PW-class laser-driven proton acceleration optimization by application of temporally asymmetric pulse shapes

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We report on experimental investigations of proton acceleration from laser-irradiated solid foils with the DRACO PW laser, where highest proton cut-off energies were achieved for temporal pulse parameters that varied significantly from those of an ideally Fourier transform limited (FTL) pulse. Controlled spectral phase modulation of the driver laser by means of an acousto-optic programmable dispersive filter enabled us to manipulate the temporal shape of the last picoseconds around the main pulse and to study the effect on proton acceleration from thin foil targets. The results show that short and asymmetric pulses generated by positive third order dispersion values are favourable for proton acceleration and can lead to maximum energies of 60 MeV at 18 J laser energy for thin plastic foils, effectively doubling the maximum energy compared to ideally compressed FTL pulses. The paper further proves the robustness and applicability of this enhancement effect for the use of different target materials and thicknesses as well as laser energy and temporal intensity contrast settings. Assuming appropriate control over the spectral phase of the laser and comparable temporal contrast conditions, we believe that the presented method can be universally applied to improve proton acceleration performance using any other laser system, particularly important when operating in the PW regime.

Laser-driven ion acceleration1,2 as a very compact accelerator technology with remarkable beam properties has been associated with a multitude of medical,3 scientific,4,5 and technical6,7,8 applications for several years now. Realizing those applications turned out to be highly complex requiring a sophisticated level of control on the laser plasma interaction process, which determines the beam quality and energy. Key to any progress on that matter is a detailed understanding of the underlying physics as well as appropriate technical control and metrology of the acceleration process, which have therefore been extensively studied both experimentally and theoretically over the last 20 years. Target normal sheath acceleration (TNSA) is the most robust and widely understood acceleration regime, and has therefore received particular attention with the context of applications. It describes the generation of electric space-charge fields ($\gtrsim TV/m$), driven by laser-accelerated prompt front-side electrons, by which particles from a contaminant layer at the target rear side get ionized and accelerated to energies of several tens of MeV per nucleon. Employing dedicated laser-target configurations (e.g. ultra-thin, low density, special shape targets) allowed for control and establishment of optimized TNSA-based as well as other advanced acceleration regimes whereby recent experiments have demonstrated that combinations of those or hybrid schemes show huge potential14,15. These efforts are complemented by a variety of laser pulse parameter scans (e.g. energy, duration, shape, temporal contrast of the pulse) to determine the optimal laser proton accelerator performance14,15.

Yet, highest proton energies were achieved with high intensity long-pulse lasers16,17 delivering only a few shots per day which prevents application-relevant high average currents. Ultra-short pulse laser systems (few tens of femtoseconds pulse duration) with high repetition rate (up to 10 Hz) could bridge this gap and given the recent progress in laser technology, numerous facilities worldwide18,19 approach or even surpass the PW-level with on target intensities between $10^{21}$ and $10^{22}$ W/cm$^2$. Furthermore, these sources provide additional options for control, modifications and diagnostics being of particular importance for the characterization of laser pulse parameters in focus at these intensities. Upon main pulse arrival, the real plasma conditions due to pre-pulses or spatio-temporal couplings may differ significantly from those assumed in idealized theoretical models. In view of exploiting the full potential of laser driven ion accelerators, on-shot diagnostics and feedback routines based on advanced computing methods, like already applied for wakefield accelerators20, might also become an option.

In this letter we experimentally demonstrate that actively manipulating the temporal pulse shape of the driver laser significantly enhances the proton acceleration performance using a state-of-the-art PW ultra-short pulse system. In a series of experiments under well-controlled contrast conditions with different target materials and thicknesses as well as laser energy and temporal intensity contrast configurations, we found that proton cut-off energies and particle numbers were consistently enhanced by changing the temporal laser profile from a Fourier transform limited (FTL) to an asymmetric pulse shape. With optimized settings we were able to routinely achieve maximum proton energies around 60 MeV which corresponds to $\sim 3.4$ MeV per Joule laser energy on...

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FIG. 1. a) Illustration of the DRACO PW laser, the experimental area, the two pick-off ports and the different diagnostics for time domain measurements of the laser pulse. b) Magnified DRACO focal spot measurement at the experimental area with logarithmic color scale for absolute intensities. The black line represents a normalized horizontal line out of the focal intensity distribution, the white dashed circles represent the FWHM, 2σ and 4σ area. c) + d) Temporal intensity contrast of the DRACO laser on the: c) ns-range (inset: 100 ps), measured with scanning TOAC (SequoiaHD), d) ps-range for intrinsic (black) and PM cleaned (red) contrast conditions, measured with single-shot time extended self-referenced spectral interferometry technique (SRSI-ETE).

target. Compared to the nominal settings, thus an effective doubling of the maximum proton energies was achieved. Based on the simplicity of the method and the stability of our results, we believe that this optimization method is universally applicable to other laser systems with particular importance when operating in the PW regime.

The presented experiments were carried out at the ultra-short pulse laser DRACO at the Helmholtz-Zentrum Dresden-Rossendorf (HZDR). DRACO is a dual beam double CPA (chirped pulse amplification) Ti:Sa laser system, designed to deliver 30 J within 30 fs on target with 1 Hz repetition rate. A simplified sketch of the laser system alongside the experimental setup can be found in FIG. 1a).

The temporal pulse structure of DRACO was characterized with rigorous care and a broad variety of scanning and single-shot diagnostics. This includes second and third order autocorrelators (AC and TOAC), field auto-correlation methods like self-referenced spectral interferometry (SRSI & SRSI-ETE) and spectral phase interferometry for direct electric-field reconstruction (SPIDER) at different positions (vacuum compressor output & just before final focusing) and pick-off methods (full-aperture ≈ 7" & 1" mirror) within the laser chain and for different energy settings (diagnostic-mode & power-mode which corresponds to non-pumped (1 J) or fully-pumped main-amplifiers (33 J), respectively). Temporal pulse contrast optimization is achieved by a series of fast pockels cells with optimized timing structure and minimal timing jitter and XPW filtering between the two CPA stages yielding an intensity contrast ratio better than 10⁻¹² up to -100 ps prior to the main pulse as depicted in FIG. 1c). The inset shows the rise of the coherent pedestal at -75 ps which persists at 10⁻⁸ until -10 ps. The few visible pre-pulse-like signatures between -500 ps and -100 ps can partially be identified as measurement artefacts typical for TOAC, reflecting the existence of post-pulses generated by internal reflections in remaining planar transmission optics (e.g. amplifier crystals). Dominantly the signatures represent the conversion of such post-pulses into pre-pulses by non-linear processes associated with the accumulated B-integral in the amplifier chain. Remaining below a level of 10⁻⁹ they can be further suppressed on-demand by inserting a re-collimating single plasma mirror (PM) setup installed close to target. The PM yields an enhancement of the intrinsic temporal contrast by almost two orders of magnitude resulting in an intensity ratio better than 10⁻⁵ at -1 ps prior to the main pulse as depicted in FIG. 1d) for the ps time window. Sub-ps pulse optimization is achieved by controlling the spectral amplitude and phase of the coherent portions of the laser beam. Therefore, two acousto-optic programmable dispersive
filters (AOPDFs), namely Mazzler and Dazzler from Fastlite/AmplitudeTechnologies, are incorporated in each CPA stage to maintain the desired spectral shape and, respectively, the spectral phase components by pre-compensation of higher order residual phase terms acquired by the laser pulses while propagating through the laser chain. After the PM, the wave-front corrected laser pulse with a total remaining energy of 18 J is focused by an F/2.3 parabola to a full width at half maximum (FWHM) spotsize of 2.6 µm yielding peak intensities of $5.4 \times 10^{21} \text{ W/cm}^2$. The high spatial quality of the focused laser beam can be seen in FIG. 1, where the dashed circles represent the FWHM, 2σ and 4σ area containing 35%, 58% and 82% of the total laser energy, respectively. The laser pulse irradiated a target at an incident angle of 45° with p-polarisation. The main particle diagnostic to detect and analyze the accelerated ion beam was a multi-channel plate equipped Thomson parabola spectrometer (TPS) aligned to the target normal direction providing an energy dependent resolution of 5 % with a minimum detectable proton energy of 7 MeV. For some selected shots stacks of calibrated radiochromic films (RCF) were inserted at a distance of 55 mm behind the target allowing for proton beam profile characterization, absolute particle number calibration and complementary maximum energy detection.

For the experimental measurements we manually varied the spectral phase terms group velocity dispersion (GVD) and third order dispersion (TOD) with the help of the Dazzler, enabling us to individually adjust the instantaneous frequencies of the electric field and thus the temporal shape of the laser pulse. First, we ensured that the automatic Dazzler feedback loop produces a flat phase over the entire laser spectrum providing almost ideal FTL pulses for all the different laser energy and PM configurations, examples of which are shown by the SPIDER measurements on the left in FIG. 2. Simultaneous measurements performed with the different redundant time domain diagnostics and pick-off ports delivered consistent results, thus all relative phase changes introduced in the following can be referenced to a 30 fs FWHM near Gaussian pulse shape. On that basis, a pure GVD change preserves the symmetric shape but stretches the pulse in time resulting in a reduction in peak intensity. A pure modification of the TOD leads to an asymmetric pulse shape, identified by a shallow rising and sharp falling edge (or vice versa depending on sign) and reduction in peak intensity due to frequency components being shifted away from the main pulse which results in post- or pre-pulse generation and reduction. Measurements with the different time domain diagnostics confirm within their resolution limits the described effects of spectral phase changes on the temporal pulse shape.

We then systematically investigated the influence of those spectral phase changes on proton acceleration for 400 nm Formvar targets. FIG. 3 shows the resulting cut-off energies and particle numbers for different phase term modifications ΔGVD and ΔTOD. While initially keeping the GVD unchanged ($\Delta$GVD=0 fs$^2$), we varied the TOD from $-20$ ks$^3$ to $+80$ ks$^3$ in 20 ks$^3$ steps (represented by different colors inside the dotted rectangle in FIG. 3). Negative TOD values degrade the acceleration performance, whereas positive TOD values generally result in higher proton cut-off energies, which increase from below 30 MeV to more than 40 MeV. However, a clear optimum is not apparent from this data set, especially since we could not further increase the TOD without producing deep and sharp modulations of the laser spectrum, critical for system safety.
To clarify whether the observed proton energy enhancement can be attributed to TOD-induced asymmetries or to the simultaneously altered length of the laser pulse, we performed an additional GVD scan for TOD values 0 fs$^3$ and 40k fs$^3$. For TOD 0 fs$^3$ the GVD was varied between -2000 fs$^2$ and +2000 fs$^2$ without having a comparable large effect on the maximum proton energies. At ±2000 fs$^2$ cut-off energies drop below 25 MeV as a result of the reduced laser intensity due to the larger pulse duration. Keeping the TOD value fixed at 40k fs$^3$, we scanned the GVD between 0 fs$^2$ and 2500 fs$^2$ which led to a further energy enhancement for higher GVD values, clearly peaking at 1750 fs$^2$ with 60 MeV, followed by a decrease for even higher GVD values. RCF measurements confirm the TPS results and prove a clear enhancement effect for the optimized spectral phase parameters in terms of particle numbers as well (right side plot in FIG. 3). The SPIDER measurements on the right in FIG. 2 reveal that the laser pulse in the optimized acceleration case ($\Delta$GVD = +1750fs$^2$, $\Delta$TOD = +40kfs$^3$) still has a well compressed but asymmetric shape, represented by a shallow rising edge followed tens of fs later by a non-negligible post-pulse structure. Higher or lower GVD values increase the pulse duration and yield lower cut-off energies as a result of the reduced peak intensity.

As the observed gain in energy and particle number is correlated with those asymmetries introduced by the TOD, we further studied the stability of this enhancement effect by applying such asymmetric pulses to various other laser-target configurations. FIG. 4 shows the effect of scanning the TOD while keeping the GVD unchanged (GVD=0 fs$^2$) on the maximum proton energy for 180 nm and 400 nm Formvar as well as 5 µm and 2 µm titanium targets, where in the latter case the PM was removed and the laser energy was reduced to 6.6 J. The obtained results reveal that the general trend of the enhancement effect exists for all studied configurations which cover a broad parameter range and hence different initial interaction conditions. Although the relative enhancement of the maximum proton energies varies for these different cases, the data show that a ~ 20% gain is always achievable. Positive TOD values thereby always lead to higher maximum proton energies while lower TOD values decrease the acceleration performance. An appropriate adjustment of the GVD (and potential even higher order phase terms) to maintain a short pulse duration is expected to increase the gain even further similar to the behaviour described before.

In conclusion, this paper shows how temporal pulse modification significantly enhances proton acceleration up to 60 MeV with a state-of-the-art PW laser system. Using an AOPDF and manually manipulating the spectral phase, notably the third order dispersion term, we experimentally demonstrated that the proton acceleration is very sensitive to the laser pulse shape on the ps-timescale around the main pulse. The highest proton cut-off energies were achieved for temporal pulse parameters well different from those of ideally compressed FTL pulses. Short, asymmetric laser pulses with a shallow rising edge can effectively double the maximum proton energy and significantly increase the particle flux as well. The demonstrated stability of this effect over parameters like target thickness and material as well as laser energy and temporal intensity contrast implies that this method could be easily transferred to other laser systems operating in the PW range. The results provide the basis for further, already ongoing experimental and numerical studies which try to explicitly resolve the different complex laser-plasma interaction processes involved on the time and intensity scale investigated in this work. Note, in perspective of future applications, automated dispersion control to optimize laser proton acceleration is a readily applicable method to be combined with real-time feedback routines based on advanced computing schemes.

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

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