

## **Laser-driven radiation sources for cultural heritage: laser-driven PIXE and XRF on historical and archaeological artifacts**

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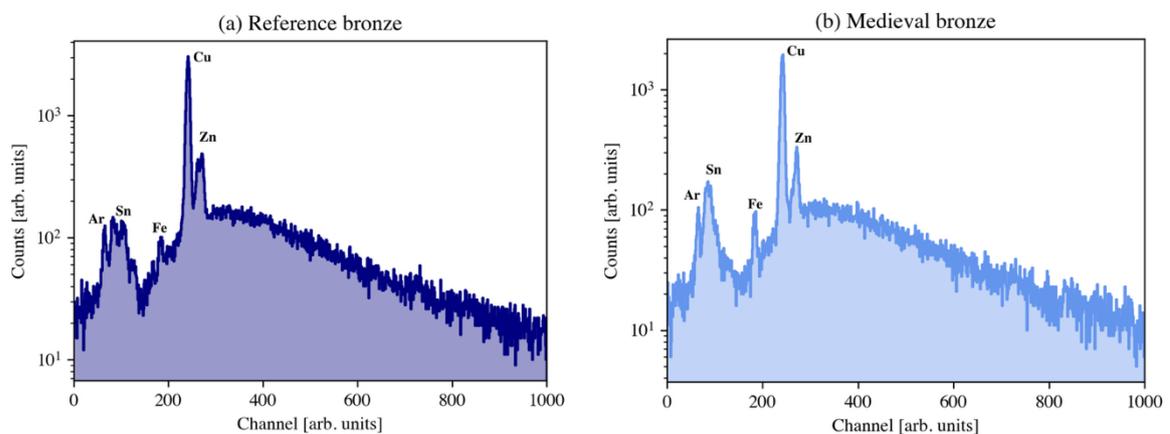
Non-destructive elemental analysis plays an important role in scientific research due to its wide application potential. As an example, in electronics applications [1], the knowledge of contaminant elements can be related to the number of defects in the crystalline structure of silicon-based devices and to the performances of the device. Non-destructive analyses can enable the detection of these contaminants during the production phase. In cultural heritage, non-destructive analyses are essential due to presence of unique and precious materials; the knowledge of elemental concentrations of major and trace elements can support historical and archaeological studies [2]. In fact, when databases allow for the connection of the trace element concentrations to the provenance of the raw materials employed for artifacts, the elemental composition can be used for provenance studies. Particle-Induced X-ray Emission (PIXE) and X-Ray Fluorescence spectroscopy (XRF) are among the most employed techniques to this aim. Their unparalleled sensitivity to trace elements together with their non-destructive nature makes them the standard in this field. PIXE employs charged particle beams (usually protons) to ionise the matter and excite X-ray emission. Particularly effective for low-Z materials, PIXE enables trace element detection at the parts-per-billion level thanks to its low bremsstrahlung background [3]. In cultural heritage, it is an effective

instrument for archaeological provenance and for artwork characterisation. In its conventional setup, PIXE employs electrostatic accelerators [4], which limits the deployments in portable configurations. In contrast, XRF uses X-rays as a source of fluorescence and is more sensitive to high-Z materials. Mostly relying on X-ray tubes, the higher bremsstrahlung source background makes the XRF limits of detection to higher than those of PIXE. Despite this, the relative simplicity of the setup enables the construction of transportable instruments [5], thus, it is a standard in cultural heritage. Only few approaches have combined the complementary strengths of PIXE and XRF into a single transportable measurement system, with a reduced background and an almost constant sensitivity to all elements. Those approaches usually rely on  $\alpha$ -emitters [6], with consequent long measurement times due to the relatively low activity of the source.

In this context, the alternative use of laser-plasma accelerators based on solid targets could offer a promising alternative. When an ultra-fast ( $\sim 10$  fs) high-intensity ( $> 10^{18}$  W/cm<sup>2</sup>) laser interacts with thin ( $\sim \mu\text{m}$ ) solid targets, the matter is ionised, and a separation of charge is generated due to the different inertia of electrons and ions [7]. This condition allows for the generation of strong electric fields ( $\sim \text{MV}/\mu\text{m}$ ) confined in a micrometric scale length that accelerate the proton-contaminants ( $\sim 10^9$ - $10^{12}$  protons per laser shot) present on the surface of the targets. The accelerated particles reach several MeV energies depending on the laser and target parameters. They possess a broad quasi-exponential spectrum [8] with a well-defined cut-off. This acceleration scheme was proved capable of providing the elemental concentrations in simple samples [9, 10, 11]. Together with ions, photons and electrons are emitted during the interaction. In addition to a low bremsstrahlung contribution [12], the ionised target emits characteristic X-rays. This emission can be combined with the accelerated protons to increase the fluorescence yield per laser shot with the addition of an XRF contribution [13]. This approach could be suitable for the development of compact, potentially transportable, table-top setups.

In this contribution, we present a proof-of-principle experiment which demonstrates a transport line-free setup for in-air combination of PIXE and XRF (i.e. PIXRF) driven by lasers on cultural-heritage relevant multi-elemental materials. We used the ELIMAIA

beamline (ELI Beamlines) driven by the L3-HAPLS laser (10 J, 27 fs) with 2  $\mu\text{m}$  copper targets to accelerate the protons; we developed a versatile setup based on a proton magnetic spectrometer [14] to allow the protons and X-rays to exit in air, while deflecting the electrons. The developed setup enabled the alternative measure of the proton spectrum or the irradiation of the sample under analysis. The irradiation point was placed at 10 cm from the proton exit with an incidence angle of  $45^\circ$  with respect to the sample surface normal. The excited X-rays were measured with an Andor iKon-M SO BR-DD CCD placed at 3.5-cm-distance/ $45^\circ$ -incidence from the sample in single-photon counting.



*Figure 1 - PIXRF spectra of two different bronzes. In panel (a) the PIXRF spectrum of the reference bronze is shown. In panel (b) the PIXRF spectrum of a medieval bronze artifact is presented. The tags in the panels represents the elements which corresponds to the spectrum peaks.*

Firstly, we characterised the proton source using the magnetic spectrometer in the measurement modality. The protons had an exponential spectrum with a cut-off of  $21.5 \pm 3$  MeV. Then we characterised the characteristic Cu K-shell X-ray emission from the target through backscattering from PMMA. Around  $10^6$  protons and  $10^5$  x-rays per laser shot impinged on the analysed sample. We used the same setup for the irradiation of a reference bronze from the BCR-691 set and a medieval age bronze fragment. The PIXRF spectra of the two bronze samples are presented in Figure 1. Figure 1-(a) shows the PIXRF spectrum of the reference, while Figure 1-(b) presents the spectrum of the medieval bronze. To compare the two, we considered the ratios between the peak intensities of different elements. One can conclude that the concentrations of tin and iron might be higher for the medieval bronze, while zinc concentration could be higher in the reference.

Element	Sn	Fe	Zn
Reference bronze [-]	$2.66 \times 10^{-2}$	$1.07 \times 10^{-2}$	$7.43 \times 10^{-2}$
Medieval bronze [-]	$6.75 \times 10^{-2}$	$2.55 \times 10^{-2}$	$2.13 \times 10^{-2}$

Table 1 – PIXRF yields of the elements normalised to copper yield for the two bronzes.

We also analysed with PIXRF an archaeological ceramic fragment of a pot from 1st - 3rd c. AD, pertaining to archaeological excavations in Milan. We were able to observe low-concentration ( $\sim 0.1$ -10% in mass) elements belonging to the clay, such as calcium or iron.

These results lay the foundation for the development of laser-based accelerators tailored for cultural heritage studies, suggesting that, with further optimisation, development of appropriate fitting procedures and reduction of the background, this approach could achieve elemental sensitivity comparable to conventional sources while maintaining the inherent compactness and versatility of laser-driven systems.

The research was carried out at the ELIMAIA beamline of the Extreme Light Infrastructure ERIC, ELI Beamlines facility

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