

## PROTON BEAMS EMISSION FROM LASER-GENERATED PLASMAS

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**ABSTRACT.** An alternative method employing large dimension ion accelerator systems to generate proton beams can be the production of ions by using a power pulsed laser, operating at high intensity, irradiating in vacuum hydrogenated targets and by extracting the ions of interest from the generated plasma. The choice of the adequate target, of the laser pulse intensity regime and of the ion current obtainable, is strongly dependent of the type of experimental apparatus, as will be discussed in the article. Proton beam emission from experiments conducted at low and high laser intensities are presented, in order to generate protons from about 100 eV, as that prepared at the Physics Department of Messina University, up to about 100 keV, as prepared at INFN-LNS of Catania, and up to about 1 MeV obtained at the international PALS Laboratory of Prague, Czech Republic.

### 1. Introduction

The production of energetic proton beams remains an important aspect of the different research fields that can employ them to reach different aims. Nuclear physics uses protons to induce nuclear reactions, nuclear excitation and deexcitation, nuclear fusion processes and nuclear activation methods [1]. Matter structure uses protons to analyze the elemental composition of different targets, such as for Rutherford scattering spectrometry (RBS), proton induced X-ray emission (PIXE), Elastic recoil detection analysis (ERDA), ion channelling investigations [2]. Microelectronics uses protons to induce chemical modifications in depth of different polymers and proton microprobes [3]. Medicine uses proton beams to deposit their energy at the tumours sites without damage the near healthy cells [4]. For these applications proton beams are generated by traditional large dimension accelerators, such as Van der Graaff, Tandem and Cyclotrons that are present in many international laboratories. An alternative method to generate proton beams by using pulsed laser-generated plasma is representing a new possibility to the proton generation and acceleration through less expensive systems, with lower dimensions, that are of high interest in the field of the new ion acceleration techniques. When a laser pulse of duration from 10 fs up to 10 ns is focused on a solid target, the fast energy release is sufficient to evaporate the irradiated matter and to induce high ionized states. Plasma is produced in front of the solid target (backward direction) or along the laser beam direction (forward direction); in both cases it is characterized by non-equilibrium processes due to the faster electron mobility and

the slower ion one. The charge separation in the plasma plume produces a high electric field, directed along the normal to the target surface, responsible of the ion acceleration in such directions [5]. Using *ns* lasers at intensities of the order of  $10^{15} \text{ W/cm}^2$  irradiating hydrogenated thick targets the protons are emitted in backward direction with kinetic energies generally below 1 MeV, while by using higher intensities, *fs* laser and thin hydrogenated films, the protons are emitted mainly in forward direction with energies above 1 MeV [6]. Plasma, generated at high temperature and density in high vacuum for times comparable with the laser pulse duration, emits photons, electron and ions in free flight. The detection of such radiations permits to characterize the plasma properties in manner to be possible to reproduce the phenomenon in the same conditions [7]. The plasma temperature can be evaluated by using the optical spectroscopy technique. It employs visible photons to measure the coronal temperature, i.e. the external plasma temperature, while X-ray detectors must be employed in order to identify the core temperature, i.e. the maximum inner temperature of the plasma volume [8]. The plasma density can be evaluated by measuring the matter emitted from a single laser shot, furnished by the surface profiler of the induced crater in the solid, and by the plasma volume, measurable with a fast CCD camera giving the image in a very low exposition time. The ratio between the number of emitted atoms or molecules and the volume permits to give the information on the atomic or molecular plasma density vs. the exposition time, but also interferometric techniques may be employed with success to map the electron plasma density [9, 10]. Successively, the measurement of average charge state of the ions present in the plasma, detectable by using a traditional charge/mass spectrometer, will permit to transform the atomic density in electron density of the plasma. The plasma proton emission can be measured in different way, for example by using ion collectors (IC), ion energy analyzer (IEA) and semiconductor devices (Si, SiC, diamond, . . .), detectors that permit to evaluate the average proton energy, the beam directionality, the energy distribution and the ion beam contamination due to different ion species target generated, as will be reported in the following.

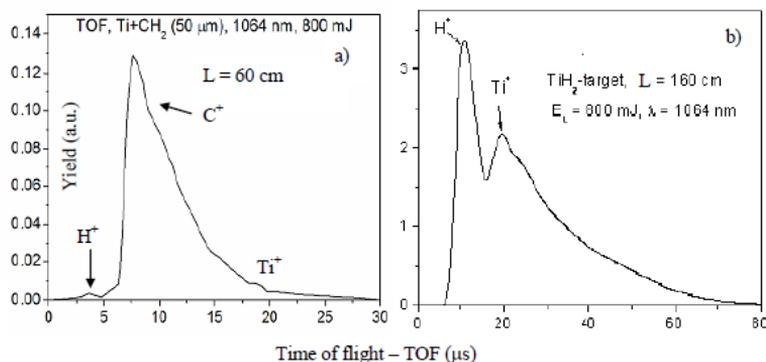
## 2. Experimental apparatus

Three main experimental apparatuses are discussed in this article; they are present at the “Laboratorio di Fisica dei Plasmi Laser” of the Physics Department of Messina University, at the National Institute of Nuclear Physics-Laboratori Nazionali del Sud (INFN-LNS) of Catania and at the International PALS Laboratory of Academy of Science of Czech Republic (ASCR) of Prague. In the first laboratory a Nd:Yag laser, operating at fundamental (1064 nm) and second harmonic (532 nm) wavelength, with 3 ns pulse duration and 300 mJ maximum pulse energy, in single mode or at 1-10 Hz repetition rate, can be focused up to about 100  $\mu\text{m}$  size spot on the solid target placed in high vacuum chamber, with a maximum intensity of about  $10^{10} \text{ W/cm}^2$  [9]. In the second laboratory a Nd:Yag laser operating up to 1 J maximum pulse energy, 1064 nm wavelength, and 9 ns pulse duration permits to generate ions and to submit they to a high voltage (HV) post acceleration system [11]. In the thirty laboratory a Iodine laser, operating at fundamental (1315 nm) and third harmonic (438 nm) wavelength, with 300 ps pulse duration and 1 kJ maximum pulse energy, in single mode, can be focused up to about 50  $\mu\text{m}$  size spot on the solid target placed in high vacuum chamber, with a maximum intensity of about  $10^{16} \text{ W/cm}^2$



**Figure 1.** (a) “Laboratorio di Fisica dei Plasmi Laser” of the Physics Department of Messina University; (b) post-ion acceleration set-up at INFN-LNS of Catania; (c) International Laboratory PALS of Prague; (d) SEM photo of typical craters in a Cu thick target irradiated by the laser pulses at PALS.

[8]. Figure 1 shows a photo of the experimental set-up at the “Laboratorio di Fisica dei Plasmi Laser” of the Physics Department of Messina University (a), of the post-ion acceleration system present at the INFN-LNS of Catania (b) and of the Vacuum chamber used for the laser-matter interaction present at the International Laboratory PALS of Prague (c). Figure 1d shows a SEM photo of typical craters produced by single laser shots in thick Cu target irradiated at PALS at 300 J pulse energy at the fundamental wavelength. In order to produce laser-generated proton beams, hydrogenated thick targets were employed. Hydrogenated polymers, such as polyethylene (PE), mylar and kapton, metals absorbing high hydrogen content, such as Pd, Ti, Ta, Cu and Au, hydrated metals, such as  $TiH_2$ , and microstructures embedded in polymers, such as multi-walled carbon nanotubes (CNT) embedded in polyethylene, were employed as targets to be laser irradiated. Carbon nanotubes store high quantity of hydrogen which can be freed and ionized by the fast and energetic laser pulse irradiations. Ion collectors (IC) connected in time-of-flight (TOF) configurations were employed to detect ions and to measure their average kinetic energy. Different IC placed at different angles, with respect to the normal to the target surface, permit to measure the angular distribution of the emitted ions. A  $90^\circ$  electrostatic deflector, as ions analyzer of their mass, energy, charge state and ion yield, was employed in direction normal to the laser irradiated target surface. The ion energy analyzer (IEA) measures the energy-to-charge ratio by fixing the deflector bias. By changing this bias it is possible to obtain the detection of different energy-to-charge ratios and consequently to plot the ion



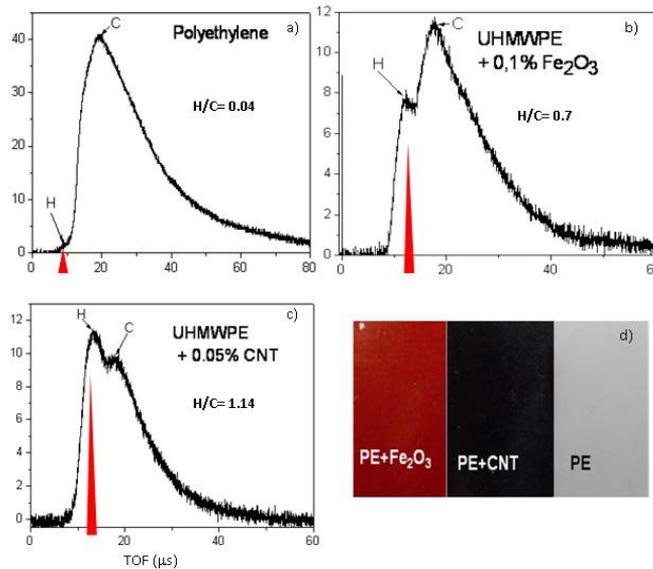
**Figure 2.** IC spectra of titanium and proton emission from laser irradiating PE-50  $\mu\text{m}/\text{Ti}$  (a) and  $\text{TiH}_2$  (b) thick targets at different flight length.

energy distributions. Details on the theoretical aspects and on the technical characteristic of the used IC and IEA are reported in literature [12].

### 3. Results

The laser irradiation of hydrogenated metals permits to generate hot plasmas producing metallic ions and proton emission. A very simply analysis permitting to measure and to compare the mean ion velocities and the relative yield emission coming from different targets consists in the use of ion collectors employed in TOF configuration technique.

Figure 2 shows two examples of IC-TOF spectra relative to the ion emission from the laser ablation of a target consisting in a pure Ti sheet covered with 50  $\mu\text{m}$  polyethylene ( $\text{CH}_2$ ) film (a) and a titanium-hydrate ( $\text{TiH}_2$ ) thick target (b). In the two cases, obtained by using a laser pulse energy of 800 mJ and a different flight length L of 60 cm (a) and 160 cm (b), respectively, a titanium and proton emission occurs and a strong emission of carbon ions is detected in the first case. The Ti ion peak reported in Figure 2b is located at a TOF time scale of about 20  $\mu\text{s}$ , from the flight length corresponding to a mean energy of about 1.6 keV. In both spectra the proton peak occurs at a kinetic energy of about 150 eV. The maximum proton energy relative to Figure 2b is calculated at the TOF time corresponding to the half of the maximum height. Results indicate that the plasma conditions (temperature and density) are comparable in the two cases because the mean  $\text{Ti}^{n+}$  and  $\text{H}^+$  energy is the same for both experiments. However, it is possible to observe that the relative proton/carbon ion yield, of the order of 0.026 in the first spectrum, increases to about 1.54 in the second spectrum (protons/titanium yield). This increment of proton production of about a factor 60 indicates that the use of the thick  $\text{TiH}_2$  may be useful for the generation of high proton fluxes [13]. Different polymeric targets, rich in hydrogen, were also investigated. In terms of proton yield better results were obtained with nanostructures embedded in polyethylene. Carbon nanotubes (CNT) and  $\text{Fe}_2\text{O}_3$  nanostructures not only absorb high hydrogen content but also increase the absorption coefficient of the

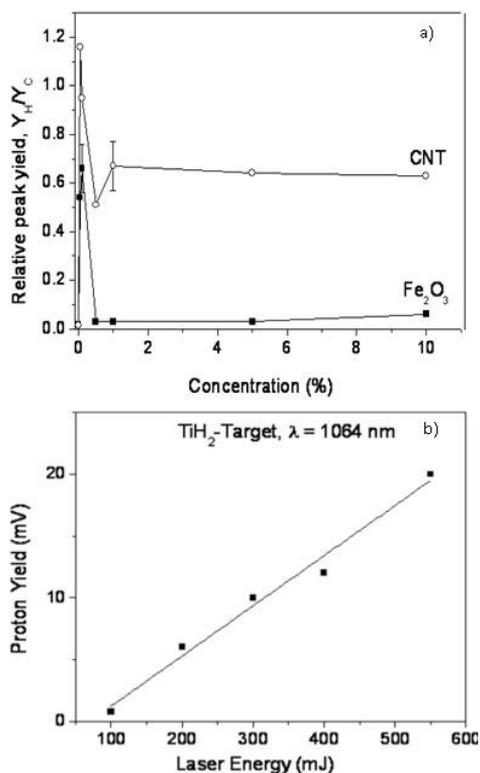


**Figure 3.** IC spectra of polyethylene as pure (a) and embedded with  $Fe_2O_3$  (b) and CNT (c) nanostructures and colors of the three polymeric targets (d).

laser visible and IR wavelengths in the polymer. Figure 3 shows three examples of IC-TOF spectra obtained, by irradiating at 532 nm wavelength and at 150 mJ laser pulse energy, a thick target of pure polyethylene (a), a target with 0.1%  $Fe_2O_3$  embedded in PE (b) and a target with 0.1% CNT embedded in PE (c). Figure 3d shows a photo of the three used targets showing the different polymer color, which is white for pure PE, red for  $Fe_2O_3$  embedded in PE and black for CNT embedded in PE. In this case TOF spectra are obtained with a flight distance  $L = 1$  m, thus the proton peak (calculated at half of its maximum height) gives a kinetic energy of 52 eV in the three cases. The carbon peak is located at about 20 μs in the three cases and it corresponds to a mean kinetic energy of 156 eV. The proton/carbon relative peak yield ratio gives a value of about 0.04 for pure PE, of 0.7 for  $Fe_2O_3$  embedded in PE and of 1.14 for CNT embedded in PE. These results indicate that the use of special nanostructures, embedded in the polyethylene matrix, increases significantly the proton flux emitted from the laser-generated plasmas.

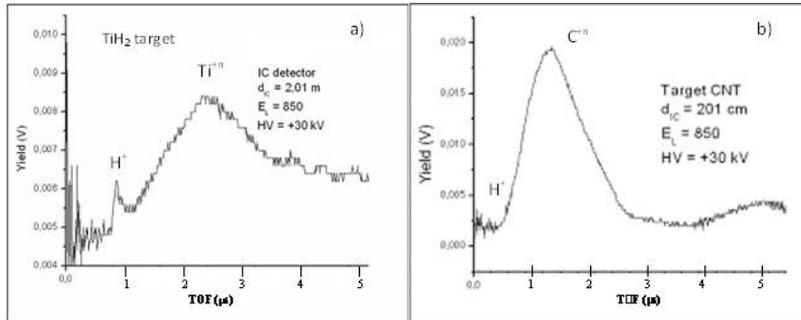
Measurements performed with higher concentrations of the embedded nanostructures indicated that the proton emission decreases instead that increase, due to the high light absorption coefficient in the polymer that reduces the penetration depth of the laser light in the target and, consequently, the number of proton atoms involved in the plasma generated at the target surface [14]. Probably effects of resonant absorption occurs due to the target nanostructures having dimensions comparable with the laser wavelength and or to the plasma frequency resonant with the laser frequency.

Figure 4a shows the results of many measurements obtained by plotting the proton/carbon yield peak,  $Y_H/Y_C$ , vs. the nanostructure concentration for  $Fe_2O_3$  and CNT. The higher



**Figure 4.** Relative proton/carbon peak yield vs. nanostructure concentration for CNT and  $Fe_2O_3$  (a) and proton yield vs. laser pulse energy for  $TiH_2$  irradiated target (b).

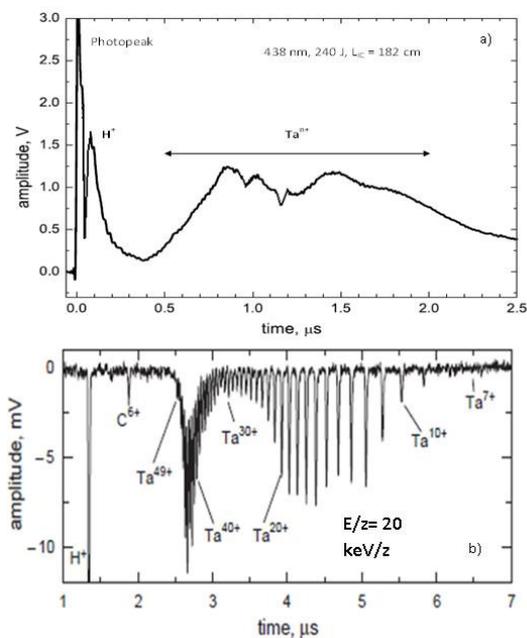
light absorption coefficient for CNT, with respect to the  $Fe_2O_3$ , induces a faster decay with the concentration content. The higher  $Y_H/Y_C$  value is obtained for a very low concentration of the nanostructures, of the order of 0.01%–1%. The ablation yield of the laser in the  $TiH_2$  target and in the nanostructured PE targets was measured through the evaluation of the crater volume generated by single laser pulses. In the cases previously reported, from the ablation yield values it is possible to calculate the proton emission yield. It is of the order of  $10^{16}$  protons/pulse both for the case of  $TiH_2$  ablation at high laser energy (800 mJ) and for the case of nanostructured PE irradiated at low laser energy (150 mJ). The ablation yield increases with the laser pulse energy, as reported in Figure 4b for the case of the  $TiH_2$  target irradiation at 1064 nm wavelength. A total proton charge of the order of 1 mC is generated per single laser pulse. This charge is emitted mainly along the normal to the target surface but its angular aperture is of about  $\pm 45^\circ$ . The correct proton yield evaluation is possible if the particle emission is due mainly to atomic species instead that to molecules and clusters. A measure of proton current is possible by using ion collectors placed near to the target in normal direction, in order to receive the total particle ejected



**Figure 5.** IC-TOF spectra of 30 kV high voltage post accelerated ions emitted from a  $TiH_2$  (a) and CNT embedded in PE (b) targets.

from plasma, and through the TOF detection of the proton ions. Generally the use of UV laser radiation, at low laser intensity, is responsible of photochemical effects producing cluster emission while the use of IR wavelengths, and high laser intensity, generates photo-thermal effects with high production of atomic emission. In order to increase the energy of the protons extracted from the laser-generated plasmas, a post-ion acceleration system can be employed with a high voltage generating electric field acceleration along the direction normal to the target surface. The accelerating setup adopted in the INFN-LNS facility consists of a plasma expansion chamber placed inside a larger vacuum chamber and containing the target to be laser irradiated [17]. A lateral input hole permits the laser to enter and strike the target. Target and expansion chamber are placed at the same electrical potential, which can be set between +10 and +100 kV by a high voltage power supply. In order to permit the ion to exit along the longitudinal chamber axis, the frontal base of the extraction chamber is opened by 8 mm diameter hole with a 10 mm long nose. In front of the nose, 12 metallic discs generate the electric field from high voltage to the ground. Generally the laser irradiations were performed at 350 mJ so that a maximum ion current extraction of 100 mA can be obtained without power supply current saturation.

A typical IC-TOF spectrum of the 30 kV post accelerated ions is reported in Figure 5 for the laser irradiation of  $TiH_2$  (a) and of CNT embedded in PE (b) targets. Spectra indicate that the proton energy is 30 keV, as expected, and that the Ti-TOF large peak, due to the different charge states, indicates a mean ion energy of 180 keV, corresponding to the acceleration of the  $Ti^{6+}$  charge state, while the C-TOF large peak indicates a mean ion energy of 120 keV, corresponding to the acceleration of the  $C^{4+}$  charge state. The high charge states of Ti and C atoms were confirmed by the IEA spectrometer which filters the ions on the base of their energy-to-charge state ratio [14]. The post ion acceleration from laser generated plasma is a method valid to accelerate ion beams in the field of the ion implantation regime ( $\sim 30$ -300 keV), especially if laser repetition rate regime is employed. In fact, ions with energies of the order of tens-hundred keV can be implanted in the first superficial layers of different substrates in order to induce, at high implanted doses, significant physical and chemical modifications, such as modification of the roughness, wetting, hardness, chemical reactivity, wear, optical, electrical and mechanical properties.



**Figure 6.** IC-TOF spectrum (a) and a typical IEA spectrum acquired for a  $E/z=20$ keV/charge state (b) obtained irradiating a Ta target at PALS Laboratory.

Although laser ion source coupled to the post ion acceleration represents a method interesting for many applications in the field of microelectronics, metallurgy and material science, it is not sufficient to accelerate protons at energies of the order of MeV, an energy request for many interesting applications, such as in the field of Nuclear Physics. To this aim it is need to use higher laser pulse intensities, above  $10^{15}$   $W/cm^2$ . Experiences carried out at PALS Laboratory of Prague, by using the 300  $ps$  Iodine-Asterix laser or by using the 10  $fs$  Terawatt Ti-sapphire laser, permit to generate protons at hundred keV up to some MeV by irradiating hydrogenated thick and thin films. As an example, the IC spectrum of Figure 6a reports the TOF ion detection obtained by irradiating a Ta target at 438  $nm$ , 300  $ps$  pulse duration, a pulse energy of 240  $J$  and a target-IC distance of 182  $cm$ . The spectrum indicates a fast proton peak, just after the laser photopulse, corresponding to a kinetic energy of about 1.1 MeV. The Ta ion energy ranges between the maximum value of about 25 MeV, acquired by the higher charge state of about  $49^+$ , up to a minimum value of about 1 MeV, acquired by the single ionized Ta ions. Considering the IC detection surface of 1  $cm^2$ , the number of protons participating to the IC peak is of the order of  $2 \times 10^{10}$  protons/ $cm^2$  per laser shot. Figure 6b shows a typical IEA spectrum acquired during the Ta ablation, by using a flight distance of 2.6 m and a  $E/z$  ratio of 20 keV/charge state. IEA shows that the Ta charge states extend up to about  $50^+$ . The Asterix laser used for this experiment operates at a low repetition rate, of a pulse per 20 min. However, another laser, a  $fs$  terawatt laser of PALS, may operate above 100 Hz repetition rate and it may produce a constant emission of high proton energy beam from hydrogenated thick and thin target.

Work is in progress in this direction in order to prepare monoenergetic proton beams, with energy above 1 MeV, in order to demonstrate that the laser-generated plasma, at intensities of the order of  $10^{20} \text{ W/cm}^2$ , may be employed as a real ion accelerator system.

#### 4. Discussion and conclusions

The acceleration of proton beams from laser-generating plasma in vacuum is based on the high electric field developed inside the plasma during the laser pulse irradiation. The basic mechanisms responsible of the high electric field development are not discussed in this article because it is devoted to present only the experimental macroscopical results. Ponderomotive forces produce ionization and protons can be generated by thermal and Coulomb interactions occurring at relatively low and high laser intensities. Both emissions in backward and in forward directions may occur but their energies at intensities of the order of  $10^{15} \text{ W/cm}^2$  are below 1 MeV in backward direction and reach values above 1 MeV in forward direction for thin targets. In this last case the emission mechanisms may be explained on the base of the "Target normal sheath acceleration" (TNSA), which assumes the maximal occurrence at very high intensities, with *fs* and TW lasers, at which proton emission in forward direction reaches energies higher than 10 MeV [16]. Of course the produced plasma emits not only protons but also other ion species coming from the target composition. Moreover the ion emissions shows an energy distribution with a Boltzmann-like shape which is shifted toward high energy by increasing the ion charge state. Thus the production of a mono-energetic and mono-specie ion beam need of a filtering technique that must remove the undesired species, such as possible by using an electromagnetic filter. Another limitation consists in the proton current that actually is too low and often not constant to be employed in real applications. From this point of view special composition and geometries of thin targets, accomplished by special fast target movements, are developing to be irradiated by laser repetition rates in order to produce a sufficient intensity of pulse current. Literature reports that using short pulses of PW lasers it is possible to obtain protons above 50 MeV with a repetition rate mode that may reach 10 Hz on thin hydrogenated targets. However, the real amount of particles for moment is of the order of  $10^{10}$  protons per laser shot and a magnetic filtering limits further this low proton flux [16, 17]. The interest for the laser plasma generation of high energetic proton beams derives from the many applications in different scientific fields. For example, in Medicine 60-100 MeV protons can be employed in protontherapy for the cure of tumours, while in the field of the energetic resources MeV deuterium ion beams, produced and accelerated by laser plasmas, can be employed to induce deuterium-deuterium and deuterium-tritium nuclear fusions to develop clean energy [18, 19]. Thus although very interesting results have been obtained in the last ten years in this field, further investigations and researches of technical apparatus are need in order to generate proton beams from laser generated plasmas useful for the different applications briefly presented in this article.

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