Laser-plasma based hadron sources for materials science applications

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Laser-driven ion sources[1] have been widely investigated for their foreseen technological applications. These sources are characterized by a relatively low average particle flux and a broad energy spectrum with a cut-off between few MeV and 100 MeV (depending on the laser). These features suggest to study laser-driven ion sources for materials characterization, either using directly the ion beam or generating a neutron beam by means of a suitable converter[2] . Materials characterization is essential for a wide variety of scientific and technological applications. A number of characterization techniques, such as Ion Beam Analysis (IBA)[3], neutron reaction analysis[4] and neutron radiography[5], rely on few MeV hadrons to probe the properties of a sample. Some of these techniques have limited requirements in terms of particle flux and energy spectrum of the probe beam. Moreover, most IBA facilities still rely on old accelerator technologies (Tandem, Van de Graaf), and neutron sources typically rely on nuclear reactors, radioistope sources or large particle accelerators (depending on the specific application). Thus, these techniques could greatly benefit from the use of an ion source driven by a compact, table-top, 10s TW laser system[6].

Here we consider Proton-Induced X-Ray Emission (PIXE), a powerful IBA technique. We develop a complete framework to describe laser-driven PIXE, and we propose a realistic design for a compact apparatus. Since neutron generation via proton conversion in Lithium or Beryllium has similar requirements in terms of proton energy, we briefly consider also this topic.

PIXE is a non-destructive IBA technique relying on x-ray emission induced by 2-5 MeV protons. PIXE is able to provide detailed information on the elemental composition of a sample, up to a depth of few μms. If different energies of the probe beam are used, as in the so-called differential-PIXE technique, it is even possible to retrieve the elemental depth profiles of non-homogeneus, complex samples (e.g. cultural heritage artifacts[7, 8, 9]). A proof-of-principle experiment of laser-driven PIXE has been recently reported[10]: the presence of Ag was detected in cultural heritage artifacts, assessing the absence of damages to the samples.

Retrieving the sample composition from the experimental x-ray yields requires an iterative procedure: (1) guess of a sample composition (2) computation of x-ray yields from the guessed composition by means of a theoretical model (3) comparison between computed x-ray yields and experimental yields (4) update of the guessed composition to reduce discrepancies (mini-

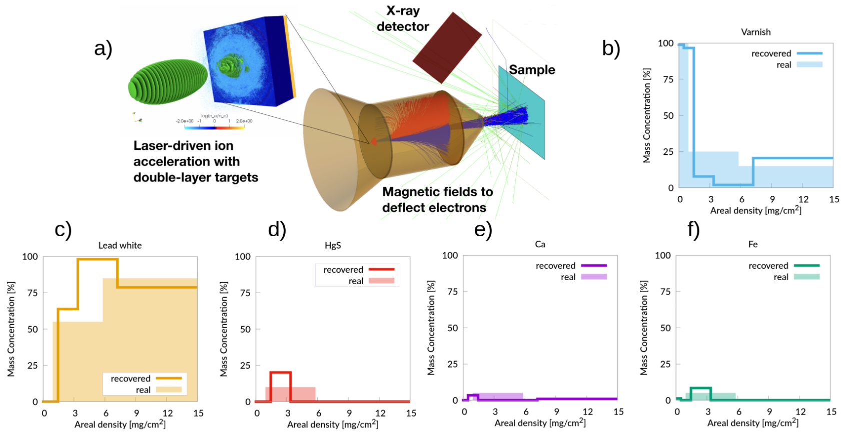

Figure 1: *a) Sketch showing a possible scheme to perform laser-driven PIXE. b)c)d)e)f) Comparison between the composition of the simulated sample and the composition retrieved by the iterative code*

mizing a suitable χ^2 function) (5) restart from point 2). However, since PIXE theory has been developed for monochromatic ion beams, in order to compute the x-ray yields from a given target composition it is necessary to modify the existing theory to allow for a broad spectrum proton beam[11, 12]. As an example we report here how the expression should be modified for the case of a multi-layer (J layers, j layer index), multi-element (I elements, i element index) sample. For ordinary PIXE (i.e. with monochromatic proton beams), assuming to perform K measurements at different proton energies E_p^k, the following expression can be written for the Y_i^k yield (i.e the yield of the i-th element obtained with the k-th energy) [8]:

$$Y_i^k = N_p^k \frac{\Delta\Omega}{4\pi}\varepsilon_i \frac{N_{av}}{M_i}\sum_{j=1}^{J-1} W_{i,j}\sum_{l=1}^{j-1} e^{-\left(\frac{\mu}{\rho}\right)_{i,l}\frac{\rho_l R_l-\rho_l R_{l-1}}{\cos\theta}} \int_{E_{p,j+1}^k}^{E_{p,j}^k} \sigma_i(E)\omega_i e^{-\mu_{i,j}\int_{E_p}^{E'}\frac{dE'}{S_j(E')}\frac{\cos\theta}{\cos\phi}} \frac{dE}{S_j(E)} \qquad (1)$$

where N_p^k is the number of protons with energy E_p^k, $(\frac{\mu}{\rho})_{i,j}$ and $S_j(E)$ are the X-ray attenuation coefficient and proton stopping power associated to the j-th layer, $\Delta\Omega$ and ε_i are the subtended solid angle and the efficiency of the detector, N_{av} is the Avogadro's number, M_i and $W_{i,j}$ are atomic weights and mass concentrations, θ is the proton impact angle and ϕ is the X-ray emission angle, $E_{p,j}^k$ and $E_{p,j+1}^k$ are the proton energies at the boundaries of the j-th layer.

Now, lets suppose to perform K measurements with different, non-monochromatic, energy distributions $f_p^k(E_p)$ for the proton source (in an experiment this could be obtained changing the energy of the laser pulse). Equation 1 should be modified as follows:

$$Y_i^k = \frac{\Delta\Omega}{4\pi}\varepsilon_i \frac{N_{av}}{M_i}\sum_{j=1}^{J-1} W_{i,j}\sum_{l=1}^{j-1} e^{-\left(\frac{\mu}{\rho}\right)_{i,l}\frac{\rho_l R_l-\rho_l R_{l-1}}{\cos\theta}} \int_{E_{p,min}^k}^{E_{p,max}^k} f_p^k(E_p) \int_{E_{p,j+1}^k}^{E_{p,j}^k} \sigma_i(E)\omega_i e^{-\mu_{i,j}\int_{E_p}^{E'}\frac{dE'}{S_j(E')}\frac{\cos\theta}{\cos\phi}} \frac{dE}{S_j(E)} dE_p \qquad (2)$$

where quantities $E_{p,max}^k$ and $E_{p,min}^k$ refer to the k-th proton energy spectrum.

In order to test the feasibility of laser-driven differential PIXE we performed a simulation of

a whole experiment[12] (see figure 1). We simulated laser-driven proton acceleration with the Particle-In-Cell (PIC) code piccante[13]. We coupled PIC results to a Monte Carlo simulation performed with Geant4[14] code in order to reproduce "synthetic" experimental data. Finally, we developed an iterative code implementing eq. 2 to retrieve sample composition. As for the laser-driven proton source we considered a solution based on enhanced-TNSA, in order to enhance the features of the accelerated ions: a double layer target consisting in a thick near-critical layer coupled to a thin solid foil[15, 16, 17, 18, 19]. The laser pulse was P-polarized, with a FWHM duration of 30 fs and 6 different intensities ($a_0 = 2, 2.5, 3, 3.5, 4, 4.5$) to generate as many different proton energy distributions. Fully 3D simulations were carried out. For Monte Carlo simulations we considered a complex multi-layer, multi-elemental sample inspired to an oil painting[8], placed in air. We modeled a beam handling system (based on a dipole magnet) to eliminate the electrons and we modeled a realistic x-ray CCD to detect x-ray emission spectrum. Simulations were performed with $\sim 10^{10}$ protons. X-ray photon counting for each characteristic line were then fed to the iterative code to retrieve sample composition.
Figure 1 shows a remarkable agreement between the original sample composition and the retrieved composition. These results strongly support the feasibility of laser-driven PIXE.
As far as laser-driven neutron sources are concerned, several possible applications have been envisaged for the future[2]. Among the possible strategies to generate neutrons from laser-driven sources, the conversion of laser-accelerated protons in Li or Be is attractive since an energy of only few MeV is required (analogously to PIXE). This means that, in principle, neutron generation could be obtained using a compact 10s TW laser system. Numerical modeling is certainly important to design and to support the experimental investigation of this scheme. However, up until recently, widely used Monte Carlo codes such as Geant4[14] have been unable to simulate reliably (p,n) processes at low projectile energies. For this reason we carried out an extensive investigation[20] of the physical models available in Geant4. Different physical models are organized in predefined *Physics Lists*. We considered all the *Physics Lists* relevant for processes involving nuclear reactions and neutrons and we compared the (p,n) conversion yield with results collected from the literature. Table 1 shows that most of the *physics lists* available in Geant4 fail to model neutron yields from (p,n) reactions in Li or Be. This is due to the fact that these *physics lists* include simplified models for nuclear reaction cross sections taylored for a very high projectile energy (> 100 MeV - 1 GeV). The only *physics list* suitable to model neutron generation (with errors < 30%) is *GCSP_BIC_All_HP*. This *physics list* is based on a combination of experimental data[23] and of evaluated cross-sections[24].
The numerical results presented here are important to assess the the feasibility of developing a

Table 1: *(p,n) conversion yield: comparison between GEANT4 simulations and literature data*

^{9}Be(p,n) at 3.7 MeV		^{7}Li(p,n) at 2.25 MeV	
Geant4 Physics List	**neutrons/mC**	**Geant4 Physics List**	**neutrons/mC**
FTFP_BERT_HP	$5.81 \cdot \mathbf{10^9}$	FTFP_BERT_HP	0
GCSP_BIC_HP	$3.56 \cdot \mathbf{10^9}$	GCSP_BIC_HP	0
QBBC	$3.50 \cdot \mathbf{10^9}$	QBBC	0
INCLXX	$4.62 \cdot \mathbf{10^9}$	INCLXX	0
GCSP_BIC_All_HP	$7.50 \cdot \mathbf{10^{11}}$	GCSP_BIC_All_HP	$4.8 \cdot \mathbf{10^{11}}$
Howard's experiment[21]	$9.61 \pm 0.48 \cdot \mathbf{10^{11}}$	Lee's theoretical yield [22]	$4.6 \cdot \mathbf{10^{11}}$

compact laser-based hadron source for materials characterization with proton or neutron beams.

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