Study of the parameter dependence of laser-accelerated protons from a hydrogen cluster source

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Abstract

We present a study on laser-driven proton acceleration from a hydrogen cluster target. Aiming for the optimisation of the proton source, we performed a detailed parametric scan of the interaction conditions by varying different laser and the target parameters. While the underlying process of a Coulomb-explosion delivers moderate energies, in the range of 100 s of keV, the use of hydrogen as target material comes with the benefit of a debris-free, single-species proton acceleration scheme, enabling high repetition-rate experiments, which are very robust against shot-to-shot fluctuations.

1. Introduction

Laser-driven particle acceleration of electrons, protons and heavier ions became an intensively studied field of research within the last decades. Promising features, like extremely high field gradients in the range of teravolt per metre (TV m⁻¹) allow a compact design of the particle acceleration setup compared to conventional accelerators, delivering at the same time a very high particle flux and ultra-short pulses, which are desirable parameters for various research purposes as well as for applications in medicine or industry. Key applications are the time-resolved probing of transient electric fields [1, 2] or the irradiation of biological samples [3].

The acceleration of electrons by schemes like laser-wakefield acceleration [4, 5], mainly from gaseous targets, allows a high repetition-rate and debris-free environment. In contrast, the acceleration of protons and ions by schemes like target-normal-sheath acceleration [6, 7] or radiation-pressure acceleration [8, 9] is based on solid density targets, which come with several drawbacks. On the one hand, the target gets destroyed by every shot and needs to be replaced. Various techniques like tape-targets, arrays or re-growing thin foils were demonstrated by different groups [10]. On the other hand, the target material evaporated during the interaction causes debris and shrapnel, which damages optics. This results in a degradation of the focus quality and therefore in a reduction of the laser intensity. A way to circumvent this problem is the use of different target materials generating less debris. Besides the use of liquids, such as water [11] or alcohol, dispersed as droplets [12] or vapour [13], liquefied and solidified gases were used recently. Especially the use of hydrogen has the big advantage that only protons as a single species ion source are present in the target material. Different experimental schemes have been realised.

Hydrogen microjet targets were demonstrated, e.g. to study superfluidity in supercooled media by Grisenti et al [14]. A hydrogen droplet source for application in laser-plasma physics was developed by Costa Frage et al [15]. In the framework of laser-driven particle acceleration Gauthier et al [16, 17] used a jet for the purpose of accelerating protons, while a further investigation of the target-geometry dependence was studied by Obst et al [18]. A drawback of these kind of structures like droplets, jets or ribbons is on the one hand the position stability as e.g. discussed in [17], where the spatial jitter overcomes the diameter of the target. On the other hand, this goes along with the general requirement to align and focus the laser reliably onto a specific position in space. A way to overcome both problems is the use of a cluster target, which generates a spray of randomly distributed clusters within a certain volume, large compared to the laser focal volume and at the same time with a sufficiently low
density to have the laser propagating into the spray. For the cluster formation, a pre-cooled gas is dispersed by an orifice resulting in a spray of statistically distributed objects. First experiments were realised more than two decades ago by Ditmire et al [19, 20], and within the last years, an increasing interest in this technique could be noticed [21, 22]. We recently presented the first proof of a multi-Hertz, kiloelectronvolt pulsed proton source as a step towards a laser-target interaction scheme, capable for applications [23]. The drawback of this first test was the limitation in cluster size due to restraints in the cooling power within the cluster jet source.

In this manuscript, we present a detailed study using a new cluster jet source, capable of operating in the gaseous and liquid regimes of hydrogen in front of the de-Laval nozzle. In addition to the systematic scan of the target parameters (incident gas/liquid temperature and pressure), we performed several scans on the laser parameters, like polarisation, energy and pulse duration in order to find the best operating condition. Utilising the high repetition rate of the target source and the laser system enabled us to record significant data statistics for all the parametric scans.

2. Setup

A new hydrogen cluster target, specially developed to study laser-plasma interactions, was used for this experiment. This new target includes a separated vacuum system inside the nozzle head and offers manual accessibility to the interaction point from many directions. Those features allow a versatile studies on the interaction as well as on the accelerated particles and generated radiation.

Following the basic principle, as presented in Grieser et al [24], purified hydrogen is used. It is pre-cooled to temperatures \( T \) as low as 24 K by using a closed-looped helium cold-head and expanded with a backing pressure \( p \) up to 16 bar through a de-Laval nozzle. The constant flow allows a continuous operation of the cluster target. Note, that depending on the temperature and the pressure conditions the hydrogen in front of the nozzle is either in the liquid or the gaseous state. In case of a gas, the adiabatic expansion of the hydrogen ejected into vacuum results in a further cooldown, allowing to form condensation centres by three-body-interactions. On these centres, more molecules are bound due to the Van-der-Waals forces, forming macroscopic clusters.

The formation process can be described empirically by the so-called Hagena-relation [25]. Based on the temperature, pressure, gas parameters and nozzle geometry, this relation gives an estimate on the cluster-formation ability and the cluster size. In order to calculate the cluster size in this regime, we calculated the Hagena-parameter \( \Gamma \) and the number of atoms per cluster \( N \) using [25]:

\[
\Gamma(p, T) = \frac{k \cdot p}{T^{2.29}} \left( \frac{0.74 d_0 (\mu m)}{\tan \alpha / 2} \right)^{0.85}
\]

and

\[
N(p, T) = A_N \cdot \left( \frac{\Gamma}{1000} \right)^{\gamma_N}.
\]

For hydrogen gas, the empirical parameters in the equations above are \( k = 184; A_N(\Gamma > 1800) = 33 \) and \( \gamma_N(\Gamma > 1800) = 2.35 \) (based on [26]) while for the given nozzle \( d_N = 42 \mu m \) and \( \alpha_{1/2} = 3.5^\circ \) are used. In a final step, the cluster diameter \( d_{Cluster, \, Hagena} \) is calculated, assuming a sphere with the number of atoms calculated in equation (2) and with the density of liquid hydrogen \( \rho_{\text{liq.} \, H_2}(16 \text{ K}) = 76.3 \text{ kg m}^{-3} \). Note, the values derived above denotes the average cluster size, while the full size-distribution is described by a log-normal function [27].

For the conditions of liquid hydrogen in front of the nozzle, the formation of a spray is described by a breakup of the liquid. The process is characterised by the Weber number \( \text{We} \) [28]. For the given nozzle geometry in the temperature range between 24 and 34 K we find \( \text{We} > 1.25 \times 10^4 \), which indicates that the breakup is induced by atomisation. Two general processes cause atomisation of a liquid in a de-Laval nozzle. First of all, the pressure gradient \( \Delta p \) occurring along the nozzle leads to a change of the capillary-pressure within the liquid, which induces Kelvin–Helmholtz instabilities, causing a primary breakup. Second, a further breakup occurs due to the aerodynamic force when the liquid interacts with the surrounding media. In our case, dispersing the liquid hydrogen into a vacuum-chamber with \( p < 10^{-1} \text{ mbar} \), the aerodynamic force can be neglected. The upper limit of the droplet size, in this case, is described by [29]:

\[
d_{\text{Cluster, atom}}(T) = 2 \cdot \frac{\sigma(T)}{\sqrt{\Delta p}}
\]

with the temperature dependent surface tension \( \sigma(T) \) and \( \Delta p = 10^8 \text{ Pa m}^{-1} \) for the nozzle geometry used. The total size distribution of the generated particles follows a logarithmic distribution, as described e.g. by Mugele et al [30]. Other processes are also occurring, of which only a few are understood and analytically described so far.
A more general overview of the field of liquid atomisation can be found, in various publications on the design and performance of diesel-fuel injector-nozzles for engines [31].

We applied the method of Mie-scattering as described in [24], deducing the cluster/particle size by characterising the angular dependent scattering of light from the cluster jet for different incident polarisations. For the Mie-measurement, the scattering signal increases nonlinearly with the particle size, which indicates, that due to the above mentioned log-normal distribution of particles, the measured size might be higher than the calculated average size. The analytical and experimental results are shown in figure 1. The Hagena-relation predicts in the gaseous regime, that a variation of nozzle temperature (green dots) or gas pressure (blue dots), results in a change of the cluster size in the range between 10 and 80 nm. Note that a variation of the temperature causes a more significant change in cluster size compared to a variation of the pressure. Generally, a lower temperature or a higher pressure leads to the formation of larger clusters. In the liquid regime, particle diameters in between 1 and 10 μm are predicted by the process of atomisation.

The results of the Mie-scattering measurements are represented by solid red dots. As can be seen in figure 1, the measured values in the gaseous regime are in agreement within a factor of 1–2 with the prospect of the Hagena-relation. However, in the liquid regime, the atomisation model predicts three orders of magnitude larger particles than the Mie-scattering measurement. We further investigated this discrepancy using the laser focus diagnostic employed in the experimental setup, imaging the central cluster plane, as shown in figure 2. With an optical resolution of about 2 μm, this imaging system should be sufficient to resolve the predicted particle size by the atomisation process, while the small clusters observed by the Mie-scattering measurement are undersized and should not affect the measurement. Employing the attenuated and defocused laser-beam as a backlighter, we recorded snapshots and could identify μm size particles for different temperature settings in the liquid regime. Note that in the gaseous regime no particles were observed. As shown in the blue insert of figure 2(b), those particles can be optically resolved within the depth of field of the used objective. Knowing the magnification factor enabled us to determine the absolute size of those particles, given by the hollow red dots in figure 1. The measured size of those large particles is in good agreement with the prediction by the atomisation model. Furthermore, knowing the particle size from above, the field of view of the objective and the laser focal volume, the average distance in between the particles and the minimum number of shots in order to hit such a particle can be calculated. While the large clusters are on an average more than 280 μm apart from each other, the interaction probability (with at least 95% confidence) was estimated to be under best conditions in between ≈1:150 for 25 K and ≈1:6 × 10^4 at 33 K. A further comparison will be done in section 4 taking also the measurements of the cut-off energies into account.

The experiment was carried out at the ARCTURUS laser system of the University of Düsseldorf, which is a Ti:sapphire based chirped pulse amplification laser system. Up to 7 J of energy before compression, at a 30 fs pulse duration in a 5 Hz operation mode can be delivered. Utilising an XPW contrast-enhancing unit, the laser contrast was measured to be 1 × 10^-8 at 80 ps before the main pulse [32]. A thin (1 mm) anti-reflex coated pellicle-window was implemented before the interaction chamber, to protect the compressor gratings from
hydrogen gas or other hydro-carbon contamination of the pump-system. The 10 cm diameter laser beam was focused by an $f/2$ off-axis-parabolic mirror to a focal spot diameter of $\omega_0 = (5 \pm 0.4) \mu m$ and a Rayleigh-length of $z_R \approx 30 \mu m$. Taking the overall energy throughput of $(31 \pm 2) \%$ from the laser amplifier exit up to the interaction chamber into account, a maximum intensity of $1 \times 10^{20} W/cm^2$ was achieved on target. The incident beam was p-polarised with respect to the breadboard of the target chamber. Figure 2 shows the experimental setup. The cluster jet nozzle is about 28 mm upstream of the interaction point, which is in the centre of the chamber. The cluster beam is dumped into a multi-stage root-pump system, keeping the pressure in the interaction chamber at --101...3 mbar level depending on the operating parameters of the cluster source. The proton beams in 0° and 45° directions, with respect to the laser axis, were simultaneously detected with two Thomson-parabola (TP) spectrometers, coupled to micro-channel plate (MCP) detectors, read out by CCD cameras. Both, the TP and the MCP were situated in separately pumped vacuum vessels, at a pressure of $p < 2 \times 10^{-5} mbar$, only connected by a small entrance pinhole to the main interaction chamber. The proton spectra were evaluated using a Matlab routine. Since protons are the only ion species present, the TPs were equipped with a magnetic field only, enabling a good energy resolution in the range of 20–300 keV.

3. Results

For the different measurements obtained for the source- and laser- parameter scans, every data point is the average of at least five consecutively recorded shots. The cut-off energy was determined for every spectrum, and the error bars denote the standard deviation in between the shots. First a variation of the source parameters was performed.

3.1. Variation of the cluster-source parameters

The cluster-source has two main parameters which can be varied, i.e. gas temperature and pressure. The temperature can be controlled by an electric heater, implemented in the cold-head. While the closed-loop helium-cryostat operates at a constant cooling power, the gas flow and the heating element induce thermal energy to regulate and stabilise the desired temperature in an equilibrium condition. Figure 3(a) shows a measurement of the proton cut-off energy depending on the cluster-source temperature, for p-polarised light at a backing pressure of 16 bar in 0° and 45° directions, with respect to the laser axis, were simultaneously detected with two Thomson-parabola (TP) spectrometers, coupled to micro-channel plate (MCP) detectors, read out by CCD cameras. Both, the TP and the MCP were situated in separately pumped vacuum vessels, at a pressure of $p < 2 \times 10^{-5} mbar$, only connected by a small entrance pinhole to the main interaction chamber. The proton spectra were evaluated using a Matlab routine. Since protons are the only ion species present, the TPs were equipped with a magnetic field only, enabling a good energy resolution in the range of 20–300 keV.
3.2. Variation of the laser parameters

In addition to the study of the cluster source parameters, the dependency of the laser parameters on the interaction (i.e. incident polarisation, energy and pulse duration) was investigated. Therefore the proton signal was recorded with the TP in the laser forward direction ($0^\circ$ direction).

Figure 4 shows the averaged proton energy spectra for 50 consecutive shots each, obtained with linear- (p-pol) and circular- (c-pol) polarised light. As the second parameter, the cluster size was varied, changing the temperature at a fixed backing pressure of 16 bar. As can be seen in figure 1, in this temperature range which corresponds to cluster diameters in between $d_{\text{Cluster}}(34 \text{ K}) \approx 113 \text{ nm}$ and $d_{\text{Cluster}}(40 \text{ K}) \approx 84 \text{ nm}$ respectively, the most significant change in the proton cut-off energy occurs. At the same time, the difference between the two polarisation states is less than 10%, i.e. not following a clear trend, that one polarisation is preferable in terms of energy enhancement.

In the second scan, utilising p-polarised light, the laser energy was varied by reducing the number of pump-lasers in the last amplifier stage. Figure 5 shows the average proton cut-off energy depending on the laser energy measured for different temperatures. Note that in the gaseous state a direct correlation between temperature (cluster size) and cut-off energy was found (figure 3(a)). The maximum cut-off energy for higher temperatures (smaller clusters) can be reached with less laser energy, compared to the case of lower temperatures (bigger clusters).
As the on-target intensity was lowered previously by reducing the laser energy, for the third scan the laser energy was kept constant, and the pulse duration was varied by de-tuning the pulse-compressor. Figure 6 shows the cut-off energy dependence for different pulse durations in between 30 and 1000 fs, assuming a timing error of 10%.

4. Discussion and numerical results

We performed a series of three-dimensional particle-in-cell simulations (PIC) employing the EPOCH code [33] to study the laser-cluster interaction and further explain our experimental results. The targets have been initialised as neutral hydrogen spheres with a diameter of 60 nm. Taking an atomic density of $4.56 \times 10^{22}$ cm$^{-3}$ corresponding to the liquid hydrogen density into account, this complies to 364 atoms/cell. The relatively low number of atoms in one cluster, here about $4.7 \times 10^6$, allows a 1:1 representation of the macro-particle to the real-particle. Starting with cold clusters, the barrier-suppression tunnel-ionisation model used by the code, was employed.

A single cluster is placed at the centre of the simulation box ($\Delta x = \Delta y = \Delta z = 300$ nm) while the laser pulse is initialised as a spatial plane wave with a wavelength of $\lambda = 800$ nm at the $x = -150$ boundary of the...
box, propagating in the positive x-direction. This approximation is justified since the focal spot in the experiment with a diameter of 5 μm is large compared to the size of the cluster. Figure 7(a) shows the motion of the electrons, induced by a linear or circular polarised, temporally Gaussian laser-pulse with a duration of τ = 25 fs (FWHM) at a maximum laser intensity of I_{max} = 1 \times 10^{20} \text{ W cm}^{-2}. For both cases, the laser-electric field strips away the electrons from the cluster within the first two oscillation periods (t < 2 T_0) of the electric field, which is in agreement with our previous observations. The Coulomb-explosion (CE) of the remaining ion core takes place on timescales of several tens of fs as displayed in figure 7(b). Here, the recording of the maximum proton energy is limited due to the size of the simulation volume.

For further understanding of the experimental results, simulations for the case of linear polarised light were performed, varying the pulse duration (25 fs; 100 fs; 300 fs (FWHM)) and the maximum laser intensity (I_{max} = 1 \times 10^{20} \text{ W cm}^{-2}; 5 \times 10^{19} \text{ W cm}^{-2}; 1 \times 10^{19} \text{ W cm}^{-2}). For comparison we used the experimentally obtained data for the cut-off energies in the 0°-direction of the energy scan (figure 5) and the pulse duration scan (figure 6). Note that we calculated the corresponding intensities for a normalised x-axis to present the results in figure 8.

For both, experiment and simulation, a reduction of intensity by an increase of the pulse duration results in lower proton energies, compared to a similar intensity reduction by decreasing the laser energy. From the simulations, this can be correlated to the longer rising-edge time of the electric field coming along with a long pulse duration. If the field ramps up slowly, the electrons are not removed quasi-instantaneously, as e.g. shown in figure 7. This results in a lower initial charge of the cluster, at the onset of the CE. By the time, all electrons are removed from the cluster, the cluster core has already started to expand, thus the distance between the ions is
larger compared to the ideal case, resulting in a smaller repulsive potential and therefore a lower maximum proton energy.

In parallel to the numerical simulations, for the ideal case of a (quasi-) instantaneous removal of all electrons, an analytical model of a homogeneously charged sphere with radius $R$, containing a number of $N$ atoms of charge $q$ can be applied to describe the CE. The electric potential $\Phi$ at a position $r$ along the sphere is given by:

$$
\Phi(r) = \frac{Nq}{4\pi \varepsilon_0 R} \left( \frac{3}{2} - \frac{r^2}{2R^2} \right).
$$

The maximum energy $E_{\text{max}}$ which can be gained by CE, is the energy of a particle of charge $q$ situated at the surface of the sphere ($r = R$) which is given by:

$$
E_{\text{max}} = \frac{Nq^2}{4\pi \varepsilon_0 R}.
$$

In the gaseous operation regime of the cluster-jet source, equations (1) and (2) are used to calculate the expected cluster size and in a second step, employing equation (5), the corresponding maximum proton energy. Figure 9 shows a comparison of the experimental data for the temperature and pressure scan with the results of the analytical model. As can be seen, especially for the temperature scan (green), the trend is well reproduced, while the analytical calculated maximum energy overestimates the measured cut-off energy constantly by 50–70 keV over the full range. This can be caused by different reasons. As mentioned above, in the analytical model assuming a charged sphere, the electrons are removed quasi-instantaneously, while in the experiment the laser electric field needs a few oscillation periods of the electric field to remove all electrons, as can be deduced from figure 7. During this time, the onset of expansion of the ion-core will reduce the maximum achievable proton energy. Another simplification in the analytical model is the estimation that the cluster is a sphere with the homogeneous density of liquid hydrogen. Since these nm size clusters contain about $10^5–6$ atoms, they might be described as a mesoscopic object, for which for example effects of reduced bonding of the surface molecules lead to an effectively lower density and therefore a reduced CE.

In the liquid regime, a discrepancy between the analytically predicted and optically measured spray sizes for the atomisation process and the results of the Mie-measurements occurred, as already seen in figure 1. While the large particles are optically detectable, it was revealed at the same time, that the number of those particles would be very small and therefore the likelihood of an interaction with the laser is small. This can be directly confirmed by the measurement of the proton energy spectra. In the liquid regime, only low energetic protons with energies in between 175 and 220 keV were measured, originating from the interaction of the laser with the small clusters, represented by the Mie-scattering signal. For a cluster size of 4–8 μm, the analytic CE-model (equation (5)) would predict particle energies in the GeV-range, assuming that the laser electric field could remove all the electrons. At a certain point the attractive potential of the remaining core will pull the electrons back, which limits the maximum energy achievable by CE. Nevertheless, energies in the MeV-range are possible, as demonstrated in the aforementioned work by [22]. Within all the measurements in the liquid regime there was not a single spectrum with significantly higher proton energy, pointing towards an interaction with one of the
large particles. Furthermore, for lower temperatures the cut-off energy slowly decreases, which indicates that the nm-sized clusters must originate from a different particle population with a different statistical distribution, compared to the μm size particles. While the size of the large particle increases with lower temperature, the size of the small cluster decreases with lower temperature.

5. Conclusion

Employing a new hydrogen cluster target, specially designed to study the laser-cluster interaction, detailed experimental scans of the laser- and cluster-parameters influencing the interaction were performed. PIC simulations and an analytical model confirmed the underlying mechanism of CE as a dominating effect over a wide intensity range. Operating the source in the gaseous regime, the resulting proton beam is very stable in energy with low shot-to-shot fluctuation but at the same time, the energy can be tuned quickly by the temperature and pressure settings of the target system. In the liquid regime, the process of atomisation, resulting in larger particles, can be observed. However, it was revealed in the same measurement, that the occurrence of those particles is significantly smaller resulting in a low interaction probability. The current combination of the cluster jet source and the given focal volume is not suitable to aim routinely for higher energies from μm size hydrogen droplets since the interaction-probability is too low. This result was confirmed by the actually measured accelerated proton energies, where no energy increase could be observed within the given statistic. While the proton energies are still in the moderate range of 100 s of keV, this system in its current stage is a versatile tool for research, delivering ultra-short bunches of protons by a continuous and debris-free generation process, from compact laser-systems at medium intensities. A further optimisation, either by an increase of the μm size droplet density or a larger focal volume could allow the reproducible production of shot-to-shot stable proton beams in the MeV-range.

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