Low-divergence MeV-class proton beams from kHz-driven laser-solid interactions

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Proton beams with up to 100 pC bunch charge, 0.48 MeV cut-off energy and divergence as low as a 3° were generated from solid targets at kHz repetition rate by a few-mJ femtosecond laser under controlled plasma conditions. The beam spatial profile was measured using a small aperture scanning time-of-flight detector. Detailed parametric studies were performed by varying the surface plasma scale length from 8 to 80 nm and the laser pulse duration from 4 fs to 1.5 ps. Numerical simulations are in good agreement with observations and, together with an in-depth theoretical analysis of the acceleration mechanism, indicate that high repetition rate femtosecond laser technology could be used to produce few-MeV protons beams for applications.

Over the past two decades, ion acceleration with intense femtosecond laser pulses has emerged as a promising alternative to conventional accelerators due to its comparatively low cost, small size and high instantaneous flux. Laser-accelerated protons are already used for targets studies for fast ignition [1], studies of warm dense matter [2] and probing fast phenomena in laser matter interaction [3]. However, societal applications of laser accelerated protons such as proton therapy [4] and nuclear physics research [5] require source parameters that are still out of reach for existing laser-based accelerators [6, 7]. Intense research efforts are dedicated to improving the source performance in terms of particle energy, average flux, beam brightness and overall source efficiency.

Ion acceleration with ultraintense lasers \( (I_{\text{las}} \gtrsim 10^{18} \text{W/cm}^2) \) has been mostly realized by irradiating thin \( (\sim 0.1 – 10 \mu\text{m}) \) foils, utilizing the well-known target normal sheath acceleration (TNSA) mechanism [8, 9]. Commercially available 100 TW-class lasers deliver TNSA proton beams with energies reaching up to few tens of MeV and \( 10^{11} – 10^{13} \) protons per shot above 1 MeV, depending on target and laser parameters [10]. These lasers typically operate at \( 1 – 10 \) Hz, yet continuous operation at this rate is very challenging to realize for thin foil ion acceleration due to target replenishing needs and debris contamination in the vacuum chamber.

One of the major shortcomings of TNSA proton beams compared to beams produced with conventional accelerators is their much larger divergence, which ranges from \( 10^0 \) to \( 30^0 \) FWHM [11–13]. The demand for high brightness has driven considerable research efforts aiming at reducing the angular spread of laser-generated beams. Such efforts include additional collimation devices [14–16], special target geometries [8, 17–30] and targets which enable other proposed acceleration mechanisms [31–35]. In many cases, target fabrication and handling, together with stringent laser contrast requirements for laser-solid acceleration, hinder the use of these targets in applications where a stable operation at a high repetition rate is necessary.

In this Letter we demonstrate a robust, efficient and easy-to-implement method for delivering highly collimated MeV-scale proton beams at a kHz repetition rate using few-mJ femtosecond pulses. The angular and energy distributions of the proton beam were detected over a wide range of interaction conditions by varying both pulse duration and plasma gradient length. The present work expands on previous studies with thick solid targets and few-mJ lasers [36, 37] with vastly improved laser performance in terms of intensity (up to 2 orders of magnitude increase), temporal contrast (up to 3 orders of magnitude increase) and density gradient control. The numerical and theoretical analysis gives a clear physical picture of the acceleration mechanism and its scalability for applications.

The experiment was carried out at the Salle Noire facility of Laboratoire d’Optique Appliquée (LOA). The laser delivers 27 fs pulses at a kHz repetition rate with high temporal contrast (\( > 10^{10} \) at 10 ps). The pulse duration can be reduced to 4 fs in a hollow-fiber compressor [38] and increased up to 1500 fs by adding intra-chain group delay dispersion (GDD), while keeping the same

Figure 1. Experimental setup.
energy and spatial intensity distribution on target. The peak intensity on target can thus be tuned from $10^{16}$ to $10^{19}$ W/cm$^2$.

A schematic layout of the experimental setup is shown in Fig. 1, where the 2.5 mJ, p-polarised pulses are focused by an f/1.3, 30° off-axis parabolic mirror (OAP) down to a $R_{\text{fwhm}} \approx 1.8$ µm FWHM focal spot at 55° incidence angle at the surface of a rotating fused silica optical substrate. A time-delayed pre-pulse is added by picking off $\approx 4\%$ of the main pulse through a holey mirror, and focusing it with the same OAP to a larger 13 µm FWHM spot. The preplasma gradient scale length $L_g \approx L_0 + c_s t_d$ can be controlled by changing the relative delay $t_d$ between the pre-pulse and the main pulse, where $L_0$ corresponds to an additional expansion induced by the main pulse pedestal due to its finite temporal contrast. The plasma expansion velocity $c_s$ is measured using spatial domain interferometry (SDI) \[39\]. The high-stability rotating target holder \[40\] keeps the target surface position stable within the few-micron Rayleigh length of the tightly focused main pulse, while refreshing the target surface for shots at 1 kHz.

The proton energy and angular distributions were measured with a Thomson parabola spectrometer (TPS) and a charge-calibrated scanning time-of-flight (TOF) detector. The TPS accepts ions in the normal direction through a 300 µm pinhole placed at about 0.6 m from the target. The TOF detector consists of a 6 mm diameter microchannel plate (MCP) placed 375 mm away from the irradiated point and connected to a 8 GHz oscilloscope. This setup provides an acceptance angle of $0.9^\circ$, such that the broadening of the measured angular profile due to convolution with the detector size is negligible. To the best of our knowledge, this is the first time a TOF detector was used to measure the angular distribution of the beam. Further details regarding both detectors are available in the Supplemental Material \[41\].

Fig. 2(a-b) shows proton energy spectra acquired with the TPS, where each spectrum shown is an average of 100 consecutive shots. In Fig. 2(a) the pulse duration is fixed at $\tau_{\text{las}} = 27$ fs and the pre-pulse delay is varied. The highest proton energies around 0.525 MeV are achieved for the steepest gradient and drop below the detection threshold of 0.41 MeV for the gradients $\gtrsim \lambda/10$. In Fig. 2(b) the gradient is kept the steepest (0 ps delay) and the pulse duration is varied. We observe an optimum around 100–300 fs, where the maximum proton energy $W_{\text{max}}$ reaches 0.48 ± 0.02 MeV. For longer pulses, the maximum proton energy slowly decreases, similar to the tendency observed in \[37\] but now with 4-5 times more laser energy on target.

Fig. 2(c-e) shows the angle-resolved TOF spectra measured for the sharpest plasma gradient and for driving pulse durations of 4 fs, 27 fs and 200 fs. For each angle, each spectrum shown is an average of 100 consecutive shots. All three measurements in Fig. 2(c-e) show similar angular profiles and in all cases the energy-integrated divergence is $\approx 3^\circ$ (FWHM). Assuming a two-dimensional Gaussian angular profile, we calculate the total charge (above 0.1 MeV) to be $12 \pm 2.4$ pC, $50 \pm 10$ pC and $98 \pm 20$ pC, respectively. The single-axis measurement is indeed fitted very well by a Gaussian and we expect the beam asymmetry to be small due to the symmetry of the system, as was observed in \[36\] (albeit with much larger beam divergence). Our estimation of 20% error originates from uncertainties in the angular distribution and the TOF calibration, as described in the Supplemental Material \[41\].

![Figure 2. TPS proton spectra in arbitrary units and logarithmic scale, for $\tau_{\text{las}} = 27$ fs and varying plasma scale length (a), and for 0 prepulse delay and varying pulse duration (b). Angular-spectral proton number distributions measured with the calibrated TOF detector for 4 fs (c), 27 fs (d) and 200 fs (e).](image-url)
Figure 3. (a) Charge density (red-blue, \( n_e \) units) and electron density (grey, normalized for visibility) for \( \tau_{\text{las}} = 200 \text{ fs} \) pulse at \( t = -240 \text{ fs} \) before peak field arrival; (b) Temporal evolution of \( z \)-averaged electrostatic field \( \langle E_z \rangle \) (red-blue colormap), maximum proton energy (blue curve) and corresponding \( z \)-coordinate (black curve), maximum proton energy from Eqs. (1) and (2) (blue dashed curve); (b) inset Zoomed \( \langle E_z \rangle \) (colors) and depth estimate Eq. (1) (black dashed curve); (c-e) Final angular spectral distributions for 9 fs (c), 27 fs (d) and 100 fs (e) laser pulse durations.

The observations in Fig. 2 show that the optimal proton acceleration occurs in the short, contrast-limited plasma gradient for laser pulses of \( \tau_{\text{las}} \sim 100 - 300 \text{ fs} \), demonstrating exceptionally low beam divergence. It is important to note, that this optimum corresponds a transition from the regime with \( W_{\text{max}} \propto \tau_{\text{las}} \) to the one with \( W_{\text{max}} \propto I_{\text{las}} \propto 1/\tau_{\text{las}} \), which was also observed in [37].

Let us consider a physical picture of the irradiation of a solid plasma slab by a short sub-relativistic laser pulse, with field strength \( a_0 = eE_{\text{las}}\lambda/2\pi m_e c^2 < 1 \), where \( E_{\text{las}} = \sqrt{2I_{\text{las}}/\epsilon_0 c} \) is the peak laser field, \( \lambda \) is its wavelength, \( e \) and \( m_e \) are the electron charge and mass, respectively, and \( c \) is the speed of light in vacuum. The laser field penetrates the preplasma gradient until it reaches the reflective layer with electron density \( n_{pe} \gtrsim n_e \), where \( n_e = \pi/r_e \lambda^2 \) is the critical plasma density and \( r_e \) is the classical electron radius. Within this layer, the laser introduces the radiation pressure \( P_{\text{rad}} = 2\cos^2 \theta I_{\text{las}}/c \), which pushes plasma electrons until it becomes balanced by the electrostatic pressure \( P_{es} \).

For a short preplasma, \( L_g \ll \lambda \), the laser and electrostatic fields are instantly screened by the dense plasma after the penetrated layer, enabling the well-known Brunel absorption [42]. Owing to this screening, the electrons to escape deep into the plasma without building up a dense negative layer, and allows ions of the target material to generate a long-range accelerating field. For the considered laser intensities, the preplasma consists of only partially ionized Si, O and C ions, and a small fraction of protons (see Sec. I.C and S-Fig. 3 in the Supplemental Material [41]). The latter have a much higher charge-to-mass ratio than heavier ions \( Z_i/M_i \ll 1/m_p \), and can thus be quickly accelerated in the electrostatic field of this positively charged preplasma before its own Coulomb explosion.

We have explored such interactions through 2D particle-in-cell (PIC) simulations using the code WarpX (v21.04-86) [43]. The initial plasma density is uniform \( n_{p0} = 250 n_e \) for \( z < 0 \), and for \( z > 0 \) it falls as \( n_p = n_{p0} \exp(-z/L_g) \), where \( L_g = 8 \text{ nm} \) (see [41] for details). Fig. 3(a) presents distributions of the charge and electron densities modulated by the laser with FWHM duration \( \tau_{\text{las}} = 200 \text{ fs} \) at \( t = -240 \text{ fs} \) before the peak field arrival \( (t = 0) \). These modulations reach down to \( z \approx 34 \text{ nm} \) and the positive charge density reaches \( \rho \approx e n_e \) (red colors in Fig. 3(a)). The thickness of the un-neutralized ion layer can be estimated theoretically from the balance condition, \( P_{es} \approx P_{\text{rad}} \), as [41]:

\[
z_0 \approx L_g \ln \left( \frac{en_{p0}L_g}{2\cos \theta} \sqrt{\frac{c}{\epsilon_0 I_{\text{las}}}} \right),
\]

where \( \epsilon_0 \) is the vacuum permittivity. At the moment shown in Fig. 3(a), the laser intensity on target is \( I_{\text{las}} \approx 2.2 \cdot 10^{15} \text{ W/cm}^2 \), corresponding to \( z_0 \approx 50 \text{ nm} \). The difference with numerical observations results from the presence of electrons confined by the transverse standing wave (blue areas in Fig. 3(a)).

The details of the simulated interaction with the 200 fs pulse are summarized in Fig. 3(b). The average accelerating field \( \langle E_z \rangle = 1/(2x_0) \int_{-x_0}^{x_0} E_z dx \) with \( x_0 = 1.5 \mu\text{m} \), (red-blue colors) appears at the laser arrival near \( z_0 \), which
agrees with the estimate from Eq. (1) (dashed black curve in the inset). For \( z > z_0 \), this field quickly grows and forms a broad plateau region, after which it diminishes and vanishes at \( z \gtrsim 0.5 \, \mu m \). This profile remains almost unchanged for \( t < 0 \), and can be estimated theoretically considering the model of a thin charged disk:

\[
E_z(t, z) = \frac{\hat{E}_{\text{las}}(t)}{2} \left[ 1 - \exp \left( -\frac{z - z_0}{L_g} \right) \right] \times \left( 1 - \frac{z}{\sqrt{z^2 + R_i^2}} \right), \tag{2}
\]

where \( \hat{E}_{\text{las}} \) is the amplitude of laser electric field, and the size of the charged ion disk is determined by the projected laser spot size as \( R_i = R_{\text{las}} / \cos \theta \). The evolution of the maximum proton energy and corresponding \( z \)-coordinate are shown in Fig. 3(b) with blue and black solid curves, respectively. Considering a test-proton motion in the modelled field Eq. (2) we can also find energy evolution that agrees well with the simulation (blue dashed curve).

In Fig. 3(b) we also see that at the later times, the field starts to expand towards vacuum with the velocity \( \nu_{\text{exp}} \approx 4.5 \times 10^6 \, \text{m/s} \) (dotted line). This process is provided by the expansion of the Si preplasma layer, and for the protons that propagate in phase with this layer this allows gaining further energy. At the same time, the field of an expanding layer acquires transverse components due to its curvature, which broadens the proton beam divergence. The simulated proton angular-spectral distributions for different pulse durations, presented in Fig. 3(c-e), show that for the 100 fs pulse, the angular divergence is indeed affected by the plasma expansion. This TNSA signature is much less pronounced in the experimental measurements (cf. Fig. 2(e)), which can be explained by the faster expansion rate in the simulations due to the 2D geometry [44], and also by a higher charge-to-mass ratio of the ions (see the relevant TPS measurement in [41]).

Let us now consider how the maximum proton energy scales with the interaction parameters. In Fig. 4(a) we compare the proton cutoff energy from the experiment (red dots), PIC simulations (blue diamonds) and the model Eqs. (1) and (2) (black solid curve) for different laser durations. In order to discard the surplus acceleration from the 2D TNSA, the simulations were truncated at \( t_{\text{end}} = t_{\text{las}} \), and for this reason the experimentally proton cutoff energies measured for longer pulses slightly exceed the ones in the simulations. All three data sets agree well, with a linear dependence \( W_{\text{max}} \propto \tau \) for the shorter pulses, which flattens as the acceleration becomes optimal for \( t_{\text{las}} \approx 200 \, \text{fs} \). In this particular limit, the acceleration length is short compared to the field’s spatial scale \( R_i \), and is long compared to the preplasma length. Discarding the geometric factors in Eq. (2), we may calculate the proton energy obtained in the field \( E_z = \hat{E}_{\text{las}}(t) \cos \theta / 2 \) over the laser duration as:

\[
W_{\text{max}} = \frac{\ln 2}{\pi} \frac{e^2}{m_p c \epsilon_0} \frac{W_{\text{las}} t_{\text{las}}}{R_i^2}, \tag{3}
\]

where \( W_{\text{las}} \) is the laser energy. Since the field in Eq. (2) vanishes on a scale \( z_{\text{cut}} = R_i / 2 \), and assuming an interaction time of the order of \( 2t_{\text{las}} \), we may define the validity condition for Eq. (3) as \( t_{\text{las}} \lesssim z_{\text{cut}} / (\nu_p) \), where \( (\nu_p) = \sqrt{W_{\text{max}} / 2m_p} \) is the proton velocity averaged over the acceleration. The scaling Eq. (3) within this validity range is shown by the dashed line in Fig. 4(a).

The observed linear growth of \( W_{\text{max}} \) with \( t_{\text{las}} \) for the short pulses corresponds to acceleration that proceeds in the uniform field and ceases before the particles reach the vanishing field region defined by the laser size. This defines the aforementioned optimal regime, which requires the laser duration to match the protons propagation time to \( z_{\text{cut}} \), and thus correlates with the validity threshold of Eq. (3). In this optimal case we obtain:

\[
W_{\text{opt}} = \left( \frac{2 \ln 2}{\pi m_p c e_0 R_i} \right)^{2/3}, \tag{4}
\]
which is shown in Fig. 4(b), together with data from three PIC simulations and the numerical integration of Eq. (2). The obtained theoretical model suggests that the final proton energy depends neither on the plasma nor on the preplasma features (as long as $L_g \ll \lambda$), but is determined solely by the laser energy and its projected spot size.

In summary, we have measured proton beams with very low divergence generated at kHz repetition rate over a wide range of laser and plasma parameters. The divergence was found to be as low as 3° FWHM, about an order of magnitude smaller than thin foil TNSA proton beams. Proton energies reaching up to 0.48 ± 0.02 MeV for an optimal pulse duration ranging from 150 fs to 300 fs were measured with the steepest plasma-vacuum interface. The total charge reached a maximum of 98 ± 20 pC above 0.1 MeV, giving an average current of ∼ 0.1 µA. The acceleration mechanism was identified in simulations and theory derived from simple physical considerations.

The demonstration of these unique proton beams is promising for numerous applications, some of which are within reach. Notable such applications are ion implantation [45, 46] and especially production of radioisotopes for positron-emission tomography (PET), which requires energies of a few MeV [47, 48]. Such energies could be reached with readily available systems providing few-ten-mJ pulses according to our analysis.

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[1] M. Roth, T. E. Cowan, M. H. Key, S. P. Hatchett, C. Brown, W. Fountain, J. Johnson, D. M. Pennington, R. A. Snively, S. C. Wilks, K. Yasuike, H. Ruhl, F. Pegoraro, S. V. Bulanov, E. M. Campbell, M. D. Perry, and H. Powell, Physical Review Letters 86, 436 (2001).
[2] P. K. Patel, A. J. MacKinnon, M. H. Key, T. E. Cowan, M. E. Foord, M. Allen, D. F. Price, H. Ruhl, P. T. Springer, and R. Stephens, Physical Review Letters 91, 125004 (2003).
[3] M. Borghesi, D. H. Campbell, A. Schiavi, M. G. Haines, O. Willi, A. J. MacKinnon, P. Patel, L. A. Gizzi, M. Galimberti, R. J. Clarke, F. Pegoraro, H. Ruhl, and S. Bulanov, Physics of Plasmas 9, 2224 (2002).
[4] V. Malka, J. Faure, Y. A. Gauduel, E. Lefebvre, A. Rousse, and K. T. Phuoc, Nature Physics 4, 447 (2008).
[5] K. W. D. Ledingham, P. McKenna, and R. P. Singhal, Science 300, 1107 (2003).
[6] E. Lefebvre, E. d’Humières, S. Fritzler, and V. Malka, Journal of Applied Physics 100, 113308 (2006).
[7] U. Linz and J. Alonso, Physical Review Accelerators and Beams 19, 124802 (2016).
[8] S. C. Wilks, A. B. Langdon, T. E. Cowan, M. Roth, M. Singh, S. Hatchett, M. H. Key, D. Pennington, A. MacKinnon, and R. A. Snively, Physics of Plasmas (1994-present) 8, 542 (2001).
[9] T. Ceccott, A. Lévy, H. Popescu, F. Réau, P. d’Oliveira, P. Monot, J. P. Geindre, E. Lefebvre, and P. Martin, Physical Review Letters 99, 185002 (2007).
[10] M. Borghesi, in Laser-Driven Sources of High Energy Particles and Radiation, Springer Proceedings in Physics, edited by L. A. Gizzi, R. Assmann, P. Koester, and A. Giuletti (Springer International Publishing, Cham, 2019) pp. 143–164.
[11] R. A. Snively, M. H. Key, S. P. Hatchett, T. E. Cowan, M. Roth, T. W. Phillips, M. A. Stoyer, E. A. Henry, T. C. Sangster, M. S. Singh, S. C. Wilks, A. MacKinnon, A. Offenberger, D. M. Pennington, K. Yasuike, A. B. Langdon, B. F. Lasinski, J. Johnson, M. D. Perry, and E. M. Campbell, Physical Review Letters 85, 2945 (2000).
[12] T. E. Cowan, J. Fuchs, H. Ruhl, A. Kemp, P. Audebert, M. Roth, R. Stephens, I. Barton, A. Blazevic, E. Brambrink, J. Cobble, J. Fernández, J.-C. Gauthier, M. Geissel, M. Hegelich, J. Kaae, S. Karsch, G. P. Le Sage, S. Letzring, M. Manoccoli, S. Meyroneic, A. Newkirk, H. Pépin, and N. Renard-LeGalloudec, Physical Review Letters 92, 204801 (2004).
[13] E. Brambrink, J. Schreiber, T. Schlegel, P. Audebert, J. Cobble, J. Fuchs, M. Hegelich, and M. Roth, Physical Review Letters 96, 154801 (2006).
[14] T. Toncian, M. Borghesi, J. Fuchs, E. d’Humières, P. Antici, P. Audebert, E. Brambrink, C. A. Cecchetti, A. Pi- pahl, L. Romagnani, and O. Willi, Science 312, 410 (2006).
[15] S. Kar, K. Markey, P. T. Simpson, C. Bellei, J. S. Green, S. R. Nagel, S. Kneip, D. C. Carroll, B. Dromey, L. Willingale, E. L. Clark, P. McKenna, Z. Najmudin, K. Krishshelnick, P. Norreys, R. J. Clarke, D. Neely, M. Borghesi, and M. Zepf, Physical Review Letters 100, 105004 (2008).
[16] S. Kar, H. Ahmed, R. Prasad, M. Cerchez, S. Brauckmann, B. Aurand, G. Cantono, P. Hadjisolomou, C. L. S. Lewis, A. Macchi, G. Nersisyan, A. P. L. Robinson, A. M. Schroer, M. Swantusch, M. Zepf, O. Willi, and M. Borghesi, Nature Communications 7, 10792 (2016).
[17] R. Sonobe, S. Kawata, S. Miyazaki, M. Nakamura, and T. Kikuchi, Physics of Plasmas 12, 073104 (2005).
[18] S. Miyazaki, S. Kawata, R. Sonobe, and T. Kikuchi, Physical Review E 71, 056403 (2005).
[19] M. Nakamura, S. Kawata, R. Sonobe, Q. Kong, S. Miyazaki, N. Onuma, and T. Kikuchi, Journal of Applied Physics 101, 113305 (2007).
[20] T. P. Yu, Y. Y. Ma, M. Chen, F. Q. Shao, M. Y. Yu, Y. Q. Gu, and Y. Yin, Physics of Plasmas 16, 033112 (2009).
[21] K. H. Pae, I. W. Choi, S. J. Hahn, J. R. Cary, and J. Lee, Physics of Plasmas 16, 073106 (2009).
Supplemental Material:
Low-divergence MeV-class proton beams from kHz-driven laser-solid interactions

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I. PROTON DIAGNOSTICS IN THE EXPERIMENT

A. TOF measurement

The TOF detector was mounted on a translation stage which moved in parallel to the rotating fused silica slab target. For each stage position, the signals of 100 consecutive shots were recorded by the oscilloscope. The angular profiles in different conditions were measured by moving the stage in steps of 0.65 mm, corresponding to about 0.1°.

A typical TOF signal is shown in S-Fig. 1(a). These measurements were taken for a pulse duration of 27 fs, no prepulse delay (0 ps) and at the target normal direction (0°). The curve is an average of the 100 consecutive shots, smoothed by a rolling average over the response time of the detector (1 ns). The shaded gray area shows the standard deviation of the shots. The first sharp peak corresponds to the radiation emitted from the laser-plasma interaction and defines the time \( t = 0 \). The first protons start to arrive at \( t \approx 50 \) ns, with a peak signal at \( t \approx 90 \) ns, followed by the heavier ions with a signal peaked at \( t \approx 180 \) ns. The noise peak at \( \sim 0.3 \) MeV was ever-present in all our measurements and is probably due to electrical noise in the circuit. At this energy and above the spectrum is more reliably measured with the Thomson parabola detector.

In S-Fig. 1(b) we show the proton energy spectrum extracted from the signal in S-Fig. 1(a) alongside the spectrum extracted from the Thomson parabola spectrometer measured under the same conditions. The TPS spectrum is also an average of 100 consecutive shots. Since the TOF detector was charge calibrated (details below) and the TPS was not, the TOF spectrum is given in units of protons/MeV/msr, while the TPS spectrum is in arbitrary units and is juxtaposed for comparison with the TOF signal. At energies beyond 0.11 MeV the two curves follow together fairly closely, showing a cutoff energy at about 0.25 MeV followed by noise. The decline in the TPS spectrum below 0.11 MeV is due to the signal reaching close to the edge of the MCP.

S-Figure 1. (a) Typical TOF signal measurement. (b)
B. TOF calibration

Determining the total emitted proton charge per shot from the TOF time-dependent signal requires calibration of the detector. This TOF detector is based on a ∼ 0.5 mm thick MCP, grounded on the front side (facing the incoming particles), and biased by a positive voltage (600 V) at the opposite anode side using a bias tee circuit. We have chosen the voltage to be as low as possible while still giving a clear signal in order not to destroy the MCP by electrical breakdown, which could potentially suffer from debris due to its proximity to the interaction point. Since the amplification factor of MCP-based systems could be sensitive to the signal level as well as duration, it is important to calibrate the detector in conditions as close as possible to the experimental conditions. We designed a pulsed calibration system to produce similar signals at the output of the TOF detector as were recorded during the experiment, both in voltage and in pulse width.

The calibration system allows simultaneously recording signals from a ring-like Faraday cup and from our TOF detector positioned concentrically with the Faraday cup. The charge transmitted by the Faraday cup is calculated from first principles and is compared with the TOF signal, thus we can calculate the amplification factor of the TOF detector.

The system is based on a HV Thyratron switch capable of switching 15 kV at ∼ 5 ns. The negative pulse output is connected to a carbon brush which emits electrons at voltages < −3 kV. The radial homogeneity of charge area density was measured by using another Faraday cup, positioned concentrically with the ring-like Faraday cup instead of the TOF detector, thus comparing the signals of the two Faraday cups (accounting for their different areas). We measured higher charge density at the periphery compared to the center, which is expected since the outer ring-like Faraday cup is placed very close to the metal chamber. The radial distribution was accounted for in the calibration.

Electron pulses are used since they are much easier to be produced in the lab. Since the stopping range of both 15 keV electrons and 100 keV protons in the MCP is much smaller than the MCP thickness (smaller than 5 µm in both cases), the error introduced by using electrons instead of protons in the calibration process is negligible with respect to other sources.

S-Figure 2. (a) Schematic diagram of the calibration setup. (b) Simultaneous measurement of TOF and Faraday cup signals. In this shot the Faraday cup signal is divided by 13 to fit the TOF signal.

S-Fig. 2 schematically shows the calibration setup alongside a simultaneous measurement of both TOF and Faraday cup signals. In order to estimate the MCP amplification factor, we divide the Faraday cup signal by some number so it fits best with the TOF signal. We estimate a 10% fitting uncertainty in deriving this constant, which dominates the other possible sources of error. We find the MCP amplification factor to be 3 ± 0.3.

C. TPS measurement

The Thomson parabola spectrometer is added as a small external chamber to the main chamber. The particles enter a 300 µm pinhole and are deflected by magnetic and electric fields. The magnetic field is produced by two permanent magnets 15 mm long with a measured average of 0.32 T over that length. The electric field is produced by two 50 × 50 mm² copper electrodes placed right after the magnets. The field strength is calculated to be 2 kV/cm.
The total distance from the far side of the magnets to the MCP is 210 mm. The MCP is composed of two plates in a chevron setup.

In S-Fig. 3 we show an example of a TPS image. The red line follows the $H^+$ ions (protons) curve. The other usual contaminant ions species of carbon and oxygen are identified. Several curves correspond to charge-to-mass ratios of various silicon ions which are likely accelerated as well.

The proton energy spectrum is obtained after averaging the values along the curve width, after subtraction of a mean background curve which is located in parallel to the signal curve below it. The background curve also has a width and both its mean and standard deviation are calculated along it. We define the cutoff energy as the lowest energy where the mean signal minus the standard deviation of the background is smaller than the mean background. The principal source of error in determining the maximum energy is geometric, i.e., the spatial extent of the ion source on the detector which manifests itself in the proton curve width. We find the relative error $E_{kin}/E_{kin}$ in our geometry to be $\Delta E_{kin}/E_{kin} \approx 0.072(E_{kin}/$MeV$)^{1/2}$.

II. THEORY AND MODELING

To gain insight on the acceleration mechanism we have started with the simulations using the particle-in-cell (PIC) code WARPX (version 21.04-86) [1]. Based on the simulation data we have considered a simple physical model to explain it.

A. Particle-in-Cell simulations

In simulations the two-dimensional numerical domain of the size $40 \times 48 \mu m$ was resolved with a grid with square cells of 6 nm size. The plasma foil was initially neutral, and its density depended only on the $z$-coordinate oriented normal to its surface. The initial density profile consisted of a uniform main plasma for $z < 0$, and an exponential preplasma $\propto \exp(-|z|/L_g)$, for $z > 0$. For simplicity, the main plasma consisted of only silicon ions pre-ionized to $Z_{Si} = 8$, and electrons with the density $n_{pe0} = 250 n_c$, where $n_c = 1.742 \cdot 10^{21} cm^{-3}$ being the critical plasma density for $\lambda = 800$ nm. The choice of the ions’ charge state is qualitative, and represents and average charge-to-mass ratio of the heavy ion species observed in Section I C. The preplasma also had a component of silicon ions pre-ionized to $Z_{Si} = 8$ with density gradient length $L_{Si}^g = 8$ nm, and an additional component of low-density proton-electron plasma. The density of this proton-electron plasma was set to reach $n_{pH} = 0.005 n_c$ at $z = 0$, and its gradient length was increased as $L_{pH}^g = L_{Si}^g \sqrt{M_{Si}/Z_{Si}m_p} = 15$ nm, due to the different ion-acoustic velocities of the ion species. Such low density the presents proton plasma as trace-particles and helps to discard the phenomena related to their space-charge, such as Coulomb explosion of the proton beam. The plasma kinetics were modelled using 64 macro-particles per cell per species in the interaction domain, $z > -140$ nm, while the “deeper” plasma bulk was resolved with a single macro-particle per cell per species.

In order to qualitatively account for non-Gaussian temporal and spatial profiles of the laser pulse, in simulations we considered energy of 1.5 mJ which is lower than in the experiment. This energy reduction was chosen by also
considering additional data of emitted electrons and reflected laser harmonics measured in the experiment (not presented here). The p-polarized pulse was launched at the angle 55° to the normal direction of the plasma surface, and was focused into a spot of $R_{\text{las}} = 1.8 \, \mu m$ FWHM size. The FWHM pulse durations are chosen according to the cases shown in the main text. For the 9 fs and 27 fs cases we consider the transform-limited pulse, while the 200 fs pulse is derived from the 27 fs adding the GDD of 1930 fs$^2$.

### B. Electrostatic field calculation

The field accelerating the protons is generated by the charge imbalance produced by the action of the laser. During this process, in the region of interest inside preplasma, the electrostatic field of the plasma ions is overlapped with the electromagnetic wave, and it cannot be directly analysed. In order to evaluate this field we reconstruct it from the instantaneous charge density distribution obtained from the PIC simulation. For this we consider the Poisson equation for the electrostatic potential and its relation with the electric field:

$$\nabla^2 \phi = -\rho/\epsilon_0, \quad E = -\nabla \phi. \quad (1)$$

The region of the laser-plasma interaction where the charge density $\rho$ is localized is small compared to the size of the simulation domain. This allows us to assume periodic boundaries and solve Eq. (1) using 2D Fourier transforms. Considering the standard fast Fourier transform technique (e.g. from numpy.fft), we can find the field as:

$$E = \text{Re} \left( \text{FFT}^{-1} \left[ -i k \text{FFT} \left[ \rho \right] / \epsilon_0 k^2 \right] \right), \quad (2)$$

where FFT and FFT$^{-1}$ are the forward and inverse Fourier transform operators and $k$ is the wave-vector in units [m$^{-1}$][2].

### C. Analytic model of proton acceleration

Let us consider plasma with a uniform electron density $n_{pe0} \gg n_c$ for $z < 0$, and the evanescent preplasma $n_{pe} = n_{pe0} \exp(-z/L_g)$ for $z > 0$ (see Section II A). A p-polarized laser field impinges on the plasma at an angle $\theta$ and reaches the depth where the electron density becomes critical for the laser $n_{pe}(z_c) \sim n_c$. For very short gradients $L_g < \lambda$, the preplasma mainly reflects the laser field and also develops various processes, including the resonant excitation of plasma electron modulations at $z \gtrsim z_c$ and the so-called Brunel absorption [3]. A description of this laser-plasma interaction can be developed with the help of numerical modelling [4], which however does not provide hints to how the acceleration process scales with the interaction parameters.

For a qualitative theoretical description, let us simplify the picture by assuming that the laser removes electrons up to some depth $z_0 \gtrsim z_c$, leaving a layer of un-neutralized heavy ions that we consider immobile during the whole process. The acceleration is produced by the electrostatic field of these ions and is thus directly related to the value of $z_0$. We further consider the electrons to be only displaced along the z-axis, which occurs as an instantaneous response to the laser field on the target. This assumption agrees with the experimental data and PIC simulations, where we see that $z_0$ is a function of the laser intensity. The simplest way to correlate $z_0$ with the laser intensity is to consider that the electron displacement is produced in a way to maintain static equilibrium between the radiation pressure of the incident and reflected laser field, $P_{\text{rad}} = 2 \cos^2 \theta I_{\text{las}}/c$, and the electrostatic pressure $P_{\text{es}}$ generated by the charge separation. Qualitatively, the latter can be estimated from the capacitor model, $P_{\text{es}} \approx \sigma_i^2 / \epsilon_0$, with $\epsilon_0$ being vacuum permittivity and $\sigma_i = e \int_{z_0}^{\infty} n_{pe} \, dz$ is the area charge density of un-neutralized ions. Note, that so far we have already discarded the ionization, heating and expansion of the heavy ion species, as well as the fast electrons ejected by the laser and present in the preplasma, $z > z_0$, (see Fig. 3(a) in the article). With this, we may write the pressure balance condition as

$$\sigma_i = \cos \theta \sqrt{2c_0 I_{\text{las}}/c} = e_0 \tilde{E}_{\text{las}} \cos \theta, \quad (3)$$

where $\tilde{E}_{\text{las}} = \sqrt{2I_{\text{las}}/e_0c}$ is the amplitude of laser electric field. For the considered preplasma profile this condition defines the penetration depth:

$$z_0 = L_g \ln \left( \frac{e n_{pe0} L_g}{2 \cos \theta \sqrt{c e_0 I_{\text{las}}}} \right) = L_g \ln \left( \frac{2 \pi n_{pe0} L_g}{a_0 \cos \theta} \right), \quad (4)$$
where the dimensionless density $\tilde{n}_{p0}$ is in units of the critical plasma density $n_c = \pi/\rho_e \lambda^2$, $\tilde{L}_g$ is in units of $\lambda$, and $r_e$ is a classical electron radius (a similar result derived in a boosted frame can be found in [5]). Note, that for the uniform plasma and normal laser incidence, this also leads to a well-known law of relativistic plasma transparency $a_0 = 2\pi \tilde{n}_{p0} \tilde{I}_0$, where $\tilde{I}_0$ is a laser penetration depth in units of $\lambda$ [6]. The obtained dependency Eq. (4) demonstrates a good agreement with the PIC simulations as presented in Fig. 3(b) of the article.

The resulting configuration can be presented as a positively charged plane (or disk in 3D) with density $n_i = en_{pe}(z > z_0)$ attached to the surface of the solid neutral plasma. This description relies on the condition that the initial preplasma is very steep, $L_g \ll \lambda$, so that for $z \lesssim z_0$ the electron density is very high ($\gtrsim n_c$) and the field of all electrons injected into the plasma by the laser is instantly screened. This phenomenon is an essential ingredient of the Brunel heating process, which allows electrons to carry away laser energy deep into the plasma, instead of building up a dense negative layer at the surface which is the case in longer preplasmas [5, 7]. From this also follows that the electrostatic field of the ion layer, which is oriented inwards the plasma at $z \lesssim z_0$, is also effectively screened. This can be observed in Fig. 3(b) in the article, where one can see only the positive component of the $E_z$ calculated accounting for all the charges of the system. The accelerating, positive component of $E_z$ grows quickly with $z > z_0$, and reaches its maximum value, which we can estimate as $E_{\text{max}} = \sigma / 2c_0 = \frac{1}{2} E_{\text{las}} \cos \theta$. We can describe its spatio-temporal distribution of the accelerating field of the ion layer as:

$$E_{\text{acc}}(t, z) = \frac{E_{\text{las}}(t) \cos \theta}{2} \left[ 1 - \exp \left( -\frac{z - z_0}{L_g} \right) \right] \left( 1 - \frac{z}{\sqrt{z^2 + R_i^2}} \right),$$

(5)

where the size of the charged ion disk is determined by the projected laser spot size as $R_i = R_{\text{las}} / \cos \theta$. The second and third factors at the right hand side of Eq. (5) account for the field distribution within the preplasma and the three-dimensional geometry of the field, respectively.

Assuming that the accelerated protons remain non-relativistic, we may solve the equations of motion:

$$d_t p_p = eE_{\text{acc}} \ , \quad d_t z_p = p_p / m_p \ ,$$

(6)

and find the maximum gained energy, $W_{\text{max}} = p_{\text{max}}^2 / 2m_p$, where $p_{\text{max}} = e \int_{-\infty}^{\infty} E_{\text{acc}} \, dt$ is the maximum proton momentum, and the integral is calculated along the proton trajectory $z_p(t)$. This integration can be done numerically, and for the parameters assumed for PIC simulations ($W_{\text{las}} = 1.5 \, \text{MJ}$, $R_{\text{las}} = 1.8 \, \mu\text{m}$, $\theta = 55^\circ$ and $n_{p0} = 250 n_c$), the model can describe the maximum gained energy as a function of the preplasma length and the laser pulse duration as presented in S-Fig. 4.

In the particular case of a very short laser pulse, the model Eqs. (5) and (6) can be further simplified. Let us assume, that the total distance travelled by a proton, $L_{\text{acc}} \sim p_{\text{max}} / 2m_p \tau_{\text{las}}$, is very small compared to the scale length of the field, $L_{\text{acc}} \ll R_i$, but is significantly longer than the preplasma gradient $L_{\text{acc}} \gg L_g$. With these conditions in mind, we may neglect the last two factors in Eq. (5), and estimate the energy gained by the protons in the uniform $E_{\text{las}}$ during the laser action. In this case, Eq. (6) can be easily integrated giving:

$$W_{\text{max}} = \sqrt{\frac{\ln 2}{\pi}} \frac{e^2 \cos^2 \theta}{m_p c \epsilon_0} \frac{W_{\text{las}} \tau_{\text{las}}}{R_{\text{las}}^2} ,$$

(7)
This scaling is shown in Fig. 4a (black dashed line) in the article along with the result of numerical integration of Eqs. (5) and (6) depicted with the black solid curve.

It is clear that Eq. (7) is only valid when $\tau \lesssim R_i/4\langle v_p \rangle$, where $\langle v_p \rangle = \sqrt{W_{\text{max}}/2m_p}$ is the proton velocity averaged over the acceleration. Assuming that this validity threshold qualitatively determines the optimal acceleration regime, we may qualitatively correlate this with Eq. (7), and find the scaling of the optimized acceleration as:

$$W_{\text{opt}} = \left( \frac{2 \ln 2}{\pi m_p} \frac{e^2 W_{\text{las}}}{4c\epsilon_0 R_i} \right)^{2/3}.$$  \hfill (8)

[1] J.-L. Vay, A. Huebl, A. Almgren, L. D. Amorim, J. Bell, L. Fedeli, L. Ge, K. Gott, D. P. Grote, M. Hogan, R. Jambunathan, R. Lehe, A. Myers, C. Ng, M. Rowan, O. Shapoval, M. Thévenet, H. Vincenti, E. Yang, N. Zaim, W. Zhang, Y. Zhao, and E. Zoni, Physics of Plasmas 28, 023105 (2021).
[2] For `numpy.fft` routines, this implies an additional factor of $k_x = 2\pi \cdot \text{numpy.fft.fftfreq}(N_x, dx)$.
[3] F. Brunel, Physical Review Letters 59, 52 (1987).
[4] M. Veltcheva, A. Borot, C. Thaury, A. Malvache, E. Lefebvre, A. Flacco, R. Lopez-Martens, and V. Malka, Physical Review Letters 108, 075004 (2012).
[5] H. Vincenti, S. Monchocé, S. Kahaly, G. Bonnaud, P. Martin, and F. Quéré, Nature Communications 5, 3403 (2014).
[6] V. A. Vshivkov, N. M. Naumova, F. Pegoraro, and S. V. Bulanov, Physics of Plasmas 5, 2727 (1998).
[7] A. Macchi, F. Cattani, T. V. Liseykina, and F. Cornolti, Physical Review Letters 94, 165003 (2005).