Laser-driven acceleration of quasi-monoenergetic, near-collimated titanium ions via a transparency-enhanced acceleration scheme

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Supplementary Material

Supplementary Note 1: Experimental details

The iWASP spectrometer is capable of measuring angularly-resolved ion energy distributions. We present in Supplementary Figure 1 the detected particles on imaging plates for different titanium target thicknesses: in rows from top to bottom, the images for a) 60 nm, b) 80 nm, c) 100 nm and d) 140 nm. We can identify: i) a horizontal band signal close to the X-ray line that is invariably found for all thicknesses; ii) a signal on target normal axis (0 °) and iii) a signal at ∼ 11 ° from target normal. Unlike signal i), both signals ii) and iii) are seen to depend on target thickness. Consequently the observations suggest the first corresponds mainly to accelerated impurities from target rear-side (protons, carbons and oxygen) and the latter two would rather correspond to heavier ions of the target bulk material. It is worth noting that the dark lobes in Fig. 1 at ∼ ±7° do not correspond to the off-axis titanium ions shown in Fig. 1(e) of the main text. They are more probably low-energy impurities since they change significantly with target thickness.

Figure 1. iWASP results for titanium targets with different thicknesses. From top to bottom: 60 nm (a), 80 nm (b), 100 nm (c) and 140 nm (d). We observe three signals on the iWASP which correspond to i) impurities (protons, carbons and oxygen), ii) heavy-ions emitted on-axis (target normal axis) and iii) heavy-ions emitted off-axis (between 8° to 15° from target normal axis). The impurities’ signal remains the same for different target thicknesses. The on-axis and off-axis heavy-ions signal corresponds to mono-energetic titanium ions of multiple ionization states.

Nevertheless, the iWASP spectrometer cannot resolve the different ionization state and mass of ion traces, since the deflection is only magnetic. To further characterized the ions traces recorded on-axis and off-axis of iWASP, we placed a pair of TP collecting ions at those two specific emission solid angles. We obtained correspondingly the q/A
parabola traces for the different species involved, as shown in Supplementary Figure 2, respectively at a) 0° and b) 11° from target normal axis. The first parabola (q/A = 1) corresponds undoubtedly to the proton signal, and despite some mono-energetic features at low energy (from 4 to 12 MeV), the signal is typical of a TNSA exponential spectrum $e^{-E/k_B T}$ with a cut-off energy of $\approx 30$ MeV and an equivalent temperature $T \sim 5$ MeV. The other parabolas with q/A lower than unity correspond to heavier ions. Noteworthy, different ion species can have the same q/A value, making the interpretation of the signal more complicated than for protons. Still, two parts can be distinguished for the two emission angles: one bright part close to the X-ray point (no deflection point) and one fainted part farthest from the X-ray point. The intense signal part closer to the X-ray point would mainly correspond to accelerated impurities, for two main reasons: i) the imaging plate sensitivity for carbon ions peaks at 50 MeV [1] and the inner lines fits well with carbon ions peaking at $\approx 60$ MeV; ii) no clear difference is observed between on-axis and off-axis signals, as suggested by the iWASP diagnostic for this part close to X-ray signal, which also did not change appreciably with target thickness. However, the presence of non fully-ionized carbon or oxygen with high deposited dose at this level of energy raises some doubts. Indeed, the presence of signal at lower q/A with stronger yield than the C$^{6+}$/O$^{8+}$ line in this high-energy region would suggest that some of these ions are actually titanium, especially for low q/A. Although there are probably some titanium ions in this high energy part, we could not distinguish them from impurities at similar q/A. In the main article we identify the energy of 5.3 MeV/amu as the point at which traces with difference in q/A of 0.02 begin to overlap. For example, beyond this point, traces with q/A = 0.42 ± 0.02 could correspond indiscriminately to Ti$^{19+}$, Ti$^{20+}$, Ti$^{21+}$, C$^{5+}$ and O$^{7+}$. Therefore, the species of high-energy ions (> 5 Mev/amu) cannot be resolved conclusively from this work and would require further investigation.

In the main article text, we consequently focus on the second signal, labeled “Ti peaks” in Fig. 2. In this signal, the lines fairly fits with titanium ions from ionization of 12+ to 21+. Besides, we can notice lines in the cluster which corresponds to values of q/A that could only be attributed to Ti$^{17+}$ and Ti$^{19+}$ ions (q/A = 0.35 and q/A = 0.40, respectively) and the differences between 0° and 11° are coherent with the iWASP results. It suggests quite convincingly that this part of the signal on IP is due to titanium ions.

Supplementary Note 2: Field ionization of titanium ions
The field ionization packages, including barrier suppression, tunneling and multiphoton ionization mechanisms, are turned on in our EPOCH[2] simulations to model the ionization of titanium ions in the laser-target interactions. The ionization process is modelled with a probabilistic approach, and the ionization rate depends on the exposure time of an ion in an electric field, in addition to the magnitude of the electric field.

The ionization energies of titanium ions with different charge state Z are illustrated in Fig. 3, in which the inherent large gaps can be observed between different electron
Figure 2. Example of Thomson Parabola (TP) results for a 100 nm-thick titanium target. The two TP were located at different angles: a) 0 degree. b) 11 degree. Each parabola trace correspond to particles having the same charge-to-mass ratio $q/A$. The dot on the bottom left of the image represent the signal created by the X-ray emitted by the target during the laser-target interaction. It represents the no-deflection point. A particular line is the charge-to-mass ratio of $q/A = 1$, corresponding to the proton signal. The other multiple lines correspond to $q/A$ less than 0.5 resulting mainly from carbon, oxygen and titanium ions.

shells (between $Ti^{12+}$ and $Ti^{13+}$, $Ti^{20+}$ and $Ti^{21+}$). These large gaps can cause the saturation of the ionization process at certain levels with proper electric field strength. For example, the ionization energy for $Ti^{13+}$ is approximately three times the ionization energy for $Ti^{12+}$. This large gap corresponds to significant difference of the electric field required to generate $Ti^{13+}$ instead of $Ti^{12+}$. Therefore, when the electric field is strong enough to generate $Ti^{12+}$ but not enough for $Ti^{13+}$, the ionization process saturates at $Ti^{12+}$. The electric field range for the saturation at $Ti^{12+}$ is wide since the field strength required for the next ionization state $Ti^{20+}$ is much higher.

Fig. 4 demonstrates the ionization saturation due to the large gaps in the ionization energy. The six figures show the time evolution of the number fraction of titanium ions with different charge states when they are exposed to different external constant electric fields. Initially all the particles are neutral titanium atoms. We find that the electric field range corresponding to ionization saturation at $Ti^{12+}$ is $1.3 \sim 7 \times 10^{12}$ V/m, and the range for saturation at $Ti^{20+}$ is $1.5 \sim 16 \times 10^{13}$ V/m.

Supplementary Note 3: Discussion on the results from 60 an 140 nm targets

The energy spectrum of $Ti^{20+}$ ions from the experiments with 60 nm thick target have very low signal along the on-axis direction, as shown in Fig. 1(b) of the main text. By contrast, the simulation shows a quasi-monoenergetic peak at $\sim 100$ MeV in Fig. 5(a). A quasi-monoenergetic ion population can also be seen for this low energy in the iWASP
Figure 3. Externally applied ionization potentials required for ionization to reach charge state Z of titanium ions, where the color-coding indicates the valence electron shell of state Z-1.

Figure 4. Calculated ionization fraction as a function of time for a low-density titanium foil (4.6 × 10^8 m⁻³) in a constant applied electric field $E$. The applied field strength ranges from $1.3 \times 10^{12}$ to $1.6 \times 10^{14}$ V/m.
results for 60 nm [Fig. 1(a)] peaking from 1 to 5°. However, at $0 \pm 1°$, where the on-axis TP is located, the signal is weaker and may not be detectable. It is possible also that such features were present but not observed in the experiment as this energy is very near the lower energy limit for a detectable signal (left gray shade of Fig. 5(a)).

In the experiment with 140 nm thick target, a quasi-monoenergetic peak with a low background [Fig. 1(b) of main text] is observed at $\sim 110$ MeV, but the simulation results show a high background spectrum with a pronounced peak at $\sim 100$ MeV and a much lower peak at $\sim 150$ MeV [Fig. 5(a)]. Despite the higher background level, the peak at 100 MeV is $\sim 800$ (arbitrary unit in Fig. 5(a)) higher than the background, comparable to the peak-background difference of $\sim 900$ for the quasi-monoenergetic peak at 145 MeV for 100 nm thick target. In what follows, we demonstrate that the higher background level for 140 nm target is relevant to the later plasma expansion compared to thinner targets.

At the end of the simulation ($t=1750$ fs), the ion energy distribution in space is chirped. Fig. 5 (c-e) show that the energy is higher for the ions farther from the target initial position ($x = 0 \mu m$). Thus, the ion energy is directly connected with the ion location, and the higher background in the ion energy spectrum for 140 nm target can be translated to higher ion density in space. This is confirmed in Fig. 5(b), which shows the higher ion density for 140 nm target compared to other targets, especially in the region with $x < 40 \mu m$. Here 40 $\mu m$ corresponds to energy of $\sim 150$ MeV for the all three targets [Fig. 5(c-e)]. Correspondingly, the Fig. 5(a) shows higher background level with energy below 150 MeV for 140 nm target. Therefore, the higher background of the $Ti^{20+}$ energy spectrum of 140 nm target is caused by the higher plasma density near the target initial position, which should results from the slower expansion of thicker target.
Figure 5. (a) Energy spectra of the $Ti^{20+}$ ions along on-axis ($-1^\circ$ to $-3^\circ$) direction for each target thickness. The white background color shows the energy range of 2 – 5.3 MeV/amu for titanium ions. This range corresponds to the white regions in the experimental spectra of Fig. 1(d)(e) of the main text, within which ions have enough energy to be detected and traces at different $q/A$ are well separated. (b) The longitudinal density profiles of $Ti^{20+}$ averaged transversely within $y = 1$ and $-1 \mu m$ at $t=1750$ fs for each target thickness. (c)(d)(e) The scatter plot of the $Ti^{20+}$ ions in space at $t=1750$ fs for each target thickness. The color-coding indicates the ion energy.
Supplementary Note 4: The time evolution of the transverse electric field $E_y$

Here we would like to demonstrate the time evolution of transverse electric fields $E_y$ which collimate the on-axis titanium ions. Fig. 6 (a) to (c) show that the $E_y$ fields mainly have stable structure in the region with $|y| > 5 \mu m$. When $y > 5 \mu m$, $E_y$ is mainly negative in all three time frames. And when $y < -5 \mu m$, $E_y$ is mainly positive. This structure can collimate the titanium ions.

![Figure 6. The transverse electric field $E_y$ on the rear side of the target near the tracked titanium ions at (a) 1150 fs, (b) 1350 fs and (c) 1450 fs.](image)

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