 5 MeV, was observed with the conduction-restricted targets (Fig. 3c), distinctive from the Boltzmann distribution obtained from the planar targets. This is another characteristic in addition to the proton patterns implying the use of a conduction-restricted foil.

Simulation of proton acceleration

We performed PIC simulations to investigate the acceleration mechanism of protons under our novel laser and target conditions, and to understand the source of the energy enhancement in conduction-restricted foils. The parameters of the laser and plasma were set according to the experimental conditions, and the detailed setup can be found in Supplementary Sec. C. Considering the 100- μm simulation domain and the 200-fs laser rising edge, and as a simple scheme to understand the physical picture, we first utilized 2D simulations to compare the two cases of planar and $D = 30 \mu\text{m}$ foils, as shown in Fig. 4a–f. In the conduction-restricted foil, the electrons spreading in the transverse direction can be pulled back around the edges of the foil and deflected to the laser direction by the magnetic field near the plasma surface¹⁰, resulting in the higher electron energy density at the centre of the foil shown in Fig. 4a. As a result, more sub-10 MeV electrons move forward for the $D = 30 \mu\text{m}$ foil, accounting for

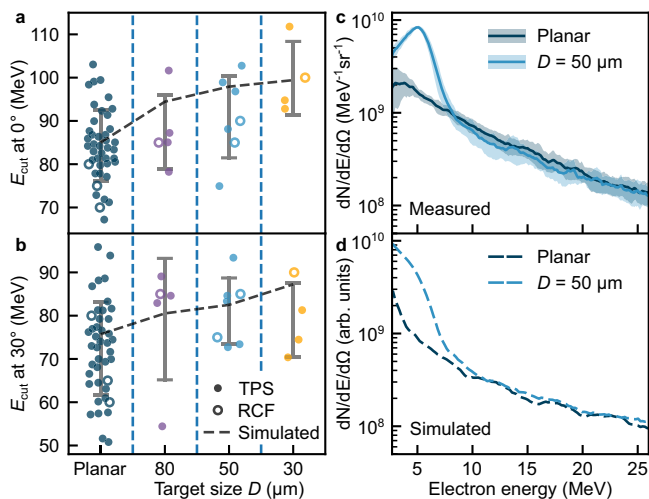


Fig. 3 | Scan of the target size. **a, b** Proton cutoff energy E_{cut} at 0° and 30°, respectively. The results of a total of 58 shots are plotted and the error bars indicate the standard deviation. The dashed lines were obtained from 2D PIC simulations, and a factor of 0.6 was multiplied to correct the dimensional effect. **c, d** Measured and simulated electron spectra at 20° from various targets, respectively. The shaded regions in **c** represent the standard errors of 10 (planar) and 5 ($D = 50 \mu\text{m}$) laser shots.

the 5-MeV peak in the measured electron spectra (Fig. 3c). These confined electrons, however, only have a limited contribution to the acceleration of the most energetic protons, as they have left behind these protons (Fig. 4c). The enhanced longitudinal electric field is mainly caused by the absence of cold electron refluxing in the conduction-restricted foil (weak J_y in Fig. 4c), decreasing the shielding effect of electrons on the Coulomb field established by the remaining carbon ions. A more explicit relationship can be found in Fig. 4e and f, indicating a close connection between the energy enhancement and the larger net charge for the conduction-restricted foil. As displayed in Fig. 5, the high-energy protons experience a hybrid mechanism of radiation pressure acceleration (RPA) during the laser irradiation and Coulomb repulsion (CR) after the end of the laser pulse. The contribution of the latter increases as the target size decreases, enhancing the proton cutoff energy as shown in Fig. 2g.

We also carried out a 3D simulation for the case of a $D = 30 \mu\text{m}$ foil to correct the underestimated electron spreading in 2D simulations. The separation of protons and carbons is pronounced in Fig. 4g, indicating the effects of carbon ions on the proton acceleration via Coulomb field. Based on a Coulomb repulsion model and using the parameters obtained in the 3D simulation, we can calculate the dependence of the maximum proton energy on the target size, matching well with that measured in experiments (Fig. 4h). Different from 2D simulations, a quantitative spectrum of all the forward protons can be derived here, which shows a reasonable fit compared to the measured spectrum (Fig. 4i), implying the reliability of the laser-plasma interaction in the 3D simulation.

Driven by a multi-petawatt femtosecond laser, the protons in our experiment experience a novel acceleration process compared to the previous works using size-reduced targets^{30–37}. Here, the key difference is the scaling factor of RPA as $a_0 \lambda_i / nd$, where λ_i , a_0 , d and n are the laser wavelength, normalized vector potential, target thickness, and normalized electron density, respectively⁴. We note that all the earlier experiments^{30–37} were performed at the condition of $a_0 \lambda_i / nd < 0.13$, and the protons are mainly accelerated by the sheath field at the target rear. The cutoff energy is determined mainly by the temperature and density of hot electrons⁵; both can be improved in size-reduced targets if the laser duration τ is larger than the electron recycling period D/c between the target centre and edges²⁸, where c is the speed of light in

vacuum. Hence, sub-picosecond driving laser pulses and μm -thick metal foils were usually utilized in these experiments. While in our experiment using 100-nm-thick conduction-restricted foils, the laser with $a_0 \approx 37$, satisfying $a_0 \lambda_i / nd > 1$, is strong enough to push the bulk of electrons forward, realizing a proton acceleration dominated by radiation pressure, instead of thermal pressure. Then after the end of the femtosecond laser, the electrostatic pressure stored in the heavy carbon ions, which increases with the decrease in foil size, governs the subsequent acceleration of protons.

Discussion

In our experiment, we utilized an ultraclean petawatt femtosecond laser and a conduction-restricted nanometre foil to accelerate protons. This is a novel approach to realizing laser-driven proton acceleration beyond 100 MeV, with a specific design of target and laser parameters, as well as a distinct acceleration mechanism, in comparison to those in the recent work¹⁵ by T. Ziegler et al. In particular, one critical difference can be found in the strict requirement of laser contrast in our experiment to ensure the survival of the initial target structure at the arrival of the main pulse. As a result, the high-energy protons are accelerated by the combination of RPA and Coulomb repulsion without the onset of relativistically induced transparency¹⁵. One main advantage of our scheme is the robust acceleration structure. Owing to the ultrahigh laser contrast, the main pulse can directly interact with a solid-density plasma, avoiding the hosing and filamentation instabilities in long preplasmas, which are primary sources of the shot-to-shot fluctuations in acceleration fields. Besides, the short laser duration (24 fs) can suppress the instabilities in RPA, and the Coulomb field established by heavy carbon ions is not so sensitive to variations of electron temperatures. Hence, we measured a stable proton acceleration, despite adjusting the shape and orientation of the foils. As displayed in Fig. 6, the cutoff energies of the protons at 0° and 30° consistently exceeded 85 and 80 MeV, respectively. Apart from protons, carbon ions were simultaneously accelerated to approximately 40 MeV u^{-1} from the $D = 30 \mu\text{m}$ foils (Supplementary Figs. 11 and 18). Compared to other isolated targets such as microsphere^{24–26} and microtube²⁷, the conduction-restricted foil also has the advantage of a large aspect ratio, ensuring a planar Coulomb repulsion field instead of an isotropic one. Hence, a small divergence angle of the high-energy protons can be maintained (Fig. 2f), which is crucial for the collection and application of protons.

Based on the demonstrated acceleration of protons to over 110 MeV from a $D = 30 \mu\text{m}$ foil, we may further improve the proton quality by advancing the target design. Using foils with a smaller D can enhance the proton cutoff energy, as predicted in Fig. 4h, if we further optimize the laser pointing stability. Employing thicker foils and stronger lasers may efficiently increase the proton energy, which requires a higher laser contrast with the better control on the spatio-temporal coupling of laser pulses and the surface roughness of mirrors⁴¹. Here, we emphasize that our scheme is sensitive to the preheating and expansion of the ultrathin foil, since both the RPA and CR require a solid-density plasma to support the acceleration field. As evidence, only low-energy protons without apparent enhancement were observed in the case of a 30-nm-thick foil (Fig. 7), due to the lower plasma density after the expansion of the foil. Besides the proton energy, some applications also pursue quasi-monoenergetic spectra⁴², which may be realized by a double-layer target consisting of a substrate and a proton-rich coating (Supplementary Fig. 26). For the material of the substrate, diamond-like carbon is more suitable than metal due to its higher damage threshold. One limitation of the current scheme is the available repetition rate at Hz scale, while a kHz source may be achieved using a liquid sheet target and double laser pulses. A transient micro-foil is first obtained by tailoring the plain sheet with a Laguerre-Gaussian prepulse (Supplementary Fig. 27), and then the protons are accelerated by the main laser pulse⁴³.

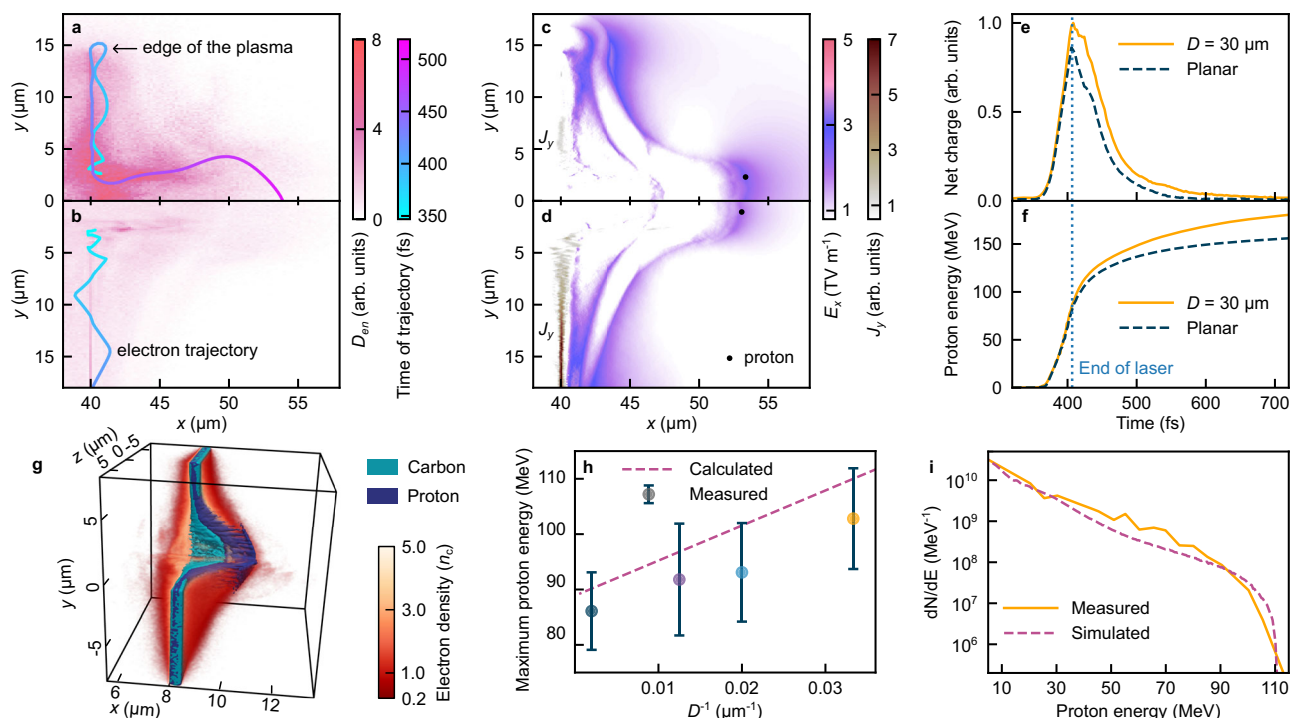


Fig. 4 | Enhanced proton acceleration in conduction-restricted foils.

a, b Distributions of the electron energy density D_{en} at the simulation time of 520 fs, as well as the trajectories of two representative electrons evolved with time. **c, d** Distributions of the longitudinal acceleration field E_x and transverse current density J_y at the simulation time of 480 fs. J_y is zoomed in by four times along the x axis to indicate the details. The dark dots represent the positions of the most energetic protons. **a** and **c** are the results of a $D = 30 \mu\text{m}$ foil, while **b** and **d** are those of a planar target, all with an initial foil position of $x = 40 \mu\text{m}$. **e, f** Temporal evolution of the net charge and of the maximum proton energy, respectively. The total

charge in the region of $38 \mu\text{m} < x < 43 \mu\text{m}$ and $|y| < 15 \mu\text{m}$ is considered.

g Isosurfaces ($1.5n_c$) of the carbon and proton densities, as well as the density distribution of electrons at the simulation time of 100 fs. Here, the critical density $n_c = 1.74 \times 10^{27} \text{ m}^{-3}$ for the 800-nm laser. **h** Dependence of the maximum proton energy on the target size. The planar targets are treated as $D = 500 \mu\text{m}$ and the error bars indicate the standard deviation. The dashed line shows the calculated proton energy in the Coulomb repulsion model. **i** Comparison of the measured and simulated proton spectra using a $D = 30 \mu\text{m}$ foil. **a–f** are obtained from 2D PIC simulations, while **g–i** are based on a 3D simulation.

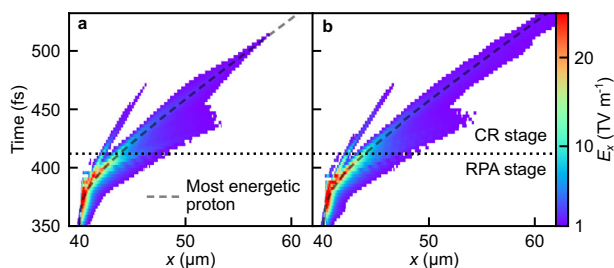


Fig. 5 | Comparison of the acceleration fields. a, b Evolution of the longitudinal electric fields E_x in planar and $D = 30 \mu\text{m}$ foils, respectively. The average E_x in the region of $0 < y < 2 \mu\text{m}$ is plotted, and the dotted lines indicate the trajectory of the most energetic proton. The acceleration stages are labelled as the time goes.

Our experiment reveals that current technologies have been developed to support the usage of multi-petawatt femtosecond lasers with sufficient temporal contrast and pointing stability for structured nanometre foils. Such powerful lasers are promising to further realize a controllable and ideal proton acceleration in well-designed 3D-structured targets, which can already be routinely fabricated through e-beam lithography⁴⁴ or 3D print based on the two-photon polymerization technique⁴⁵. Moreover, driven by a multi-petawatt femtosecond laser, unprecedented electric and magnetic fields with designed configurations can be established inside the 3D-structured plasma, providing a compact platform for the study of high-energy-density physics and laboratory astrophysics, such as magnetic reconnection⁴⁶, resonant x-ray emission⁴⁷ and gamma-ray burst⁴⁸.

Methods

Laser system. The 4-PW Ti:sapphire laser at the Center for Relativistic Laser Science (CoReLS) was utilized to accelerate protons. In the front end, a cross-polarized wave (XPW) system and an optical parametric chirped-pulse amplification (OPCPA) stage were employed. The compression gratings were carefully aligned to reduce the rising edge and spatio-temporal coupling of the laser. After compression, the laser contrast was further enhanced by more than five orders of magnitude in a double plasma mirror (DPM) system³⁹. In the target chamber, the cleaned p-polarized laser pulse was focused by an $f/2.8$ off-axis parabolic mirror (OAP) on a target of a polymer foil at an incidence angle of 20° . The on-target laser energy and duration were $32 \pm 2 \text{ J}$ and $24 \pm 2 \text{ fs}$, respectively. Using a random-pinhole attenuator to attenuate the full-energy laser after the triggering of the DPM, we measured the full width at half maximum (FWHM) of the focal spot as $3.6 \pm 0.6 \mu\text{m}$, with the energy concentration of 22% in the FWHM (Supplementary Fig. 1). Hence, the laser peak intensity I was about $3 \times 10^{21} \text{ Wcm}^{-2}$, corresponding to a normalized vector potential of $a_0 \approx 37$. Here $a_0 = 8.5 \times 10^{-10} \sqrt{I(\text{Wcm}^{-2})\lambda_L(\mu\text{m})}$. To finely match the focal spot and the size-reduced foil, the laser pointing stability was improved by reducing the vibration of the optical components. Moreover, before every full-energy shot, one test shot using an attenuated laser with μJ energy after the DPM was first performed, and the position of the focal spot was confirmed according to the tiny damaged hole on the foil.

Conduction-restricted target. We chose nanometre-thick polymer (F8BT, $\text{C}_{35}\text{H}_{42}\text{N}_2\text{S}$) foil as the target due to its high damage threshold exceeding 10^{12} Wcm^{-2} for femtosecond prepulse. The foils were fabricated through spin coating, with the thicknesses and mass density

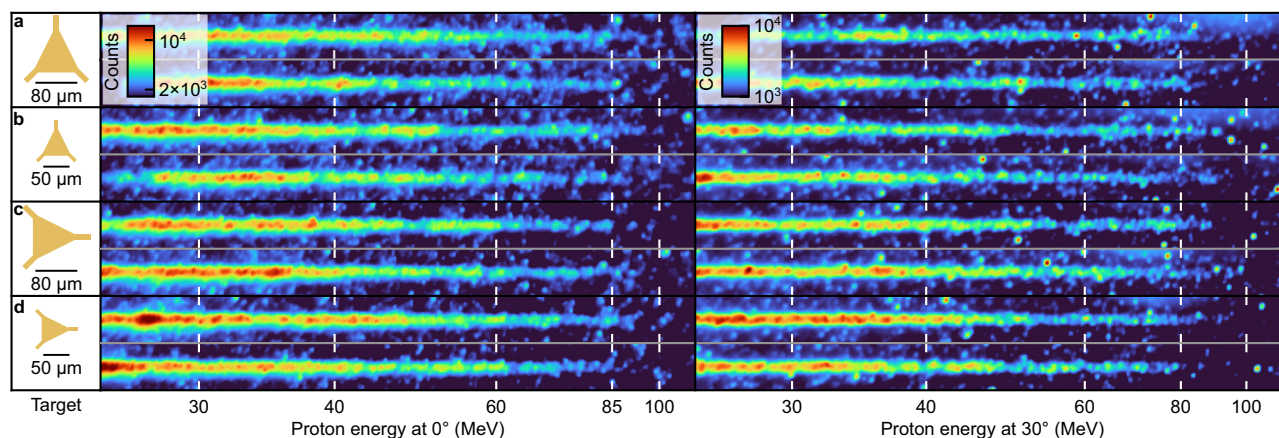


Fig. 6 | Fluctuation of the protons. a–d Proton spectra measured by the TPSs at 0° and 30° from eight consecutive shots using 100-nm-thick triangular foils with various sizes and orientations.

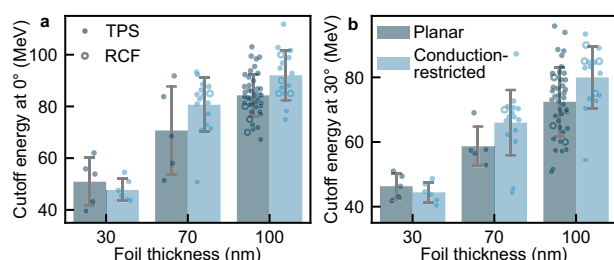


Fig. 7 | Influence of the target thickness. a, b Cutoff energies of the protons at 0° and 30° to the laser direction, respectively. The results of $D = 30$, 50, and 80 μm targets with the same thickness are plotted together. The error bars show the standard deviation.

($1.2\text{ g}\cdot\text{cm}^{-3}$) calibrated by the X-ray reflectivity measurement. The plasma density is $210n_c$ if all the atoms are ionized. We utilized a 100-kHz, μJ , 250-fs laser to tailor the free-standing foil on the target holder, whose transverse size and shape could be simply modified by adjusting the movement of the laser focus on the foil surface. For the square target, four stalks with widths of $<15\mu\text{m}$ remained to support the conduction-restricted foil (Supplementary Fig. 4). Thanks to the isolated structure preventing the propagation of surface shock, a higher survival rate of nearby targets was observed after a laser shot on a conduction-restricted foil.

Diagnostics. The accelerated electrons and protons were measured online by one electron spectrometer (ES) and two TPSs, respectively. The ES, composed of a 0.6-T magnet to deflect the electrons and a phosphor screen (Lanex Fast B) to record the signals (Supplementary Fig. 10), was installed inside the vacuum chamber. A microchannel plate (MCP) and a charge-coupled device (CCD) were equipped in the TPS to detect the trajectory of the protons and carbon ions. Quantitative spectra of proton and carbon ions were obtained according to previous calibrations⁴⁰. The main limitations of TPSs are the tiny acceptance angle (Supplementary Table 3) and the low resolution in the high-energy region. Hence, a stack detector is essential to accurately characterize the accelerated protons.

The stack detector, composed of aluminium, copper, CR-39 and RCF, was prepared with the help of the Monte Carlo code FLUKA⁴⁹. In the stack (Supplementary Table 1), twenty-one pieces of RCFs (Gafchromic) were employed with the proton Bragg peaks equally separated by 5 MeV. To eliminate the potential noise on the RCFs caused by laser-accelerated electrons⁴⁰, seven copper plates were used for the radioactivation measurement. Considering that secondary gamma-rays and neutrons can also radioactivate copper, we assembled seven

pieces of CR-39 (Track Analysis Systems Ltd.) to unambiguously verify the proton cutoff energy at the expense of a low saturation fluence.

During the experiment, the prepared stack, installed on a linear stage, was inserted before the selected laser shot and aligned parallel to the target rear surface with a gap of 14 mm. After the shot, we quickly removed the stack from the vacuum chamber and placed the copper plates on a BAS-MS imaging plate (Fujifilm). After a 20-min exposure, the IP was scanned with a resolution of 100 μm , a dynamic range of $L = 5$, and a sensitivity of $S = 4000$. The RCFs, after waiting for longer than 24 h, were also scanned by a 48-bit colour scanner (Perfection V750 Pro, Epson) in transmission mode. The CR-39 plates were first etched for 4 h in a 6.25N NaOH solution at 70 °C, and then scanned by an upright microscope (BX53M, Olympus) to record the pits on the front and rear surfaces.

To determine the proton spectra from the stack data, we applied a differential evolution algorithm to find the best fit between the calculated values from the assumed spectra and the measured values, including the total deposited energy (TDE) in each RCF or photo-stimulated luminescence (PSL) value of every IP. The deposited energy curves of the RCFs and the nuclear reaction yields of the copper plates were obtained from the FLUKA simulations (Supplementary Figs. 6 and 7) and utilized in the calculations. The measured TDE and PSL values were calculated from the optical densities (OD) in the red channel of the RCFs and the count values of the IPs, respectively. Here, uncertainties were introduced from the calibrations of RCF and IP we utilized⁴⁰, which have been included in Fig. 2g. The spectral intensities of the protons were also deduced from the front and rear surfaces of the last two CR-39 plates, demonstrating good consistency with those measured by the RCF and radioactivation methods. Note that due to the scattering effect, protons with the same initial energy will have a broadened energy distribution when they reach the CR-39 surface (Supplementary Fig. 8), which should be considered during the calculation.

Particle-in-cell simulation. To investigate the underlying physics in proton acceleration, we performed PIC simulations using the code Smilei⁵⁰. For Figs. 3 and 4a–f, 2D simulations were carried out with a box of $128\lambda_i \times 192\lambda_i$ in the x - and y -axes, respectively. An 800-nm laser travelling at 20° to the x -axis was shined on a plasma composed of $105n_c$ electrons, $15n_c$ protons, and $15n_c$ fully ionized carbon ions. The laser with a normalized vector potential of $a_0 = 35$, a waist width of 3.2 μm , a duration of 24 fs, and a rising edge from 10^{17} Wcm^{-2} (Supplementary Fig. 20), was set according to the experiment. The transverse widths of the plasmas were $192\lambda_i$, $100\lambda_i$, $64\lambda_i$, and $36\lambda_i$ for the planar, $D = 80\mu\text{m}$, $50\mu\text{m}$, and $30\mu\text{m}$ foils, respectively. For Fig. 4g–i, a 3D PIC simulation for the 30- μm -wide foil was performed

with similar laser and plasma parameters but a reduced grid resolution. A preplasma was directly set instead of the laser rising edge used in the 2D simulation. More details can be found in Supplementary Sec. C.

Coulomb repulsion calculation. The enhancement of the proton cutoff energy in conduction-restricted foils comes from the stronger Coulomb field established by the larger net charge. In the RPA stage, almost all the electrons inside the focal spot region on the nanometre foil are pushed away; then, in the planar foil, the majority of these electrons are pulled back by the ions after the end of the laser, while a portion of the net charge is compensated by the refluxing of cold electrons along the target surfaces. In the conduction-restricted foil, however, the refluxing is suppressed, and the net positive charge $\eta Z n_c e \pi w_0^2 d$ improves the proton cutoff energy from E_0 to $E_0 + \Delta E$, where the carbon density $n_c = 30 n_c$, $Z = 6$, and $\eta Z \approx 1$ according to the approximately 15% more net charge as shown in Fig. 4e. We can assume this positive charge equally distributes along the transverse directions of the foil, and the most energetic proton locates at $x = D/2$ after the charge equilibrium time. Then ΔE can be estimated by the potential of the proton as $\int_{-D/2}^{D/2} \int_{-D/2}^{D/2} \frac{\sigma}{4\pi\epsilon_0} (D^2/4 + y^2 + z^2)^{-1/2} dydz$, where ϵ_0 is the permittivity in vacuum. Considering $\sigma = \eta Z n_c e \pi w_0^2 d / D^2$ and $\int_{-1}^1 \int_{-1}^1 (1 + y^2 + z^2)^{-1/2} dydz = 3.17$, we have $\Delta E = 3.17 \sigma D / 8\pi\epsilon_0 \propto D^{-1}$. From the 3D PIC simulation for the $D = 30 \mu\text{m}$ foil, we can estimate $E_0 = 88.9 \text{ MeV}$ and obtain the dependence of $E_0 + \Delta E$ on D^{-1} as shown in Fig. 4h.

Data availability

Source Data file has been deposited in Figshare under accession code DOI link⁵¹. Additional data are available from the corresponding authors upon request.

Code availability

The open-source code Smilei is available at <https://github.com/SmileiPIC/Smilei>. The detailed setup of the simulations that supported the plots and other findings of this study is presented in the Supplementary Information.

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Author contributions

I.W.C. and C.H.N. conducted the work. The experiments were performed by Y.S., G.E.A., S.Y.K., X.W., S.H.K., J.W.Y., J.H.S., S.K.L. and I.W.C. Y.S., X.W. and K.H.P. carried out all the simulations. Some details of the physics were clarified by Z.G. and X.Y. The manuscript was written by Y.S. and reformed by I.W.C. and C.H.N. All the authors reviewed the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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