





Muonic Atoms X-Rays Spectroscopy for non-destructive analysys of cultural heritage

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on behalf of CHNET_TANDEM_Collaboration

UNIMIB - Dipartimento di Fisica "G. Occhialini" e INFN sezione di Milano Bicocca

Outline

Introduction

Methodologies used in Archeometry Elemental analysis with Muonic X-rays Negative muon and Muonic atoms ChNET_Tandem Experiment

Facility

RIKEN-RAL PORT4 beam line @ISIS Muon Facility

Experimental Set-up

Germanium detectors Monitor Beam: Hodoscopes made of Scintillating fibers and SiPM New DAQ

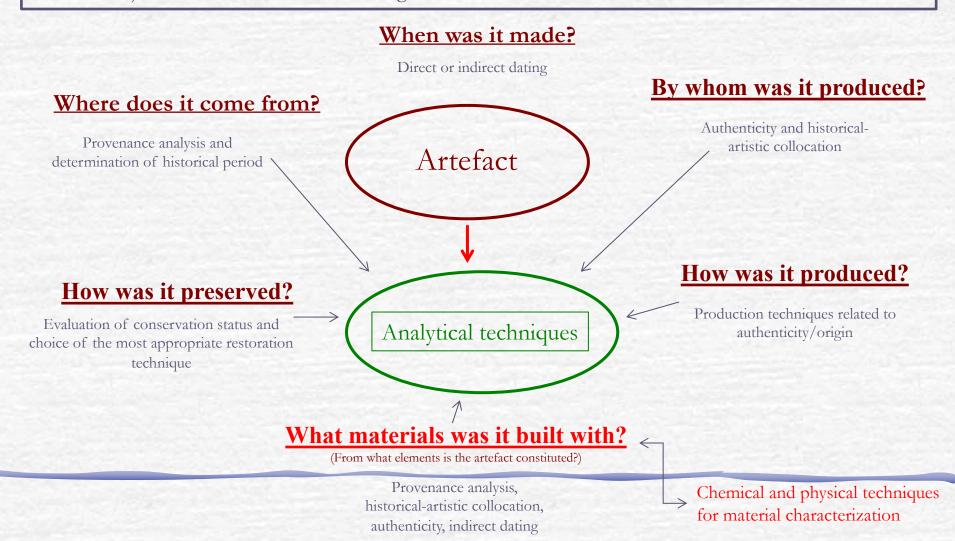
Measurements

Multi Layer sample for muon beam momentum scan "Calibration Curve" with alloys and pure elements used as reference material Archeological samples: Bronze *nuragic* Votive lamps

Future perspectives and summary

Archaeometry: the measurement on ancient or artistic objects

It is the area of applications of scientific disciplines, which have as their object the measurements of ancient/artirtic finds. An example: dating. By extension, the term "archaeometry" includes the methods of analysis useful for the characterization of the material or the diagnosis of the state of conservation of the objects (ancient and otherwise) of interest for the cultural heritage.



Exist the "perfect" archaeometric analytical technique?

What are the characteristics that an analytical technique must have in order to be considered an effective analytical technique?

ACCURACY

Concordance between data and true value

SPECIFICITY

Absence of interferents

"Perfect" analytical technique

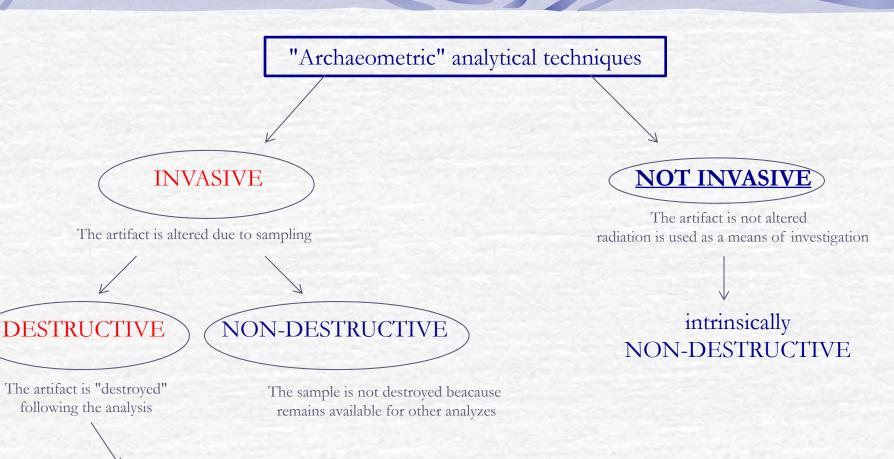
PRECISION

Repeatability

SENSITIVITY

Ability to measure low concentrations

Types of analysis



MICRO-DESTRUCTIVE or MICRO INVASIVE

alterations not perceptible to human eye at a certain distance: size <1mm, weight <10 mg

CHNET-TANDEM 2017-2019 sezioni MIB, LNS-UniSS, PV, RM3

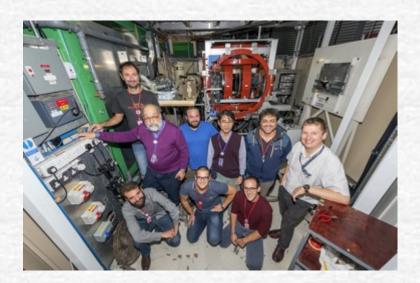
Tecniche Analitiche Non Distruttive per l'archEoMetria

The idea behind the experiment called CHNET_TANDEM is to implement, develop, optimize non-destructive and non-invasive analysis techniques to be used in the archaeometric field for the elemental characterization of finds for Italian Cultural Heritage. Funded and supported by INFN CSNV and Cultural Heritage NET of INFN, LENA, RIKEN-RAL (ISIS-STFC)

Muonic Atom X Rays Spectroscopy (MAXRS)

PGNAA

PORT4 RIKEN-RAL@ISIS-STFC



TRIGA MARK II @ LENA (UNIPV)



Both techniques are based on the detection of **prompt** electromagnetic radiation emissions in the first case of atomic (muonic) type, in the second of nuclear type.

Progetto TANDEM: Tecniche Analitiche Non Distruttive per l'archEoMetria

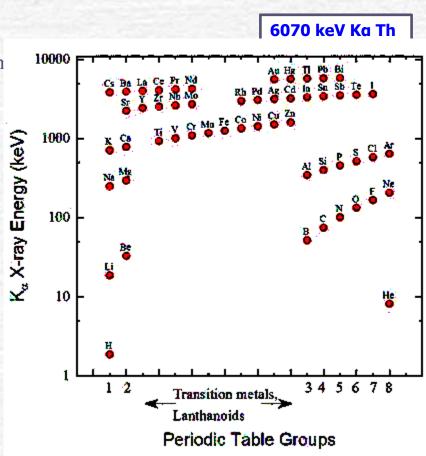
Muonic Atom X-Rays Spectroscopy (MAXRS)

(RIKEN-RAL)

Muonic Atom X-Rays Spectroscopy (MAXRS)

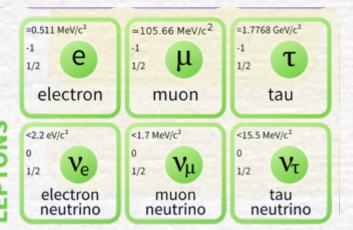
This technique consist in the sample exposure to a **collimated muon beam** (at the ISIS Muon source @Rutherford Appleton Laboratory) and the detection of **characteristic X-rays** emitted following the implantation/formation of **muonic atoms** inside the target.

- High energy characteritics X-rays 10 keV to 10 MeV observable from outside of sample No need of vacuun
 -> applicable to matrices and several shapes;
- Applicable to **every element from (Li to U)** simultaneous multi-elemental
- Possibility to perform depth profile by changing muon beam momentum/energy, Site selective,
 3D mapping
- No sampling Non-destructive techniques pratically no activation



What are Muons? Are leptons

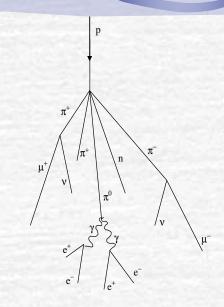
Leptons:



Muon Production:

$$\begin{array}{c} \pi^+ \! \to \mu^+ \! \nu_\mu \\ \pi^- \! \to \mu^- \! \overline{\nu}_\mu \end{array}$$

 π^{+} are produced by p ($\leq 280 \text{ MeV}$) + nucleus



$$m_{\mu} = 207 m_e \sim 1/9 m_p$$

$$\frac{m_{\mu}}{m_e} = 206.7683$$

Elementary particle:

- Charge (-1) same of e-
- Spin: 1/2
- Life time 2.2 usec
- No strong interaction
- Mass 106 MeV/c^2

Muons in materials:

μ+: light proton

μ-: heavy electron

$$\mu^- + p \rightarrow n + \nu_{\mu}$$

$$\mu^- \to e^- \bar{\nu}_e \nu_\mu$$

 $\mu^+ \to e^+ \nu_e \bar{\nu}_\mu$

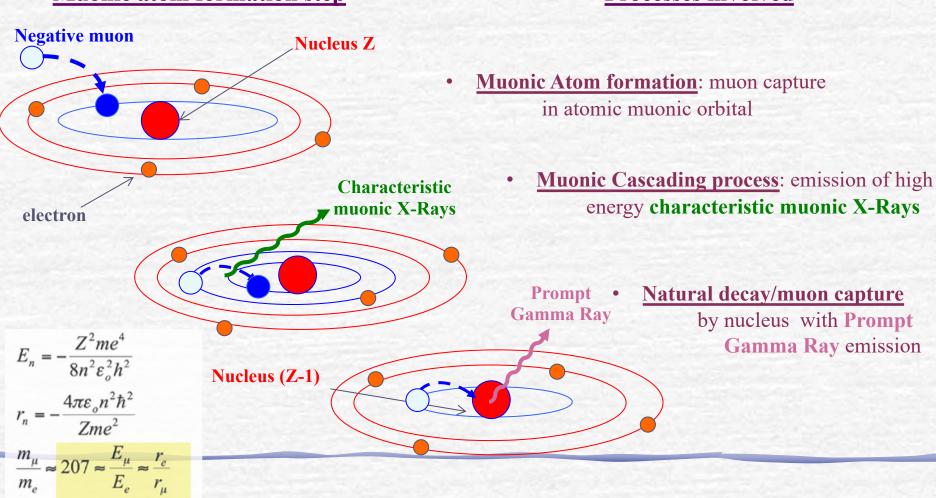
$$\tau_{\mu} = 2.19703 \pm 0.00004 \; \mu sec.$$

Negative Muon and Muonic Atom

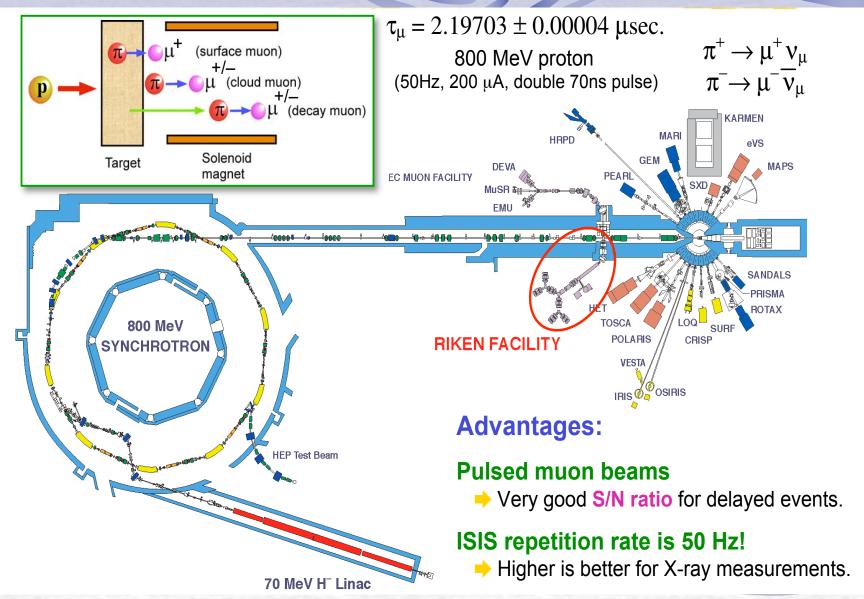
This technique involves **samples exposure** to a **collimated muon beam** (at the ISIS Muon Source @Rutherford Appleton Laboratory) and **characteristic X-rays detection**, emitted following the formation/Implatation of muonic atoms inside the sample.

Muonic atom formation step

Processes involved



Muonic X-rays: RIKEN-RAL facility (UK)



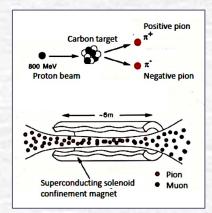
Muonic X-rays: PORT4 ISIS STFC facility (ex-RIKEN-RAL)

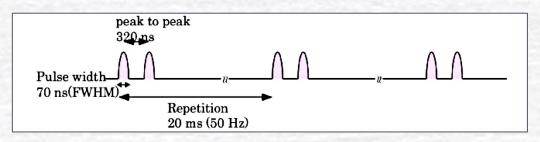
Beam Structure@ PORT4 ISIS Muon source

 8×10^4 muons /sec @60 MeV/c

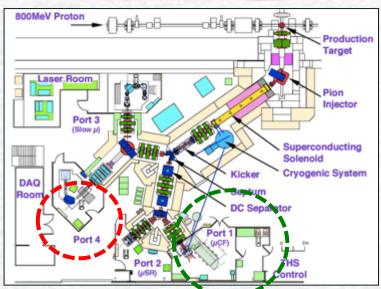
Beam properties surface μ⁺ (20~30MeV/c) and decay μ ⁺/ μ ⁻ (20~120MeV/c)

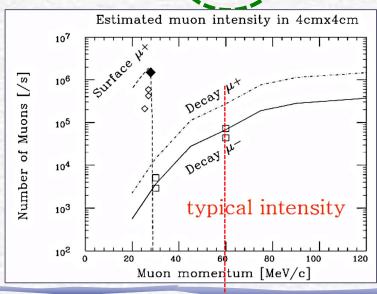
choice of double or single pulse
with magnetic kicker (<60 MeV/c)
Δp/p FWHM 10%(decay), 5%(surf)
typical beam size 10cm²





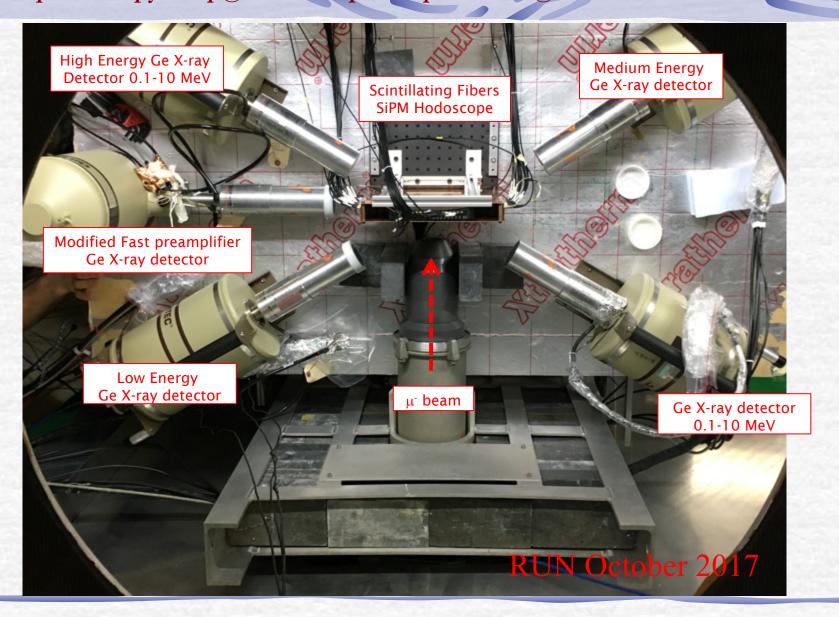
Pulsed proton beam with 50 Hz repetition double pulse with 70 ns width, 320 ns peak to peak on Graphite Target 10 mm thick





Thanks to K. Ishida (RIKEN)

Muon Spectroscopy setup@ RALSet-up of Exp. 1720283 @PORT4 ISIS MUON SOURCE



Spettroscopia Muonica: Attività 2017-2018 presso il RAL

Spin-off dell'Esperimento FAMU CSNIII (raggio di Zeemach del protone tramite spettroscopia laser del µp spokeperson A. Vacchi)

RUN "FAMU" marzo 2017:

- RUN Esperimento FAMU (sono stati testati Odoscopio e Rivelatori al Germanio in vista del RUN di TANDEM)

RUN "TANDEM" ottobre 2017:

- Odoscopio: monitor del fascio e "posizionamento" dei campioni
- Germanio con pre-amplificatore "custom", per misure spettroscopia X
- Irraggiamento di targhette di materiali certificati
- Irraggiamento di un campione multistrato per effettuare lo scan del momento

RUN "TANDEM" giugno-luglio 2018:

- Irraggiamento prolungato di 4 frammenti di Navicelle Nuragiche
- Implementazione di un nuovo sistema di acquisizione dati DT5780 (Multicanale/Digitizer)

RUN "TANDEM" giugno 2019:

- Test beam per misure reperti archeologici "navicelle Nuragiche»

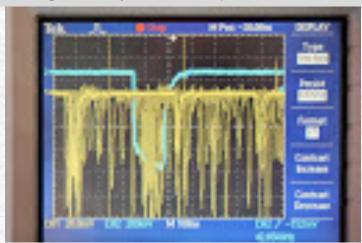
RUN Ottobre 2020:

- Irraggiamento Moneta Portoghese del XVIII sec intercomparison IAEA

Hodoscope Performances at RiKEN/RAL

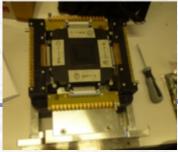
- The system is based on 2 different hodoscopes Fiducial area is 3.2 x 3.2 cm² (10 x 10 cm²) (one with 1 mm and one with 3 mm pitch) with similar design
- Mechanics is realized with a 3-D printer
- 2 X/Y Bicron BCF12 square single clad scintillating fiber planes (32+32 channels) read by 3x3 mm² (or 1x1 mm2) Hamamatsu (Advansid) SiPM
- EMA coating (Al film wrapping) for 1 mm (3 mm) fibers to avoid light cross-talk
- Both have electronics based on CAEN V1742 FADC (waveform → peak, area, time) and common HV for a fiber plane (V_{brk} in a large SiPM sample is very similar)
- Measure X/Y profile and monitor beam intensity

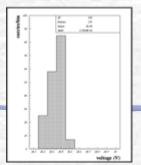
Pulsed high intensity muon beam (~70 muons/fiber/70 ns)



RUN October 2017

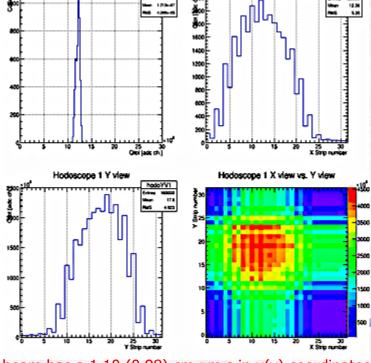






A typical run at 60 MeV/c: 8×10^4 μ -/sec

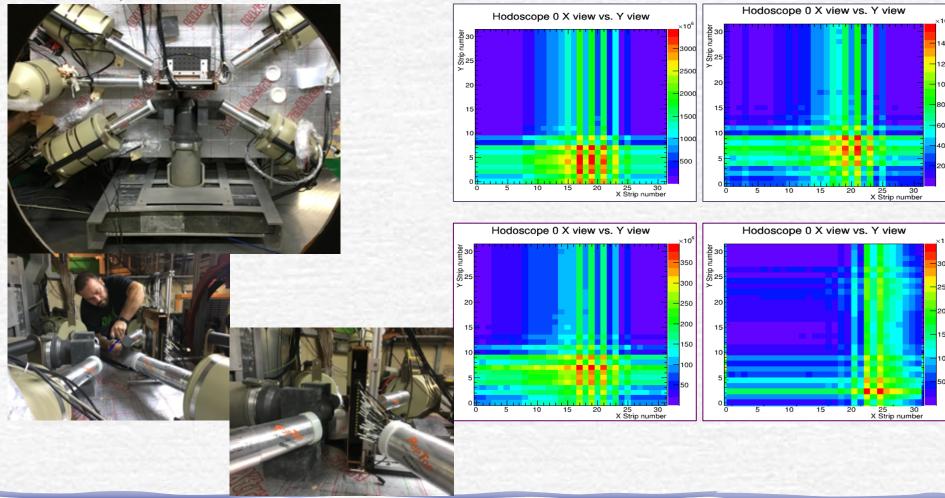
Hodoscope 1 Qtot distribution



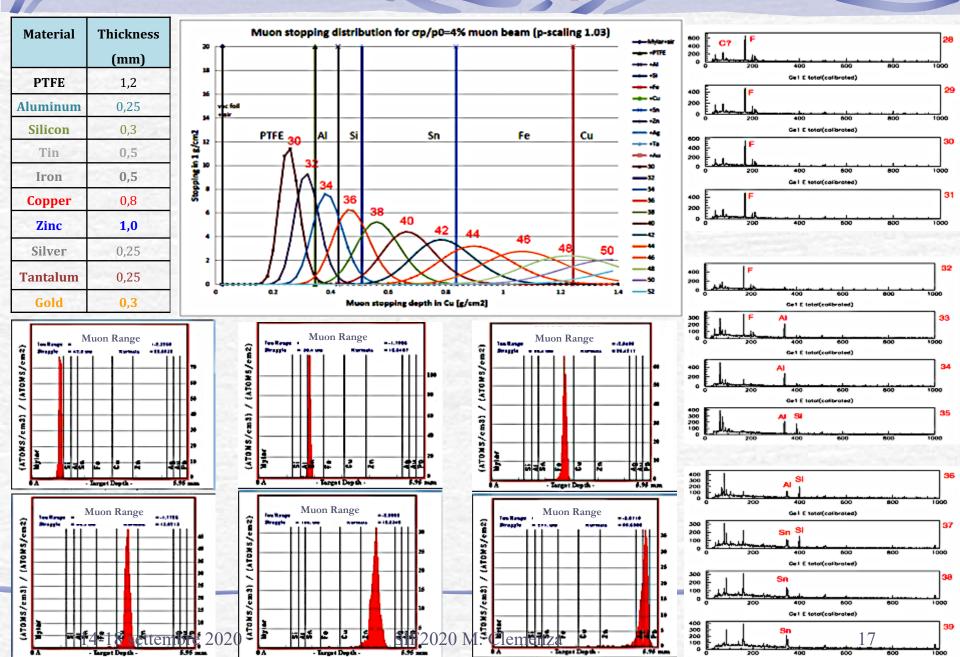
The beam has a 1.10 (0.98) cm r.m.s in x(y) coordinates

SiPM-Scintillating Fibers Hodoscopes: october 2017

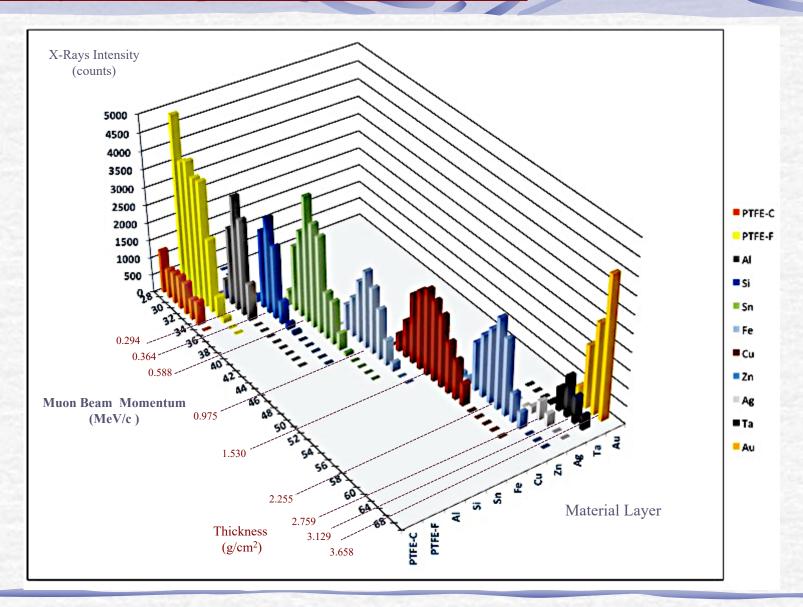
Use of scintillating optic fiber sensors read with SiPM for positioning the sample by measuring the muons flux before and after the sample, allowed us to optimize its position with respect to the center of the beam;



TRIM simulation of a MultilAyered sample: october 2017

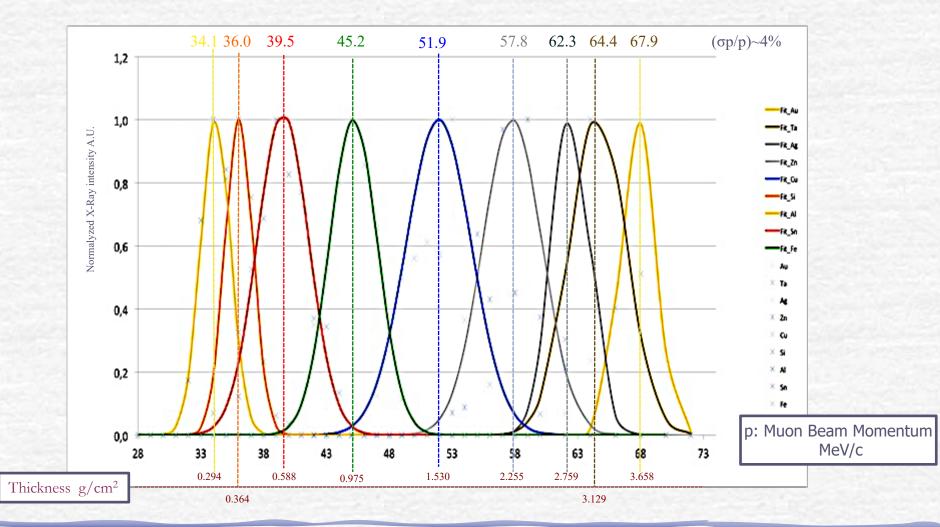


Multi-Layered sample Results october 2017



Multi-Layered sample Results october 2017

Optimization of Beam momentum



Best fit with gaussian curve of the normalyzed intensity of muonic X-rays distributions

June-July 2018: implementation of new DAQ DT5780 CAEN

Dual Digitizer Multi Channel Analyzer - Desktop

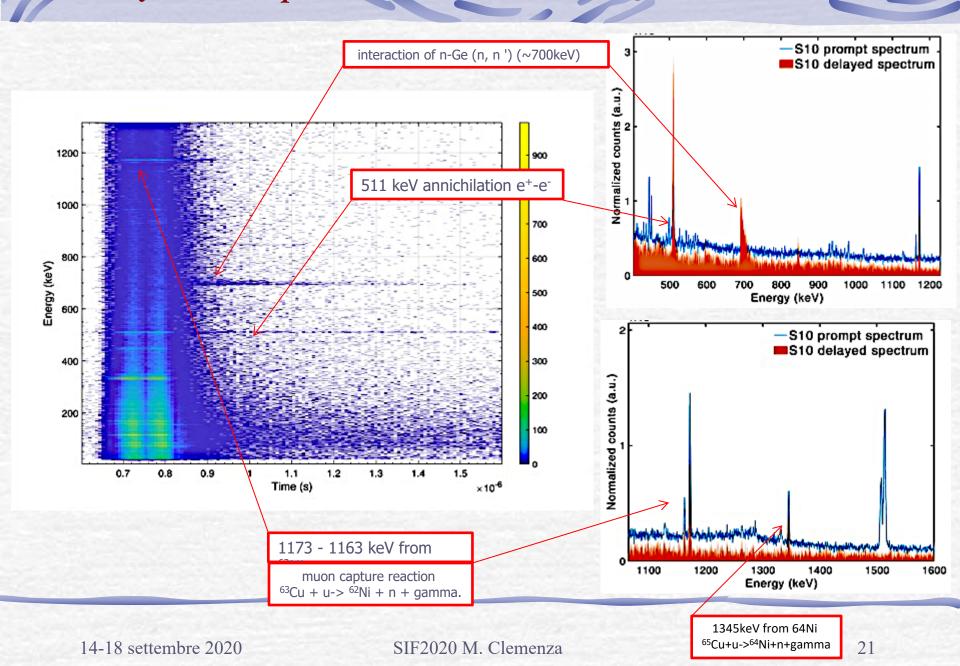


The DT5780 is a compact desktop system integrating 2 Independent 16k Digital MCA and featuring HV/Preamp capabilities for digital nuclear Spectroscopy. 2x 100 MS/s 16-bit waveform digitizer (based on 724 series)

Operating modes:

- "Pulse Height Analysis": pulse height histogram (1k-2k-4k-8k-16k) built at software level
- "List": pulse height and time stamp for each event: 10 ns resolution
- "Oscilloscope": input and internal filters waveforms

June-July 2018: implementation of new DAQ DT5780 CAEN



June-July 2018: Calibration Curves

"Reference" Materials:

Silver 99.95 Cu99.9 Al 99.999 Al77.9SiC17.8Cu3.3Mg1.0 Fe65Cr18Ni10Mo3 Fe54Ni29Co17 Carbon 99.95 Cu55/Ni45 Cu63/Zn37 Cu62/Ni18/Zn20 Zn99.95Sn98.8 Ag72/Cu28 Cu94/Sn 6

"N. of values" for Calibration curve

Cu:

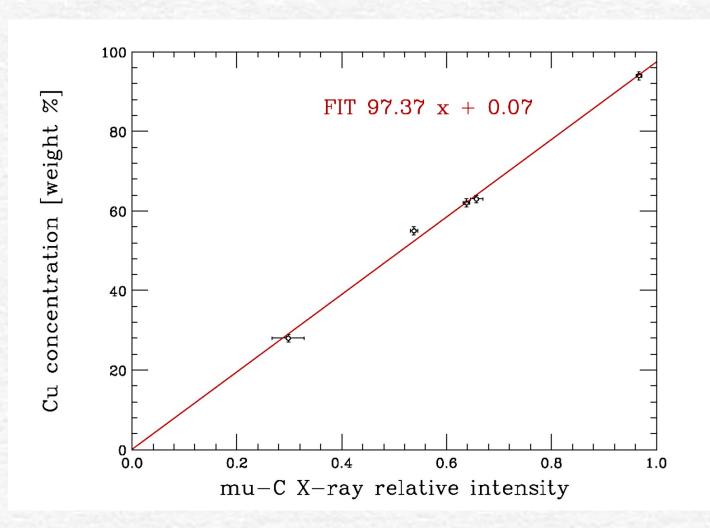
99.9-94-63-62-55-28-3.3

Ni: 45-29-18-10

Zn: 99.95-37-20

Sn: 98.8 – 6 Ag: 99.95-72

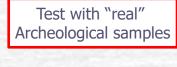
Al: 99.995-77.9



Elemental Characterization of Nuragic Bronze Age Votive Ships: June-July 2018

Questo sistema di misura è stato utilizzato per l'analisi approfondita di 4 reperti archeologici (frammenti di navicelle in bronzo votive di età nuragica) che hanno portato all'evidenza dell'utilizzo del piombo come addittivo nella lega di Rame e stagno soprattutto in alcune particolari zone di giunzione (prua della navicella).











Elemental Characterization of Nuragic Bronze Age Votive Ships: June 2019

Test beam per misure reperti archeologici "Navicelle Nuragiche" (lucerne a olio)

Museo Archeologico Nazionale di Firenze:

2 navicelle nuragiche in bronzo

«Tomba delle tre navicelle» a Vetulonia (GR)



MOSTRA: "Laboratorio di analisi di Oeforo
UUGGO E DATA: Oeford

201 121

DMT 27/6/3059 WT 38/06/3059

PESO NG.: GR. 500
MATERIALE: BRONZO FUSO

DATAZIONE VI SEC.A.C.

MSURE: IL OM. 13.5; LUNGH, CM. 20.5; LARGH, CM. 6
COLLOCAZIONE: DEPOSITI M.A.N.F. ARMADIO 119 85
PROVENIENCA: VETALIONIA. TOMMA DELLE THE NAVOELLE.

VALORE ASSICURATIVO: € 50,000,00

STATO DI COMERNAZIONE: OTTIME CONDIZIONI, INTEGNA, LA PRESA E' INCLINIZIA VERSO LA PRUA, PATINA VERSOAZINA.

P.S. VEDI DOCUMENTAZIONE POTOGRAFICA

NOTE: IL REPERTO DEVE ESSERE IMBALLATO CON LE DOVUTE PRECALIZIONI DA PERSONALE SPECIALIZZATO, L'ESPOSIZIONE DOVIRA REVENIRE IN AMBIENTE CON CONTROLLO DI LIMIDITA E

TEMPERATURA

LUGGO, DATA, FRIMA: FRENZE 13/03/20

IL DIRETTORE DEL MUSEO: DOTT. MARIO IOZZO RESTAURATORE : STEVANO SARI

Musico

Polo Museale della Toucana

Tac della Prograia, 455 – 30121 Firmoy – sol. +33 03323575 – Jan. +33 0332425.







"Laboratorio di analisi di Oxford"

LUGGO E DATA: Oxford

DAL 19040019 AL 19040019

PESO NG: 67.500
MATERIALE: BROAZO FUSO

MISURE H. CM. 6.5; LUNGH. CM. 17.5; LARGH. CM. 6
COLLOCATIONE: DEPOSIT M.A.R.F. ARAMONO 119 8.5
MICHIGANIA CAMPA DOLL THE ABANCHLE.

VALORE RESIDURIENO: € 40.000,00

STATO DI CONSERVAZIONE: BUONE CONDIZIONI, MANCANTE DELLA PRESA, DELLA POPPA, MANCANTE DELLE CORNA TERMINALI DELLE FIGURA ANIMALE A PRUA, METNA VERDALITRA

P.S. VEDI DOCUMENTAZIONE FOTOGRAFICA

NOTE: IL REPERTO DEVE ESSERE IMBALLATO CON LE DOVUTE PRECAUZIONI DA PERSONALE SPECIALIZATO L'ESPOSIZIONE DOVER AVVENIRE IN AMBIENTE CON CONTROLLO DI UMIDITA E TEMPERATURA

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Mustage

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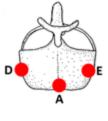
Pelo Manuele della Terrana la dela Papsia, el – 10031 Finanza – al + 30 00020071 – da: + 30 00020231 /

Elemental Characterization of Nuragic Bronze Age Votive Ships: June 2019

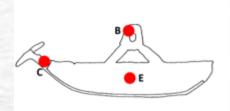
Test beam per misure reperti archeologici "navicelle Nuragiche"



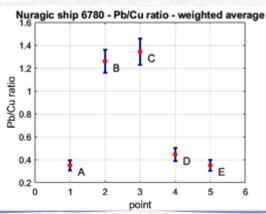


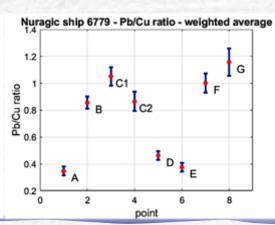


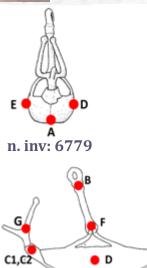
n. inv: 6780



Museo Archeologico Nazionale di Firenze







Summary of features and drawbacks

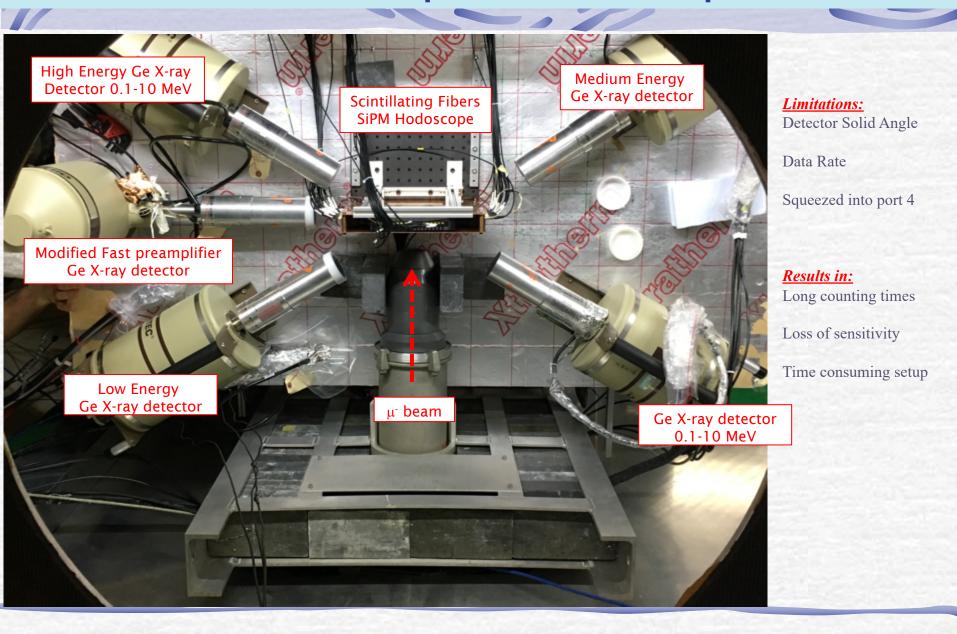
Muonic atom X-rays spectroscopy has proved to be:

- a non-invasive, simoultaneous multielemental analytical technique
- **High energy characteristic X-rays** emission (**small auto absorption** also for low Z elements) potentially applicable for **all elements** (from Li to U) without vacuum system
- **Depth** (site) **selective** (easy muon beam momentum scan) possibility to perform depth profile elemental characterization (**3D mapping**)
- Very high specifity (energy and temporal signatures) and negligible radioactivation
- **Complementary technique** to other non-invasive analytical techniques for bulk (PGNAA) or surface analysis (XRF)

Drawbacks:

- Poor sensitivity (only major elements)
- Long counting/irradiation time (12-18 h to reach sensitivity to 0,5% m/m !!!)
- Very low solid angle coverage (caused by detector pile-up)

Current experimental Setup

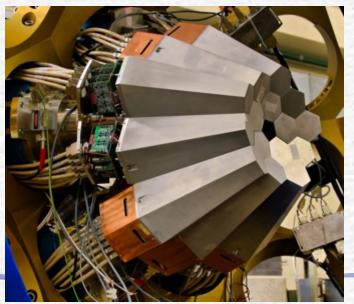


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A Possible Solution



By Shantonu Biswas



New set-up like "**AGATA experiment":

Pixelated Ge detectors

Ge crystals are tapered hexaconical shape electronically into 36 segments

Possible design

upstream and downstream banks gives open access for large cultural heritage samples

^{*}The Advanced GAmma Tracking Array (AGATA) is a European gamma-ray spectrometer used for nuclear structure studies

Spettroscopia muonica: Prospettive future

- RUN Ottobre 2020: Irraggiamento Moneta Portoghese del XVIII sec inter-comparison IAEA nuclear techniques





PhD Novembre 2020: co-finanziato da RIKEN-RAL e DFO-UNIMIB

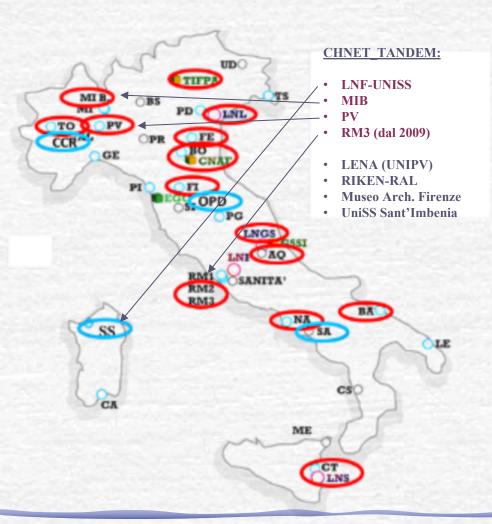


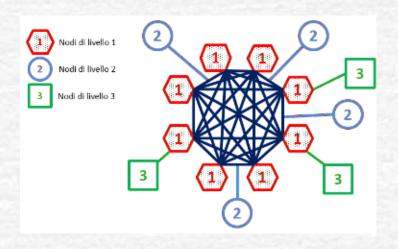


Slide di backup

CHNET_TANDEM: R&D CHNET e CSNV

Born to coordinate the cultural heritage activities of INFN facilities

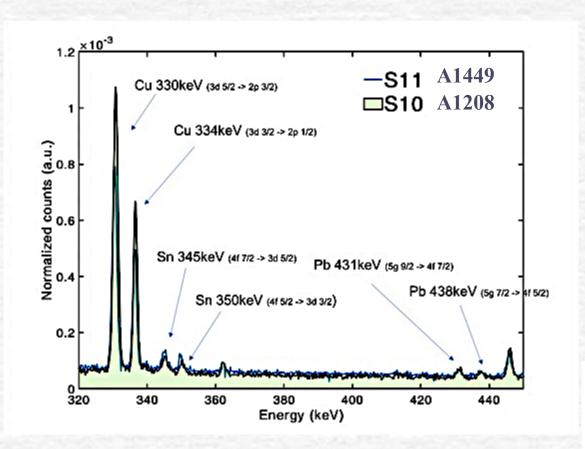




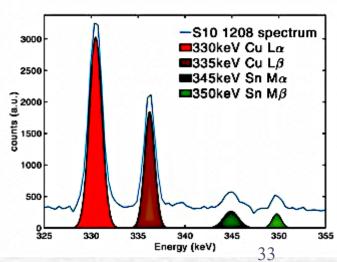
CHNET

- Common R&D lines and activities
- Sharing funds from the Institute and projects
- Common strategies for **recruitment** solutions
- Technology & knowledge transfer
- Answering the issues of the Italian public Institutions devoted to the preservation of the Cultural Heritage
- Expanding the network worldwide

June-July 2018: implementation of new DAQ DT5780 CAEN



The analysis showed interesting differences in the ratio of Sn/Cu and in the presence of Pb, which give the possibility of dividing the set of samples into two set one with Cu/Sn content and the other with Cu/Sn/Pb



Elemental Characterization of Nuragic Bronze Age Votive Ships

Journal of Radioanalytical and Nuclear Chemistry (2019) 322:1357-1363 https://doi.org/10.1007/s10967-019-06927-6



Muonic atom X-ray spectroscopy for non-destructive analysis of archeological samples

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Abstract

The implementation in the RIKEN-RAL negative muons facility of a new muon beamline monitoring and novel digital data acquisition system for gamma and X-ray spectroscopy are presented. This work also shows the high potential of the muonic atoms X-ray spectroscopy technique in non-destructive elemental characterization of archaeological samples.

Keywords Muonic atom X-ray spectroscopy · Pulsed muon beam · Non-invasive elemental analysis · SIPM-fibers scintillating hodoscope



Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

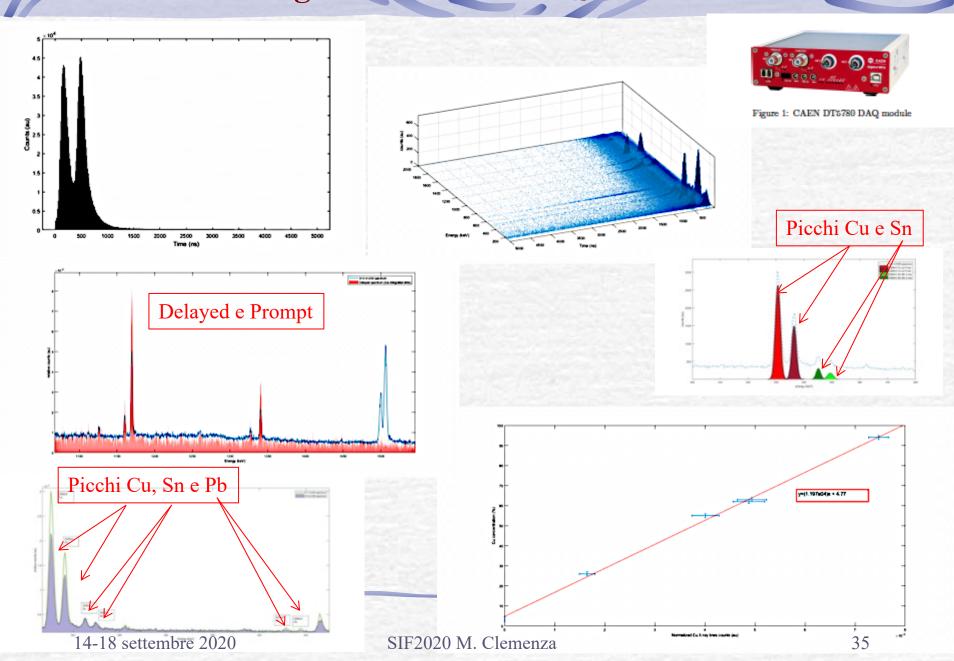


Journal homepage: www.elsevier.com/locate/nima

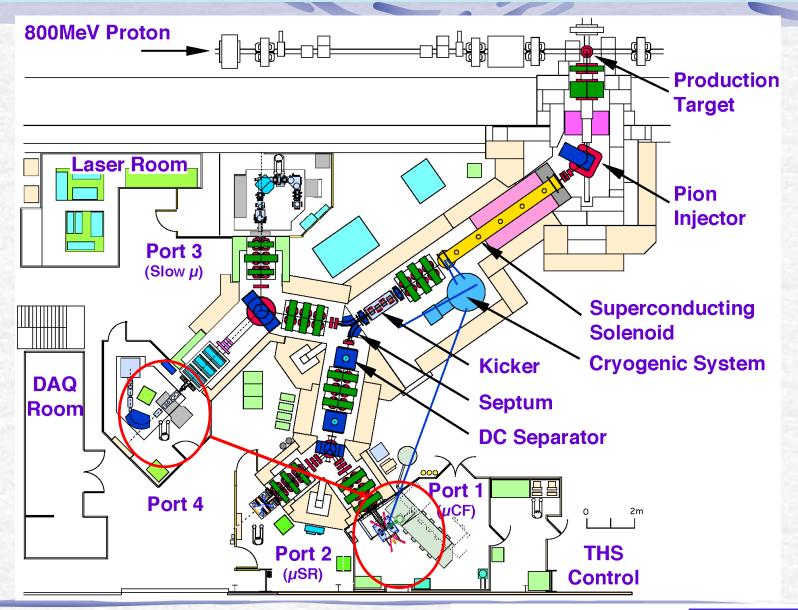
CHNET-TANDEM experiment: Use of negative muons at RIKEN-RAL Port4 for elemental characterization of "Nuragic votive ship" samples

M. Clemenza **, G. Baldazzi **, G. Ballerini **, M. Bonesini *, M. Carpinelli **, O. Cremonesi *, E. Di Stefano **, E. Fiorini **, G. Gorini **, A. Hillier *, K. Ishida *, A. Menegolli **, M. Minoja **, E. Mocchiutti **, P. Oliva **, M. Prata *, A. Pullia **, M. Rendeli **, L.-P. Rignanese **, M. Rossella *, V. Sipala **, M. Soldani **, L. Tortora *, A. Vacchi ***, E. Vallazza **

RUN TANDEM Luglio 2018: nuovo DAQ DT5780 CAEN



A Possible solution











Dr. Adrian Hillier
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23 July 2018

Elemental analysis deep beneath the surface

To whom it may concern.

Science and Technology Facility Councils Rutherford Appleton Laboratory is located on the Harwell Science and Innovation Campus in Oxfordshire, it provides a thriving and collaborative environment for research in different fields like Condensed Matter, Biological system, Chemical analysis, particle physics, space science, materials, astronomy etc... On the campus, we have a neutron and muon facility (ISIS), together with a light source (DIAMOND) and these facilities are used to investigate the material world. This can also be applied to the conservation, preservation and study of cultural heritage that is a field of concern within Italy and Europe.

In particular, it is possible to perform the non-destructive depth analysis of a variety of materials using the intense muon beam @RIKEN-RAL (a 33 year collaboration between ISIS and RIKEN, Japan). By tuning the momentum of the muon the implantation depth can be easily controlled and can penetrate into materials much deeper than electrons without any residual radioactivity. The characteristic muonic X-rays have about 200 times higher energy than that of characteristic X-rays generated by electron beam analysis. Therefore, it becomes possible to obtain information about chemical composition inside materials, from 100 microns to centimetres thick (with a resolution of +/-20%), in a non-destructive manner.

Currently, a successful collaboration with INFN and other research bodies, in particular with the research group of Massimiliano Clemenza of the University of Milano Bicocca, a series of experiments to optimize muonic spectroscopy as a non-invasive and non-destructive probe for quantitative "bulk" analysis for the use in the archaeometric field for elemental characterization of metal artefacts and ancient objects.

This project aims to perform an in-depth study with a negative muon beam of metallic artefacts from the late Bronze-age found in different area of the Italy (Sardinia, Tuscany, Lazio and Campania regions). The ultimate goal is to achieve a series of measurements on homogeneous type objects from three different areas of the Italian peninsula, all affected by contact with the Sardinian population. In particular, it would be analyzed similar objects from different contexts:

The possibility to investigate the miniaturist boat from the grave 74 Montevetrano (Salerno), the miniaturistic shaped pail from Pontecagnano (Salerno), the analogue miniaturist object from the tomb of Sardinian bronzes Cavalupo Vulci and a boat would be of great interest. In addition, possible objects of an interesting study would be from the Tyrrhenian Etruria, one could choose from the Gravisca sanctuary (Viterbo) or that of the recent discovery by the waters of Lake Trasimeno (San Feliciano, Perugia). It would be of great interest to include the analysis of similar objects from the Sardinian territory, with regard to this we propose the following possible artefacts such as pail of Santa Vittoria di Serri and/or that of Santa Cristina of Paulilatino, the miniaturist boat from Villagrande-S'Arcu' and forros and / or the Sorridile boat and / or Baunei boat with a monkey. If there is the possibility of further extending this project thought should be given to the statue of "centaur"



of Nule, as try to provide useful input to the on going research on the technical characteristics of its

The research work is proposed and supported by STFC, INFN, UNIMIB, UNISS and CNR.

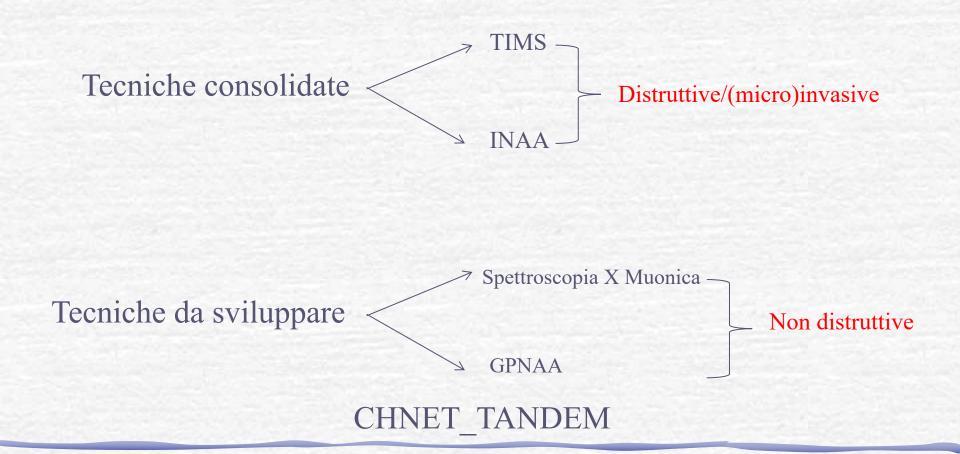
Yours sincerely,

Adrian Hillier Muon Group Leader ISIS Pulsed Neutron and Muon Facility STFC Rutherford Appleton Laboratory

Grazie per l'attenzione

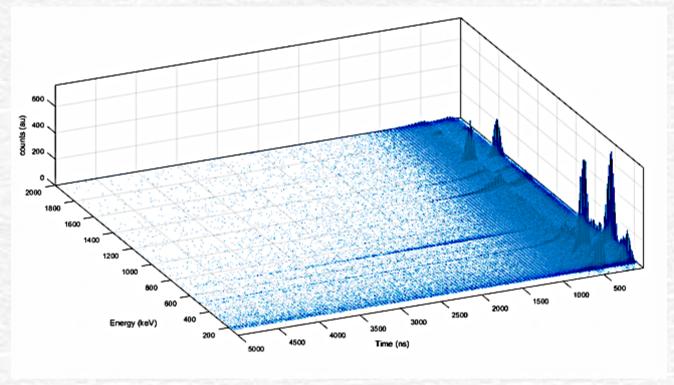
Progetto TANDEM: Tecniche Analitiche Non Distruttive per l'archEoMetria

L'idea che sta alla base del progetto **TANDEM** è quella di implementare, sviluppare e ottimizzare tecniche di analisi da utilizzarsi in ambito archeometrico per la caratterizzazione di manufatti



June-July 2018: implementation of new DAQ DT5780 CAEN

Energy spectrum and Time distribution



Research and development of analytical techniques CHNET_TANDEM experiment: X rays spectroscopy with Muonic Atoms

Non-destructive elemental analysis by X-ray spectroscopy of Muonic Atoms

PORT4 RIKEN-RAL@ISIS

Collaboration:

INFN-MIB - LNS- UNISS- UNIPV-LENA - ISIS-RIKEN RAL Depth of Implantation as a function of muon beam moment/energy

Surface and bulk elemental characterization without activation!!

Muon Momentum (MeV/c)

