Title
Acceleration of high charge ion beams with achromatic divergence by petawatt laser pulses

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I. INTRODUCTION

Laser-driven acceleration of ion beams [1–3] has great potential for numerous applications such as heavy ion therapy of oncological diseases [4], injectors for conventional accelerators [5], radiography [6], studies of radiation damage [7] and single event effects in electronics [8,9], as well as fast ignition inertial confinement fusion [10–13], and drivers and probes for the studies of warm dense matter [14,15]. Such ion beams are accelerated in the form of neutralized bunches by means of spatially localized and temporally synchronized fields from relativistic laser plasma interactions, and exhibit exceptional beam properties [6] (e.g., low emittance, short bunch duration and high charge).

For applications including those above and detailed physics studies of the relativistic laser-plasma interaction, high repetition rates (Hz to kHz) are essential. During the past few years, several petawatt laser systems with short pulse duration (< 100 fs) [16] and high repetition rate (up to 1 Hz [17]) have become available. Such short pulse lasers are scalable to higher repetition rates and have a much smaller footprint. Thus, the path towards multipetawatt systems with intensities in excess of 10²² W/cm² (required to reach e.g., 400 MeV/u carbon ion energies for deep tumor carbon therapy) becomes much more cost effective [18].

The acceleration mechanism studied most extensively—target normal sheath acceleration (TNSA) [19–21]—has matured to a high level of robustness and reliability. This has made studies possible that cover a wide range of laser and target parameters from many different experiments [2,3]. However, not all the laser-plasma and ion parameters can be measured at the same time in a single experiment, especially at petawatt-class laser systems where shot rates have historically been at or below a few shots/hour [16]. Providing comparable experiment conditions at different facilities is hardly possible, mainly due to the unique temporal intensity structure of each laser system. To date, proton energies of up to 85 MeV [22] with TNSA, or 94 MeV protons [23] and 1 GeV carbon ions [24] with advanced mechanisms were generated using petawatt lasers with pulse durations exceeding 0.5 ps at ultrarelativistic intensities of ~10²⁰ W/cm². Proton beam divergences in TNSA are chromatic with typical values ranging from 400 to 900 mrad at half of the maximum energy.

We report on the generation of proton beams with achromatic, narrow divergence at unprecedented charge density by reducing the curvature of the hot electron sheath field on the rear side of the target. In contrast to earlier work [25,26], this is achieved by increasing the laser spot size 2w₀ incident on the targets front side to values well above the target thickness (l ≪ w₀), with w₀ being the radius at 1/e² of the peak intensity. As the laser spot size is increased (at constant laser intensity) and hence, the virtual source size of the ion beam, the number of accelerated ions is increased accordingly. If the laser is incident at a 45 degree angle to maximize laser absorption, the laser-target interaction is prolonged by 2w₀/c, where c is the speed of light, as the laser sweeps across the target. In the large spot size regime, this prolongation of the interaction is significant as it exceeds the initial laser pulse duration by a factor of 6.
Geometric considerations suggest that the initial laser pulse duration has to be increased to $2w_0/c$ to optimize the acceleration process as well as generating a flat-top-shaped electric field that leads to an achronmic divergence.

These findings are enabled by the first study of TNSA with statistical significance at petawatt laser power, consisting of several hundred shots. We applied a tape drive target system [27] that allows for conducting the experiments with titanium (Ti) tape of 5 μm thickness at repetition rates up to 0.5 Hz. Such ion beams are well suited for subsequent high gradient and emittance preserving beam transport which typically have narrow angular acceptance, such as active plasma lenses [28] and many applications in cell biology, high energy density physics and material sciences.

II. EXPERIMENTAL RESULTS

For these experiments, laser pulses with durations $t_L = 35–700$ fs at a central wavelength $\lambda_L = 815 \pm 20$ nm were generated at the BELLA PW Ti:sapphire laser [17] with a repetition rate of 1 Hz. The laser pulses were focused using a 13.5 m focal length off-axis parabolic mirror to a focal spot size of $w_0 \approx 52$ μm. Deformable mirror (DM) based feedback technology enabled a high focal spot quality with a Strehl ratio $> 0.8$. The laser energy delivered at the focal location was $E_L = 35$ J, corresponding to peak intensities $I_L$ in the range of $I_L = 0.6–12 \times 10^{18}$ W/cm$^2$ (depending on laser pulse duration), i.e., in terms of the normalized laser vector potentials $a_0 = 0.5–2.4$. Freestanding Ti foils of thickness $l = 1–10$ micron were irradiated at a 45 degree angle of incidence in the polarization plane of the laser. The accelerated ion beams were analyzed using a Thomson parabola spectrometer in target normal direction. The characteristic parabolas were visualized by a CCD coupled to a microchannel plate (MCP) with phosphor screen [29].

The measured maximum energies for proton and carbon (C$^{4+}$) ions are plotted in Fig. 1(a). For each thickness, the focus location relative to the target surface was varied within a Rayleigh length of the laser using the DM as shown in Fig. 1(b) exemplary for the 10 micron target. The highest ion energies were obtained with the focal plane close to the target surface, i.e., with the highest laser intensity. An additional five shots were taken at the optimum focus location for each thickness for the data in Fig. 1(a). The highest energies for protons and C$^{4+}$ ions were approximately 7 and 8.5 MeV, respectively.

A feedback-controlled tape drive system [27,30,31] spooling Ti tape of 5 micron thickness (and 100 mm width) was employed to increase the number of shots, enabling ion beam generation near the repetition rate of the laser. With this tape drive, uninterrupted operation at up to 0.5 Hz was possible for hours of run-time, but was limited to several hundred shots due to maximum length of the Ti tape (5 m). The tape position was referenced by polished metallic rods and it was tensioned before each shot to provide a wrinkle-free surface with a surface position repeatability $< 10$ micron [27]. In Fig. 2(a) proton spectra

**FIG. 1.** Measured maximum ion energies for proton (blue) and carbon (C$^{4+}$) ions (red) in MeV as a function of Ti-target thickness (a) and relative focus-target distance (b) adjusted with the deformable mirror for the 10 μm target. The vertical error bars represent the standard deviation from shot-to-shot variation over five consecutive shots [(a) and (b)]. The horizontal error bars represent the accuracy of the focus positioning using the DM (b).

**FIG. 2.** Results from tape drive: (a) Waterfall plot of proton spectra as a function of relative compressor separation, i.e., laser pulse duration (positive grating separation corresponds to negative chirp and vice versa). The green line gives the integrated number of protons with energies above 3 MeV. (b) Proton (blue line), C$^{4+}$ (red line) spectra averaged over 15 shots for a pulse duration of 120 fs and proton counts from radiochromic film (RCF) on the spectrometer axis (blue squares). The standard deviation from shot-to-shot variations is shown in grey for each spectrum.
FIG. 3. Processed radiochromic film data: proton beam profiles originating from 5 micron (a) and 2 micron (b) thick Ti targets. The energy of the fully stopped protons is given in MeV for each film layer. The deposited dose (Gy) in each layer is given by the color scale of the individual image. The divergence was derived as the FWHM of a fitted Gaussian profile to the measured dose distribution and shown for 5 micron (green squares) and 2 micron (purple circles) for each layer, i.e., proton energy in (c). The solid lines are piecewise linear fits of the data. The visible holes in the RCF film do not correspond to the target normal direction.

are shown as a function of laser pulse duration, ranging from 35 to 660 fs obtained with the Ti tape drive target system. The laser pulse duration was varied by changing the relative grating separation in the final vacuum compressor and thereby adding positive or negative chirp to the laser pulse. The maximum proton energy increased linearly with decreasing laser pulse duration, starting with 4.5 MeV at 660 fs and reaching an optimum of 7.5 MeV at around 180 fs. For shorter pulse durations, the energy decreased to 6 MeV at 35 fs. This behavior was independent of the laser chirp direction within our experimental accuracy.

The deviation from a linear intensity scaling [32–34] for laser pulses shorter than $2w_0/c$ can be illustrated by considering the following three cases at a 45 degree angle of incidence: (i) $t_L \ll 2w_0/c = 204$ fs (FWHM): The laser interacts with the target and heats the plasma very localized ($\sqrt{2t_L}c$) as it sweeps across the focal plane. At the time when the spatiotemporal intensity peak arrives at the target, the interaction is already terminated at the leading edge. The interaction time at a single point on the surface is insufficient to create plasma profile that allows for efficient coupling. (ii) $t_L = 2w_0/c$: After the initial sweep across the focal plane, the target is illuminated and heated uniformly. (iii) $t_L \gg 2w_0/c$: The scenario is similar to (ii) but at reduced laser intensity and hence, reduced electron temperature $T_e$ according to $T_e \propto a_0$ (see Fig. 2).

Two-dimensional energy-resolved proton beam profiles were measured for target thicknesses of 5 and 2 micron [Figs. 3(a) and 3(b)] by means of radiochromic film (RCF) stacks at a laser pulse duration of 100 fs. Those RCF stacks ($5 \times 5$) cm² were positioned normally 10 cm downstream of the target rear surface and consisted of eight layers GafChromic film, allowing sufficient sensitivity and resolution for the whole detectable energy range of the proton beams. A 15 micron thick aluminum foil was placed in front of the RCF stacks to block laser light as well as heavy ions. Each of the individual films of the layer records the deposited energy and the whole stack constitutes an energy resolved measurement. The dose response of the films and scanner unit was calibrated at the NDCX-II [35] facility with proton beams at 1.1 MeV. Reconstructed proton beam profiles are shown in Fig. 3 for shots on targets with 2 and 5 micron thickness up to the seventh layer of RCF, corresponding to proton energies of 8.8 MeV. Similar to [36], the spatially resolved proton spectra were reconstructed from the single shot dose distribution and is in agreement with our Thomson spectrometer data [see Fig. 2(b)]. In case of the 5 micron targets, the divergence decreases linearly with energy up to 5.4 MeV, followed by a constant divergence of 100 mrad for energies ranging from 6.3 to 8.8 MeV. Such an achromatic divergence was observed as well for 2 micron targets with an earlier onset of the plateau for energies $\geq 3.2$ MeV at a divergence of 125 mrad. This is a direct consequence of the larger laser spot size compared to the accelerating distance, i.e., $w_0 \gg \lambda_D$. The contribution to the divergence by scattering of the hot plasma electrons propagating through the solid target and their recirculation [37] is increased for thicker targets. As a result, the rear-side sheath field is transversally extended, leading to reduced proton beam divergence for thicker targets. Additionally, the dose deposited in the individual layers is almost twice as high for the 2 micron shot compared to the 5 micron shot with a total of $(7 \pm 2) \times 10^{11}$ protons with energies $\geq 1.2$ MeV.

Compared to other published experiments with PW lasers in the TNSA regime, the proton beam divergence was significantly reduced by a factor of 3–5 [36], reaching values of advanced acceleration schemes [38–40]. Together with the increased proton numbers, the charge density reached values so far only accessible with single shot laser systems and/or advanced acceleration mechanisms [3].

III. PARTICLE-IN-CELL SIMULATION

The relativistic fully electromagnetic particle-in-cell (PIC) simulation code, WARP [41,42], was used to model
The electron density of the target was initialized preformed plasmas with exponential density profiles and scale lengths $L$ of $0.01\lambda_L$ and $0.01L_0$, at the front and back side, respectively, at a temperature equal to zero. To account for surface contamination, we added contaminant species, protons and C$^+$ ions on both sides of the target, with electron densities of $n_e = 50n_{\text{crit}}$ for the protons and $100n_{\text{crit}}$ for the carbon ions. Both have exponential profiles with the same gradient scale length $L$. Convergence tests were used to determine numerical parameters, resulting in use of 120, 40, and 20 particles/cell for Ti, carbon, and protons, respectively and electrons/cell according to the ionization state given above. The center of the laser was initialized ahead of the target at $z = 96\, \mu m$ and hence, the spatiotemporal peak of the laser arrived at the target at $t = 614\, \text{fs}$. The simulation results are shown in Figs. 4 and 5.

FIG. 4. (a) Simulated Energy vs angle distribution of protons in logarithmic color scales. (b) Sweeping effect shown schematically for three different laser pulse lengths as indicated in the figure. (c) Acceleration time ($t_{\text{acc}}$) defined as the $1/e$ duration of $E_z$ acting on the protons with highest energy (see inset) as a function of a laser pulse duration (blue squares). Estimated interaction time $2w_0/c + t_{\text{acc}}$ (blue line). Impulse experience by a proton, electric field $E_z \times t_{\text{acc}}$ (black circles). (d) $E_z$ as a function of simulation time at different transversal locations $x$ as indicated in the figure for laser pulse durations of 30 fs (blue), 140 fs (red) and 300 fs (green). (e) Maximum proton energy for laser pulse durations of 30 fs (blue), 140 fs (red) and 300 fs (green) as a function of $L$.

The size of the simulation box was ($L_z, L_x$) = $(146\, \mu m, 357\, \mu m)$ and the cell size was $0.0121\lambda_L \times 0.0242L_0$, where $\lambda_L$ is the laser wavelength. The main target was composed of Ti$^{1+}$ ions and electrons, initialized at $z = 96\, \mu m$ with a thickness of 1 $\mu m$ along the $x$ direction. The electron density of the target was $n_e = 300n_{\text{crit}}$, where $n_{\text{crit}} = e_0m_e\omega^2/e^2$ is the electron critical density, with $e_0$ the vacuum permittivity, $m_e$, $e$ electron mass and charge and $\omega$ the laser frequency.

To account for the effects of laser prepulses, we initialized preformed plasmas with exponential density profiles and scale lengths $L$ of $(0.05)\lambda_L$ and $0.01\lambda_L$, at the front and back side, respectively, at a temperature equal to zero. To account for surface contamination, we added contaminant species, protons and C$^+$ ions on both sides of the target, with electron densities of $n_e = 50n_{\text{crit}}$ for the protons and $100n_{\text{crit}}$ for the carbon ions. Both have exponential profiles with the same gradient scale length $L$.

FIG. 5. (a) Evolution of longitudinal electric field $E_z$ and electron temperature $T_e$ at three different times during the interaction with $L = 0.01\lambda_L$, shown for three laser pulse duration in (a) 30 fs, (b) 140 fs and (c) 300 fs. The time $t = 636\, \text{fs}$ corresponds to the peak time of the spatiotemporal of the laser pulse at the front side of the target.
obliquely incident [Fig. 4(b)]. The temporal evolution of $E_z$ and $T_e$ (Fig. 5) demonstrates this sweeping effect. Where electron heating occurred very localized in case of the 30 fs laser pulse, it was homogenized and most effective in the 140 fs case that meets the condition $t_L = 2w_0/c$. Here, $E_z$ was almost constant along the transverse coordinate $x$ at the time of the maximum spatiotemporal laser intensity ($t = 636$ fs). This is also evident in the lineouts of the temporal evolution of $E_z$ in the field at different transverse coordinates $x$ [Fig. 4(d)]. To quantify this effect, we defined the acceleration time $t_{acc}$ as the $1/e$ duration of $E_z$ [see inset Fig. 4(c)], experienced by the high energy protons and compared it to the laser target interaction time given by $2w_0/c + t_L$ in Fig. 4(c). We found very good agreement for $t_L \geq 2w_0/c$ and $t_{acc} \ll 2w_0/c + t_L$ for $t_L < 2w_0/c$. The latter is due to transversely localized interaction of the short laser pulse. The impulse the proton experienced $\propto (E_z \times t_{acc})$ is shown in Fig. 4(c) for $L = 0.01\lambda_e$ in case of preplasma gradients with $L \leq 0.02\lambda_e$, the interaction time is the dominant factor, since laser absorption mainly occurs in plasma created by the main laser pulse. For longer preplasma scale lengths, the maximum $E_z$ field amplitude becomes more important, i.e., the initial preplasma conditions dominate the laser absorption process.

IV. CONCLUSIONS

In conclusion, we have presented an experimental study of TNSA with large laser spot size, where an increased laser focal spot leads to sheath field geometries that accelerate ion beams of narrow and achromatic divergence at very high charge densities. In order to optimize the laser pulse duration, laser pulse sweeping across the target needed to be considered which leads to increased interaction times and optimum laser pulse duration of $2w_0/c$. Such ion beams were generated at repetition rates of up to 0.5 Hz, for the first time with lasers at petawatt peak power. It was shown that the accelerator performance allows tailoring of particle energy and numbers to the requirements of specific applications in the field of cancer cell biology, high energy density and material sciences. In particular, this could for the first time enable emittance preserving ion beam transport with a high-gradient active plasma lens to achieve a high ion peak fluence at nanosecond pulse durations.

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