Proton stopping measurements at low velocity in warm dense carbon

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Ion stopping in warm dense matter is a process of fundamental importance for the understanding of the properties of dense plasmas, the realization and the interpretation of experiments involving ion-beam-heated warm dense matter samples, and for inertial confinement fusion research. The theoretical description of the ion stopping power in warm dense matter is difficult notably due to electron coupling and degeneracy, and measurements are still largely missing. In particular, the low-velocity stopping range, that features the largest modelling uncertainties, remains virtually unexplored. Here, we report proton energy-loss measurements in warm dense plasma at unprecedented low projectile velocities. Our energy-loss data, combined with a precise target characterization based on plasma-emission measurements using two independent spectroscopy diagnostics, demonstrate a significant deviation of the stopping power from classical models in this regime. In particular, we show that our results are in closest agreement with recent first-principles simulations based on time-dependent density functional theory.

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Ion stopping in warm dense matter (WDM) is an important topic in inertial confinement fusion (ICF) for the ignition of small-margin ICF targets by α-particle self-heating and for ICF schemes using ion beams as the main driver, like heavy-ion fusion or ion-driven fast ignition. A precise knowledge of ion stopping in WDM is also essential for understanding proton transport in matter and for experiments where dense plasma states are generated using ion beams, in particular proton iso-degenerate conditions large discrepancies between the predictions of different stopping-power models are reported.

Experiments probing the Bragg peak are also more challenging, and the few measurements carried out for classical plasmas support models that include close binary collisions in the beam-plasma interaction description.

For WDM target conditions, theoretical modelling is more difficult due to electron coupling and degeneracy, and requires more advanced theories like quantum many-body approaches and first-principles calculations. This leads to even larger theoretical discrepancies than in classical plasmas, which increase for low projectile velocities and culminate near the Bragg peak. At low velocities, temperature and/or degeneracy effects are expected to be important on the stopping power and significant deviations from classical theories are predicted.

Measurements in WDM are also more challenging because of shorter sample lifetimes and a more difficult target characterization due to high plasma densities. A few indirect stopping measurements in degenerate conditions have been extracted from ICF implosions by using tertiary neutron spectra, but these data do not allow a precise benchmarking of stopping models. The only direct ion-stopping measurements in WDM reported so far have been performed at the OMEGA laser facility. Probes were quasi-monoenergetic protons of around 14.6 MeV energy created from DH3 fusion reactions during exploding-pusher implosions. The target was a warm dense beryllium sample isochorically heated by multi-kilojoule laser-driven X-rays over few nanoseconds, reaching Tem ≈ 30 eV at solid density, corresponding to Γ ≈ 0.3 and Θ ≈ 2. However, as the beam-plasma interaction was in the high-velocity limit (vP/vTh ≈ 13), temperature and degeneracy effects on the stopping power were negligible, and the latter could be described by a simple Bethe-like formalism. Moreover, no detailed target characterization could be carried out, and only a small number of trials were able to be taken due to the scale of the laser facility. Meanwhile, lower-velocity regions (vP/vTh ≤ 10) in WDM have not been experimentally investigated until now, not to mention the Bragg peak region (vP ≈ vTh).

The parameter domain investigated so far is illustrated in Fig. 1, that shows a selection of reported stopping experiments displayed as a function of the velocity ratio vP/vTh and the electron coupling parameter Γ. Experiments performed in gas-discharge and Z-pinch targets are limited to low plasma densities...
(n_e \approx 10^{17}–10^{18} \text{ cm}^{-3}) and the high-energy probing ion beams on the MeV/u scale. Low to moderate velocity ratios v_p/v_{th} \leq 3 can be obtained in laser-generated plasma and exploding-pusher experiments, which are essentially limited to hot, ideal plasmas. Cold and dense plasma conditions can be achieved with X-ray driven targets. While the plasma density remains \approx 10^{20} \text{ cm}^{-3} in ref. 10, the experiment of ref. 13 does reach solid-density WDM conditions. However, the reported measurements involve high velocity ratios v_p/v_{th} \geq 10 due to the use of fast projectiles. Our goal is to simultaneously reach WDM states with moderate to strong electron coupling \Gamma \approx 0.1–1 and to probe them with low to moderate velocity ratio (v_p/v_{th} \ll 10), which remains an unexplored parameter domain approaching the conditions of α-particles in an ICF fuel shell and constitutes a step further towards the Bragg-peak region. Measurements at low velocity require well-characterized WDM samples and projectile ions with energies of a few hundred keV. Such low probing energies require thin samples which can experience a significant hydrodynamic expansion within tens to hundreds of picoseconds. Precise stopping measurements thus require a probing beam duration comparable or shorter than the sample lifetime. These requirements are difficult to achieve with accelerator ion beams because usual bunch durations lie on the nanosecond time scale. Exploding-pusher sources are limited by the relatively high (\geq 1 \text{ MeV}) reachable projectile energies as well as proton pulse durations \approx 100 \text{ ps} and by the availability of short-duration heater beams. This makes the WDM sample generation. On the other hand, laser-generated proton beams, that feature short pulse lengths and broadband energy spectra, offer the required flexibility to overcome these limitations. Therefore, they have been used in several recent stopping experiments and are planned to be used in future experiments, in general in association with an energy filtering device to select a narrow energy band. Moreover, as the stopping power at low velocity has a stronger temperature dependence, precise target temperature measurements are needed in order to both benchmark the plasma conditions and to interpret the energy-loss data.

In this work, we use an experimental approach based on a laser-generated proton selection platform operated at high-repetition rate at a short-pulse laser facility. Using this platform, we have measured the proton energy loss in a low-velocity regime in a warm dense carbon target that was heated by a second short-pulse laser. The projectile energy of around 500 keV led to velocity ratios v_p/v_{th} between 3 and 10, significantly lower than in previous experiments (see Fig. 1). For these conditions, discrepancies between first-principles stopping-power calculations and classical predictions reach up to 20% and can be resolved experimentally. Our energy-loss measurements, in association with a detailed characterization of the WDM conditions using two complementary spectroscopy diagnostics, provide a first test of ion stopping models in this unexplored regime.

Results
Experimental setup. The experiment was performed at the PW-class VEGA laser facility at the Centro de Láseres Pulsados (CLPU), Salamanca, Spain. The experimental setup is shown in Fig. 2. The initial 200 TW VEGA2 laser beam was split into two short pulses, respectively called the main and the heater beam. The setup consists of four main stages: (i) the generation of the proton beam by the main laser beam, (ii) the generation of the WDM sample by the heater beam, (iii) the measurement of the downshifted spectrum of the proton beam that passed through the WDM target using a magnet-based spectrometer and (iv) the characterization of the WDM conditions by using two independent spectroscopy diagnostics.

Simulations of the WDM target. The WDM conditions were simulated using the two-dimensional (2D) radiation-hydrodynamic code RALEF2D widely used for simulations of different experiments, assuming local thermodynamic equilibrium (LTE), over a 500 ps time span after the proton beam hit the target. The target ionization is deduced by post-processing the density and temperature profiles with the LTE version of the FLYCHK code. The profiles of mass density \rho, electron temperature T_e and mean ionization Z_\bar{e} along the target central axis are plotted in Fig. 3 for various times of the target evolution, where the target thickness is reported in areal-density units. The reached conditions are \rho \approx 0.1 \text{ g/cm}^3 and T_e between a few eV and a few tens of eV, which correspond to carbon ionization degrees \approx 4. The resulting values of \Gamma \approx 0.1–2 and \Theta \leq 10 (in most of the target) are also shown in Fig. 3, indicating moderately to strongly coupled, and moderately degenerate target conditions. The velocity ratio values corresponding to the projectile energy of 500 keV are also plotted, with v_p/v_{th} \approx 10 over the considered time domain, and v_p/v_{th} \approx 2–3 in the first tens of picoseconds of the target evolution, which is significantly lower than in previous experiments. As also appears on the graphs of Fig. 3, the target areal density remains remarkably constant, which indicates a one-dimensional target evolution over the time range of 500 ps.
In addition, the interaction of the short-pulse heater beam with the target is likely to generate significant transient electric fields which may impact the proton beam and thus the energy-loss measurement. The effect of such fields was estimated by using a dynamic model of target charging by short-pulse laser interaction\(^5\). The target charging was estimated to dissipate within the first 10 ps after the heater beam onset on the target, which thus may influence only a small fraction of \(\approx 2\%\) of the beam protons and does not perturb the rest of the beam.
WDM target characterization. The temporal evolution of the target temperature extracted from the SOP data averaged over 80 shots is shown in Fig. 4a and compared with the time-dependent temperature extracted from the RALEF2D simulation. Both the experimental and the simulated temperature are determined at the critical density for the 532 nm wavelength used for the measurements and averaged within a 50 μm emission diameter around the central plasma axis corresponding to the proton beam probing area. The experimental error results from the statistical error on the measurements and from the detector calibration uncertainty and is estimated as ±30%. The error band on the simulation curve accounts for the signal variation due to shot-to-shot pointing fluctuations of the heater beam estimated to be below 50 μm. The temperature determined from the SOP data is slightly lower than the one predicted by the RALEF2D simulation, while agreeing, in average, within the ±30% experimental error bar. The experimental temperature is also compared to the temperature extracted from a hydrodynamic simulation performed with the one-dimensional (1D) MULTI-fs code in LTE. The MULTI-fs prediction, also determined at the critical density for the 532 nm wavelength, overestimates the measured temperatures by around 30%. The RALEF2D prediction is clearly more accurate as the simulation was performed using the experimentally measured spatial distribution of the heater focal spot intensity.

The X-ray emission spectra over the whole target emission area measured with the XPHG diagnostic are presented in Fig. 4b. They are compared to the spatially and temporally integrated spectra obtained with the PrismSPECT code53,54 assuming LTE and using the density and temperature profiles extracted from both the RALEF2D simulation (Fig. 3a, b respectively) and from the MULTI-fs 1D hydrodynamic simulation carried out over a weighted range of intensities matching the experimentally measured focal spot. The measured spectra agree within 10–30% with the spectra predicted by the RALEF2D and the MULTI-fs codes, which is on the order of the experimental error bar estimated as ≈20%. In contrast to the SOP data, the XPHG measurement shows an X-ray emission at higher energies than simulated, which corresponds to an experimental temperature higher than simulated.

Based on the RALEF2D simulation, a mass-weighted and time-integrated temperature of 7.5 eV is estimated within the 50 μm proton diameter spot. Taking the average of both diagnostics, it can be concluded that the measured temperature is within 15% agreement with the RALEF2D simulation. The good agreement of the XPHG data with the RALEF2D prediction also shows that the target electron density is known with a reasonable accuracy. Moreover, the overall agreement of both measurements with the simulation over the whole considered time range indicates that the target expansion and thus the target areal density, are correctly simulated. In particular, the agreement of the experimental data with both the 2D and the 1D hydrodynamic simulations confirms that the target expansion is nearly one-dimensional as predicted by the RALEF2D code. Therefore, the presented spectroscopy measurement data set enables to validate the target parameters over the time domain of interest of a few hundred of ps. It is worth mentioning that this WDM sample characterization has been carried out simultaneously with the proton energy-loss measurements, which has not been done in previous stopping-power experiments.

Stopping-power calculations. For estimating the discrepancies between stopping-power models for typical conditions of the experiment, various predictions for protons in carbon are compared in Fig. 5a for a density $\rho = 0.5 \text{ g/cm}^3$ and a temperature $T_e = 10 \text{ eV}$. The proton stopping power in solid carbon according to the SRIM database is plotted as a reference.

Firstly, we use ad hoc calculations combining a free-electron and a bound-electron contribution that are obtained separately knowing the target ionization56–58. The free-electron term is calculated using several models that have the same Bethe-like high-velocity limit determined from dielectric stopping theory: the Li-Petrasso (LP) model59, the Brown-Preston-Singleton (BPS) model60, the T-matrix (TM) model with velocity-dependent screening61, the dielectric random phase approximation (RPA) model62 and the Zimmerman parametrization63 of the Maynard-Deutsch dielectric stopping power64, the latter being very similar to the RPA description. In all cases, the bound-electron stopping term, which is specifically plotted in Figs. 5a and b, is calculated using a model by Casas et al.37 that is valid for all projectile velocities.

Secondly, we use a self-consistent average-atom method in the local density approximation that simultaneously calculates the plasma ionization and the total stopping power using the method presented in refs. 65,66 and using the quantum average atomic model (QAAM) described in ref. 67 under the LTE assumption.
Thirdly, we employ an ab initio approach based on a recently developed time-dependent density functional theory (TD-DFT), including an orbital-free (TD-OF-DFT) version\textsuperscript{30,31} and a full Kohn–Sham approach (TD-KS-DFT) utilizing a mixed basis of deterministic and stochastic orbitals\textsuperscript{68}. The target ionization in TD-DFT is defined self-consistently, as localized and delocalized electrons are naturally determined by the mean-field theory of DFT. Hence, there is no ad hoc separation between ionization and stopping-power physics. Moreover, the many-orbital representation of TD-KS-DFT allows for an exact treatment of Fermi-Dirac statistics, i.e. electron degeneracy, while TD-OF-DFT accounts for the degeneracy effects through a kinetic energy functional. It has been shown in refs. \textsuperscript{30,31} that the TD-OF-DFT theory agrees with the high-velocity data of ref. \textsuperscript{34}, but predicts deviations of up to 20% from classical stopping-power predictions for WDM conditions at low projectile velocities. The TD-OF-DFT and TD-KS-DFT values are determined with an uncertainty estimated to \pm 10%.

The ad hoc and the QAAM calculations are in close agreement for proton energies \(E_p \geq 500\text{ keV}\) and predict a significant increase of the stopping power compared to the solid, that reaches \(\approx 20\%\) at \(E_p = 500\text{ keV}\). Discrepancies between these models increase at lower energies. In contrast, the TD-OF-DFT and the TD-KS-DFT theories predict a stopping power very close to the solid level for \(E_p \geq 400–500\text{ keV}\). The more precise TD-KS-DFT predictions are smaller and within better than 10% agreement with the TD-OF-DFT values. A stopping enhancement relative to the solid is also predicted but at lower velocities than according to other calculations, in the close vicinity of the Bragg peak. Hence, in the probing range of 500 keV energy, a 20% reduction of the stopping power is predicted by the TD-OF-DFT and TD-KS-DFT theories compared to the other models, which we attribute to the electron coupling and quantum degeneracy more precisely included in the TD-DFT calculations.

The effect of the target temperature on the stopping power is shown in Fig. 5b for an ad hoc calculation (namely with the Zimmerman model), and for the QAAM and TD-OF-DFT models, respectively, for the same density \(\rho = 0.5\text{ g/cm}^3\) and for temperatures \(T_e = 10, 20,\) and \(30\text{ eV}\). For these conditions, the ionization degree according to FLYCHK is \(Z_e = 1.43, 2.31\) and 2.95, while the one estimated with QAAM is \(Z_e = 1.56, 2.40\) and 2.86, respectively. As is shown, the stopping-power variation with temperature is very small for proton energies above 200–300 keV, reaching few percent at the experimental projectile energy of 500 keV. This also shows that the variation of the ionization degree with temperature on the one hand and the small ionization differences used for the various stopping-power calculations on the other hand are negligible for the studied conditions. These estimates thus suggest that the stopping power in the studied beam-target parameter range is not impacted by thermal effects, but that it is significantly impacted by coupling and/or degeneracy effects as shown by the discrepancy between the first-principle TD-OF-DFT calculation on the one hand and the classical and average-atom calculations on the other hand.

For comparison with the experimental measurements, we calculated the energy loss \(\Delta E_{\text{sim}}\) at each time step of the hydrodynamic simulation as the integral of the stopping power along the ion trajectory through the target

\[
\Delta E_{\text{sim}} = -\int \frac{\partial E}{\partial \rho(x)} [p(x) \, dx],
\]

where the stopping power, expressed as an energy loss per unit of areal density, is calculated with the parameter profiles as shown in Fig. 3. Each energy-loss value is averaged over the target parameters in a temporal range of 400 ps corresponding to the duration of the proton bunch interacting with the target, as well as in a spatial range of 50 \(\mu\text{m}\) corresponding to the probing proton beam diameter. The calculation was respectively performed for the cases where the proton beam is centered on the target central axis and where the proton beam is deviated by 50 \(\mu\text{m}\) from the central axis, which corresponds to the maximum estimated pointing fluctuation between the proton and the heater beam in the experiment.

For computational effectiveness, the energy loss is only calculated as follows. First, it is estimated in an ad hoc manner, using the Zimmerman, Li-Petrasso and T-Matrix models for the free-electron stopping and the Casas model for the bound-electron stopping. These three calculations predict very similar values within 1% (as also suggested by Fig. 5), and are simply designated as “classical calculation” in the following. Second, a TD-KS-DFT stopping-power fit is generated as a function of the target density and the projectile energy assuming a constant temperature \(T_e = 10\text{ eV}\) (see methods) for calculating a DFT-predicted energy loss.

The plasma parameters for our energy-loss calculation are well-characterized. Indeed, we neglect the experimental uncertainty of \pm 15% on the \(T_e\) measurement due to the low sensitivity of the stopping power to temperature in the studied conditions. Moreover, the uncertainty on the target areal density is negligible because of the 1D target expansion.

**Energy-loss results.** Firstly, the energy loss of the proton beam was measured in solid carbon foils over 35 shots to estimate the measurement accuracy and provide a reference energy-loss value in the solid target \(\Delta E_{\text{sol}}\). The downshifted proton energy after passing the solid target was measured to be 449 \(\pm 5\text{ keV}\), where
the error $\sigma = \pm 5$ keV results from the standard deviation at $1\sigma$ over all shots and from systematic measurement uncertainties as is explained in the methods. This results in an energy loss of $\Delta E_{\text{sol}} = 49 \pm 5$ keV, which is in good agreement with the energy loss of 48.1 keV predicted with the SRIM database.

Subsequently, the proton energy loss in the sample was measured on shots with the heater beam driving the target, at respective time delays of $-316 \pm 100$ ps, $-116 \pm 100$ ps and $86 \pm 100$ ps relative to the onset of the heater laser pulse on the sample. The experimental data acquired over several shots are presented at each time delay in Fig. 6a–c, where each data point corresponds to an individual shot. The blue band on each graph corresponds to the experimental error interval of $\pm 5$ keV of the reference energy-loss measurement in the solid target $\Delta E_{\text{sol}}$.

In Fig. 6a, b, the energy loss is measured before the laser heating of the sample, for protons still probing the solid target. The obtained data points are consistent with the previous reference energy-loss measurement in the solid target $\Delta E_{\text{sol}}$.

In contrast, in Fig. 6c, the energy-loss measurement is performed when the temporal center of the proton beam is at 86 ps after the beginning of the sample heating, so that protons almost fully probe the WDM state. The measured energy loss reaches values between $36 \pm 5$ keV and $43 \pm 5$ keV depending on the shot, with an average value $\Delta E_{\text{WDM}}$ of $39.4 \pm 5$ keV over four shots. This corresponds to values of 13–26% lower than the measurement in the solid target $\Delta E_{\text{sol}}$ of 49 keV $\pm 5$ keV, with an average percentage difference of 20 $\pm$ 9%.

A comparison of the averaged proton spectra acquired respectively after free propagation in vacuum (averaged over 20 shots, green curve), the downshifted proton spectrum after passing the solid target (averaged over 35 shots, blue curve), and the downshifted proton spectrum after passing the WDM target (averaged over 4 shots, red curve). The vertical bars mark the spectra maxima positions.
These values are 15% and 12% higher than the SRIM energy loss in the solid target and 12% and 10% higher that the measured energy loss in the solid at. Hence, the energy loss measured in the WDM sample, with an average value $\Delta E_{\text{WDM}} = 39 \pm 5$ keV, is at least 15 keV lower than the classical prediction. These differences are greater than the error bars and thus suggest that the classical calculation $\Delta E_{\text{cm}}$ overestimates the measured energy loss by 41%.

The experimental data are also compared with the results of the TD-KS-DFT energy-loss calculations $\Delta E_{\text{DFT}}$. The energy loss predicted at the time of proton probing in WDM $\Delta E_{\text{DFT}} = 51 \pm 2.5$ keV is only 6% higher than the energy loss measured in the solid target. Consequently, it overestimates the energy loss measured in WDM by 22.7 ± 14%. Therefore, the TD-KS-DFT calculations provide a twice better agreement with our experimental data than classical stopping-power models, which predict a much higher stopping-power enhancement for the considered WDM conditions and appear to be not valid in the probed parameter range.

In summary, our proton energy-loss data at 500 keV energy in warm dense carbon, at a velocity ratio down to $v_p/v_{th} \geq 3$, provide measurement in the unexplored regime of low-velocity stopping in coupled and degenerate plasma conditions. When comparing these experimental measurements to existing stopping-power models, we find that the closest agreement is with the Density Functional Theory (TD-OF-DFT and TD-KS-DFT) calculations. This highlights the effect of electron coupling and degeneracy at near-Bragg-peak conditions. Several developments of this experimental measurement in order to provide more accurate comparisons as described.

Magnet spectrometer. The magnet spectrometer was designed and characterized at CLPU and was used to measure the proton beam energy. It was positioned at a 3.5 cm distance from the WDM sample along the proton propagation axis. The spectrometer consists of a 0.2 T, 10.4 cm long dipole magnet. It deflects protons upwards to a microchannel plate (MCP) detector, which is coupled with a phosphor screen located 10 cm from the end of the magnet and imaged onto a CCD camera. The 2D magnetic field of the spectrometer was measured with a Hall effect probe and was used to calculate the predicted proton deflection on the MCP. The resolution of the spectrometer at 500 keV proton energy is 2 keV per pixel of the image. A horizontal slit of 1 mm height and 1 cm length was inserted in front of the spectrometer entrance aperture of 1 cm diameter to ensure that only protons within the horizontal plane of the propagation axis enter the spectrometer. This effect provides the “zero height” (zero beam) reference position on the detector. The vertical positioning uncertainty of the proton beam of 125 nm results in an energy uncertainty of ±2.5 keV on the MCP that constitutes a systematic error $\sigma_{\text{sys}}$ on the energy measurement.

Examples of raw images obtained with the MCP detector for individual shots are shown in Fig. 7, which shows a reference signal of the selected proton beam (a) and selected proton beam signals after passing through target samples (b–d). As visible in Fig. 7b, the angular straggling of the proton beam through the target, which is estimated to be around 2° using FLUKA simulations, results in a 3 cm spot at the spectrometer entrance. This signal broadening introduces an error in the center values of the energy loss of the downshifted proton spectrum. In order to mitigate this error, we mounted a horizontal slit of 1 mm height in front of the spectrometer for reducing the beam spot size on the MCP detector as illustrated in Fig. 7c. Using this slit, a systematic error is added on the energy measurement in the sample due to partial collection of protons on the detector, which is estimated as $\sigma_{\text{sys}} = \pm 3.5$ keV.

The total error on the energy-loss measurement is estimated as

$$\sigma_{\text{tot}} = \sqrt{\sigma_{\text{stat}}^2 + \sigma_{\text{sys}}^2 + \sigma_{\text{exp}}^2},$$

where $\sigma_{\text{stat}} = \sigma/N$. Here, $\sigma$ is the standard deviation and $N$ is the number of shots, while $\sigma_{\text{sys}} = \pm 2.5$ keV and $\sigma_{\text{exp}} = \pm 3.5$ keV are the systematic errors coming respectively from the proton beam alignment and from the detector. The energy loss in the target is estimated as the difference between the measured central energies of the reference and of the downshifted beam, and can be written as

$$\Delta E_{\text{down}} = E_{\text{ref}}(\sigma_{\text{stat}}), E_{\text{sys}} - E_{\text{down}}(\sigma_{\text{stat}}), E_{\text{sys}},$$

where

$$E_{\text{down}} = E_{\text{ref}}(\sigma_{\text{stat}}), E_{\text{sys}} - E_{\text{down}}(\sigma_{\text{stat}}), E_{\text{sys}}.$$
from the proton source and it is rotated by 14.5° for pointing the selected proton beam in straight axis with respect to the WDM sample. The entrance slit, of 20 μm width and 3 mm height, is attached in front of the dipole magnet yoke for reducing the horizontal acceptance of the incoming TNSA proton beam. The selected pencil-like proton beam undergoes a horizontal energy spread after entering into the magnetic field region. The exit pinhole of 20 μm diameter, positioned at 1 cm after the exit of the magnet, selects a narrow bandwidth of proton beam energies that freely propagates up to the carbon sample. The selector is designed to be fully operational at high repetition rate with a motorization of the dipole magnet moving in and out and a holder for the exit pinhole with horizontal and vertical motorization. The design and the optimization of the energy selector are presented in detail in ref. 41. In this work, we selected a proton beam with a central energy of 498 ± 4 keV and an energy bandwidth of 44 ± 4 keV at FWHM, where 4 keV is the total uncertainty for a single shot. The selected proton energy was found to be highly reproducible from shot to shot with a statistical error of ±0.6 keV (given by $e_{\text{stat}} = \sqrt{\sigma_x^2 + \sigma_y^2}$, where $\sigma_x = 12$ keV is the measurement standard deviation and $N = 20$ is the shot number). Such small error also suggests a low sensitivity of the selected proton beam parameters to the laser shot-to-shot instability (pointing stability of ~ 12 μm, energy variation ~ 3%).

The energy spectrum of the selected proton beam measured with the high-resolution monochromator spectrometer is shown in Fig. 8 and compared with a synthetic spectrum obtained with a FLUKA Monte-Carlo simulation using the experimental configuration for the proton energy selector (black dashed curve). The shaded area around the red curve represents the measurement error.

**Synchronization between the proton and the heater beam.** The sub-ns time synchronization was performed for both laser beams at their respective interaction points (proton target and WDM target position) accounting for the time-of-flight (TOF) for 500 keV protons between these points. The proton trajectory was calculated analytically based on the experimental geometry and verified using Monte-Carlo simulations. The heater beam was delayed in respect to the main beam by the proton TOF of 9.2 ns up to the WDM sample position. This was achieved with the help of a 3 m long delay line for increasing the heater beam path. This main delay line was coupled with a smaller motorized delay line of 20 cm length enabling a fine adjustment with a minimum time step of 10 ps. The main and the heater beams were synchronized with a 9.2 ns delay by using photodiodes positioned at their respective interaction points. Both pulse signals were adjusted on a 1 GHz oscilloscope with identical cable lengths using the smaller delay line. The required delay value was obtained with a precision of ±10 ps, calculated as $\sigma_{\text{stat}} = \sqrt{\sigma_x^2 + \sigma_y^2}$. Where $\sigma_{\text{stat}} = 70$ ps is a statistical error and $\sigma_y = 50$ ps is the error on the proton TOF calculation.

**Fig. 7 Selected proton beam spectrum.** Comparison between the measured selected proton beam spectrum (red solid curve) and the selected proton beam spectrum obtained from an initial broadband TNSA-like spectrum with energies of 0 - 2 MeV simulated with the FLUKA Monte-Carlo code using the experimental configuration for the proton energy selector (black dashed curve). The shaded area around the red curve represents the measurement error.

XUV pinhole grating (XPHG) diagnostic. The XPHG diagnostic, based on a free-standing multi-pinhole X-ray transmission grating, was used to measure the broadband XUV emission from the plasma.

- **Proton energy-loss calculations.** The energy-loss calculations were performed using the FLUKA code.
- **SOP diagnostic.** The Streaked Optical Pyrometry (SOP) diagnostic was used for measuring the time-resolved black-body temperature of the WDM target with a 10 ps resolution. Due to its sensitivity to low temperatures, this diagnostic is well-suited for measuring the temperature of WDM samples. The SOP diagnostic had a target view of 25° in relation to the target normal on the heater beam side.

The emission of the WDM target was collected by the optical system, imaging a region of ~ 400 μm onto a Hamamatsu S20 streak camera with a magnification of 5. The interferometric filter was centered at a 532 ± 0.6 nm wavelength with a FWHM bandwidth of 3 ± 0.6 nm (F1532). An additional color-glass bandpass filter for wavelengths of 360–580 nm (BG39) was used to mitigate the laser light at 800 nm wavelength propagating along the collection axis. The wavelength-dependent response of the SOP system within the 3 nm bandwidth was provided by the manufacturers. The transmission of the optical system for SOP has been measured with a 532 nm continuous diode laser and the streak camera was absolutely calibrated at the selected wavelength using the calibration of ref. 71. The latter was carried out with the same streak camera employed in this experiment and with the filter data set BG38 and F1532 that are similar to the ones we used. The data were acquired inside a time window of 2 ns. The temporal evolution of the temperature was determined using a vertical line-out of the central target region of 50 μm diameter corresponding to the proton beam size entering the WDM sample. The curve average of the temperature is presented in Fig. 4. The error bar is estimated as $\sigma_{\text{stat}} = \sqrt{\sigma_x^2 + \sigma_y^2 + \sigma_z^2}$ that includes the standard error $\sigma_{\text{stat}} = \sigma / \sqrt{N} \approx \sigma_{\text{stat}}$ where $\sigma$ is the standard deviation from the mean, a 25% statistical error $\sigma_{\text{st}}$ and a 5% uncertainty in the detector calibration $\sigma_{\text{calib}}$.

**Modelling**

**RALEF2D and MULTI-fs hydrodynamic simulations.** The RALEF2D simulation was performed in axi-symmetric geometry using the experimentally measured laser parameters, namely an energy of 0.45 J, a gaussian-shaped temporal laser pulse profile with a 217 fs width at FWHM and a spatial distribution profile of the focal spot of ~ 150 μm radius.

The density and temperature profiles are sampled with a 5 ps time step for $t = 0$ to 100 ps and a 10 ps step for $t = 100$ to 500 ps. The spatial sampling is of ~ 350 points for longitudinal rays (along the proton propagation axis) over the target areal density, and of 5 μm in the transverse direction up to a radius of 150 μm.

The MULTI-fs 1D simulation was performed using the same laser energy and pulse duration. In order to represent the radial intensity profile of the focal spot, four separate simulations were performed using the input intensity of the heater and WDM target position accounting for the time-of-flight (TOF) for 500 keV protons between these points. The proton trajectory was calculated analytically based on the experimental geometry and verified using Monte-Carlo simulations. The heater beam was delayed in respect to the main beam by the proton TOF of 9.2 ns up to the WDM sample position. This was achieved with the help of a 3 m long delay line for increasing the heater beam path. This main delay line was coupled with a smaller motorized delay line of 20 cm length enabling a fine adjustment with a minimum time step of 10 ps. The main and the heater beams were synchronized with a 9.2 ns delay by using photodiodes positioned at their respective interaction points. Both pulse signals were adjusted on a 1 GHz oscilloscope with identical cable lengths using the smaller delay line. The required delay value was obtained with a precision of ±10 ps, calculated as $\sigma_{\text{stat}} = \sqrt{\sigma_x^2 + \sigma_y^2}$. Where $\sigma_{\text{stat}} = 70$ ps is a statistical error and $\sigma_y = 50$ ps is the error on the proton TOF calculation.

**Proton energy-loss calculations.** The energy-loss simulations are performed similarly as in ref. 24. The ionization distribution of the plasma is calculated using the collisional-radiative FLYCHK code in local thermal equilibrium, which provides the ion densities ($n_e$, ..., $n_{\text{ion}}$) of the different plasma charge states ($C^{0+}$, ..., $C^{9+}$) for
exchange correlation functional. 74 trajectories per velocity point were used to each point of the considered profile. The free electron density is calculated as \( n_{\text{e}} = 6 n_{\text{e}} + 5 n_{\text{e}} + 4 n_{\text{e}} + 3 n_{\text{e}} + 2 n_{\text{e}} + n_{\text{e}} \). The mean plasma ionization degree \( Z^\text* \) is then determined from the relation \( n_{\text{e}} = Z^\text* n_{\text{e}} \). Here, \( n_{\text{e}} = p N_{\text{e}}/A_{\text{e}} \) is the total ion density, where \( A_{\text{e}} = 12 \) is the molar mass of carbon and \( N_{\text{e}} \) is the Avogadro number. The free-electron stopping power is calculated using the density \( n_{\text{e}} \) with the Zimmerman, Li-Petrasso and T-Matrix models. The bound-electron stopping power is determined using the ion densities \( n_{\text{e}} = n_{\text{e}} \ldots, n_{\text{e}} \) and the Casas model. The carbon atomic quantities required for the bound electron calculation are taken from ref. 79. The total stopping power is obtained as the sum of these contributions:

\[
\frac{dE}{dx_{\text{total}}} = \frac{dE}{dx_{\text{ion}}} + \frac{dE}{dx_{\text{bound}}}
\]

\( (3) \)

The projectile charge state is modeled using the effective charge state predicted by Gusakov et al.\textsuperscript{72}, which is valid in plasma at any projectile velocity. At 500 keV projectile energy, it reaches values \( 0.98-0.99 \) depending on the target conditions. The projectile slowing down inside the target is taken into account for each step along the proton propagation path for the beam charge state and the stopping-power calculation. An illustration of stopping-power profile calculation is shown in Fig. 9 for the plasma conditions along the target central axis at \( t = 50 \) ps after the beginning of the laser target heating. The target density, temperature, ionization and free electron densities are shown in Fig. 9a, the corresponding \( T, \Theta \) and \( n_{\text{e}}/n_{\text{e}} \) values are shown in Fig. 9b, and the resulting stopping power for one proton of initially 500 keV energy is shown in Fig. 9c. The three ad hoc calculations provide almost identical results, consistently with Fig. 5. Meanwhile, the result of our KS-DFT fit (which is explained below) lies between the SRIM curve and the classical values. The energy loss for one proton at this time step is obtained as the integral of the represented stopping power. The energy loss values obtained for all time steps are then convoluted with the spatial and temporal profiles of the probing proton beam. For this purpose, a Monte–Carlo calculation is performed assuming a beam energy bandwidth of 44 keV at FWHM, i.e. a temporal width of 400 ps at FWHM, and a spatial width of 30 μm at FWHM. The simulation is performed with the approximate estimated proton number per bunch of 1000.

**TD-DFT stopping-power calculations.** The time-dependent orbital-free density functional theory (TD-OF-DFT)\textsuperscript{30,31} formulation included a nonadiabatic, temperature-dependent kinetic-energy density functional and an exchange-correlation contribution in a local density approximation as well as the usual Hartree and external terms with a local all-electron pseudopotential for carbon. Rectangular prisms of 512 atoms with dimensions 70.0 × 17.5 × 17.5 Å were employed as reference cells with the atomic configurations determined from an equilibrium orbital-free molecular dynamics simulation. For a given projectile velocity, the total electron stopping power is determined by the work on the proton as a function of the distance travelled averaged over 2–3 atomic configurations, 10–15 initial positions for the proton, and 3–4 passages of the proton through the cell. For the 10 eV case, TD-OF-DFT calculations with 2048 atoms and \( 70 \times 35 \times 35 \) Å cells were also performed to conclude that the approximate estimated proton number per bunch of 1000.

**XPHG and SOP diagnostic modelling.** For the comparison with the experimental XUV spectra obtained with the XPHG diagnostic, the PrismSPECT atomic code was used to postprocess target profiles extracted from the RALEF2D and MULTI-fs 1D hydrodynamic simulations. Using the RALEF2D simulation, we considered target profiles at radius \( r = 30, 60, 90, 120, 150 \) μm from the proton propagation axis (target center), over the expansion time of 0–300 ps, with time steps of 10 ps for \( t = 0–100 \) ps, and 50 ps for \( t = 100–300 \) ps. At each time step, the spatially-integrated emission spectrum was obtained by summing the area-weighted emissivity at each radius. The spectra were then integrated over time to obtain the total space- and time-integrated emission as measured by the XPHG diagnostic on the heater beam side of the target. As for the MULTI-fs simulation, we used target profiles of each of the four simulations with different heater intensities over the expansion time of 0–300 ps, with time steps of 10 ps for \( t = 0–100 \) ps, and 50 ps for \( t = 100–300 \) ps to calculate the emissivity with PrismSPECT. For obtaining an area-weighted emissivity at each time-step, a simple model was employed to numerically determine a radius for each average ring of intensity used in simulations and calculate the area. In order to compare the experimental temperature temporal evolution obtained with Streaked Optical Pyrometry, we considered the RALEF-2D expansion profiles averaged over the central 50 μm diameter area and the MULTI-fs expansion profiles of the simulation with the intensity that corresponds to the effective proton probe spot radius of 25 μm. For this purpose, we considered the temperature with a time step of 10 ps at the critical density \( n_{\text{e}} = 3.88 \times 10^{21} \) cm\(^{-3} \) that corresponds to the wavelength \( \lambda = 532 \) nm.
Fig. 9 Plasma conditions and stopping power along the plasma central axis at a time $t = 50$ ps. a Mass-density, electron temperature, free electron density and mean ionization degree. b Electron coupling $\Gamma$, electron degeneracy $\Theta$ and velocity ratio $v_p/v_e$ for a 500 keV energy proton. c Corresponding stopping-power profiles. The bound-electron contribution as well as the stopping power in the solid target according to the SRIM database are also represented. The x-axis is reported in areal-density units ($\mu g/cm^2$).

Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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References

1. Hurricane, O. et al. Approaching a burning plasma on the NIF. Phys. Plasmas 26, 052704 (2019).
2. Zylstra, A. et al. On alpha-particle transport in inertial fusion. Phys. Plasmas 26, 080706 (2019).
3. Callahan-Miller, D. A. & Tabak, M. Progress in target physics and design for heavy ion fusion. Phys. Plasmas 7, 2083–2091 (2000).
4. Hofmann, I. et al. Review of accelerator driven heavy ion nuclear fusion. Matter Radiat. Extremes 3, 1–11 (2018).
5. Both, M. et al. Fast ignition by intense laser-accelerated proton beams. Phys. Rev. Lett. 86, 436–439 (2001).
6. Fernández, J. C. et al. Fast ignition with laser-driven proton and ion beams. Nucl. Fusion 54, 054006 (2014).
7. Kim, J. et al. Self-consistent simulation of transport and energy deposition of intense laser-accelerated proton beams in solid-density matter. Phys. Rev. Lett. 115, 054801 (2015).
8. Kim, J. et al. Anomalous material-dependent transport of focused, laser-driven proton beams. Sci. Rep. 8, 17538 (2018).
9. Sharkov, B. et al. High energy density physics with intense ion beams. Matter Radiat. Extremes 1, 28–47 (2016).
10. Patel, P. et al. Isochoric heating of solid-density matter with an ultrafast proton beam. Phys. Rev. Lett. 91, 125004 (2003).
11. McGuffey, C. et al. Focusing protons from a kilojoule laser for intense beam heating using proximal target structures. Sci. Rep. 10, 9415 (2020).
12. Mancic, A. et al. Picosecond short-range disordering in isochorically heated dense aluminum at solid density. Phys. Rev. Lett. 104, 035002 (2010).
13. Ping, Y. et al. Heat-release equation of state and thermal conductivity of warm dense carbon by proton differential heating. Phys. Rev. E 100, 043204 (2019).
14. McKevey, A. et al. Thermal conductivity measurements of proton-heated warm dense aluminum. Sci. Rep. 7, 7015 (2017).
15. White, T. et al. Observation of inhibited electron-ion coupling in strongly heated graphite. Sci. Rep. 2, 889 (2012).
16. Mackinnon, A. et al. Proton radiography of a laser-driven implosion. Phys. Rev. Lett. 97, 045001 (2006).
17. Volpe, L. et al. Proton radiography of laser-driven impeding target in cylindrical geometry. Phys. Plasmas 18, 012704 (2011).
18. Hoffmann, D. H. H. et al. Energy loss of heavy ions in a plasma target. Phys. Rev. A 42, 2313–2321 (1990).
19. Dietrich, K.-G. et al. Energy loss of heavy ions in a dense hydrogen plasma. Z. Phys. D. At. Molec. Clust. 16, 229–230 (1990).
20. Belyaev, G. et al. Measurement of the Coulomb energy loss by fast protons in a plasma target. Phys. Rev. E 53, 2701 (1996).
21. Frank, A. et al. Energy loss and charge transfer of argon in a laser-generated carbon plasma. Phys. Rev. Lett. 110, 115001 (2013).
22. Gericke, D. O. & Schlanges, M. Energy deposition of heavy ions in the regime of strong beam-plasma correlations. Phys. Rev. E 67, 037401 (2003).
23. Cayzac, W. et al. Predictions for the energy loss of light ions in laser-generated plasmas at low and medium velocities. Phys. Rev. E 92, 053109 (2015).
24. Cayzac, W. et al. Experimental discrimination of ion stopping models near the Bragg peak in highly ionized matter. Nat. Commun. 8, 15693 (2017).
25. Frejje, J. A. et al. Experimental verification of low-Z ion-stopping formalisms around the Bragg peak in high-energy density plasmas. Phys. Rev. Lett. 122, 015002 (2019).
26. Gericke, D. et al. Stopping power of nonideal, partially ionized plasmas. Phys. Rev. E 65, 036406 (2002).
27. Edie, D. J. et al. a-particle stopping and electron-ion energy relaxation in highly compressed ICF fuel. Euro. Phys. J. Web Conference 59, 5013 (2018).
28. Deutsch, C. et al. Multiple scattering in electron fluid and energy loss in multi-ionic targets. Nucl. Instrum. Methods Phys. Rev. A 733, 39–44 (2014).
29. Ding, Y. H. et al. Ab initio studies on the stopping power of warm dense matter with time-dependent orbital-free density functional theory. Phys. Rev. Lett. 121, 145001 (2018).
30. White, A. T. et al. Time-dependent orbital-free density functional theory for electronic stopping power: Comparison to the Mermin-Kohn-Sham theory at high temperatures. Phys. Rev. B 98, 144302 (2018).
31. Hayes, A. C. et al. Reaction-in-flight neutrons as a test of stopping power in degenerate plasmas. Phys. Plasmas 22, 082703 (2015).
32. Hayes, A. C. et al. Plasma stopping-power measurements reveal transition from non-degenerate to degenerate plasmas. Nat. Phys. 16, 432–437 (2020).
33. Zylstra, A. B. et al. Measurement of charged-particle stopping in warm dense plasma. Phys. Rev. Lett. 114, 215002 (2015).
34. Chen, S. N. et al. Experimental evidence for the enhanced and reduced stopping regimes for protons propagating through hot plasmas. Sci. Rep. 8, 14586 (2018).
35. Ren, J. et al. Observation of a high degree of stopping for laser-accelerated intense proton beams in dense ionized matter. Nat. Commun. 11, 5157 (2020).
36. Zylstra, A. B. et al. Platform development for $dE/dx$ measurements on short-pulse laser facilities. High. Energy Density Phys. 35, 100731 (2020).
37. Jahn, D. et al. Focusing of multi-MeV, subnanosecond proton bunches from a laser-driven source. Phys. Rev. Accel. Beams 22, 013101 (2019).
38. Volpe, L. et al. Generation of high energy laser-driven electron and proton sources with the 200 TW system VEGA 2 at the Centro de Laseres Pulsados. High. Power Laser Sci. Eng. 7, 25 (2019).
39. Wilks, S. et al. Energetic proton generation in ultra-intense laser-solid interactions. Phys. Plasmas 8, 542 (2001).
40. Apiñaniz, J. et al. A quasi-monoenergetic short time duration compact proton source for probing high energy density states of matter. Sci. Rep. 11, 6881 (2021).
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Author contributions
L.V., S.M. and W.C. conceived the idea, designed the experiment and wrote the paper. L.V. and S.M. led the experiment. S.M. coordinated the analysis of the experimental results and wrote the simulation. W.C. coordinated the stopping-power calculations and performed the energy-loss simulations. J.V. provided support for the RPA stopping-power calculations. G.F. performed the QAMA stopping-power calculations. I.A.C., A.J.W., K.N. and S.X.H. provided theoretical support and performed the TD-DFT stopping-power calculations. A.T. performed the RALFED hydrodynamic simulation. S.M., L.V., W.C., R.F., J.A.P., I.J.A., V.O., K.B., M.B., X.V., C.M., M.H., F.N., G.P. and G.V. participated to the experimental campaign and the preparation campaign. S.M., J.A.I. and L.V. operated the proton detector diagnostic. S.M., C.M. and X.V. analyzed the proton diagnostic data. R.F. and K.B. operated the XPHG spectrometer and analyzed the XPHG measurements. V.O. operated the SOP diagnostic. V.O. and M.B. analyzed the SOP data. S.M. performed the modelling and the simulations for both plasma emission diagnostics. D.L., J.A.I. and G.G. built the magnetic selector. R.F., J.S., D.B., J.V., L.R. and F.N.B. provided theoretical support.

Competing interests
The authors declare no competing interests.

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