Quasi-monoenergetic proton beams by laser-plasma X-rays

D Delle Side1,2, J Krása3, V Nassisi1, L Velardi1

1LEAS – Dipartimento di Matematica e Fisica “Ennio De Giorgi”, Università del Salento and INFN Section of Lecce, Via per Arnesano s.n., 73100, Lecce, Italy
2Institute of Physics, ASCR, v.v.i., Na Slovance 2, 182 21 Praha 8, Czech Republic

E-mail: domenico.delleside@le.infn.it

Abstract. We report the details of a technique for the production of proton beams with very low energy spread exploiting the short soft X-rays obtained by laser ablation. These beams have been generated by the dissociation and ionization of an hydrogen buffer gas induced by the laser-plasma X-rays and then accelerated by means of an electrostatic accelerator. Their properties have been analyzed through the time-of-flight method applying different accelerating voltages. The resulting energetic spread ranges between 6 and 11%, as a function of the applied voltage. Such a system could be extremely useful for producing quasi-monoenergetic proton beams.

1. Introduction

Laser induced plasmas (LIPs) revealed over the years as a prolific field, both for scientific and applicative purposes. Since the early days of lasers, researchers all over the world investigated the effect of focusing high power radiation onto solid targets in high vacuum, obtaining as a result high density plasmas. In these experiments, plasmas expanding rapidly along the normal to the target surface were known to originate the emission of energetic ions since the early 1960's[1]. These findings evolved rapidly, suggesting that these beams could be used for the development of new ion sources[2,3]. Indeed, laser ion sources (LIS) proved to be an useful tool both in the scientific and industrial fields[4-7]. One of the main problems that researchers have to consider when planning the use of a LIS is the broad energy distributions of the ejecta, particularly when used as injectors for particle accelerators. In this case, ad hoc systems[8] should be used to reduce the energy spread of the ion beams.

In this work, we addressed the issue of the energetic spread of ion beams originating from LIPs, tackling the problem from a different point of view. In particular, as proton beams are a field of current scientific interest[9], we developed a technique through which a population of quasi-monoenergetic proton beams is effortlessly obtained. In order to reach our scope, we exploited the short soft X-rays emitted in LIPs, using them to dissociate and ionize a rarefied hydrogen buffer gas; then we were able to accelerate the resulting ions within an electrostatic accelerator. In this context, the LIP is used as a source of vacuum ultraviolet (VUV) and soft X-ray (SXR) radiation. In the past, in effect, it has been shown that LIPs offer the possibility to easily generate this kind of radiation[10].

3 To whom correspondence should be addressed
The efficiency of the production of such radiation and its emission spectrum depend on several factors: laser beam and target properties[11]. We show that our setup is well suited for generating proton beams with small energetic spread and easily tunable characteristics.

2. Materials and methods

The measures reported here were performed at the LEAS laboratory in Lecce (Italy). The setup used was based on the PLATONE device[12], which consists of a laser ion source (LIS) coupled to a double stage electrostatic accelerator. The LIS uses a KrF excimer laser (Lambda Physik, Mod. COMPEX 205) and a stainless steel vacuum chamber as accelerating chamber (AC), see figure 1. The laser generates pulsed beams of 248 nm wavelength and a duration (FWHM) of 23 ns. The laser beam, guided by a 15 cm focal length lens, enters the AC through a quartz window with an angle of 70° with respect to target normal. The accelerating chamber has inside an expansion one (EC) that enables the hydrodynamic expansion of the plasma before the ion extraction. The EC forms a hermetic contact with the target support T. At the opposite EC side there is a 1.5 cm diameter hole in order to extract ions. A base of the EC, together with T, is fixed to the AC by an insulating flange (IF) that allows the application of a positive high voltage to the EC for both extraction and electrostatic acceleration purposes. The distance T-EC hole is fixed at 18 cm ($L_{T-EC}$). At a distance of 3 cm from the EC hole ($L_{EC-GE}$), it is placed a grounded electrode (GE), having the central part drilled by a hole of the same diameter of the EC one. After the GE, at a distance of 98 cm ($L_{GE-FC}$), it is placed a further third electrode, connected to a power supply of negative polarity. This electrode, biased at -100 V, was used as Faraday cup (FC). The vacuum was operated through the use of two turbo-molecular pumps, reaching a pressure of $8.6 \times 10^{-6}$ mbar. A fast photodiode sensible to the UV radiation coming from a beam splitter near the entrance window was used to trigger measurements on a digital oscilloscope.

![Figure 1. Cross section of the PLATONE accelerator. IF: insulation flange; AC: accelerating chamber; GE: ground electrode; EC: expansion chamber; FC: Faraday cup; C₁: stabilizing capacitors; C₂: insulation capacitor. The small circles represent H₂, while corrugated lines the X-rays.](Image)

The target used was a commercial tantalum disk, pure at 99.9%. Tantalum was chosen because, together with the VUV radiation, offers a high conversion efficiency of laser light into SXRs[13]. Moreover, due to its high atomic mass, it allows to easily separate time resolved currents of lighter ions from those of Ta ions. Since tantalum is a known hydrogen absorber material[14], before the main experiments, the target has been laser cleaned in order to remove surface contaminants via laser induced desorption. This would allow to sensibly reduce the contribution originating from these contaminants. In particular, we operated around 36000 laser shots at an irradiance of $3.5 \times 10^7$ W/cm², obtained defocusing the laser beam. The laser pulses heated the samples in vacuum and the temperature rise $\Delta T(r, z, t)$ at distance $r$ from the spot center, depth $z$ and time $t$ was estimated through the equation[15]

$$\Delta T(r, z, t) = \frac{I_{max}}{K} \left( \frac{D}{\pi} \right)^{1/2} \int_0^t \frac{p(t-\tau)}{(4D\tau + d_z^2)} \exp \left( -\frac{z^2}{4D\tau} - \frac{r^2}{4D(\tau + d_z^2)} \right) d\tau , \quad (1)$$
where $d$ is the average spot radius (0.3 cm in our case), $I_{max}$ is the peak irradiance value, $p(t) \sim t \exp(-\beta t)$ is the temporal pulse shape (where $\beta$ takes into account for the FWHM length of the pulse), while $K$ and $D$ are respectively tantalum thermal conducibility and diffusivity. Equation (1) is obtained in the hypothesis of a large absorption coefficient, so that it could be assumed that light absorption happens mainly at the surface. This represents a good approximation for the case under consideration. Solving numerically equation (1), we found that $\Delta T$ could be safely considered zero at 0.2 s (the time between two consecutive laser pulses). In other words, there is no residual temperature rise. Consequently, the surface temperature of the target as a function of time follows the shapes showed in Fig. 2, approaching zero as $t$ reaches 0.2 s. The heating of the target induces thermal desorption of surface contaminants. Moreover, the use of an UV laser gives the opportunity to stimulate also photodesorption[16].

![Figure 2](image) Temperature on the target surface ($z = 0$) at various distances from the spot centre as a function of time. The environment temperature considered was 20 °C.

After the cleaning, a series of measures were performed both in vacuum and with a constant flux of molecular hydrogen (Linde Gas Hydrogen 6.0) maintained within AC using a finely graduated valve. In presence of the hydrogen, the internal pressure was raised to $5.0 \times 10^{-4}$ mbar. At such a pressure, plasma expansion is not significantly influenced by the presence of the background gas[17]. Furthermore, we refocused the laser beam on the target, obtaining an irradiance of $3.0 \times 10^8$ W/cm$^2$ on a 0.001 cm$^2$ spot, while on EC it was applied a positive high voltage up to 40 kV.

Our aim is to dissociate and ionize the background H$_2$ available in the EC-GE gap through the SXRs generated from the LIP and then to accelerate the resulting ions. In these conditions, in effect, we found[13] that the LIP SXRs have an average energy around 180 eV, a value suitable to reach our scope.

The data was collected on a digital oscilloscope and then processed. In general, the time $t_{TOT}$ spent by ions flying from T to FC could be divided in 3 different contributions[18],

$$t_{TOT} = t_{T-EC} + t_{EC-GE} + t_{GE-FC}. \tag{2}$$

In effect, this originates from the three different motions an ion experiences in our setup: one with constant velocity from T to the EC hole; a uniformly accelerated one from the EC hole to GE; and a uniform motion again from GE to FC. Since we are interested in considering only the protons generated in the gap EC-GE by the LIP induced SXRs, we could safely consider $t_{T,EC}$ to be zero (a rough estimation of the time spent by SXRs to reach the EC hole), while protons start to be accelerated in EC-GE with a non zero initial velocity $v_0$, transferred by the interaction with SXRs. Consequently, we could rewrite the total drift time as
\[ t_{\text{TOT}} = \frac{m L_{\text{EC-GE}}}{\alpha e V \left( \sqrt{v_0^2 + 2\alpha \frac{eV}{m}} - v_0 \right)} + \frac{L_{\text{GE-FC}}}{\sqrt{v_0^2 + 2\alpha \frac{eV}{m}}}, \]  

(3)

where \( L_{\text{EC-GE}} \) and \( L_{\text{GE-FC}} \) are the distances the protons travel in the corresponding sections of our setup as in figure 1, \( m \) is the proton mass, \( e \) the elementary charge, \( V \) the voltage applied to EC and \( \alpha \) is a parameter that takes into account the Debye shielding effect on the protons plasma. In effect, this is known to induce lower acceleration efficiencies with respect to the applied potential[18,19]. In equation (3) the only unknowns are \( v_0 \) and \( \alpha \). Although \( \alpha \) is known to slightly reduce increasing \( V \), we could in first approximation consider it fixed and use the recorded current peak times of our interest at each accelerating potential to estimate the unknowns. In effect, fitting the couples \( (V, t_{\text{TOT,peak}}) \) with equation (3), an estimation for \( v_0 \) and \( \alpha \) could be obtained. With this knowledge, solving equation (2) for \( t_{\text{GE-FC}} \), we could analyze the data through the procedure described in [20] for time-of-flight (TOF) analysis. This corresponds to consider protons as originating at GE. In particular, protons TOF signals has been fitted using the function

\[ S(t; L_{\text{GE-FC}}) = \frac{L_{\text{GE-FC}}^2}{t^3} \sum_i S_{0,i} \exp \left[ -\frac{m}{2k_B T_i} \left( \frac{L_{\text{GE-FC}}}{t} - u_i \right)^2 \right], \]  

(4)

where \( t_{\text{GE-FC}} \) was relabelled as \( t \), \( k_B \) is the Boltzmann constant, while \( T_i, u_i \) and \( S_{0,i} \) are respectively the Knudsen layer temperature, the center of mass velocity and the signal amplitude of the expanding proton population. Moreover, the energy spread \( \sigma_E \) at full width half maximum (FWHM) has been computed using the relation[20]

\[ \sigma_E = \frac{\Delta E}{E_{\text{peak}}} \approx 2 \frac{\Delta t}{t_{\text{peak}}}. \]  

(5)

Since it is quite impossible to remove carbon impurities from the signals collected on FC (the carbon comes from the walls of the chamber), we proceeded with the experiments taking care of performing a comparative evaluation of the effect of a double laser pulse (i.e. a pulse followed by a second one after a few ms) over the single pulse.

3. Results and discussion

In the following, we’ll be interested in reporting the results obtained for protons. For this reason, TOF spectra will be shown emphasizing their details using logarithmic scales in abscissas. Experimental results showed that without the application of an extraction potential, it wasn’t possible to detect other protons than those coming from the target. In effect, the corresponding voltage had to be raised to 5 kV on T to observe the appearance of an additional fast peak on the currents recorded through FC (data not shown), when the H\(_2\) was fluxed in AC. In particular, increasing the applied voltage resulted in the occurrence of fast and sharp protons signals when a background hydrogen is present, together with a slower population of protons coming from the target, as shown in figure 3. The estimation of the initial velocity of these protons through equation (3) resulted in \( v_0 = 140 \pm 54 \) km/s, corresponding to an energy of \( 101 \pm 30 \) eV. The \( \alpha \) parameter obtained was \( 0.28 \pm 0.01 \).

Figure 3 clearly reports also the dramatic effect of the double pulse on the TOF signals. In single pulse waveforms, in effect, it is evident the occurrence of slower peaks (with respect to protons) due to carbon impurities. These peaks are severely reduced in the waveforms obtained with the double pulse technique. Despite of this, no other significant effect was observed on TOF signals in double
pulse mode. For this reason, it seems that this option could be used mainly when it is advantageous to obtain beams with a low impurities content.

**Figure 3.** TOF signals for different extraction voltages: a) 10 kV, b) 20 kV, c) 30 kV and d) 40 kV. Abscissas are in logarithmic scale after the break.

**Figure 4.** Details of the observed and computed fast proton signals for different extraction voltages: a) 10, b) 20, c) 30 and d) 40 kV. Waveforms in single pulse mode only, time has been rescaled as explained in section 2.
Careful analysis of the fast proton TOF signals by means of a fit using equation (4) revealed that they are composed by two different populations: one highly peaked around a maximum value, the other with a broader velocity distribution (respectively 1P and 2P in figure 4). The narrower peaks (1P) result from direct ionization by laser-plasma SXRs of the hydrogen available in the gap EC-GE near the EC hole, in effect they have a duration comparable to that of the photopeak. The broader peaks (2P), although resulting in the same way from SXRs interaction with ambient H$_2$, should come from the volume between the target and the extraction hole. This population is extracted thanks to the penetration inside EC of the electric field between EC-GE[12], that causes the observed broader velocity distribution.

Figure 5 reports, as a function of the extraction/accelerating voltage, some of the properties of the fast proton peaks obtained by the fits (in single pulse mode). It is interesting to note the small energetic spread of the proton beams (Fig. 5a), computed through relation (5), which is smaller than those obtained in [8] with a dedicated system. Its value starts from about 6% at 5 kV and increases up to about 11% at 40 kV with a trend similar to a power law ($\propto x^{1.3}$, in the range of voltages tested). The center of mass energy, as expected, depends linearly on the applied voltage (figure 5b). The most notable feature of this linear dependence is in the slope $z$ of the line. In effect, one would expect that it is roughly equal to the charge state of the proton, instead it is about a third. Such a small acceleration efficiency is not surprising, since it is an expected characteristic of laser ion sources[18,19]. This depends on the fact that the protons plasma is very efficient in screening the applied voltage to preserve its neutrality. It is also interesting to note that the value of slope is in good accordance with the $\alpha$ parameter considered above. Furthermore, the intercept, taken with its error, is in accordance with the initial energy obtained from the fit with equation (3).

Figure 5 reports, as a function of the extraction/accelerating voltage, some of the properties of the fast proton peaks obtained by the fits (in single pulse mode). It is interesting to note the small energetic spread of the proton beams (Fig. 5a), computed through relation (5), which is smaller than those obtained in [8] with a dedicated system. Its value starts from about 6% at 5 kV and increases up to about 11% at 40 kV with a trend similar to a power law ($\propto x^{1.3}$, in the range of voltages tested). The center of mass energy, as expected, depends linearly on the applied voltage (figure 5b). The most notable feature of this linear dependence is in the slope $z$ of the line. In effect, one would expect that it is roughly equal to the charge state of the proton, instead it is about a third. Such a small acceleration efficiency is not surprising, since it is an expected characteristic of laser ion sources[18,19]. This depends on the fact that the protons plasma is very efficient in screening the applied voltage to preserve its neutrality. It is also interesting to note that the value of slope is in good accordance with the $\alpha$ parameter considered above. Furthermore, the intercept, taken with its error, is in accordance with the initial energy obtained from the fit with equation (3).

Figure 5. Properties of the fast protons as a function of the applied voltage: a) energetic spread, b) center of mass energy (together with a linear regression), c) Knudsen layer temperature and d) fast protons yield.

The Knudsen layer of the expanding protons plasma front has a temperature that increases faster than linearly with respect to the applied voltage (Fig. 5c). Indeed, in the range of voltages explored it follows a power law of type $x^{2.7}$. This behaviour is the result of the fact that we are considering the protons as being generated at GE. Consequently, the complex interactions of the protons plasma with the accelerating potential in the gap EC-GE result in the KL temperature dependence. It is worth noticing that double pulse waveforms are characterized by slightly smaller temperatures (data not
shown), that translates also to slightly smaller energetic spreads. This should be related to the absence of the carbon impurities from the target surface that would otherwise induce small differences in the generation of the X-rays. The yield per pulse (Fig. 5d) is not very high, it is of the order of $10^7$ protons per pulse, with an increase with respect to the applied voltage that is quite logarithmic. A similar trend is shown also by the total yield of the fast protons, obtained by computing the total area of the reconstructed proton signals. In this case, the yield reaches a maximum of $1.8 \times 10^8$ protons per pulse at 40 kV of applied voltage. It should be noted, however, that the low yields obtained have to be considered a result of the long observation distance, since it is known that the total charge $Q$ extracted from a LIS depends inversely on it as $Q \propto L^{-2}$ [21]. Consequently, using shorter distances would result in sensibly higher yields.

4. Conclusions
We have shown an effective method to obtain a population of quasi-monoenergetic proton beams with a simple laser ablation setup. In the voltage range examined, the energetic spread varied from 6% up to 11%. This result is important since it offers a viable and simple solution to fulfil a need diffused in the scientific field. These beams, for example, could be used for calibration of a Thomson parabola spectrometer. Moreover, the properties of the beams could be easily determined as a function of the applied voltage. Future work will be devoted to a detailed study of the parameters (e.g. target geometry, material and interaction conditions) that could lead to a further reduction of the energy spread.

Acknowledgment
We are grateful to Dr. Domenico Doria for the insightful discussion with him.

References
[1] Linlor W I 1963 Appl. Phys. Lett. 3, 210.
[2] Peacock N J and Pease R S 1969 J. Phys. D: Appl. Phys. 2, 1705.
[3] Byckovsky Y A, Eliseev V F, Kozyrev Y P and Silnov S M, Sov. Patent 324 938, Oct. 1969.
[4] Okamura M, Pikin A, Zajic V, Kanesue T and Tamura J 2009 Nucl. Instrum. Meth. Phys. Res. A 606, p. 94-96.
[5] Noli F, Lagoyannis A and Misaelides P 2008 Nucl. Instrum. Meth. Phys. Res. B 266, p. 2437–2440.
[6] Rosinski M, Badziak B, Parys P, Wolowski J and Pisarek M 2009 Appl. Surf. Sci. 255, p. 5418–5420.
[7] Chrisey D B and Hubler G K, Pulsed Laser Deposition of Thin Films (John Wiley and Sons, New York, 1994).
[8] Tambini J 1996 SPIE Proc. 2767, p. 67.
[9] Velardi L, Delle Side D, Krása J, Nassisi V 2014 Nucl. Instrum. Meth. Phys. Res. B 735, p. 564.
[10] Giuliani D and Gizzi L A 1998 Riv. Nuovo Cimento 21, p. 1.
[11] J. M. Bridges, C. L. Cromer and Thomas J. McIlrath 1986 Appl. Optics 25, 2208-2214.
[12] Nassisi V, Delle Side D and Velardi L 2013 Appl. Surf. Sci. 272, p. 114.
[13] Siciliano M V, Lorusso A, Velardi L and Nassisi V 2009 X-Ray Spectrom. 38, p. 544-547.
[14] Ferrin P, Kandoi S, Udaykumar Nilekar A, Mavrikakis M 2012 Surf. Sci. 606, p. 679.
[15] Ready J F, Effects of High-Power Laser Radiation (Academic Press, Orlando, 1971).
[16] Chuang T J 1983 Surf. Sci. Rep. 3, p. 1-105.
[17] Geohegan D and Puretzky A A 1995 Appl. Phys. Lett. 67, p. 197.
[18] Yeates P, Costello J T and Kennedy E T, 2010 Phys. Plasma 17, 123115.
[19] Trinczek M, Werdich A, Mironov V, Guo P, Gonzalez Martinez A J, Braun J, Crespo Lopez-Urrutia J R and Ulrich J 2006 Nucl. Instrum. Meth. Phys. Res. B 251, p. 289.
[20] Krása J 2013 Appl. Surf. Sci. 272, p. 46.
[21] Krása J, Parys P, Velardi L, Velyhan A, Ryč L, Delle Side D and Nassisi V 2014 Laser Part. Beams 32, p. 15-20.