Experimental observation of transverse modulations in laser-driven proton beams

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Abstract

We report on the experimental observation of transverse modulations in proton beams accelerated from micrometer thick targets which were irradiated with ultra-short (30 fs) laser pulses of a peak intensity of $5 \times 10^{20}$ W cm$^{-2}$. The net-like proton beam modulations were recorded using radiochromic film and the data suggest a dependence on laser energy and target thickness for their onset and strength. Numerical simulations suggest that intensity-dependent instabilities in the laser-produced plasma at the target front side lead to electron beam break-up or filamentation, then serving as the source of the observed proton beam modulations.

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1. Introduction

The advent of a new generation of high repetition rate petawatt laser systems, in combination with recent experimentally achieved proton energies of up to 45 MeV from ultra-short pulse ($\leq 50$ fs) facilities [1, 2], should advance the application of laser–plasma based accelerators. In addition to the demand for increased proton energies, a key challenge in the field is the development of methods to spectrally tailor the accelerated proton or ion distribution at

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the source. Application experiments presently performed with laser-accelerated protons such as proton radiography [3–6] or radiobiological studies [7–13] would immediately benefit therefrom. In the case of the latter, for instance, the dosimetry protocols applied rely on a stable spectrally shaped proton distribution, currently guaranteed through passive filtering at the cost of experimental flexibility and achievable dose rate [8, 14, 15]. Several promising approaches to spectral tailoring have been realized, among them target preparation [16–18] and laser pulse modification methods [19, 20]. Moreover, novel acceleration schemes such as radiation pressure acceleration are predicted to inherently produce particle energy distributions with quasi-monoenergetic features [21–27].

At the ultra-short pulse Draco laser, using laser intensities exceeding $10^{20}$ W cm$^{-2}$ and micrometer thick foils as interaction targets, the present work was performed in the regime of target normal sheath acceleration (TNSA) [28], leading to exponential proton energy spectra [8, 29]. Aiming at further development of our ongoing radiobiological studies [13], we investigated the influence of picosecond prepulses on the spectral distribution for different micrometer thick targets and laser energies, similar to the approach of Dollar et al [20]. Depending on the choice of the laser and target parameters, we obtained a spectral modulation of the accelerated proton distribution. Coinciding with this spectral shaping, we observed the onset of reproducible and strong transverse spatial modulations in the proton beam. This observation will be investigated in detail in this paper. Whereas proton beam modulations were reported for a number of laser and target conditions recently [30–34], this is to our knowledge the first observation for micrometer thick target foils irradiated with ultra-short laser pulses. Concerning medical applications, spatial beam modulations and consequently a spatial variation of the dose would be detrimental and must therefore be understood in greater detail.

After presenting our experimental results, we investigate how far the observed transverse spatial proton beam modulations can be linked to the ultra-short pulse, high intensity laser–plasma dynamics for a micrometer thick target using numerical simulations. These simulations suggest that filamentary instabilities occurring in the laser-produced target front side plasma can translate into spatial modulations in the proton beam profiles during the energy transfer from electrons to protons.

2. Experiment

The experiment was performed at the ultra-short pulse ($\tau_L = 30$ fs full-width at half-maximum (FWHM)), 150 TW laser facility Draco at the Helmholtz–Zentrum Dresden–Rossendorf. Compared to the setup of the laser system as presented in [38], the temporal intensity contrast was altered in two ways for this experiment (figure 1). Firstly, the prominent side lobe in the temporal contrast (red curve), extending from $-2$ to $-1$ ps, was eliminated through an optimized spectral phase correction procedure to provide for a cleaner and steeper rising edge of the laser pulse (blue curve). Afterwards, the clean pulse was modified by introduction of several prepulses on the timescale of 10–4 ps before the main pulse to investigate their influence on the energy spectrum of laser-accelerated protons. The prepulses were artificially created as post-pulses in front of the stretcher and then translated into prepulses by nonlinear effects in the amplification chain [39] so that their duration approximately matched the main pulse duration. The prepulse intensity contrast with respect to the peak laser intensity of $\approx 5 \times 10^{20}$ W cm$^{-2}$ ranged from $5 \times 10^{-8}$ to $8 \times 10^{-7}$.
Figure 1. Temporal intensity contrast for two different setups. The amplified spontaneous emission (ASE) defines the base contrast level. In the range of $-10$ to $-4$ ps the introduced prepulses are visible. For the temporal range $\leq -2$ ps two sets of spectral phase correction are compared. The prominent side lobe in the former temporal contrast was eliminated through an optimized spectral phase correction procedure. Generally, the spectral phase is adjusted by the application of phase correction terms to the stretched pulse by an acousto-optic filter that operates in a closed feedback loop with a spectral phase measurement device. Improved phase correction was achieved using a WIZZLER [35, 36], which features a higher dynamic range, instead of a spectral phase interferometry for direct electric-field reconstruction (SPIDER) [37] for phase characterization.

The linearly polarized laser pulse was used to irradiate the targets under normal incidence and the laser energy on target was scaled between 0.9 and 2.9 J, being adjusted after the last amplification stage with an attenuator consisting of a wave plate and a pair of thin film polarizers. In that way, the energy scaling was performed without changing particularly the focal spot characteristics.

The energy resolved spatial distribution of the accelerated proton beam was recorded using stacks of radiochromic film (RCF, Gafchromic HD810/EBT2) placed 45 mm behind the non-irradiated target rear side. Their high spatial resolution make RCF an ideal tool to measure fine structures in proton beam profiles. Through a hole in the RCF stack, the central part of the proton distribution propagated into a Thomson parabola spectrometer 60 cm downstream from the target, covering a solid angle of 0.02 $\mu$sr [29]. The spectrometer was used for the online measurement of high resolution spectra of all ion species produced in the laser–target interaction.

As targets, we used large titanium foils with thicknesses of 2 and 3 $\mu$m, which will be referred to as 2 and 3 $\mu$m Ti in the following. Additionally, 2 $\mu$m thick titanium foils were spin-coated with a layer of $\approx 0.8$ and $\approx 3.5$ $\mu$m of photo resist (PR) at the rear side to provide for a low density surface layer. These targets will be referred to as 0.8 $\mu$m PR and 3.5 $\mu$m PR.

Representative data for the 3 $\mu$m Ti target is summarized in figure 2, presenting proton beam profiles measured with RCF for different laser energies (figure 2(a)) and proton spectra measured with the spectrometer for a laser energy of 0.9 and 2.9 J (figure 2(b)). Based on these
data, we will exemplify the observed coincidence of spectral and transverse spatial modulations before the discussion will focus on the characterization of the observed spatially modulated proton beams. As figure 2(b) shows, for a laser energy of 0.9 J an exponential energy distribution is observed, which is in accordance with the TNSA process [28]. For a laser energy of 2.9 J, the spectrum is modulated with the proton distribution featuring a broad shoulder for the high particle energies in addition to an exponentially decreasing low-energy part. This spectral feature is consistent with the depth dose profile in the RCF which shows a reduced dose in the central part of the beam at the energy layer of 4.5 MeV Bragg peak energy.

Comparison with the corresponding proton beam profiles recorded with RCF in figure 2(a) indicates a coincidence of this spectral modulation with a strong transverse spatial modulation of the proton distribution which will be analyzed in detail in the following. Whereas the beam profile carries only some random inhomogeneities for the low laser energy case, it is modulated with a net-like fine-scale pattern for the highest laser energy. The modulations persist up to...
the highest proton energies without major changes of the pattern so that the RCF layers shown are representative for the proton beam. In agreement with the visual impression, the modulation strength in the proton beam profiles increases with increasing laser energy, as quantified through the dose variation $\Delta D$ (figure 2(c)). $\Delta D$ is the standard deviation of the mean dose averaged over the complete beam profile (excluding the central hole) and is hence a measure for the global beam profile (in)homogeneity. We have not observed such strongly modulated proton beam profiles in former experimental studies at the Draco laser system with a temporal pulse contrast corresponding to the red curve in figure 1 [38].

Comparing the characteristics of different targets (figure 3), two major observations were made: firstly, all targets featuring spatially modulated beam profiles, consistently display the same net-like fine-scale pattern above a certain laser energy threshold. Comparison of the proton beam profiles for the 2 $\mu$m Ti at 1.5 J of laser energy (figure 3(a)) and the 3 $\mu$m Ti target at 2.9 J (figure 3(b)) demonstrates that for a thicker target the onset of the net-like fine-scale pattern is only achieved for an increased laser energy. In accordance with these observations, for the 3.5 $\mu$m PR target tested (figure 3(d)), we observed a smooth beam profile even for the highest laser energy.

Secondly, in terms of target properties, the proton beam modulation is predominantly a function of the target thickness, while the target composition seems to play no significant role. Accordingly, the $\approx$3 $\mu$m thick 0.8 $\mu$m PR target, consisting of a 2 $\mu$m thick titanium foil and 0.8 $\mu$m of PR, features almost the same laser energy dependence of the transverse beam profile modulation as the 3 $\mu$m Ti target, no influence of the different target rear sides is
observed (figure 2(c)). The same holds true for the maximum proton energy measured for both targets. Figures 3(b) and (c) show the corresponding beam profiles for a laser energy of 2.9 J. It should be noted that the coincidence of spectral and transverse spatial modulations of the proton distribution shown for the 3 µm target in figure 2 was consistently observed for all target configurations.

The occurrence of spatially transversely modulated proton beams can hint at many possible effects in the laser–target interaction. Based on the fact that our data show a laser energy dependent evolution of the proton beam profiles for a fixed target type, we can very likely exclude the possibility of the modulation being imprinted into the proton beam from a target rear surface structure, as reported in [40, 41]. To further confirm this hypothesis, the target rear surfaces were characterized with scanning electron microscopy (SEM) and atomic force microscopy (AFM) techniques. The results are summarized in figures 3(e)–(g). Whereas the 2 and 3 µm Ti foils appear smooth in the SEM (figures 3(e) and (f)), a strong surface structure with a transverse frequency of roughly 1 µm and an amplitude of a few 100 nm is visible in the SEM picture for the 0.8 µm PR target (figure 3(g)). We assume that this surface structure stems from the target preparation process in which we cooled the foil fixed in an aluminum frame using liquid nitrogen so that it stretched during warming. This, apparently, caused tension and disruption in the formerly smooth PR layer. The AFM characterization of the pure 2 µm Ti target reveals surface structures with a spatial frequency below 200 nm and a modulation amplitude below 10 nm peak-to-valley, therefore being orders of magnitude smaller as compared to the coated target. In conclusion, given that uncoated as well as coated Ti targets show a comparable behavior regarding the proton beam modulation despite the very different target rear surface topography, we believe that the transverse modulations observed cannot be traced back to a target rear surface structure.

3. Numerical modeling

Particle-in-cell (PIC) simulations were performed to study dynamic processes in the high intensity laser–target interaction that can result in spatial modulations of the ion beam qualitatively comparable to our experimental findings.

The simulations were carried out with the collisional 2D3V iPICLS code [42] using a simulation box of a 10 λ × 10 λ size with 192 cells per wavelength λ and 192 time steps per laser period. For analysis purposes, the laser pulse was modeled as a plane wave with periodic boundary conditions. Tests for the influence of focusing however yielded equal transverse filamentation patterns in the electron energy density (see supplementary figure, available at stacks.iop.org/NJP/16/023008/mmedia). The laser intensity or normalized field strength, respectively, and the pulse duration were set to approximately match the parameters of the Draco laser system with \( I_L = 2 \times 10^{20} \text{W cm}^{-2} \) (\( a_0 = 10 \)) and \( \tau_L = 50 \text{fs} \) (FWHM). The pulse had a Gaussian rising edge and a 40 fs wide flat top to provide a long period of constant intensity for analysis.

The interaction targets were modeled with ions preionized to \( q/A = 1/2 \) or 1/6 with a resulting electron density of 100 \( n_c \), \( n_c \) being the critical density. This density is lower than that of a fully ionized solid foil, which is 716 \( n_c \) for titanium, but test simulations for different electron densities showed qualitatively comparable electron dynamics once the electron density was large enough to constitute an opaque foil. In order to model different preplasma conditions as resulting from a variation of the temporal pulse contrast, the laser-irradiated target front
Figure 4. Simulation results. (a)–(c), (f)–(i) Snapshots of the electron energy density 33 fs after the pulse has reached its maximum for different laser intensities and preplasma scale lengths. In contrast to the electron density, the electron energy density allows to distinguish the energetic electrons accelerated at the target front surface from the background electrons within the target volume, hence increasing the visibility of spatial modulations. Each snapshot is normalized to $2/3$ of its individual maximum value. The inset in (b) illustrates the observed surface rippling. The black arrows below (g)–(i) mark the position of the energy density maximum and the dashed lines correspond to the position of the target surface without the preplasma gradient. (d), (j) Transverse profiles of the electron energy density taken 1.5 $\lambda$ inside the target. (e), (k) Transverse profiles of ions at the rear surface with at least 50% of the maximum ion energy in the simulation, averaged in $z$ direction. The point in time is 80 fs after the pulse has reached its maximum. The vertical scales in all profile plots are linear and the same for all four graphs (green, blue, red and black) within each plot. The profile plots represent the (energy) density fluctuation around the respective mean value for the profile which is shown as a solid line in the plots.

The side was either composed of a step-like or an exponentially decreasing density gradient with a variable scale length $L_p$. The target rear side was covered with a 0.5 $\lambda$ thick layer of ions preionized to $q/A = 1/2$ for both preionization cases in order to allow for direct analysis of the dynamics of ions accelerated from the target rear side. Excluding the preplasma gradient, the total target thickness was set to 2 $\lambda$.

The simulation results summarized in figure 4 show that in the parameter range studied, transverse spatial modulations of the rear side accelerated ions can indeed be observed. They originate from a filamentation of the target front side accelerated electrons which are subjected to different plasma instabilities, depending on the initial plasma conditions.

In a first set of simulations, we studied the influence of the laser intensity for targets with a step-like density gradient. The chosen preionization of $q/A = 1/2$ matches the case of light target ions (e.g. titanium) which are ionized almost completely at the given laser
intensities. Snapshots of the electron energy densities for the normalized field strengths $a_0 = 5$, 10 and 25 at 33 fs after the pulse has reached its maximum are presented in figures 4(a)–(c). Whereas the electron energy density is almost homogeneous for $a_0 = 5$, transverse filaments reach throughout the target for $a_0 = 10$ and 25. Lateral profiles of the snapshots taken at 1.5λ inside the target (figure 4(d)) reveal an increased modulation strength (peak-to-valley ratio) with increasing laser intensity. Translation of the spatial modulation into the ion density is confirmed by the lateral profiles in figure 4(e) corresponding to the spatial distribution of ions at the target rear side with at least 50% of the maximum ion energy in the simulation, averaged in z direction at 80 fs after the pulse has reached its maximum. The profile plots represent the ion density fluctuation around the profile average (solid line in the plot) because this parameter closely correlates with an experimentally observable (e.g. with RCF) spatial dose variation originating from lateral proton beam modulations.

Analysis of the electron dynamics shows that the spatial modulation within the electron distribution originates directly from the target front surface which develops a lateral rippling itself (inset figure 4(b)). Hence, the $2\omega_0$ electron jets injected into the target by the $\mathbf{v} \times \mathbf{B}$ force are spatially bunched in lateral direction, similar to [43], and propagate ballistically within the target after being driven beyond the critical density, preserving a lateral modulation. A detailed study presented by Kluge et al [44] suggests as a possible scenario for the surface ripple formation the decay of the plasma oscillation at $\omega_p$ (plasma frequency) into electron surface waves. They then form a standing wave pattern and application of the results from [45] yields a node spacing of $\approx \lambda/10$, in accordance with the simulations (inset figure 4(b)). This structure supposedly seeds a Rayleigh–Taylor-like instability which would otherwise have a much slower growth rate [46].

In a second set of simulations, the influence of a preplasma gradient was investigated for $a_0 = 10$ and targets preionized to $q/A = 1/6$. This choice suppresses the aforementioned rippling mechanism because the heavier target ions (low charge state) now inhibit surface acceleration and reduce the instability growth rate. Scale lengths of $L_p = 0$, 0.1, 0.3 and 1.0λ were tested, corresponding to the parameter range relevant at Draco. PIC simulations for Draco including the temporal pulse contrast of the red curve in figure 1 predicted a preplasma scale length of a few 100 nm [38].

The electron energy densities for the preplasma scan at 33 fs after the pulse has reached its maximum are presented in figures 4(f)–(i) and (j). For the case of $L_p = 0$, a weak lateral spatial modulation of the electron energy density can be observed at the target front side. Comparison with the plasma dynamics for the case of lighter ions ($q/A = 1/2$) shows that the front surface plasma stays much flatter during the laser–target interaction due to the suppression of the surface rippling, as discussed above. With the introduction of preplasma at the target front surface, a considerable change in the plasma dynamics is observed: whereas the directionality of the electron filaments is diffuse for a steep density gradient (figure 4(b)), for $L_p = 0.1$λ and 0.3λ, the electron filaments are strongly directed in laser direction and accompanied by surrounding magnetic fields (see supplementary figure, available at stacks.iop.org/NJP/16/023008/mmedia). Hence, we conclude that the more efficient electron acceleration due to the preplasma generates strong currents that drive a Weibel-like instability (WI) [47].

As the simulations show, the WI critically depends on the preplasma scale length with the modulation depth being maximized for $L_p \approx 0.1$λ (figure 4(j)). The optimum can be explained as follows: on one hand, a thin preplasma shelf at the target front side allows the laser to efficiently collect electrons and generate a dense current of energetic electrons inside the target
which then filaments due to WI. On the other hand, the laser can suffer strong depletion in
the presence of a long underdense plasma in front of the target, a process that reduces the
electron currents driven into the targets and hence the growth of WI. Figures 4(f)–(i) illustrate
this aspect: whereas the energy density peak (marked by a black arrow) is localized close to
the target surface (black dashed line) for $L_p = 0.1 \lambda$, indicating efficient transport of front-side
accelerated electrons into the target, the energy density peak lies far out in the underdense
plasma for the longer preplasmas.

Translation of the lateral spatial modulation in the electron distribution into the target rear
side accelerated ions is also strongest for the optimum preplasma scale length of 0.1 $\lambda$ because
in that case the electron filaments reach furthest into the target. For the longer preplasma scale
lengths, the electron filaments are partly washed out due to merging of neighboring filaments
during propagation. Filament merging also yields different spatial modulation frequencies for
the electron energy density (figure 4(j)) as compared to the ion distribution (figure 4(k)).

In summary, the two simulation sets demonstrate that lateral ion beam modulations cannot
only be created during propagation of the accelerated ion pulse behind the foil [33] but can
as well be produced during their acceleration process through the imprinting of filamentary
structures in the electron distribution. Within the relevant laser and target parameter regime, we
identified two instabilities that can trigger a break-up of the electrons at or close to the target
front surface, resulting in modulated ion distributions at the target rear side. The simulation sets
investigated are limiting cases showing two distinct break-up processes for the hot electrons, a
possibly surface ripple seeded Rayleigh–Taylor-like and a WI plasma instability. Additionally,
intermediate cases were studied, e.g. targets preionized to $q/A = 1/2$ with a preplasma gradient
present, yielding a simultaneous occurrence of both instabilities.

Particular relevance for the experimental observations lies in the results of the preplasma
scan: even though the preplasma scale length was not varied in the experiment but just changed
compared to former setups through the introduction of picosecond prepulses, the simulation
results indicate the relevance of even subtle changes in preplasma conditions for both the
occurrence and mechanism of filamentation as well as the translation into the ion distribution.

Finally, the modulations’ spatial scale at the source for our experimental data can be
estimated: assuming that the modulations observed in the proton distribution directly stem from
the foil surface, we project the patterns recorded on RCF back onto the focal spot region,
yielding an approximate size of $\lesssim \lambda/2$ which is in good agreement, e.g with the results for
$L_p = 0.1 \lambda$ in figure 4(k). Note however that a quantitative comparison of experiment and
simulation is not only challenging as parameters such as the exact preplasma scale length but
also more subtle parameters such as bulk temperature profile, transverse gradients and target
surface roughness have to be matched with the experimental conditions.

4. Conclusion

In this paper, we have reported on the experimental observation of spatially modulated proton
beams emitted from micrometer thick metal foils which were irradiated with a 30fs laser pulse
having a peak intensity of $\approx 5 \times 10^{20}$ W cm$^{-2}$. The transverse net-like fine-scale modulations
observed show a dependence on laser energy, and hence laser intensity, and target thickness
for their onset and strength. Our numerical analysis suggests the spatial modulations to be
a signature of electron instabilities in the ultra-fast laser–target interaction dynamics and a
surface ripple seeded Rayleigh–Taylor-like instability or a WI instability were found to be likely
candidates. Both instabilities can be detected via distinctive signatures in the simulations, such as their growth rates or a prominent surface rippling. However, experimental access to those signatures is challenging and demands the development of new diagnostic tools. It has recently been considered that small-angle x-ray scattering with x-ray free electron laser (XFEL) pulses may allow to resolve spatial modulations of the electron density within the target volume [48] and may therefore enable the discrimination of different instabilities. Furthermore, a recent paper showed that high-order harmonics carry specific signatures when emitted from a target surface that is affected by a plasmon decay instability [49].

Besides a further investigation of their origin, for application experiments it is especially important to consider the consequences of spatially modulated proton beams for laser proton and ion acceleration. Firstly, the transverse modulations can of course be detrimental for applications that demand a homogeneous or at least well-characterized spatial distribution of particles, such as proton radiography or radiobiological studies. On the other hand, the simulation results indicate that the electron filaments can indeed be beneficial for the absorption of laser energy into the ions.

Another aspect to consider is how spectrally modulated proton beams recorded within a small solid angle (e.g. with a Thomson parabola spectrometer) have to be interpreted when occurring in coincidence with complex spatial patterns. Even though we attribute the spectral modulation to a global beam feature in our case, a simple estimation emphasizes the challenge: projecting the size of the entrance aperture to the spectrometer onto the RCF, we obtain a diameter of 7.5 µm which is extremely small compared to the average feature size of the modulation pattern of ≈2–3 mm at the position of the RCF. Hence, the spectrometer very likely records the spectrum of a singular feature within the pattern which is therefore not necessarily representative of the entire data. Diagnostic devices such as RCF or wide-angle spectrometers [50], giving a better representation of the global characteristics of the accelerated particle distribution, present important alternatives here.

Finally, for the pursuit of higher ion energies at higher laser intensities, e.g. available at (future) Petawatt laser systems, a thorough understanding and further characterization of laser intensity dependent plasma instabilities will be essential for a reliable scaling of present acceleration mechanisms (e.g. TNSA) and associated proton or ion beam properties.

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