

Laser-driven high-energy proton beams from cascaded acceleration regimes

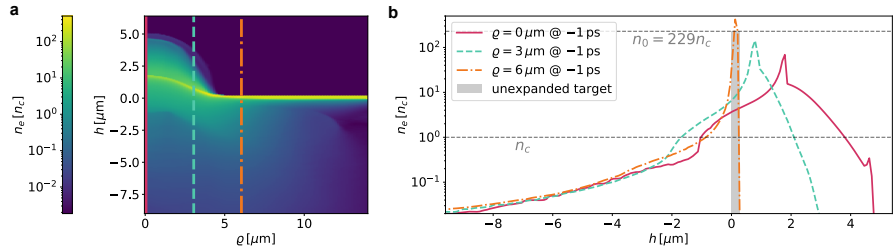
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1 Initial plasma density distribution for particle-in-cell simulation

The target pre-expansion from the onset of laser-induced dielectric breakdown (≈ -100 ps) up to -1 ps before the main pulse arrives at the target was modelled using the FLASH code (v4.6.2) in a 2D radially symmetric geometry. The resulting density distribution in ρ - h coordinates at the end of the FLASH simulation is shown in Supplementary Fig. 1a. The cylindrically symmetric density profile from the FLASH simulation was mapped into 3D, serving as the initial density profile for the subsequent 3D particle-in-cell simulation. The spatially varying pre-expansion of the target can be seen in Supplementary Fig. 1b, showing the simulated electron density for three different lineouts through the target.



Supplementary Fig. 1 Electron density from FLASH simulation 1ps before the arrival of the main laser pulse. **a** 2D electron density in ρ - h coordinates at the end of the simulation. **b** Density lineouts for $\rho = 0$ μm , 3 μm and 6 μm , respectively (indicated by the corresponding lines on the left) along with the initial target density (grey shaded area).

2 Contribution of different acceleration mechanisms

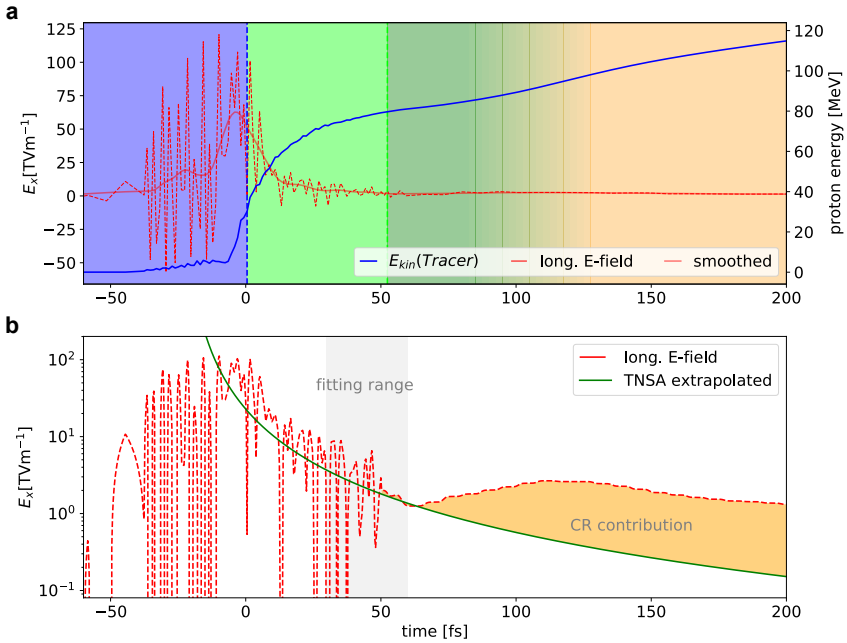
To distinguish contributing mechanisms within the acceleration cascade, the evolution of the electric field experienced by the tracer protons along their direction of propagation and the target density at their position were evaluated. The particle energies at the specific time steps are then assigned to the corresponding mechanism.

The discrimination between acceleration occurring at the target front side (FSA component) and at the target rear side is based on the particle's position within the plasma density profile. FSA ends at the target rear-side boundary, which is defined at the position where the plasma density falls below the relativistic critical density γn_c .

Beyond this boundary, the sheath acceleration phases (prompt and thermal) are separated according to local field fluctuations. The field oscillations in the prompt

phase are detected by a relative comparison to the temporally smoothed (over half a laser period) field evolution. The prompt phase ends when this relative difference becomes smaller than a defined threshold value. Since the transition from oscillating fields to a constant field is smooth, the choice of this threshold is somewhat arbitrary; in this study, it is set to 1, indicating that the field energy in the oscillating field component should be half of the constant one.

Finally, the Coulomb repulsion (CR) phase is identified to begin when the proton charge in the expanding sheath exceeds that of the electrons and the accelerating field increases over time. The CR field then partially overlaps with the sheath acceleration field. A distinction between the two components is conducted by comparing the field evolution at late times with an extrapolated decreasing sheath field, estimated with a power law $E(t) \propto t^{-n}$ as expected for target normal sheath acceleration (TNSA). The energy gain by CR is then calculated using the temporally integrated difference of the actual field and the extrapolated TNSA field up to the end of the simulation. The timeline of the kinetic energy together with the longitudinal electric field projected onto the axis of the current propagation direction is shown in Supplementary Fig. 2a for the most energetic tracer proton. The different acceleration phases as identified of the cascade as described above are indicated by the same colours as used in the main text (FSA - blue, prompt/thermal - light/dark green, CR - yellow). Supplementary Fig. 2b shows the time evolution of the electric field (red dashed line), including the extrapolated sheath field (green solid line).



Supplementary Fig. 2 Acceleration of most energetic proton. **a** Time history of kinetic energy (blue line) and longitudinal electric field (red line) experienced by the most energetic tracer proton. Colour shaded areas belong to different mechanisms of the acceleration cascade. **b** Sheath field extrapolation (green line) showing the electric field increase due to the Coulomb repulsion contribution (orange shaded area).