

## CHNET TANDEM: Research and development of non-destructive analytical techniques

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

#### Tecniche Analitiche Non Distruttive per l'archEoMetria

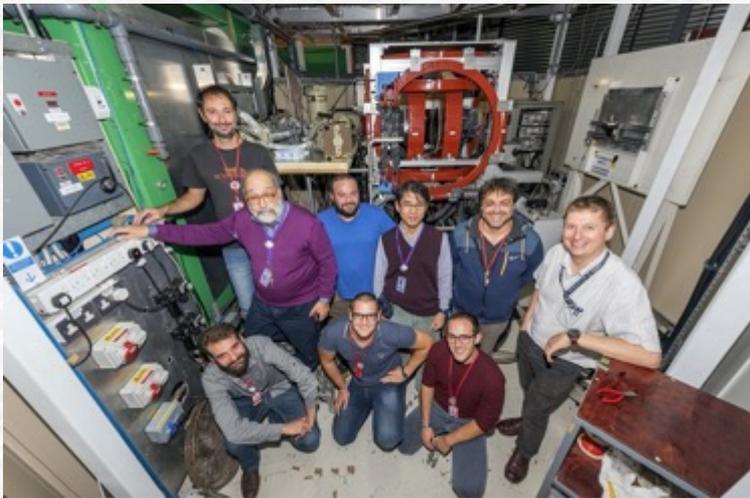
The idea behind the experiment called CHNET\_TANDEM is to implement, develop, optimize **non-destructive** and **non-invasive** analysis techniques to be used in the archaeometric field for the **elemental characterization** of finds for **Italian Cultural Heritage**. Funded and supported by INFN CSNV 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)

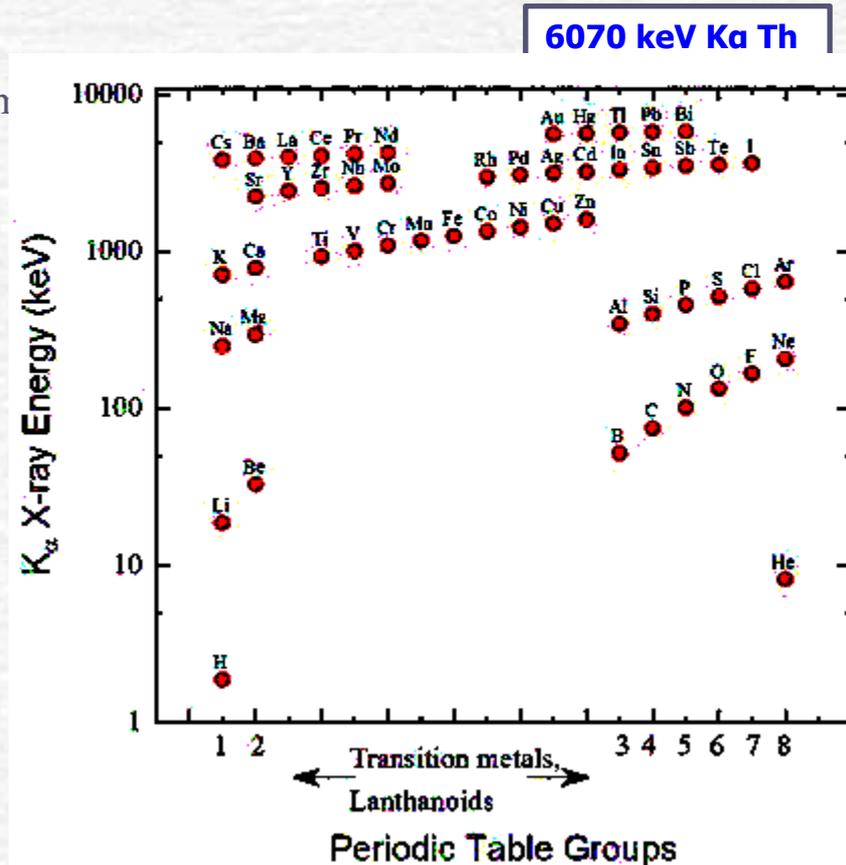


**Both techniques** are based on the detection of **prompt** electromagnetic radiation emissions in the first case of **atomic** (muonic) type, in the second of **nuclear** type.

# 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.

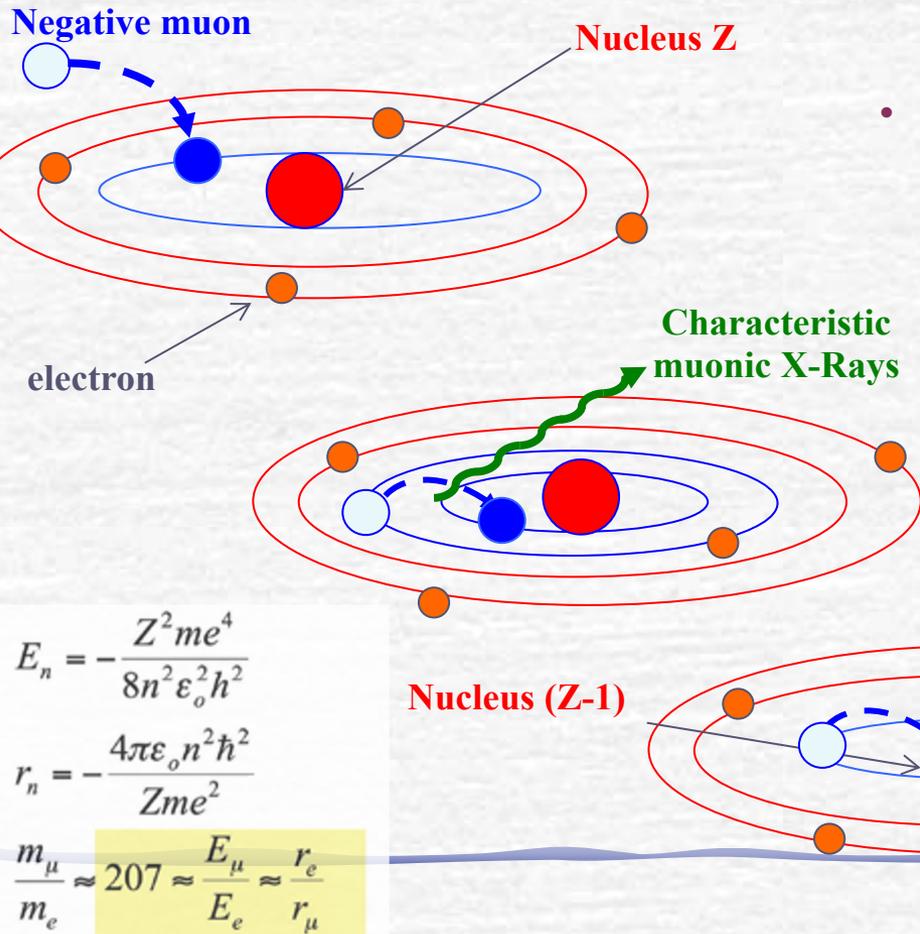
- High energy **characteristics** X-rays **10 keV to 10 MeV** observable from outside of sample - No need of vacuum  
-> applicable to matrices and several shapes;
- Applicable to **every element** from (Li to U) *simultaneous multi-elemental*
- Possibility to perform **depth profile** by changing muon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



# 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/Implantation of muonic atoms inside the sample.

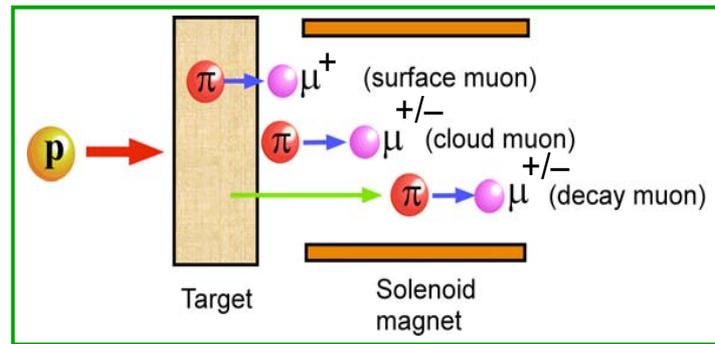
## Muonic atom formation step



## Processes involved

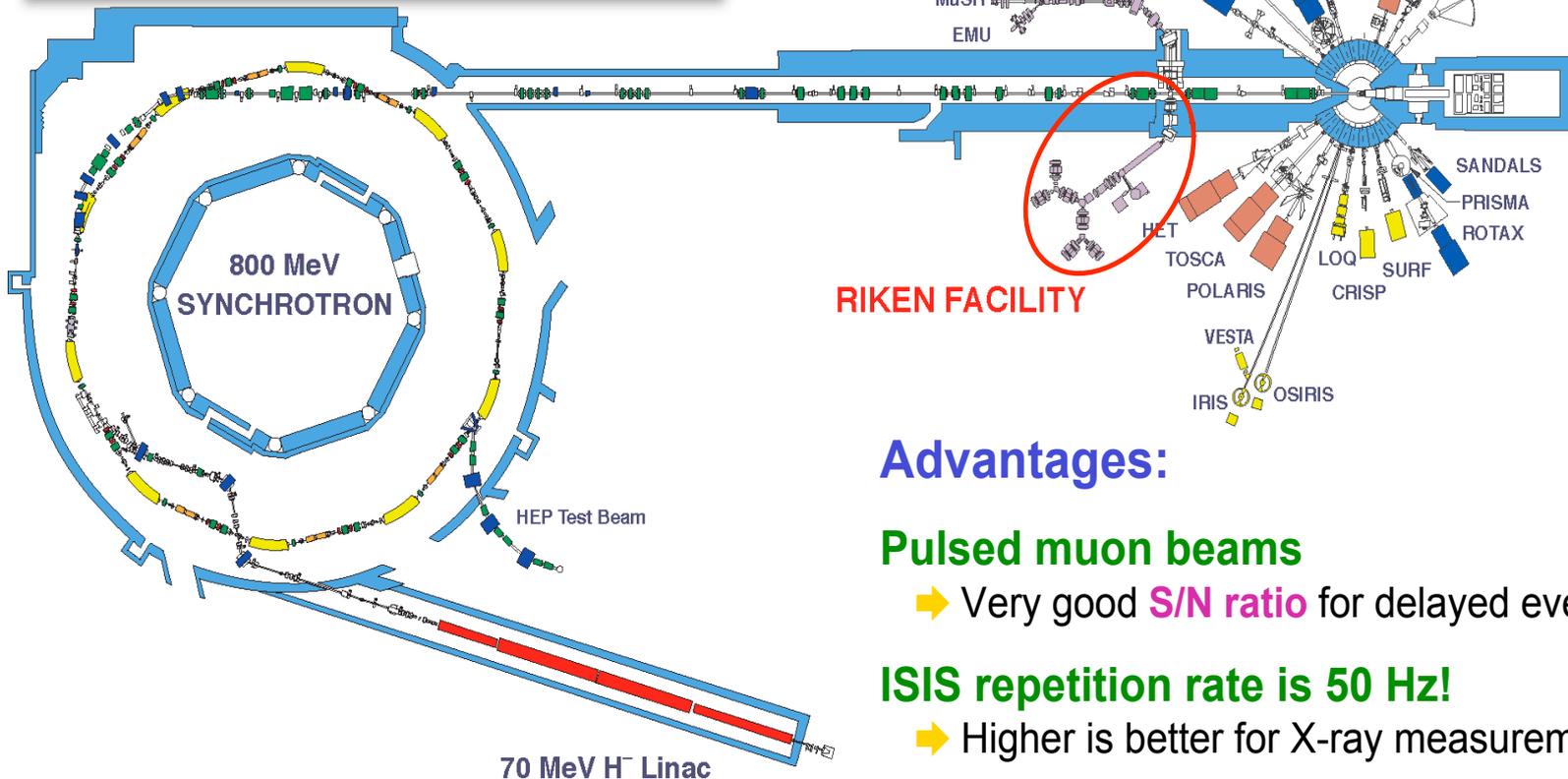
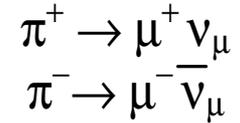
- **Muonic Atom formation**: muon capture in atomic muonic orbital
- **Muonic Cascading process**: emission of high energy **characteristic muonic X-Rays**
- **Natural decay/muon capture** by nucleus with **Prompt Gamma Ray emission**

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



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

800 MeV proton  
(50Hz, 200 μA, double 70ns pulse)



## Advantages:

### Pulsed muon beams

➔ Very good S/N ratio for delayed events.

### ISIS repetition rate is 50 Hz!

➔ Higher is better for X-ray measurements.

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

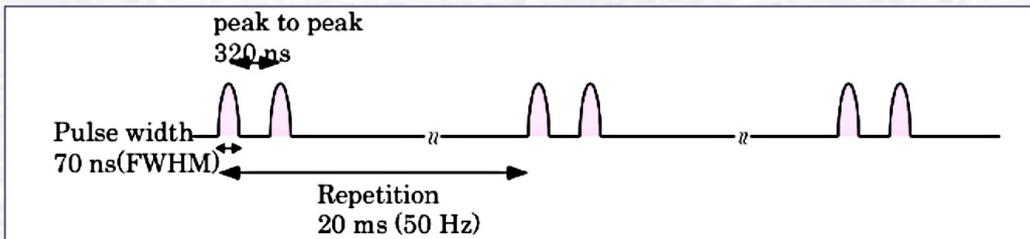
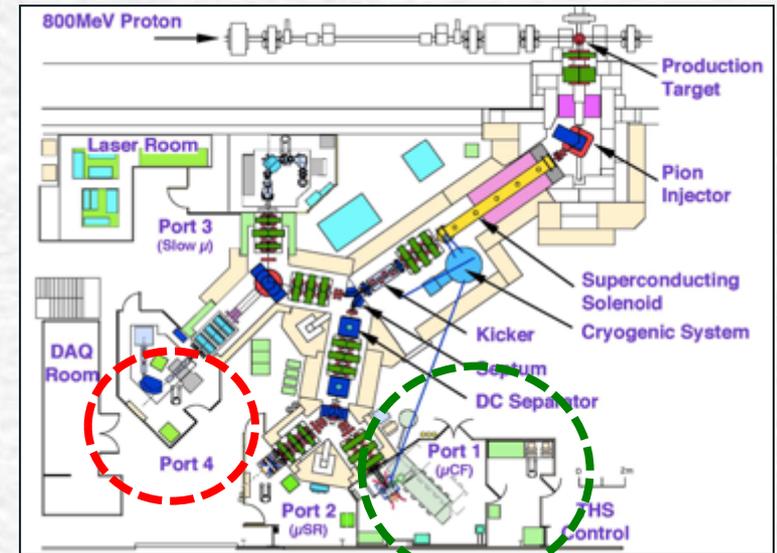
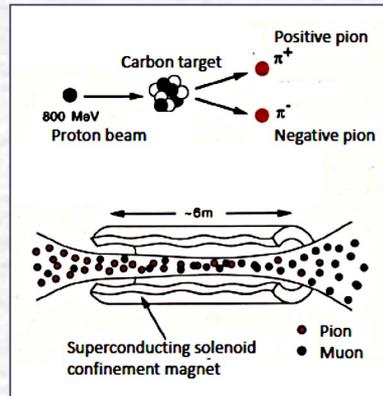
## Beam Structure @ PORT4 ISIS Muon source

$8 \times 10^4$  muons /sec @60 MeV/c

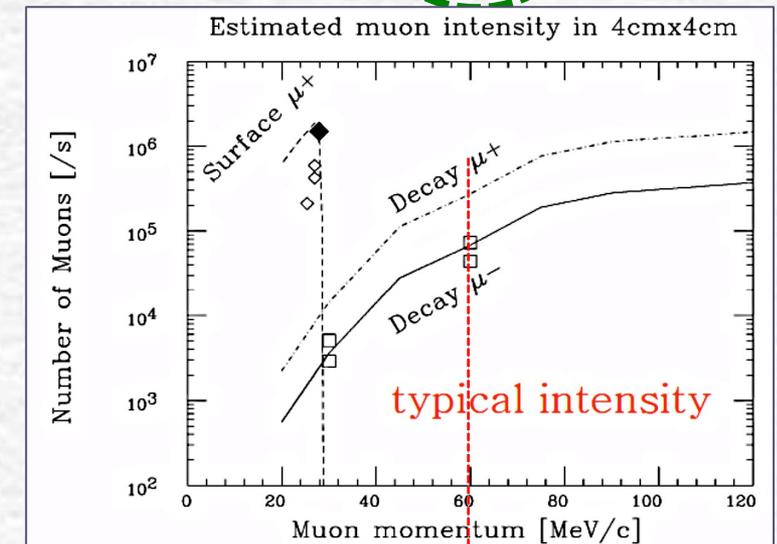
### Beam properties

surface  $\mu^+$  (20~30 MeV/c) and  
decay  $\mu^+ / \mu^-$  (20~120 MeV/c)

choice of double or single pulse  
with magnetic kicker (<60 MeV/c)  
 $\Delta p/p$  FWHM 10%(decay), 5%(surf)  
typical beam size 10cm<sup>2</sup>



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



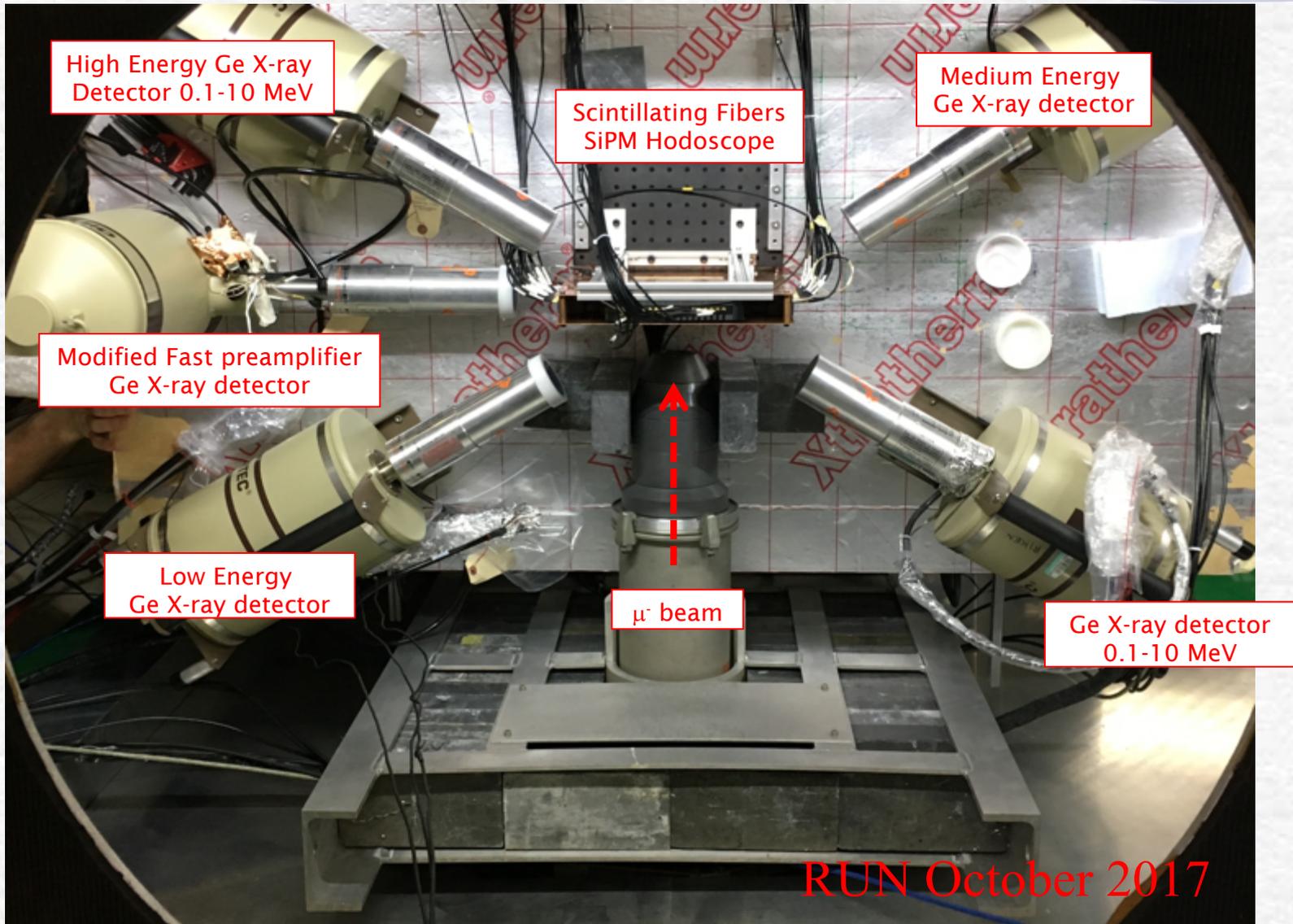
Thanks to K. Ishida (RIKEN)

14-18 settembre 2020

SIF2020 M. Clemenza

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



RUN October 2017

Spin-off dell'Esperimento FAMU CSNIII (raggio di Zeemach del protone tramite spettroscopia laser del  $\mu p$  spokesperson 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»*

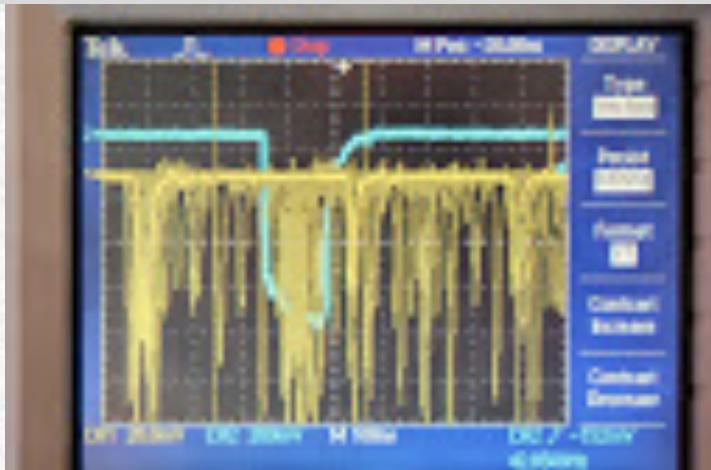
### **RUN Ottobre 2020:**

- *Irraggiamento Moneta Portoghese del XVIII sec intercomparison IAEA*

# Hodoscope Performances at RIKEN/RAL

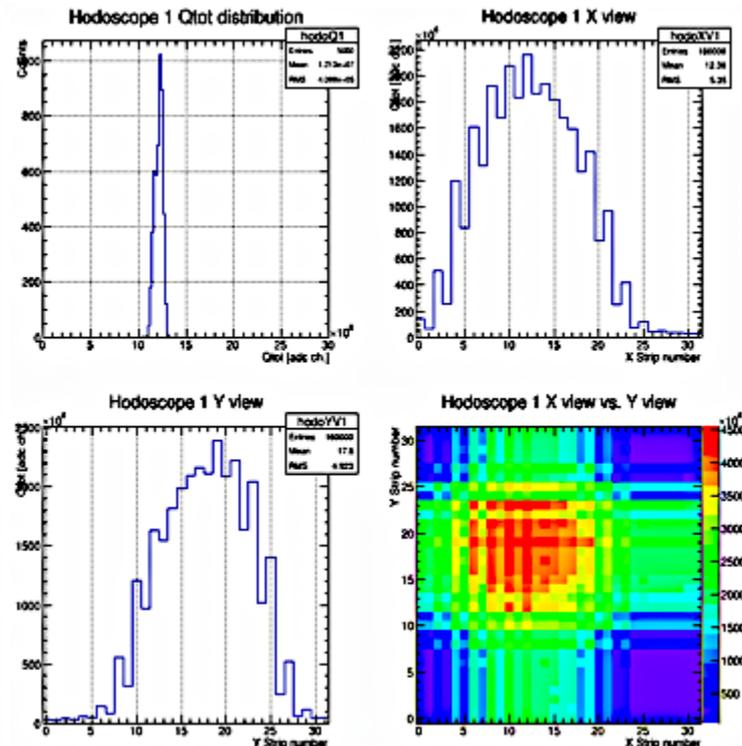
- The system is based **on 2 different hodoscopes** Fiducial area is  **$3.2 \times 3.2 \text{ cm}^2$  ( $10 \times 10 \text{ cm}^2$ )** (one with **1 mm** and one with **3 mm** pitch) with similar design
- Mechanics is realized with a 3-D printer
- 2 X/Y **Bicron BCF12** square single clad **scintillating fiber** planes (32+32 channels) read by  $3 \times 3 \text{ mm}^2$  (or  $1 \times 1 \text{ mm}^2$ ) 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  $\rightarrow$  peak, area, time) and common HV for a fiber plane ( $V_{\text{brk}}$  in a large SiPM sample is very similar )
- Measure X/Y profile and monitor beam intensity**

Pulsed high intensity muon beam ( $\sim 70$  muons/fiber/70 ns)

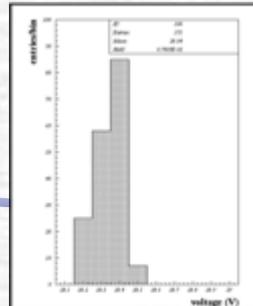


RUN October 2017

A typical run at 60 MeV/c:  $8 \times 10^4 \mu\text{-sec}$

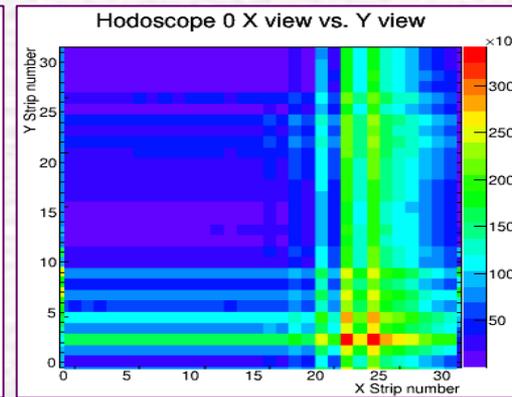
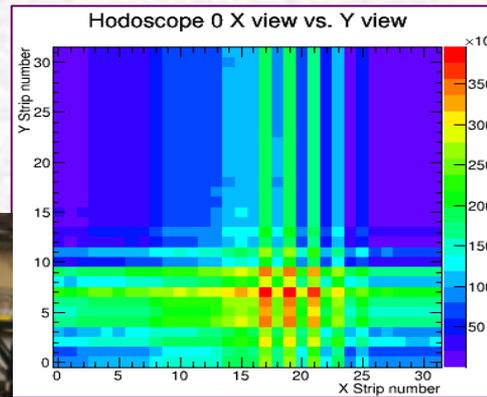
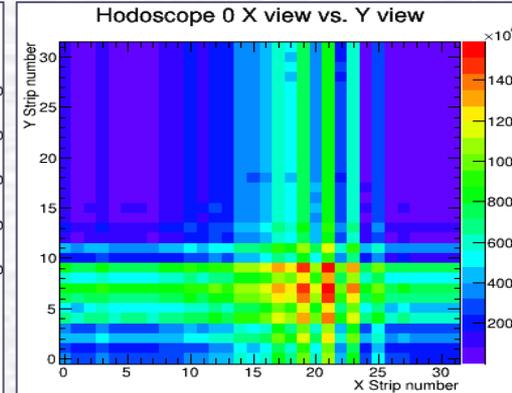
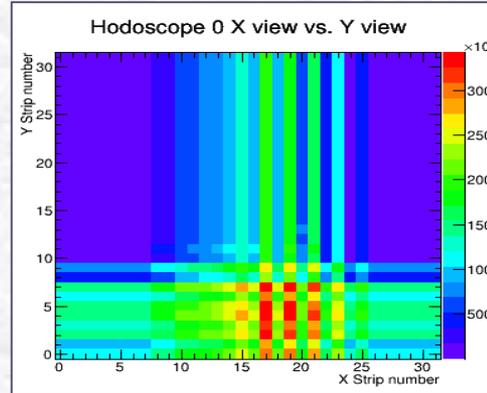
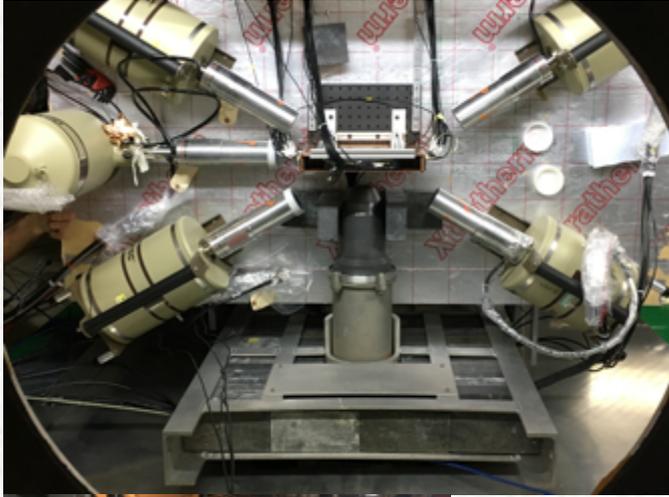


The beam has a 1.10 (0.98) cm r.m.s in x(y) coordinates



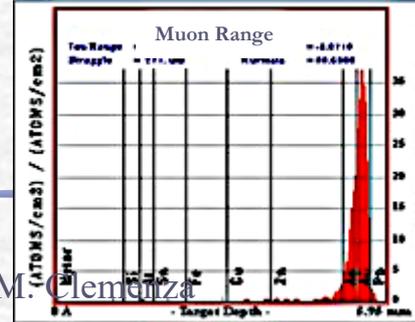
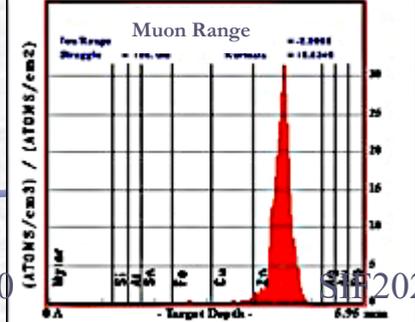
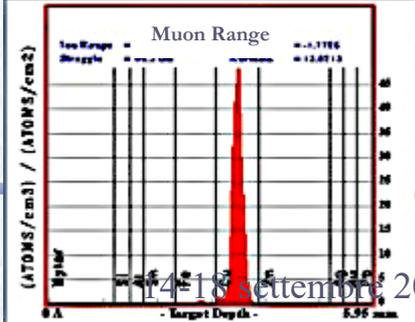
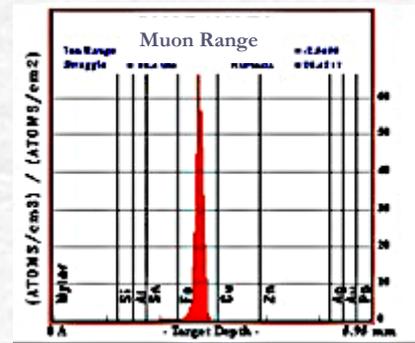
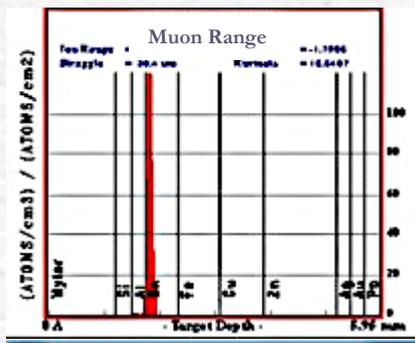
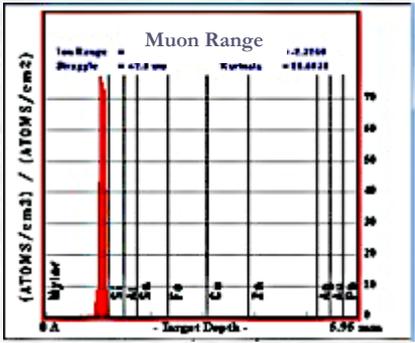
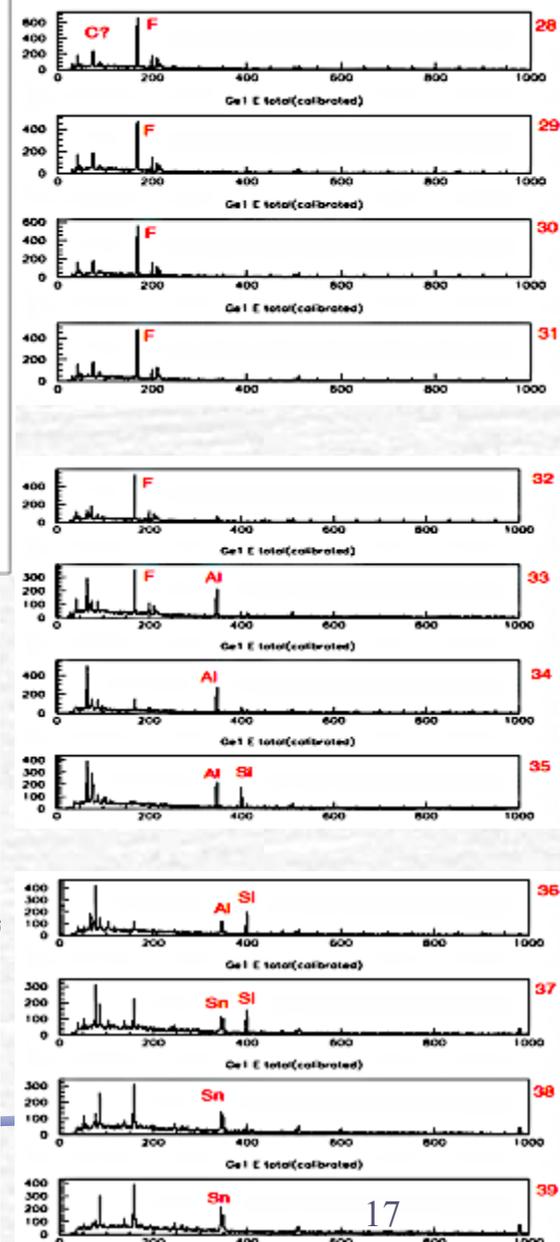
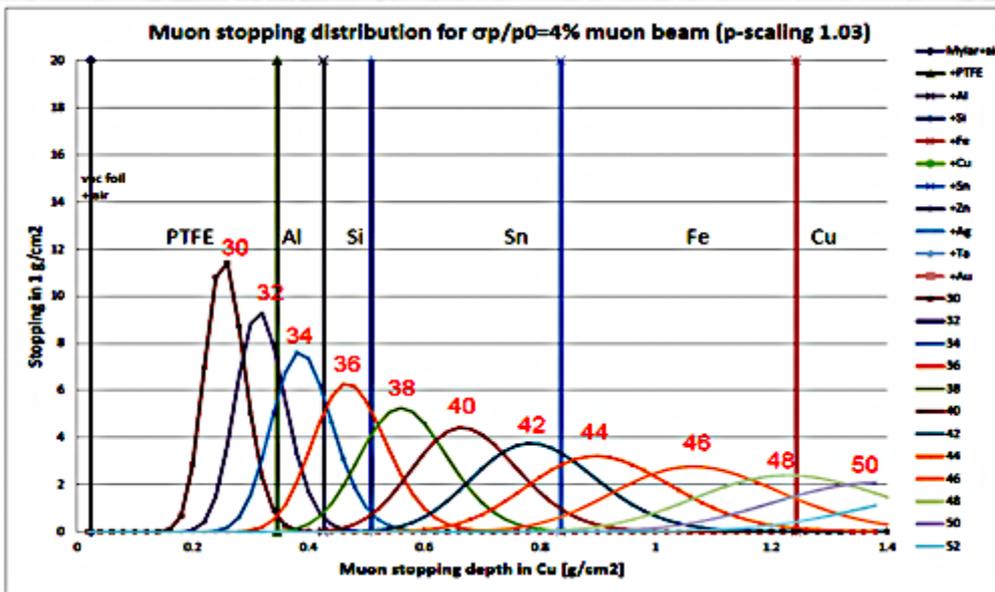
# SiPM-Scintillating Fibers Hodoscopes: october 2017

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;

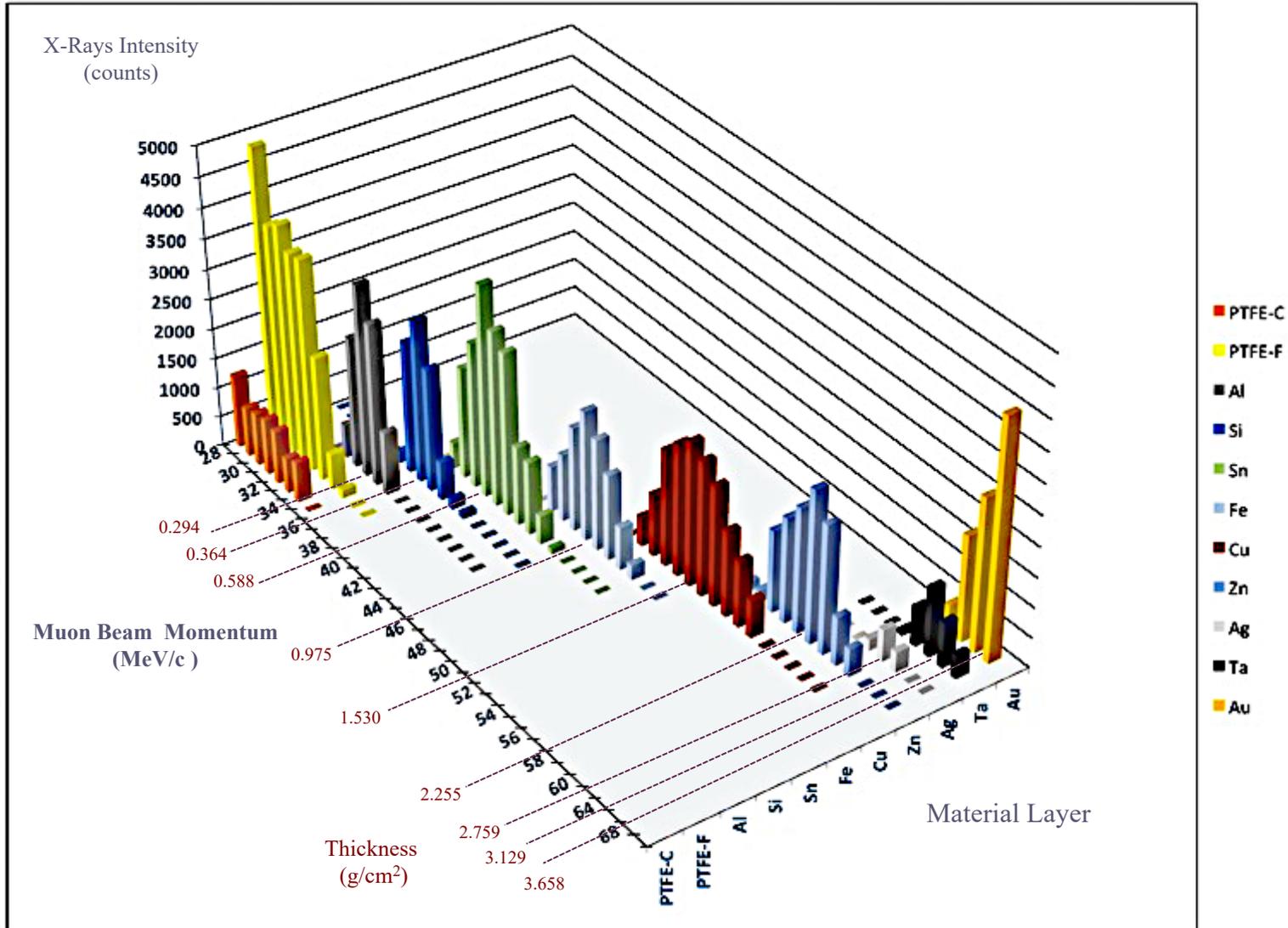


# TRIM simulation of a MultiLayered sample: october 2017

Material	Thickness (mm)
PTFE	1,2
Aluminum	0,25
Silicon	0,3
Tin	0,5
Iron	0,5
Copper	0,8
Zinc	1,0
Silver	0,25
Tantalum	0,25
Gold	0,3

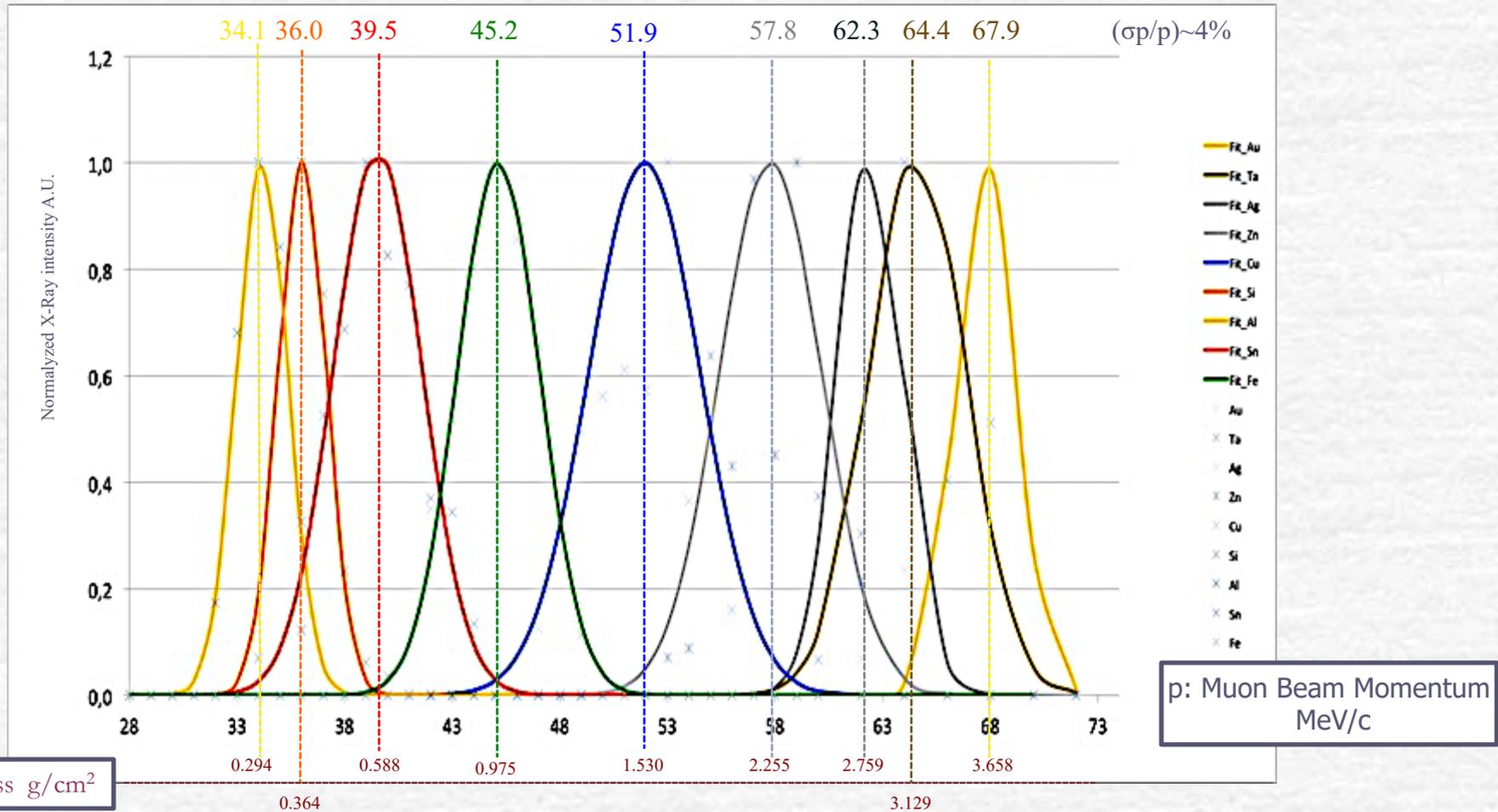


# Multi-Layered sample Results october 2017



# Multi-Layered sample Results october 2017

## Optimization of Beam momentum



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

# June-July 2018: implementation of new DAQ DT5780 CAEN

## Dual Digitizer Multi Channel Analyzer - Desktop



CAEN DT5780 DAQ module

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

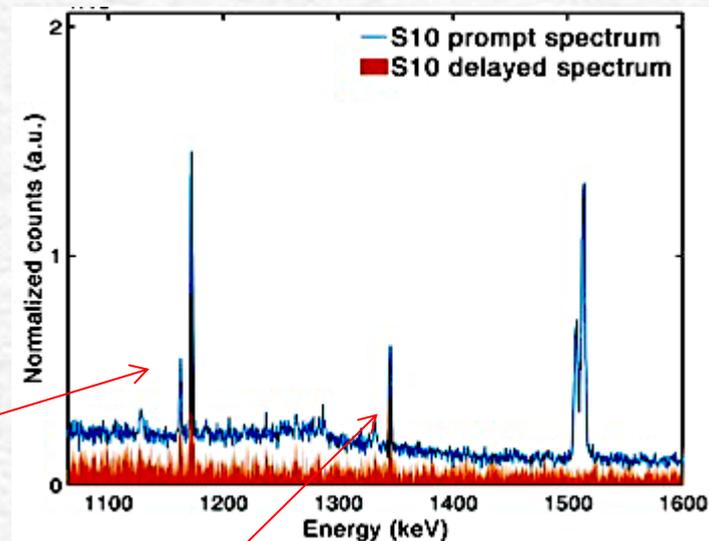
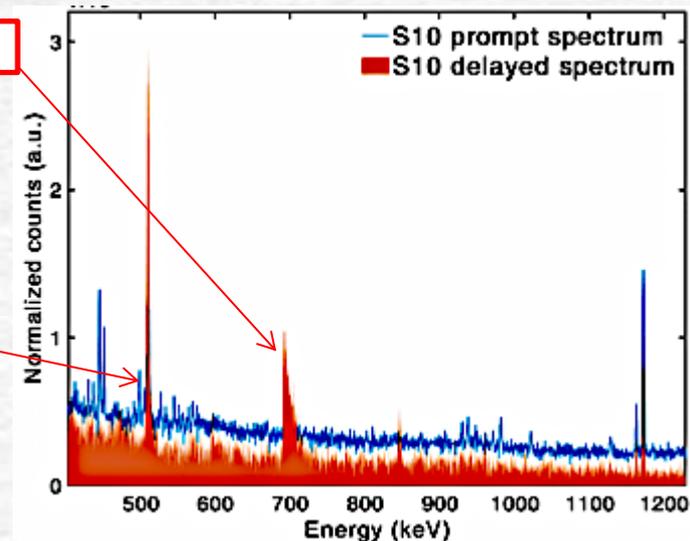
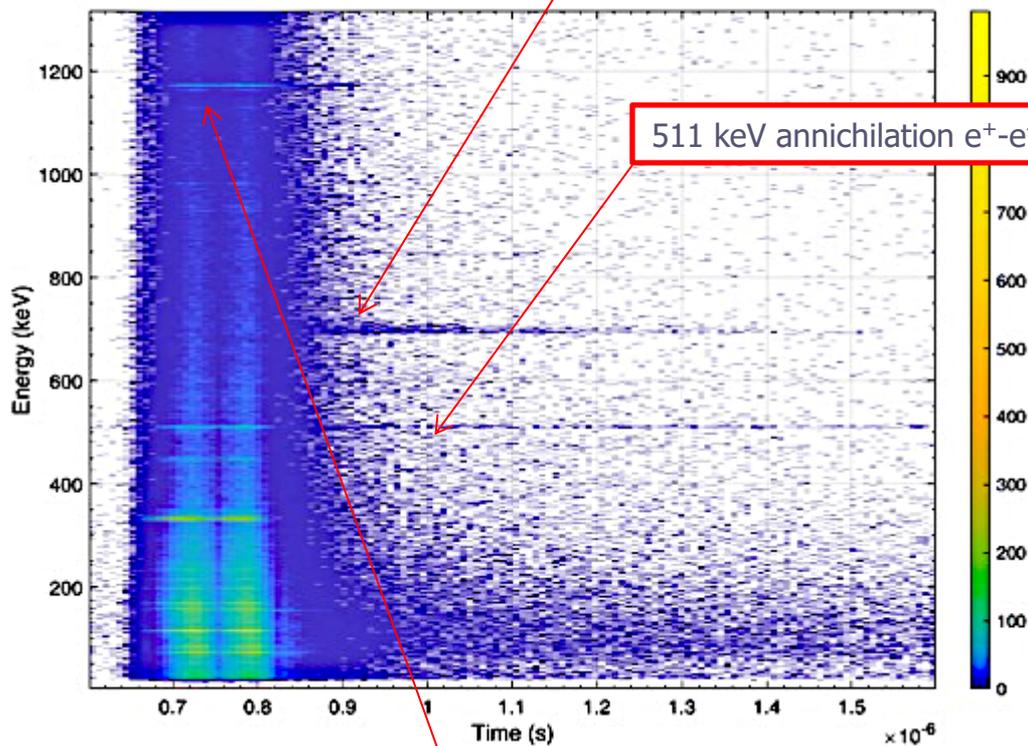
interaction of n-Ge (n, n') (~700keV)

511 keV annihilation  $e^+e^-$

1173 - 1163 keV from

muon capture reaction  
 $^{63}\text{Cu} + u \rightarrow ^{62}\text{Ni} + n + \text{gamma}$ .

1345keV from  $^{64}\text{Ni}$   
 $^{65}\text{Cu} + u \rightarrow ^{64}\text{Ni} + n + \text{gamma}$



# June-July 2018: Calibration Curves

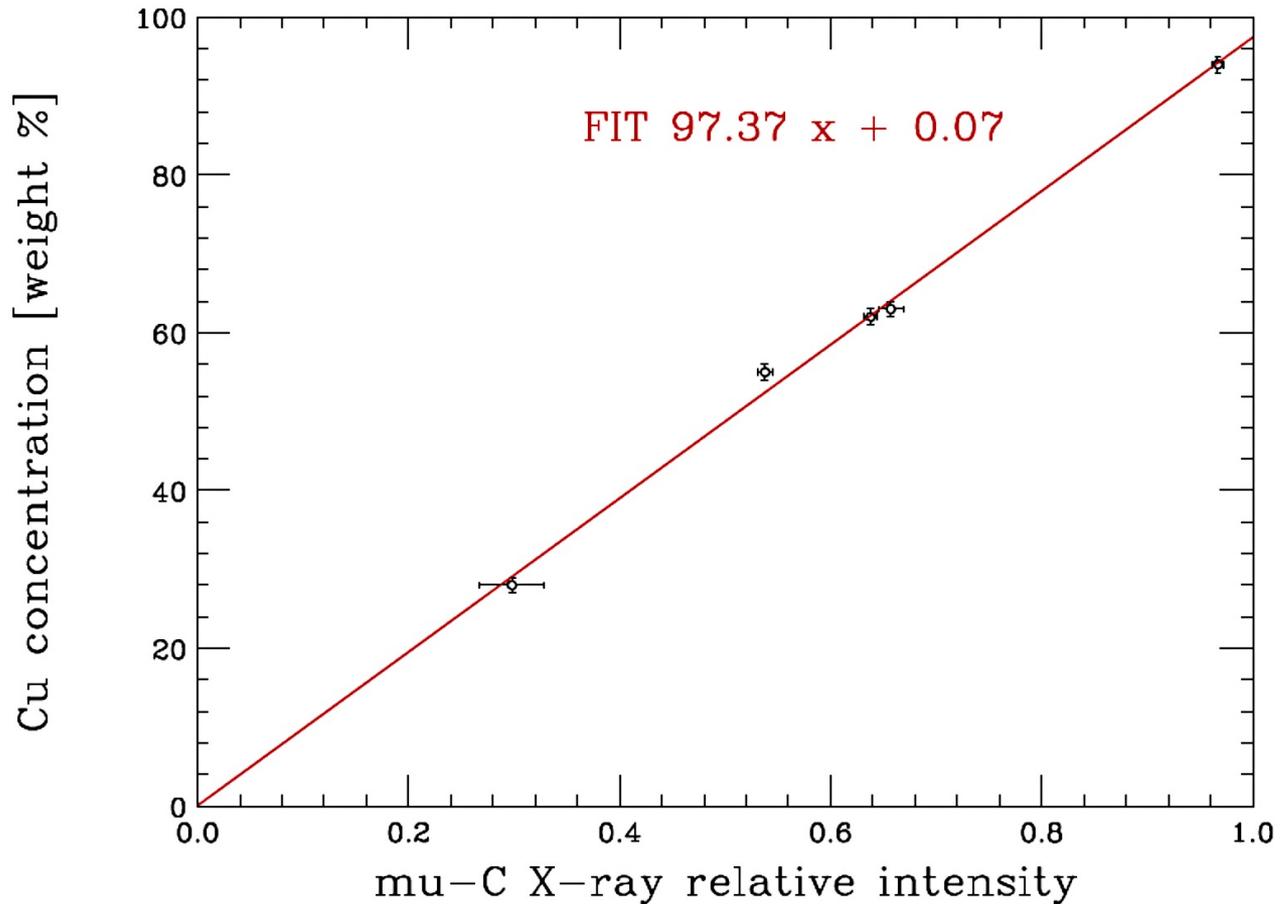
## “Reference” Materials:

Silver 99.95  
Cu99.9  
Al 99.999  
Al77.9Si17.8Cu3.3Mg1.0  
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

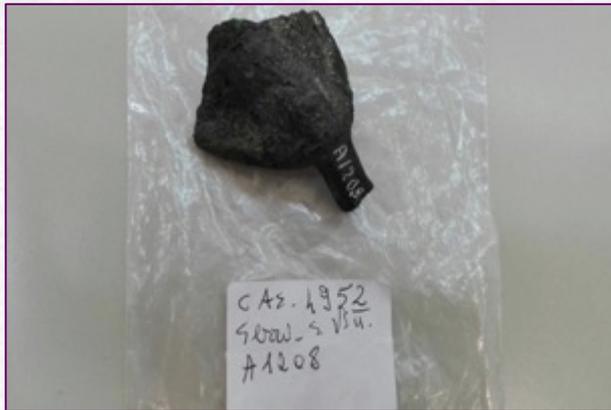
Cu:

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 additivo nella lega di Rame e stagno soprattutto in alcune particolari zone di giunzione (prua della navicella).



Test with "real"  
Archeological samples



*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)

  
Ministero per i Beni e le Attività Culturali e del Turismo  
**POLO MUSEALE DELLA TOSCANA**  
**MUSEO ARCHEOLOGICO NAZIONALE DI FIRENZE**  
SCHEDA CONSERVATIVA

MOSTRÀ: "Laboratorio di analisi di Oxford"  
LUOGO E DATA: Oxford  
DAL 13/06/2019 AL 19/06/2019

OGGETTO: BARCHETTA  
INVENTARIO: 6779  
PESO KG.: GR. 500  
MATERIALE: BRONZO FUSO  
DATAZIONE: VII SEC. A.C.  
MISURE: H. CM. 13,5; LUNGH. CM. 20,5; LARGH. CM. 6  
COLLOCAZIONE: DEPOSITI M.A.N.F. ARMADIO 119 B 5  
PROVENIENZA: VETULONIA TOMBA DELLE TRE NAVICELLE  
VALORE ASSICURATIVO: € 50.000,00

STATO DI CONSERVAZIONE: OTTIME CONDIZIONI, INTEGRATA, LA PRESA E' INCLINATA VERSO LA DESTRA, PASTINA VERDASTRA.

P.S. VEDI DOCUMENTAZIONE FOTOGRAFICA

NOTE: IL REPERTO DEVE ESSERE IMBALLATO CON LE DOVUTE PRECAUZIONI DA PERSONALE SPECIALIZZATO, L'ESPOSIZIONE DOVRA' AVVENIRE IN AMBIENTE CON CONTROLLO DI UMGIDITA' E TEMPERATURA

LUOGO, DATA, FIRMA: FIRENZE 13/06/2019  
IL DIRETTORE DEL MUSEO: DOTT. MARIO IOZZO      RESTAURATORE: STEFANO SARRI

**Polo Museale della Toscana**  
P.le della Repubblica, 47 - 50127 Firenze - tel. +39 055 231117 - fax +39 055 231211



  
Ministero per i Beni e le Attività Culturali e del Turismo  
**POLO MUSEALE DELLA TOSCANA**  
**MUSEO ARCHEOLOGICO NAZIONALE DI FIRENZE**  
SCHEDA CONSERVATIVA

MOSTRÀ: "Laboratorio di analisi di Oxford"  
LUOGO E DATA: Oxford  
DAL 13/06/2019 AL 19/06/2019

OGGETTO: BARCHETTA  
INVENTARIO: 6780  
PESO KG.: GR. 500  
MATERIALE: BRONZO FUSO  
DATAZIONE: VII SEC. A.C.  
MISURE: H. CM. 6,5; LUNGH. CM. 17,5; LARGH. CM. 6  
COLLOCAZIONE: DEPOSITI M.A.N.F. ARMADIO 119 B 5  
PROVENIENZA: VETULONIA TOMBA DELLE TRE NAVICELLE  
VALORE ASSICURATIVO: € 40.000,00

STATO DI CONSERVAZIONE: BUONE CONDIZIONI, MANCANTE DELLA PRESA, DELLA POPPA, MANCANTE DELLE CORNA TERMINALI DELLE FIGURA ANIMALE A PRUA, PASTINA VERDASTRA.

P.S. VEDI DOCUMENTAZIONE FOTOGRAFICA

NOTE: IL REPERTO DEVE ESSERE IMBALLATO CON LE DOVUTE PRECAUZIONI DA PERSONALE SPECIALIZZATO, L'ESPOSIZIONE DOVRA' AVVENIRE IN AMBIENTE CON CONTROLLO DI UMGIDITA' E TEMPERATURA

LUOGO, DATA, FIRMA: FIRENZE 13/06/2019  
IL DIRETTORE DEL MUSEO: DOTT. MARIO IOZZO      RESTAURATORE: STEFANO SARRI

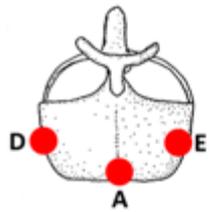
 

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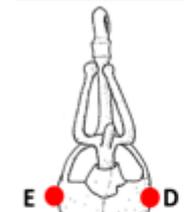
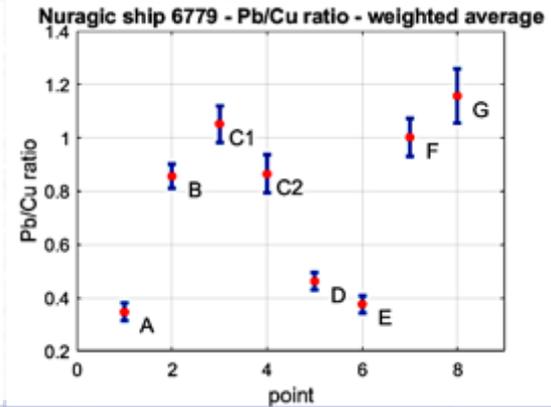
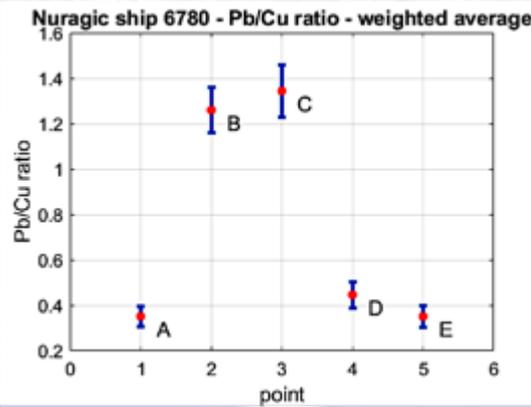
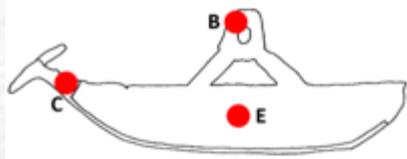
*Test beam per misure reperti archeologici "navicelle Nuragiche"*



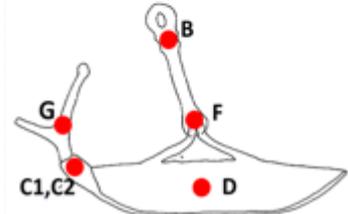
Museo Archeologico Nazionale di Firenze



n. inv: 6780



n. inv: 6779



# Summary of features and drawbacks

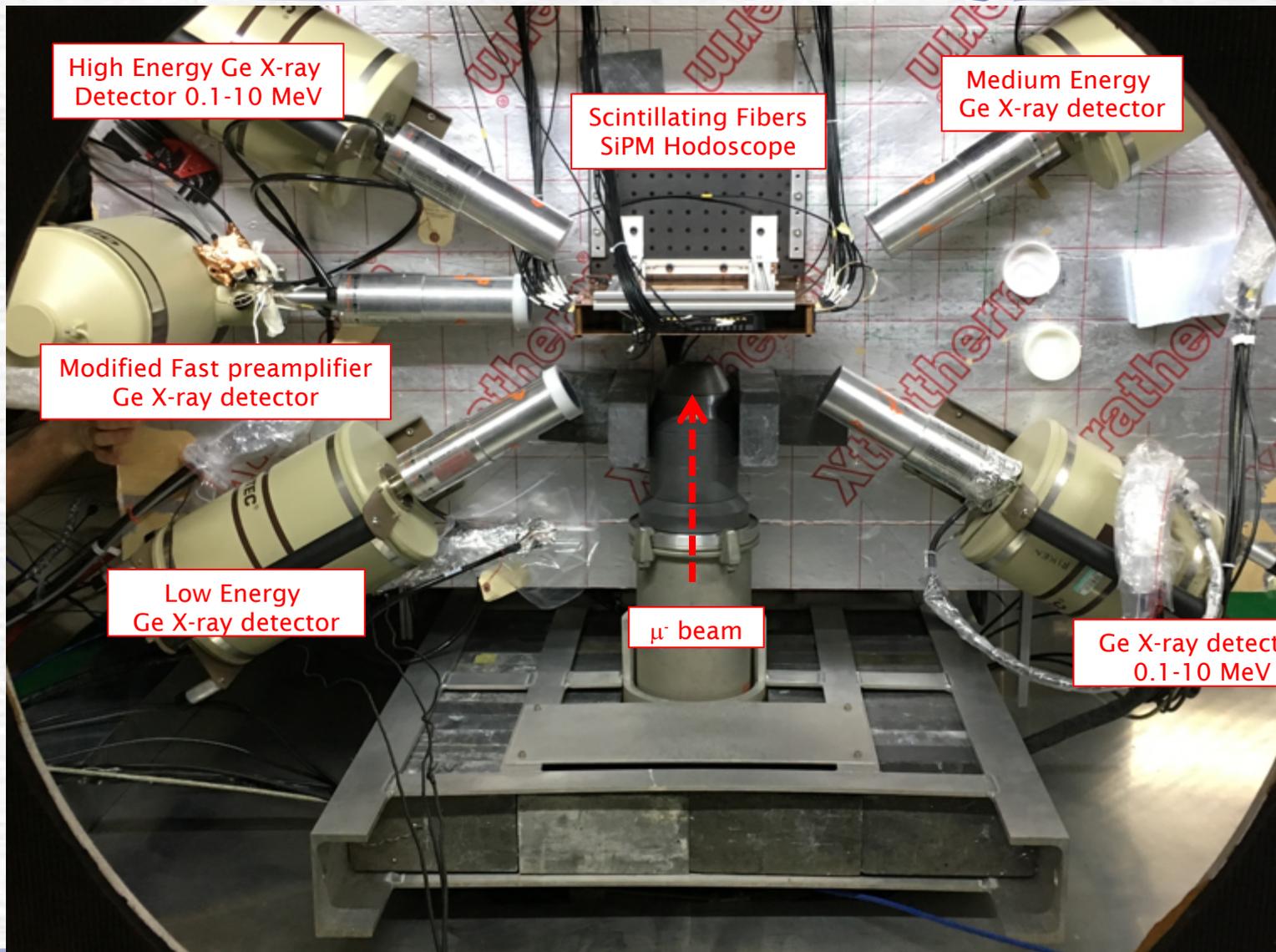
Muonic atom X-rays spectroscopy has proved to be:

- a **non-invasive, simultaneous 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 specificity (**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



High Energy Ge X-ray  
Detector 0.1-10 MeV

Scintillating Fibers  
SiPM Hodoscope

Medium Energy  
Ge X-ray detector

Modified Fast preamplifier  
Ge X-ray detector

Low Energy  
Ge X-ray detector

$\mu$  beam

Ge X-ray detector  
0.1-10 MeV

## Limitations:

Detector Solid Angle

Data Rate

Squeezed into port 4

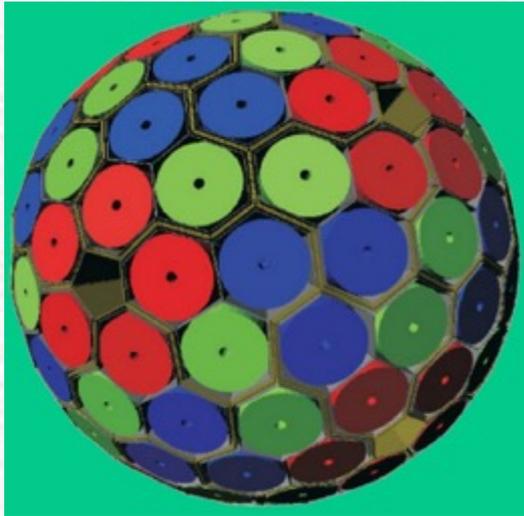
## Results in:

Long counting times

Loss of sensitivity

Time consuming setup

# A Possible Solution



By Shantonu Biswas

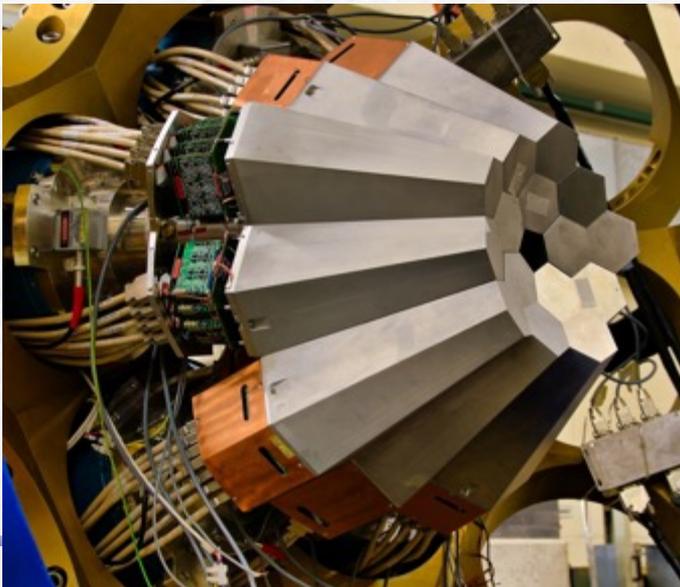
New set-up like “\**AGATA experiment*”:

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

A screenshot of an email from the ISIS Facility regarding a PhD studentship proposal. The email is dated 25 November 2019 and is addressed to Dr Clemeza and Dr Hillier. The subject is "ISIS Facility Development Studentship Proposal". The email text includes a welcome message, a statement of appreciation for the proposal, and details about the funding and the proposal's assessment. It mentions that the proposal was assessed by a panel consisting of ISIS management, senior ISIS scientists, and external members of the ISIS user community. The email also includes contact information for the ISIS Facility and a signature from Philip King, Science Director Head, ISIS Facility Studentships.

ISIS Facility  
STFC Rutherford Appleton Laboratory  
Harwell Oxford  
Didcot, OX11 0QX  
United Kingdom  
www.stfc.ac.uk

25 November 2019

Dr Clemeza and Dr Hillier

Dear Dr Clemeza and Dr Hillier

**ISIS Facility Development Studentship Proposal**

Very many thanks for your proposal for an ISIS co-funded Facility Development Studentship.

Proposals were assessed by a panel consisting of ISIS management, senior ISIS scientists and external members of the ISIS user community. We are very pleased to announce that we would like to support your studentship application, and co-fund this project with you.

There was a very strong field of proposals – a total of 30 studentship requests, all with very good science aims and facility development projects. From these submissions we are intending to fund 6 projects, including your proposal.

Could I now ask you to work with your ISIS co-supervisor to select a student, if you do not already have one in mind? The student should be appointed by 1/10/2020. If you are unable to find a student to fill this position within this timeframe ISIS will consider the studentship to have lapsed and the offer of funding will be withdrawn.

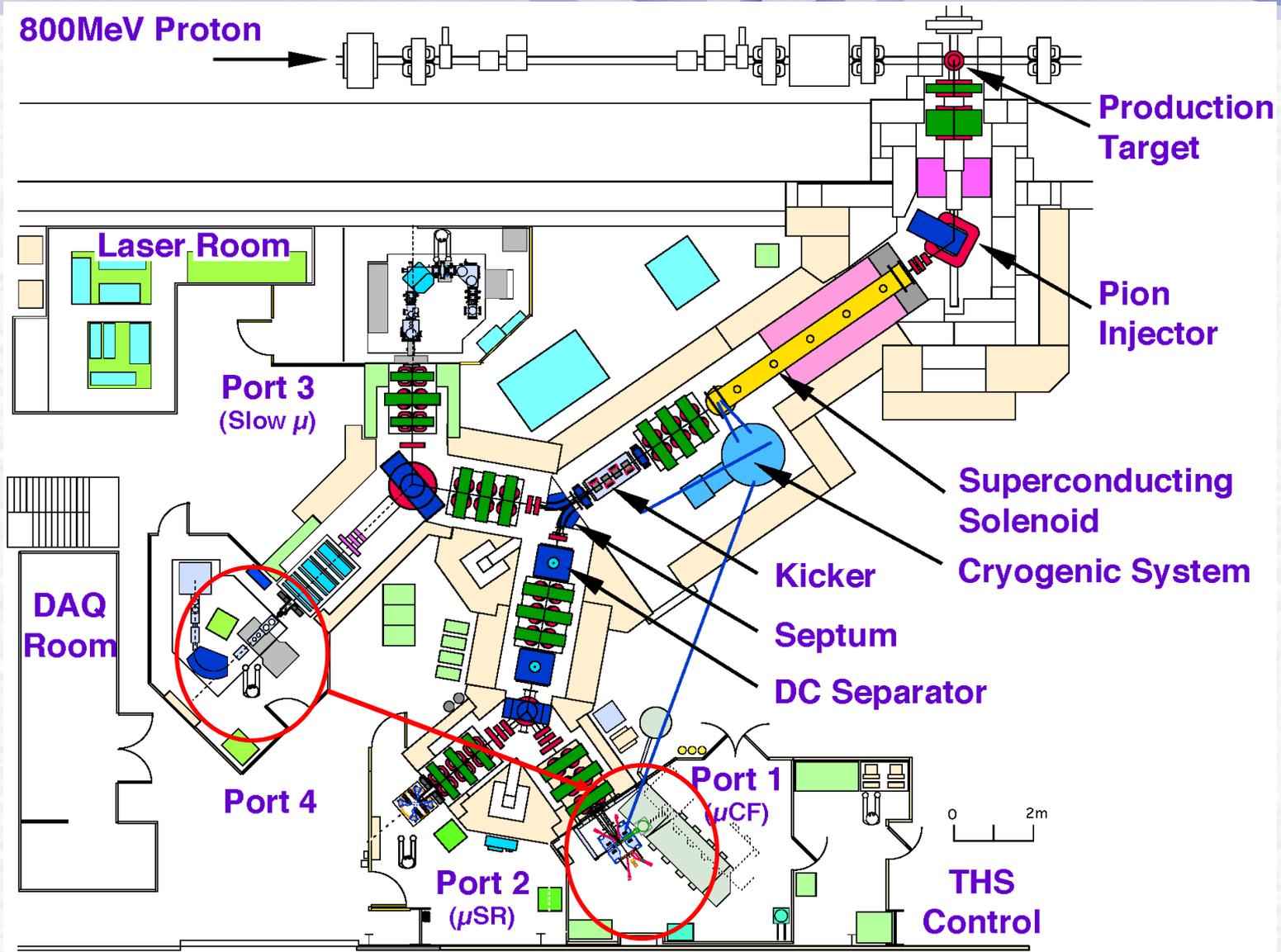
I have also attached here a copy of the call requirements to remind you that the ISIS staff scientist must play a full and equal role in the studentship project and supervision, including the interviewing of potential candidates. Furthermore, we request that copies of all standard PhD progression qualifications (1st / 2nd year reports etc.) all publications arising from this collaboration and details of conferences attended by the student or where work arising from this collaboration is reported should be sent to the ISIS co-supervisor and to Professor Martin Jones at ISIS (Martin.jones@stfc.ac.uk).

You will also need to put in place an agreement between ISIS (STFC) and your University to specify the arrangements for the studentship including how payments will be made. We have model agreements designed for this purpose which your ISIS colleague will be aware of. The ISIS supervisor will need to issue a purchase order for the studentship, for which guidance is also attached.

With best regards

Philip King, Science Director Head, ISIS Facility Studentships  
Science Director Head, ISIS Facility and Mass Source and Secretary, ISIS Facility Studentships  
[Philip.King@stfc.ac.uk](mailto:Philip.King@stfc.ac.uk) [Tina.Lawrence@stfc.ac.uk](mailto:Tina.Lawrence@stfc.ac.uk) and [Philo@isis.stfc.ac.uk](mailto:Philo@isis.stfc.ac.uk)

# A Possible solution



Move to Port 1  
14-18 settembre 2020

SIF2020 M. Clemenza



Science & Technology Facilities Council

ISIS



RIKEN



Dr. Adrian Hillier  
ISIS Pulsed Neutron and Muon Facility  
Science and Technology Facilities Council  
Rutherford Appleton Laboratory  
Harwell Science and Innovation Campus  
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+44 (0) 1235 446001  
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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 analysed 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 di 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 realization.

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

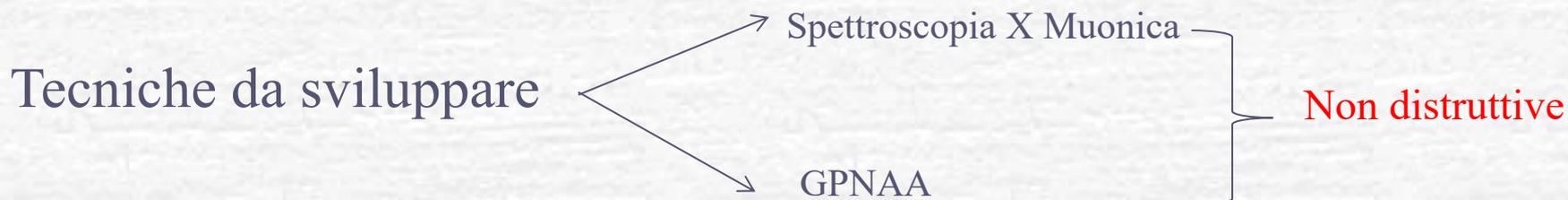
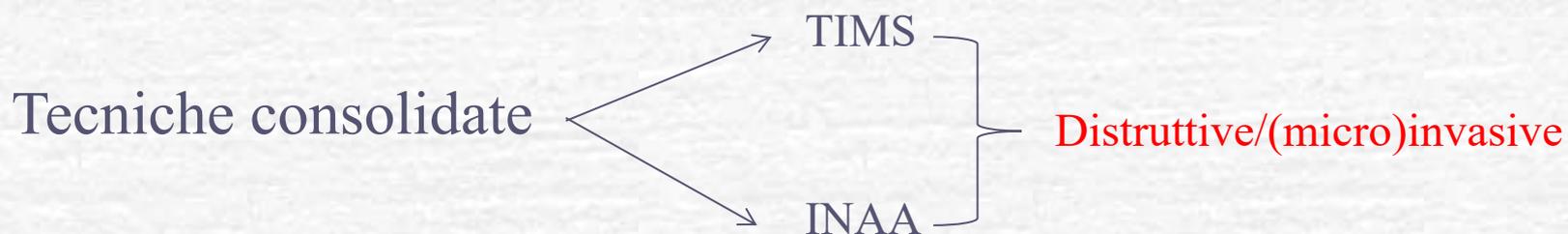
Yours sincerely,

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

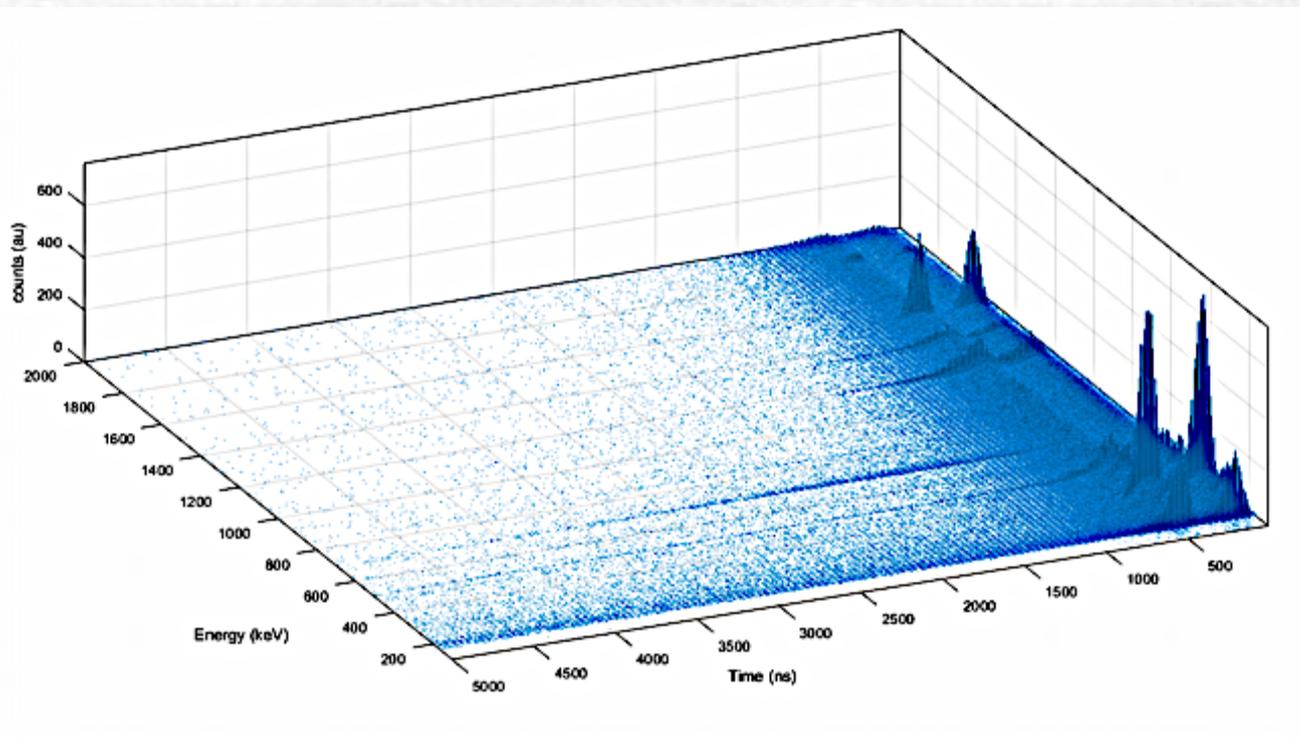
L'idea che sta alla base del progetto **TANDEM** è quella di implementare, sviluppare e ottimizzare tecniche di analisi da utilizzarsi in ambito archeometrico per la caratterizzazione di manufatti



CHNET\_TANDEM

# 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

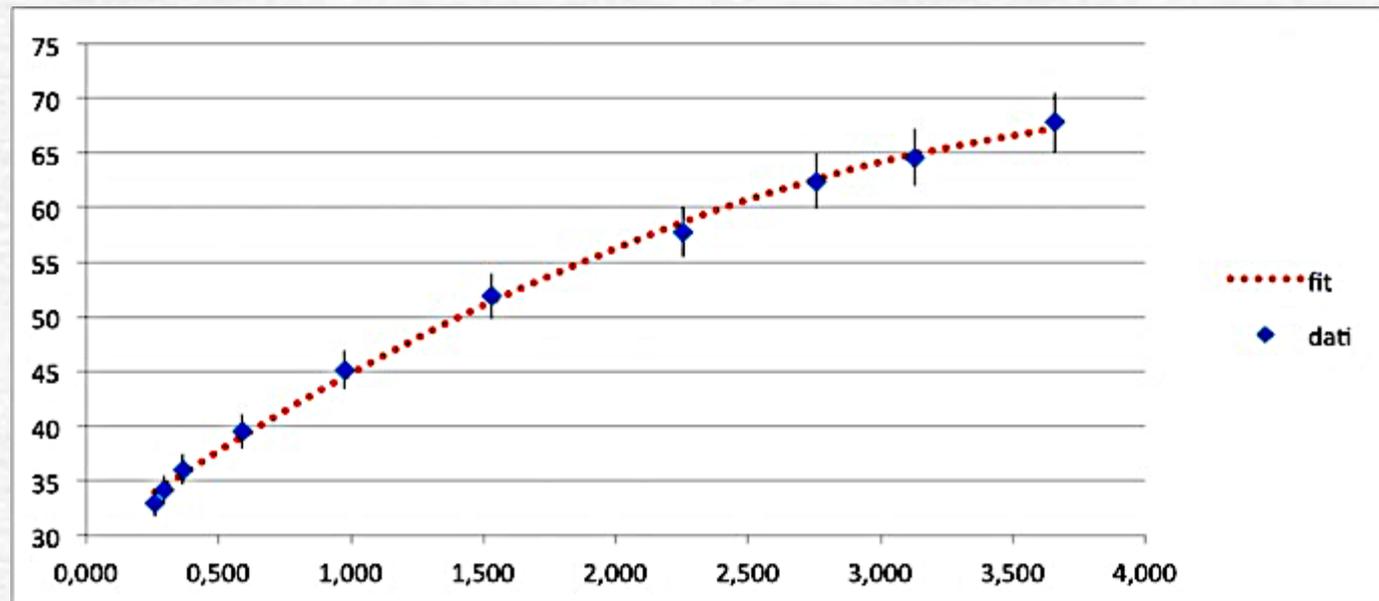
**Collaboration:**

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)



Cumulative mass thickness (g/cm<sup>2</sup>)