A Proton Source via Laser Ablation of Hydrogenated Targets

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Abstract. In this work we present results on the extraction of proton beams from a plasma generated by pulsed laser ablation of solid hydrogenated targets. The laser used was an excimer KrF operating at low irradiances ($10^8$–$10^9$ W/cm$^2$) and nanosecond pulse duration. The ablated targets were disks obtained by compression of TiH$_2$ powder. The ion emission was analyzed by the time-of-flight technique using a Faraday cup as ion collector. In order to improve the ion yield, an electrostatic extraction system was applied. Studies on the produced plasma for different laser irradiances and accelerating voltages have been performed. The results obtained show that this setup is suitable for a high yield proton source.

1. Introduction
Today it is possible to easily arrange laser beams at irradiances of the order of $10^8$ – $10^{10}$ W/cm$^2$ and nanosecond pulse duration that, interacting with solid matter in vacuum, produces hot plasmas[1] by pulsed laser ablation (PLA). From these plasmas, ion beams of moderate energy can be extracted[2]. These beams have a number of interesting applications. Laser ion sources (LIS), indeed, proved over the years to be an useful tool both in the scientific and industrial fields.

Many projects across the world use them as injectors for common particle accelerators[3]. They are also successfully employed as devices for surface modification of materials[4] or for semiconductor doping[5]. Moreover, they constitute the basis for the quite ubiquitous pulsed laser deposition technique[6].

It is known in literature that hydrogenated materials are good sources of protons and heavy ions using infrared (IR) PLA setups[7-9]. IR-PLA is generally used for ion generation due to the broad use of Nd:YAG lasers in the laboratories. Moreover, at low/medium irradiances ion energy is known to scale as $I\lambda^2$, where $I$ is the laser irradiance and $\lambda$ the wavelength[10], hence the fundamental wavelength of Nd:YAG lasers is an efficient option to generate energetic ions. Furthermore, PLA performed with IR lasers generates ions with high charge states[11].

Although ultraviolet (UV) lasers induce plasmas with lower charge states, their high energy photons (4 - 5 eV) allow an efficient photoionization of the target material and direct bond breaking, leading to higher plasma densities[2]. For this reason, UV wavelengths are suitable for high flux ion and proton beams. In this work, we present the results of an UV LIS aimed at proton generation from TiH$_2$ targets. Our goal is to study the proton production by UV-PLA, comparing the results obtained with those already available.

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2. Experimental setup
The experimental apparatus used was the PLATONE[12] setup. PLATONE is a LIS composed by a Compx 205 KrF excimer laser (\( \lambda = 248 \text{ nm} \), \( \tau_{\text{FWHM}} = 23 \text{ ns} \)) and an electrostatic extraction system, consisting of an accelerating gap, as shown in Fig. 1. In particular, the extracting voltage (that could reach values up to 60 kV in DC mode) was applied to the first electrode, T+EC anode. In front of it, a second grounded electrode GE is placed. In this way, an intense electric field is obtained in the region EC-GE. The extraction apertures of EC and GE are coaxial and 1.5 cm in diameter. The laser beam was focused through a thin lens on the target surface. As targets we used compressed disks of TiH\(_2\) powder, pure at 99%.

![Figure 1. Sketch of the experimental apparatus. IF: insulating flange, C: HV capacitors, GC: generating chamber, T: target support, EC: expansion chamber, GE: ground electrode, FC: faraday cup.](image)

The disks were obtained by compression of 230 mg of TiH\(_2\) powder at a pressure of \( 10^5 \text{ N/cm}^2 \) for 30 minutes. This powder is relatively cheap (prices are around 1 k€ per kg) and easily available. As an example, from 1 kg of powder more than 4000 disks could be produced. Moreover, they will last for thousands of laser shots.

The targets, mounted on the support T, were irradiated in high vacuum (\( 10^{-6} \text{ mbar} \)) at different laser irradiances (0.6, 1.3, 2.5 and 5.1 GW/cm\(^2\)) and for different extracting voltages, ranging from 0 (free expansion) to +15 kV, in steps of 5 kV.

We characterized the resulting ion beams by means of a Faraday cup (FC), a 7.7 cm diameter aluminium disk, positioned at the right end of GC and connected to a digital oscilloscope. The FC lacked of any suppressor for secondary electrons, but the ion and proton yield obtained were corrected, taking into account secondary electrons emission (see Appendix). The total fly length available for ions, from T to FC, was 28.0 cm (17.5 cm of free expansion inside EC, 3 cm of acceleration between EC-GE and 7.5 cm between GE-FC of free drift).

3. Results
During nanosecond laser ablation, a high density plasma \( (10^{17} - 10^{19} \text{ cm}^{-3}) \) is obtained as a result of the laser-matter interaction. These plasmas are heated to high temperatures by the inverse bremsstrahlung and photoionization processes, expanding rapidly, perpendicularly to the target surface[13]. This expansion give rise to characteristic TOF signals when the ionic components impinge on Faraday cups. From these signals, numerous information could be obtained. During the experiments, we obtained well defined and separate TOF peaks both for protons and “Ti plasma” (Fig. 2). Applying an extraction potential, these peaks increased in amplitude, denoting a better charge extraction[14].
Figure 2. Ion currents recorded on FC by a digital oscilloscope at 0.6 (a), 1.3 (b), 2.5 (c) and 5.1 (d) GW/cm$^2$, for different accelerating voltages.

The fastest peaks (zoomed in Fig. 2) represent fast protons escaped from the main plasma plume. In order to obtain a proof of their correct identification with respect to possible impurities (Carbon, Nitrogen), we used an electrostatic barrier (EB) as particle analyzer. This device, able to select particles depending on their charge-to-mass ratio, was positioned inside GC, before FC, reducing the total fly length to 23.0 cm from T.

Figure 3. TOF proton currents for different stopping voltages applied to EB at 5.1 GW/cm$^2$ in free expansion.

In Fig. 3 are shown typical fast proton TOF signals for different stopping voltages $V_b$ applied to EB, in order to halt particles with the charge and the mass of the proton. The precise value of $V_b$ could be assessed using the following relation[15]
\[ V_b = \frac{m L^2}{2 Ze t_{TOF}^2}, \]  

where \( m \) is the ion mass (proton in our case), \( L \) is the fly length, \( Z \) is the ion charge state (1+ in our case), \( e \) is the elementary charge and \( t_{TOF} \) is the TOF value of the particle to be stopped. These results confirmed the correct identification of protons. For example at 5.1 GW/cm\(^2\) in free expansion, 70 V were sufficient to reduce the major part of the proton peak, as shown in Fig. 3.

The amplitudes of the Ti plasma signals are sensibly higher than those of the fast protons. The former are indeed the result of the convolution of the signals of different charge states of Ti ions present in the plasma plume (in our case, the principal[16,17] is 1+) and protons trapped within the main plume. In effect, according to the target stoichiometry, one could expect a greater charge extraction for protons. Nevertheless, for example in free expansion at 0.6 GW/cm\(^2\), the experimental results showed that the total charge obtained for the fast protons was 0.01 nC, while for Ti plasma was 2.32 nC, suggesting that a large share of protons is in the main plume. A similar behavior was observed also under the effect of the extraction potential, for all laser irradiances used. To confirm this circumstance, we performed a numerical deconvolution of the TOF signals, using the well known function introduced by Kelly and Dreyfus[18,19]

\[ J(t) \propto \frac{L^2}{t^5} \exp \left\{ -\frac{m}{2kT_{KL}} \left( \frac{L}{t} - u \right)^2 \right\}, \]  

where \( L \) is the fly length, \( k \) is the Boltzmann constant, while \( m, u \) and \( T_{KL} \) are respectively the mass, the center of mass velocity and the Knudsen layer temperature of the expanding specie. It is worth to note that we performed the deconvolutions only on the TOF signals obtained in free expansion, in this way we could safely neglect the effect of SEE, as shown in the appendix. The result obtained for free expansion at a laser irradiance of 0.6 GW/cm\(^2\) is shown in Fig. 4.

As it could be seen from the deconvolution of the TOF spectra, three distinct curves could be observed, corresponding to the contributions of protons, Ti\(^{1+}\) and Ti\(^{2+}\) ions. Computing their relative charges, we obtained 1.60 nC, 0.75 nC and 7 pC respectively. Using the computed values, the average charge ratio H/Ti obtained for the cases analyzed resulted 1.8 ± 0.3.

Using the TOF signals we obtained information about the fast protons kinetic energy in free expansion. Both maximum and minimum kinetic energy values were calculated using full width half maximum (FWHM) time values (Fig. 5).
The energy spread at FWHM, calculated using the relation

$$\sigma_E = \frac{E_{\text{max}} - E_{\text{min}}}{E_{\text{max}}} \times 100,$$  \hfill (3)

varied from 56% (at 0.6 GW/cm$^2$) to 62% (at 5.1 GW/cm$^2$).

For what concerns the charge of the fast protons, integrating the corresponding TOF signal we obtained values up to 3.52 nC (at 5.1 GW/cm$^2$ and 15 kV) per laser shot. Using these values we computed the proton yield per pulse, shown in Fig. 6.

The data of Fig. 6 stress a general behavior of the extraction system. At the lowest laser irradiance, the application of the extracting potential significantly increases the proton yield; while, as the irradiance increases, the effect of the potential decreases. This arise from the fact that the extracted current depends on the plasma density near the meniscus\cite{20,21} formed at the extraction electrode (EC). The lower the plasma density, the higher is the efficiency of the extraction electrode, since the plasma is less effective in screening the extracting electric field.

The maximum extraction of protons was reached at the maximum value of laser irradiance and extracting voltage. This depends on the fact that an higher irradiance induce an higher plasma density available for extraction near the meniscus.

It is worth to note that these results were very stable and reproducible at each shot.
4. Conclusion
This work shows the potentiality of the use of UV lasers to realize high yield proton sources from TiH$_2$ targets. Indeed, the use of these targets is very interesting not only in IR-PLA setups, as already shown in literature[7-9], but also in the UV ones. A comparison with the results just cited is presented in Table 1, where the corresponding proton yield is shown together with some relevant experimental parameters. These results show an increased proton yield, particularly if compared to the laser irradiances used.

| Target   | Laser wavelength (nm) | Laser irradiance (W/cm$^2$) | protons/pulse |
|----------|-----------------------|-----------------------------|---------------|
| Current work | TiH$_2$               | 248                         | $5 \times 10^9$ | $3.8 \times 10^{10}$ |
|          | MgH$_2$               | 1064                        | $2 \times 10^9$ | $4.0 \times 10^8$ |
|          | ZrH$_2$               | 1064                        | $1 \times 10^9$ | $7.1 \times 10^8$ |
| Torrisi et al.$^8$ | TiH$_2$               | 1064                        | $2 \times 10^{10}$ | $2.8 \times 10^9$ |

As shown, it is possible to enhance the proton yield both increasing the laser irradiance and the extracting voltage. Indeed, we obtained fast proton bunches with fluxes up to $10^{10}$ proton/pulse and this represent an enhancement with respect to the literature. Moreover, applying a magnetic particle filter, it is possible to extract also the protons trapped in the main plasma plume. Their yield, shown in Table 1 within parentheses, is notable and could be even increased by means of the extracting potential. Further work will deserve more attention to the main plume composition and to comparative analysis with other hydrogen-rich targets.

As a general conclusion, we can assert that LIS aimed at low energy proton production using TiH$_2$ are more effective using lasers in the UV range. Indeed, these sources are intended to provide protons to devices that will accelerate them to substantially higher energies. In this context it is important to obtain high fluxes, in order to reduce the effects of charge losses during the beam transport. As shown, high fluxes can be obtained by taking advantage of the higher ionization fraction induced by the UV radiation[2].

Appendix
It is worth to note that the FC used was lacking of any suppressor to prevent secondary electron emission (SEE) due to ion impact. We know that this could lead to misleading estimation of quantities. It is known that the main parameters that determines the yield of SEE are the ion energy and its charge state[22]. In the experiments under exam we dealt with low ion energies, obtained as the result of the applied accelerating voltage (for a maximum of 15 kV). Moreover, as stated above, the UV laser is known to induce ion with low charge states; consequently due both to the laser used and to the target composition we dealt mainly with singly charged particles.

Another key parameter in the yield of SEE is the presence of oxides on the surface of the FC. These impurities, indeed, are known to greatly increase this undesired effect. For this reason, before any measurement we performed a prior degassing together with a cleaning process through sputtering of ions on the FC surface.

In these conditions it is reasonable to expect that SEE is low, although not negligible. Consequently, using a suppressor, we estimated the SEE yield. The effect of SEE yield on the charge $Q$ collected on FC could be represented by the equation
\[ Q_{\text{measured}} = (1 + \gamma)Q_{\text{ions}}, \]  
\hspace{1cm} (A.1)

where \( \gamma \) is the SEE coefficient. In particular, restricting the measures to the fast protons, we found that coefficient is lower with respect to the main plasma, owing to the higher kinetic energy of Ti ions. In Table A1 are presented the values obtained for \( \gamma \). The values of \( \gamma \) have been used to correct the yields that we deduced from the experimental data.

**Table A1.** Values obtained for the SEE coefficient.

| Accelerating Voltage (kV) | \( \gamma \) for fast protons | \( \gamma \) for main plasma |
|--------------------------|-------------------------------|-----------------------------|
| 0                        | 0.01                          | 0.01                        |
| 5                        | 0.06                          | 0.25                        |
| 10                       | 0.10                          | 0.41                        |
| 15                       | 0.13                          | 0.50                        |

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