

NUCLEAR PHYSICS
FOR CULTURAL HERITAGE



NUCLEAR PHYSICS FOR CULTURAL HERITAGE

A TOPICAL REVIEW BY

the Nuclear Physics
Division of the European
Physical Society

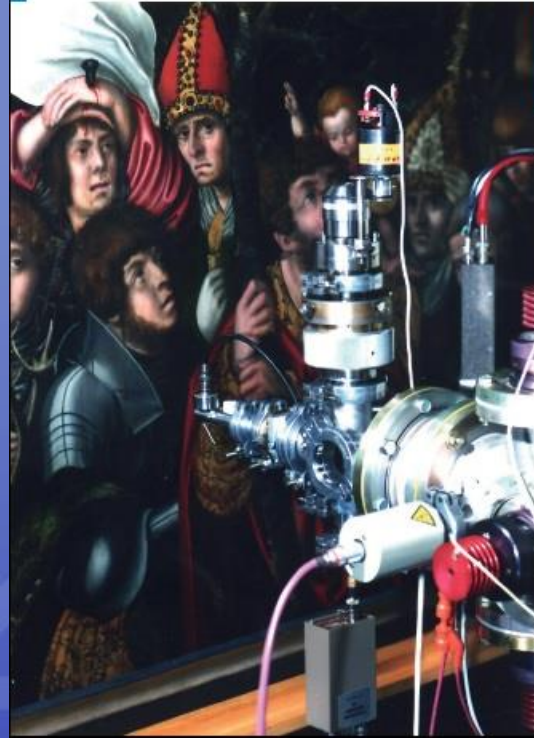
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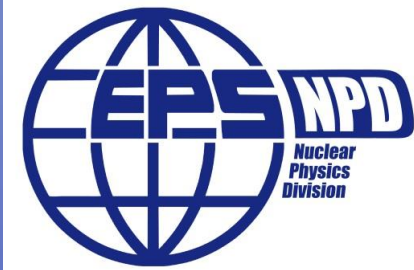
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European Physical Society

Nuclear Physics for Cultural Heritage, Topical paper of the Nuclear Physics Board of the EPS, (2016) Eds. A. Mackova, D. MacGregor, F. Azaiez, J. Nyberg, E. Piassetzky, <http://www.edp-open.org/images/stories/books/fullIdl/Nuclear-physics-for-cultural-heritage.pdf>

OUTLINE



- Motivation
- Editors and Authors
- Topical paper content
- Ion Beam and Neutron Beam analysis
- Cultural heritage characterization - examples
- Conclusions

MOTIVATION

- The idea of this topical paper was first discussed 6 years ago at the Nuclear Physics Division meeting in Bucharest, Romania 2012. The title chosen was “Nuclear Physics for Cultural Heritage”
- Key advances in cross-disciplinary techniques are explained and illustrated using examples from archaeology, pre-history, history, geography, culture, religion and curation.
- Great advances have been made in recent years in the use of nuclear physics techniques to study, characterize and preserve cultural heritage artefacts.
- The goal was to prepare a paper for the public to bring this work to the attention of a wide non-specialist audience.
- The final version was published in 2016 www.edp-open.org.
- 200 printed copies were distributed.

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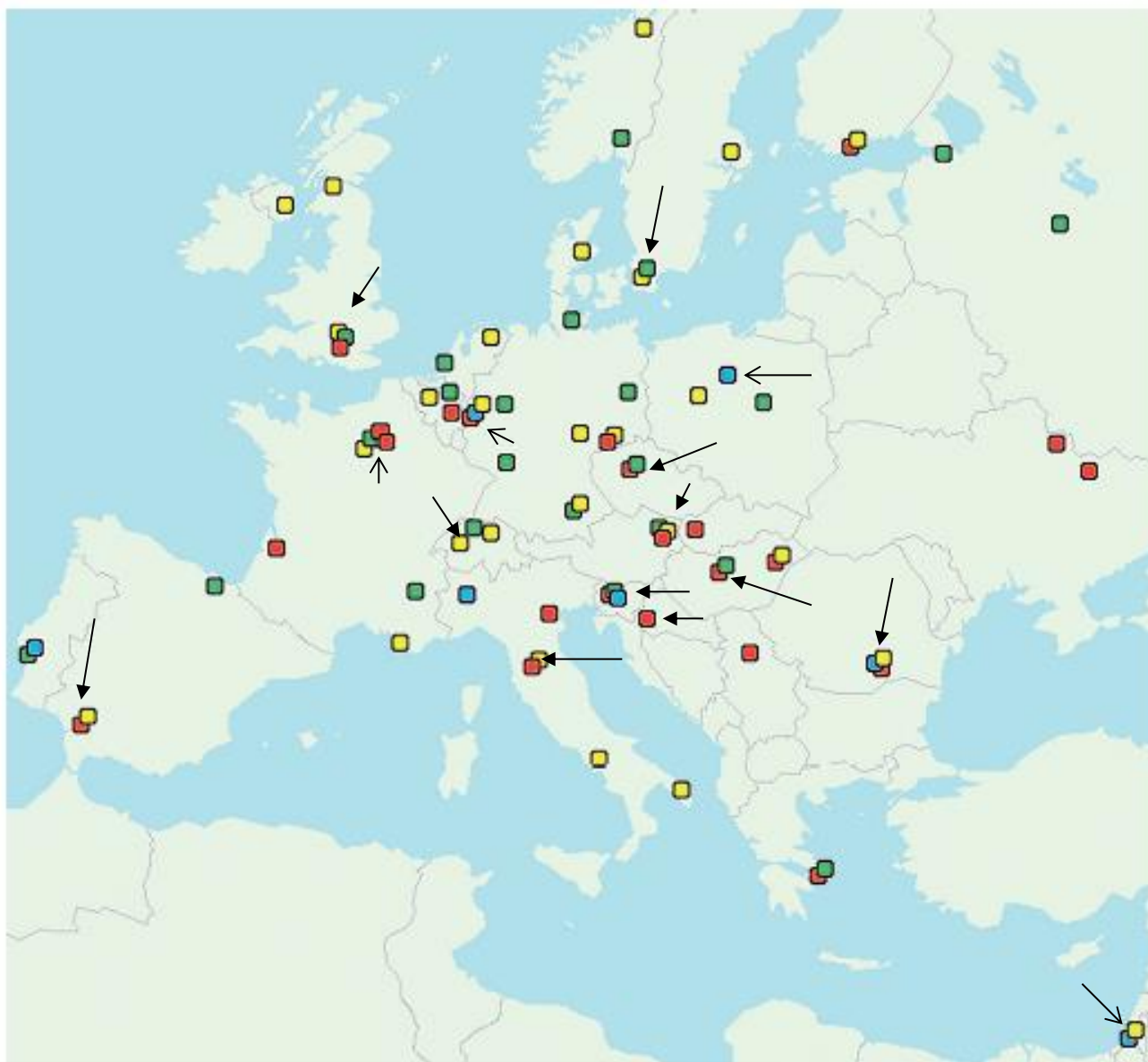
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Institut für Technische und Makromolekulare Chemie (ITMC), RWTH Aachen University, Germany. Physicist studying low-field NMR, instruments, methods, and applications. Coauthored the authoritative monograph "Compact NMR" (De Gruyter Berlin, 2014) and is currently working towards miniaturised NMR sensors for everyday use.

The map below shows laboratories and centres with facilities relevant to nuclear physics studies of Cultural Heritage objects. These are grouped into four categories:

- Ion Beam Analysis Facilities in Europe
- European Neutron Sources

- European Accelerator Mass Spectrometry Facilities
- Other European Centres, Facilities and Laboratories



TOPICAL PAPER CONTENTS

FOREWORD

1. IMPORTANCE OF NUCLEAR PHYSICS FOR CULTURAL HERITAGE STUDY AND PRESERVATION

- 1.1. INVESTIGATION OF CULTURAL HERITAGE OBJECTS
- 1.2. PRESERVATION OF CULTURAL HERITAGE OBJECTS
- 1.3. PRESERVE THE OLD, BUT KNOW THE NEW

2. ION BEAM ANALYTICAL METHODS

- 2.1. BASIC PRINCIPLES OF ION BEAM ANALYSIS (IBA)
- 2.2. INSTRUMENTATION OF IBA
- 2.3. APPLICATIONS OF IBA

3. NEUTRON BEAM ANALYTICAL METHODS

- 3.1. BASIC PRINCIPLES OF NEUTRON BEAM ANALYSIS
- 3.2. INSTRUMENTATION OF NEUTRON BEAMS
- 3.3. APPLICATIONS OF NEUTRON BEAMS

4. DATING METHODS - LUMINESCENT DATING AND ACCELERATOR MASS SPECTROMETRY

- 4.1. BASIC PRINCIPLES OF DATING METHODS
- 4.2. INSTRUMENTATION OF DATING METHODS
- 4.3. APPLICATIONS OF DATING METHODS

TOPICAL PAPER CONTENTS

5. COMPLEMENTARY METHODS: γ -BEAM TECHNIQUES, X-RAY FLUORESCENCE (XRF) AND NUCLEAR MAGNETIC RESONANCE (NMR)

5.1. BASIC PRINCIPLES

5.2. INSTRUMENTATION OF COMPLEMENTARY METHODS

5.3. APPLICATIONS OF COMPLEMENTARY METHODS

6. PRESERVATION OF CULTURAL HERITAGE

6.1. BASIC PRINCIPLES

6.2. INSTRUMENTATION OF NUCLEAR PRESERVATION METHODS

6.3. APPLICATIONS OF NUCLEAR PRESERVATION METHODS

7. CONCLUSION

APPENDIX A: EUROPEAN FACILITIES USING NUCLEAR TECHNIQUES TO STUDY CULTURAL HERITAGE

APPENDIX B: GLOSSARY OF TERMS

APPENDIX C: EXPERTISE OF AUTHORS

REFERENCES

ION BEAM ANALYTICAL METHODS

Ion beams of several MeV, produced by small accelerators, penetrate into matter, interact with the atoms of the sample and produce, among other phenomena, X-rays and γ rays, which carry information about the investigated artefact.

The small accelerators can provide a wide range of ion beams (protons, alphas and heavy ions), with flexible energy range (and thus adjustable probed depth) and diameter of the beam (from millimeter to micron size), and thus can give us tailored tools for the study of the diverse objects of Cultural Heritage.

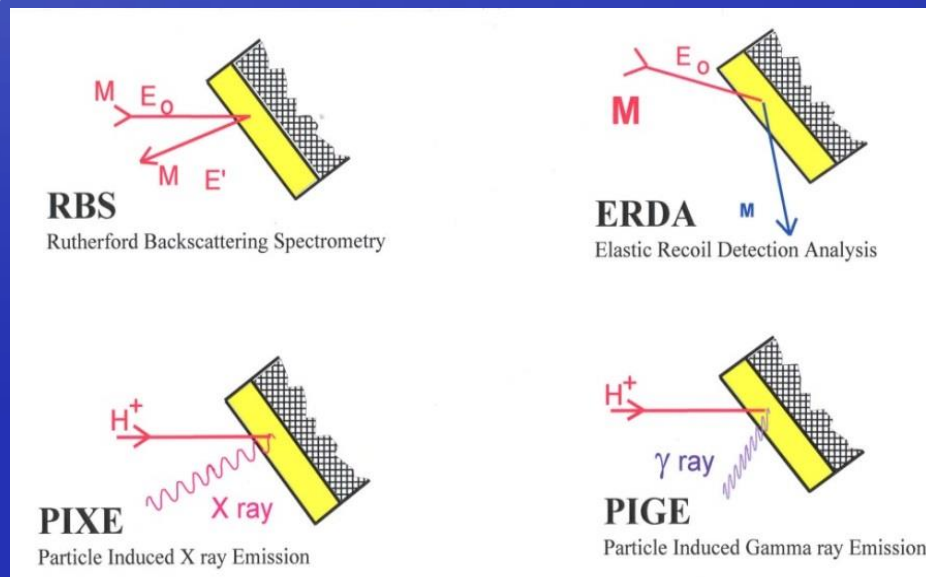
Nuclear analytical methods successfully applied in archaeometry

Archaeometry involves non-invasive surveys of the terrain, science-based dating methods and analytical techniques for object characterization.

Nuclear physics contributes significantly to the dating

methods (radiocarbon dating) and to analytical methods with techniques sensitive to practically all the elements of the periodic table and capable of reconstructing the spatial distribution of the elements present in the sample

J.-C. Dran et al., (2004), Ion beam analysis of art works: 14 years of use in the Louvre, Nucl. Instr. and Meth. in Phys. Res. B 219–220, 7–15.



ACCELERATOR FOR ION BEAM ANALYSIS

RBS (Rutherford Back-Scattering spectrometry)
ERDA (Elastic Recoil Detection Analysis)
PESA (Proton Elastic Scattering Analysis)
PIXE (Particle Induced X-ray Spectroscopy)
PIGE (Particle Induced Gamma-Ray Spectroscopy)
NRA (Nuclear Reaction Analysis)
TOF-ERDA (Time of Flight ERDA)
RBS-channeling
Ion microprobe
External beam

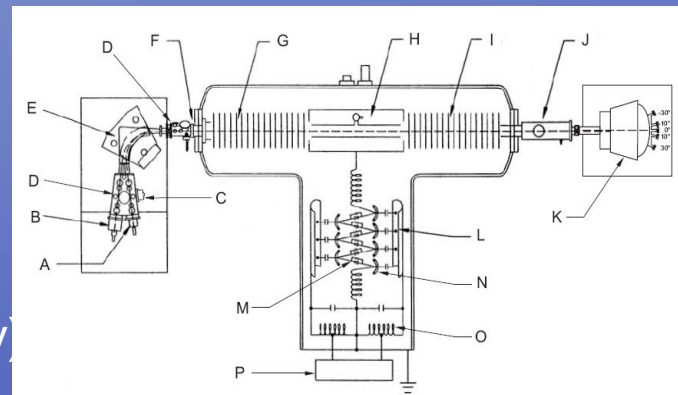
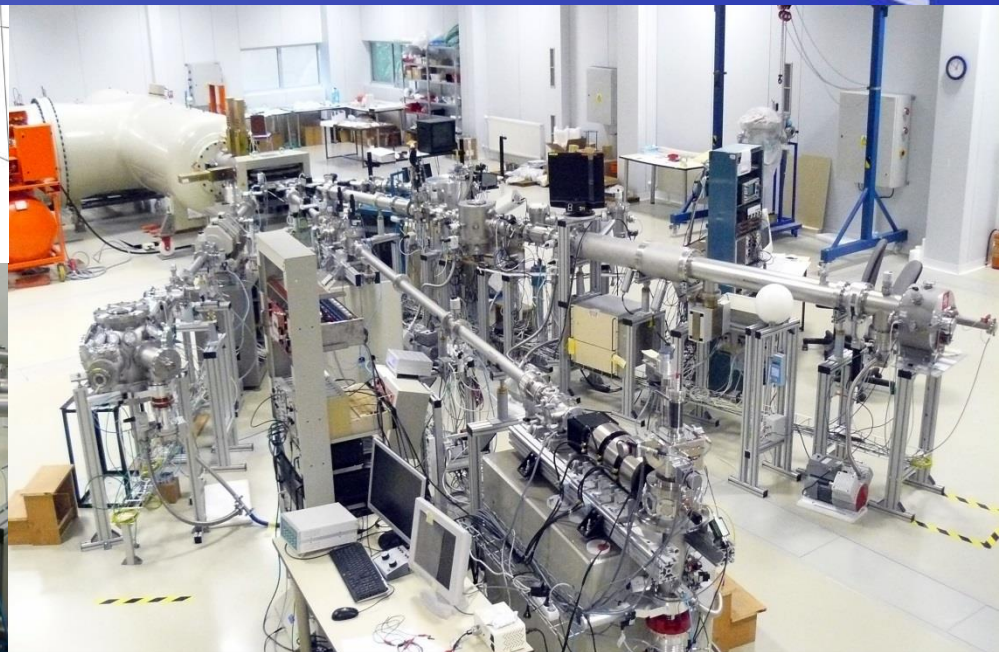
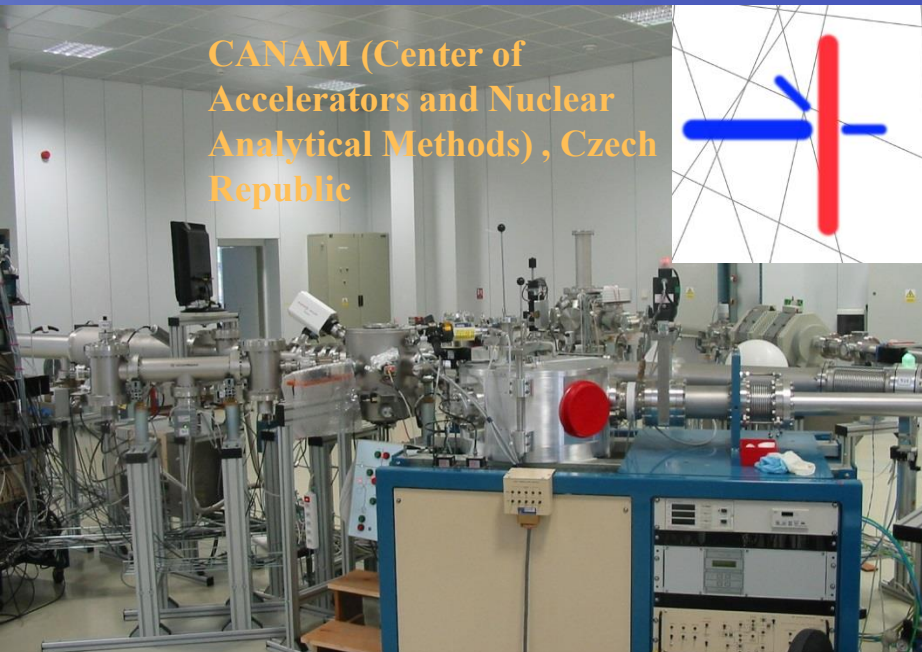
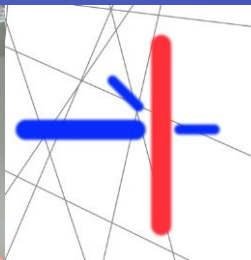


Figure 2.2: The scheme of the Tandatron 4130 MC. Labeled parts: Duoplasmatron ion source (A), Cs sputter ion source (B), Li charge exchange canal (C), Ion optics elements (D), 90° switching/analyzing magnet (E), Q-snout lens (F), Low-energy accelerator tube (G), HV terminal with gas stripper (H), High-energy accelerator tube (I), Electrostatic quadrupole triplet lens (J), High-energy switching/analyzing magnet (K), Rf driver electrode (L), Rectifier stack (M), Capacitor coupling ring (N), Rf oscillator coil (O), Rf driver (P). Ref. [9].

CANAM (Center of Accelerators and Nuclear Analytical Methods), Czech Republic

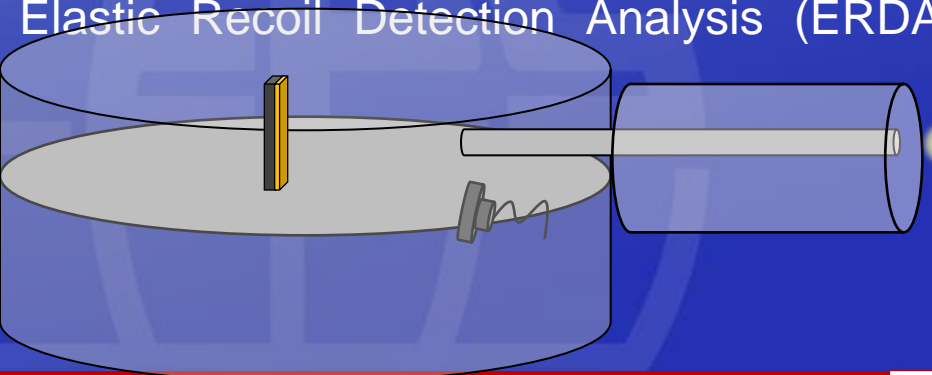
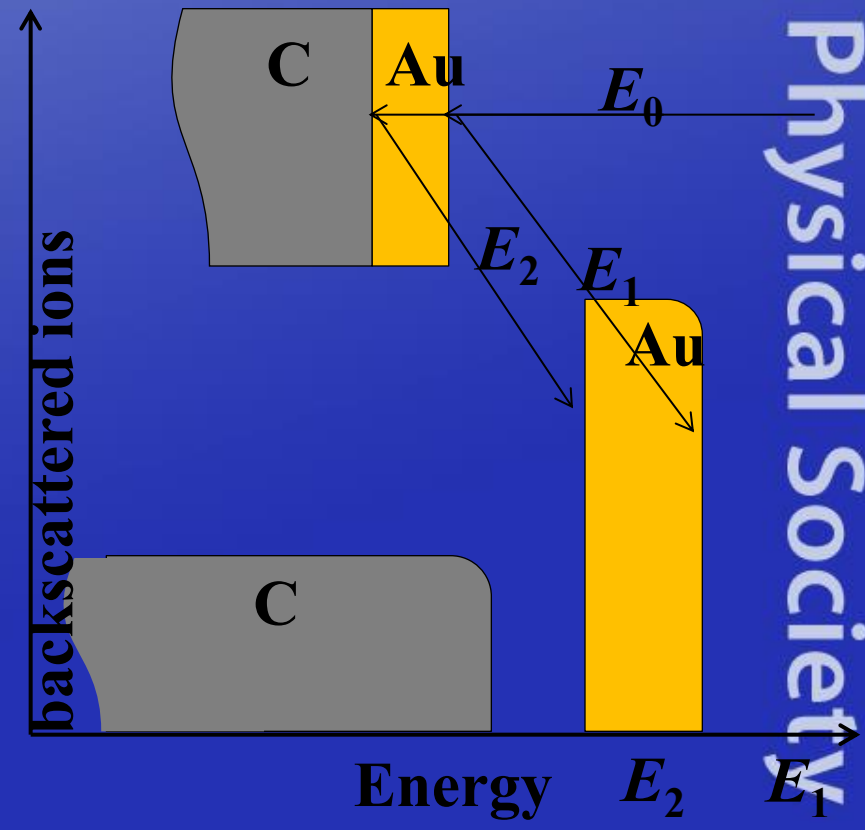
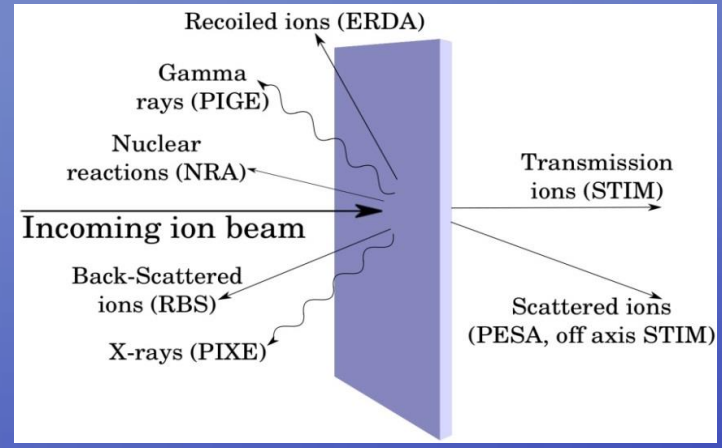


ION BEAM ANALYTICAL METHODS

As a result of ion beam irradiation of a material, two types of collision occur:

In inelastic collisions two phases exist. In the first phase particles are emitted (NRA – Nuclear Reaction Analysis). This is followed in the second phase by the emission of γ -rays (PIGE – Particle Induced Gamma-ray Emission spectroscopy) or X-rays (PIXE – Particle Induced X-ray Emission spectroscopy).

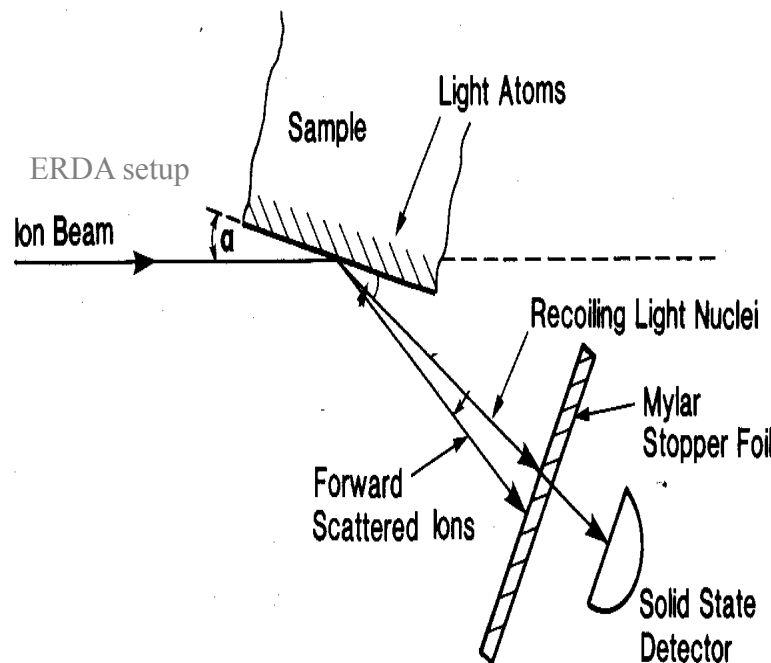
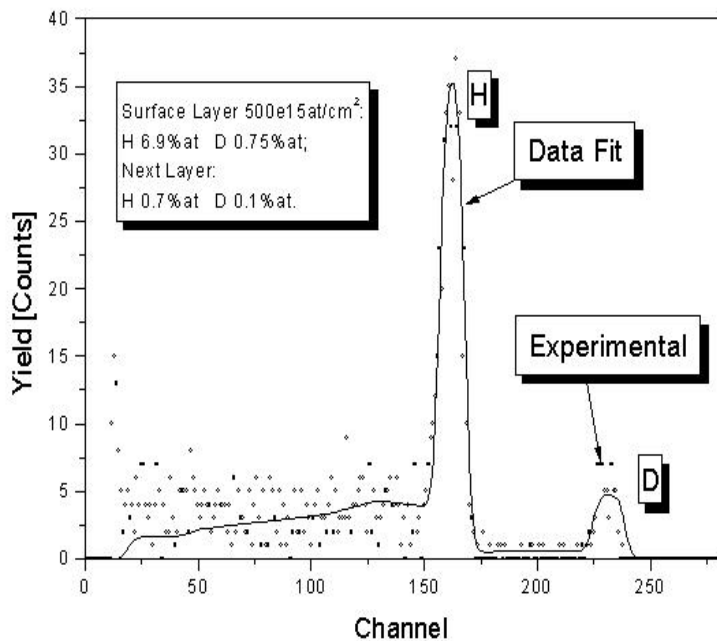
In elastic collisions two main phenomena are taking place: (i) the primary ion beam is back-scattered and is used in Rutherford Back-Scattering spectrometry (RBS) and (ii) lighter atomic nuclei can be ejected, recoiling from the heavier projectile ions in Elastic Recoil Detection Analysis (ERDA).



ELASTIC RECOIL DETECTION ANALYSIS - ERDA

The elastic-recoil detection analysis (ERDA) is one of the IBA methods suited for the non-destructive depth profiling of light elements in bulk samples. It is based on the detection of atoms which are knocked out from the sample by incoming heavy ions. When only kinetic energy is measured, ions of different elements coming from various depth within the sample can produce the same signal in the energy detector.

In addition, also elastically scattered primary ions can be detected which further complicate the acquisition and evaluation of the energy spectra. To overcome this difficulty Time-of-Flight ERDA (TOF-ERDA) was developed.



Measurement of the time of flight of ions through the telescope serves for distinguishing the outgoing ions and recoiled atoms according to their mass. The time flight t is given by the non-relativistic formula.

$$t = l \sqrt{\frac{m}{2(E_{out} - E')}}$$

l ... Fixed distance of flight

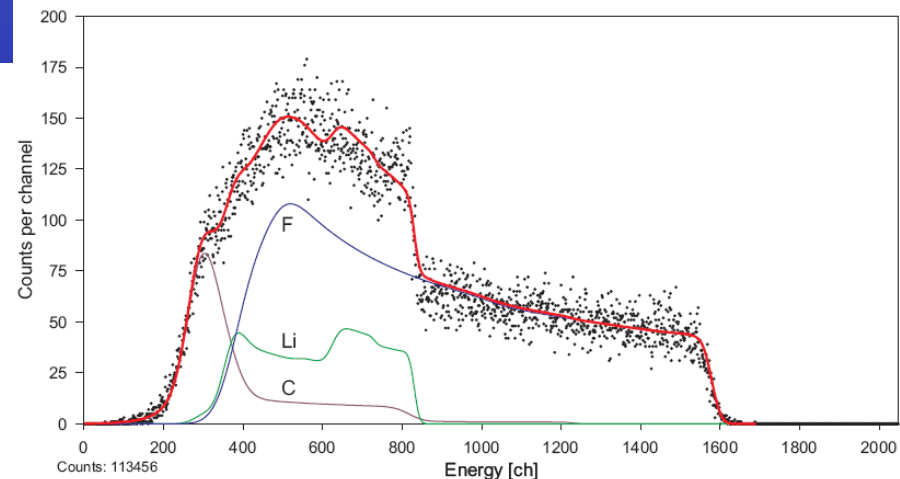
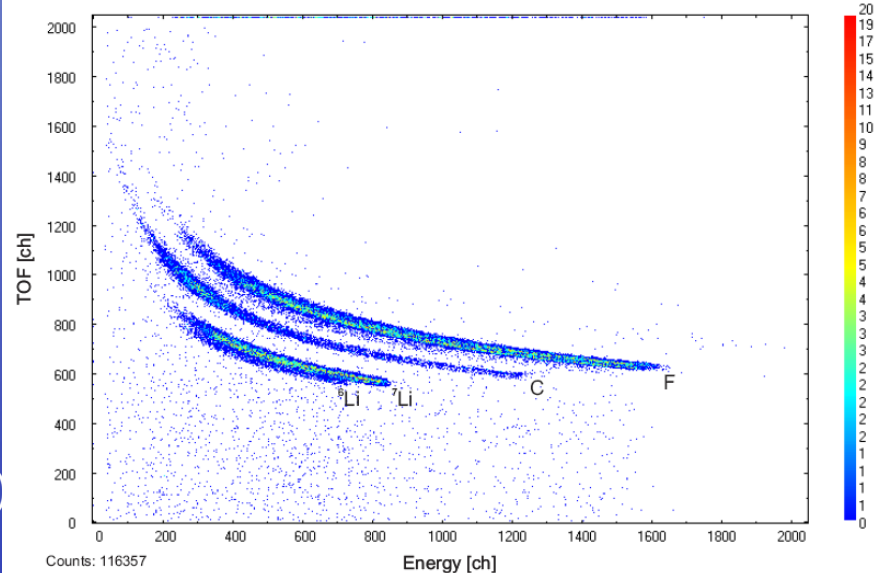
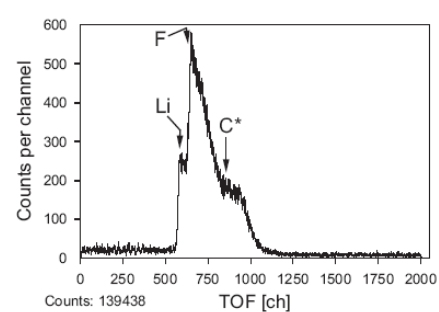
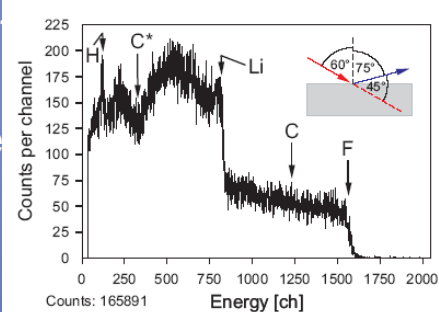
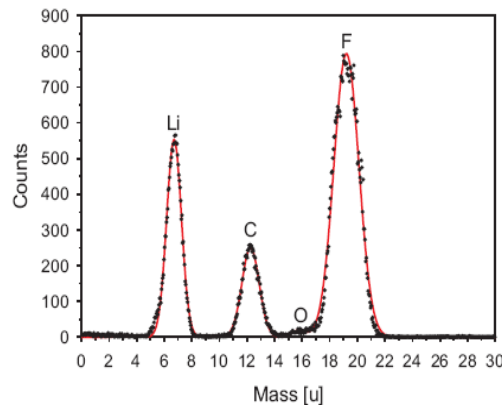
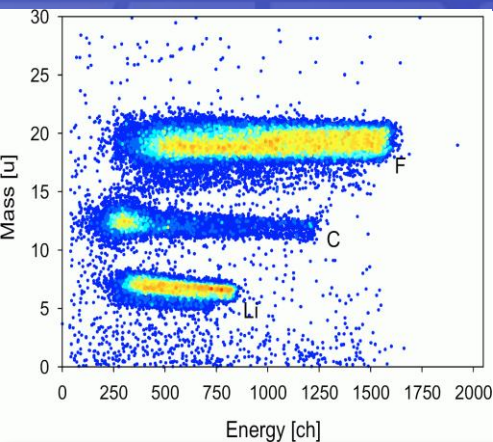
m ... Recoiled atom mass

E ... energy loss of recoiled atom in the time detector

Testing of TOF spectrometer

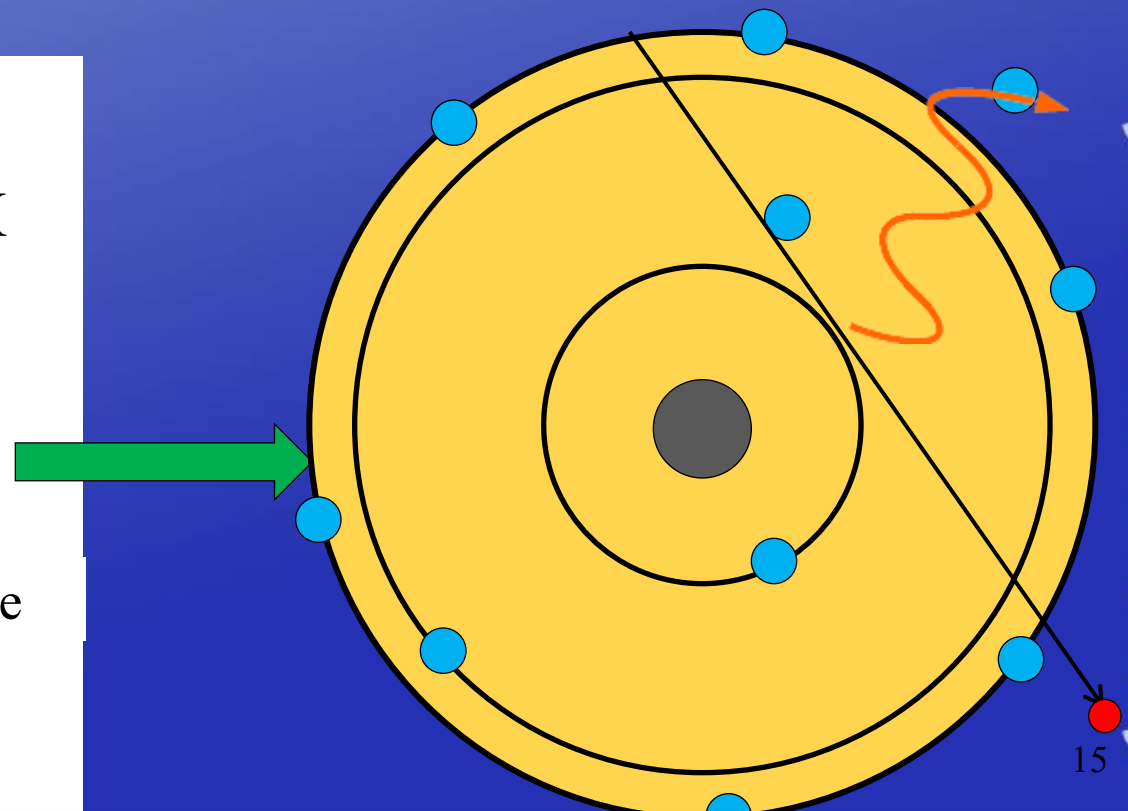
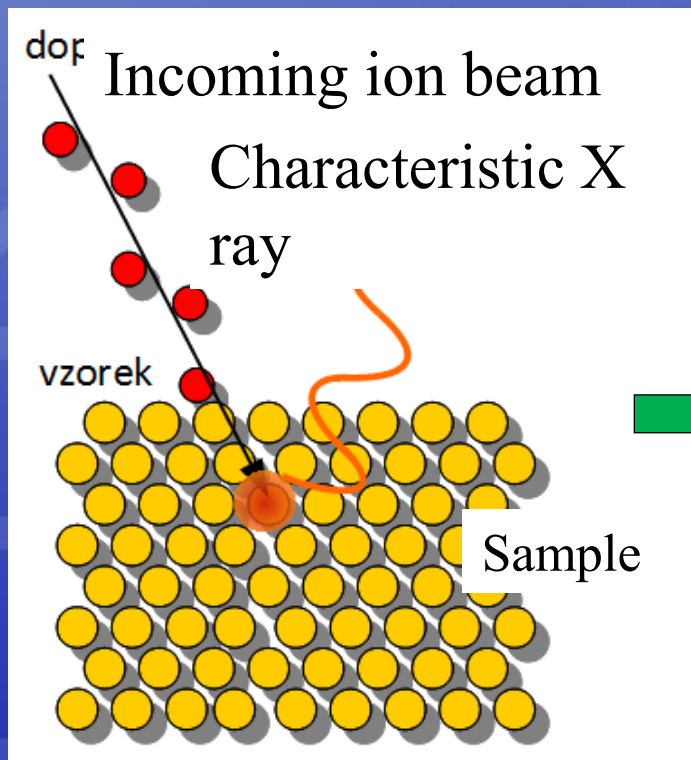
Used parameters

- ion beam: 15,4 MeV Cu^{6+} (terminal voltage: 2,2 MV)
- Used sample: 200 nm LiF layer deposited on glassy carbon



PARTICLE INDUCED X-RAY EMISSION SPECTROSCOPY (PIXE)

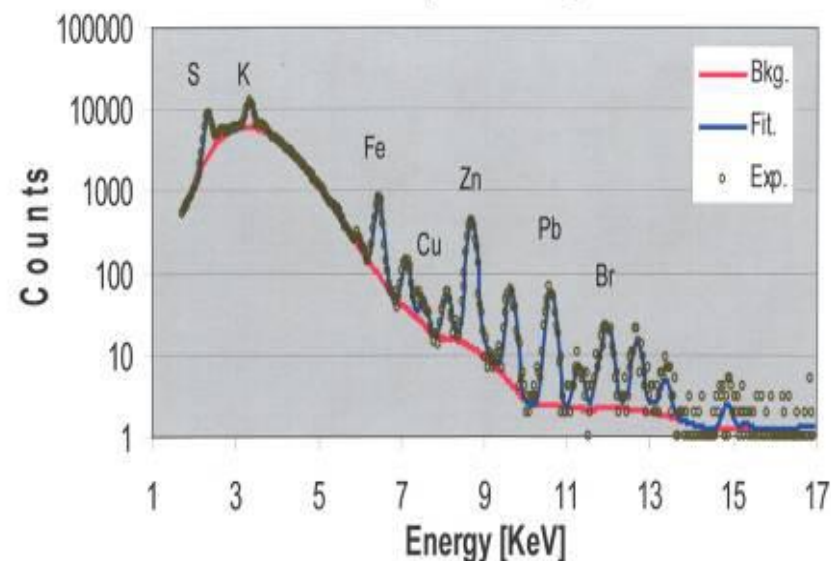
- PIXE uses X-ray emission for elemental analysis . Samples are irradiated by an ion beam from an accelerator and characteristic X-rays are then detected.
- Ions, or protons, with energies of a few MeV ionize atoms in the sample and induce the emission of characteristic X-rays.
- The X-ray yield depends on the number of atoms in the sample, the ionization cross section, the intensity of the ion beam.
- Depending on the sample type and measuring apparatus, the concentration of elements with $Z > 5$ can be determined with sensitivities of $0.1\text{--}1 \mu\text{g.g}^{-1}$.



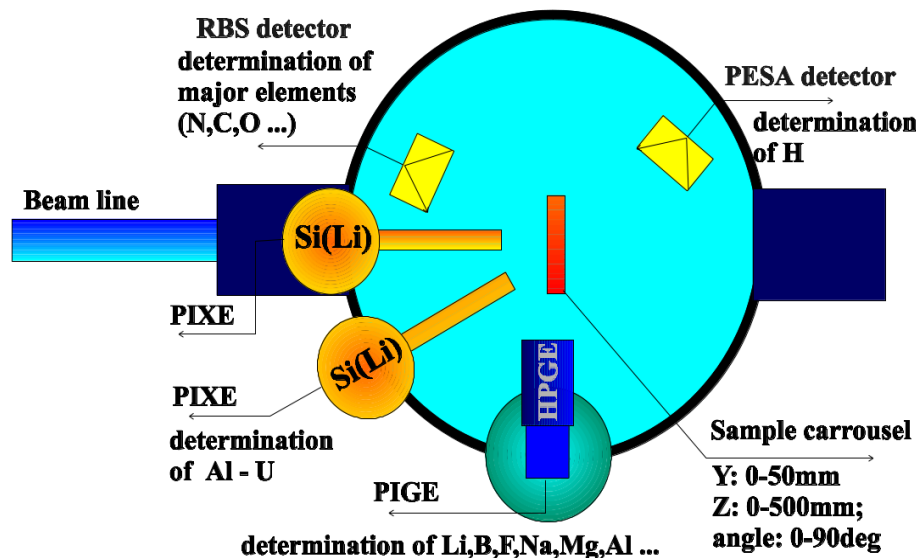
Nuclear reaction methods are suitable for identifying a range of isotopes from ^1H to ^{32}S . The most frequently used reactions are (p,α) , (d,p) , and (d,α) which provide useful alternative methods for determining isotopes such as ^2D , ^{12}C , and ^{16}O , compared with Rutherford Back-Scattering spectrometry (RBS) or Elastic Recoil Detection Analysis (ERDA).

Isotopes up to ^{32}S can be determined in heavier matrices at $\mu\text{g.g}^{-1}$ levels depending on the maximum beam current that the sample can withstand. The use of glancing measurement geometries or heavy incident ions make possible depth profiling with typical resolutions at the surface of 10–100 nm.

The PIXE spectrum of the BLPI impactor sample
one spot from the stage 5

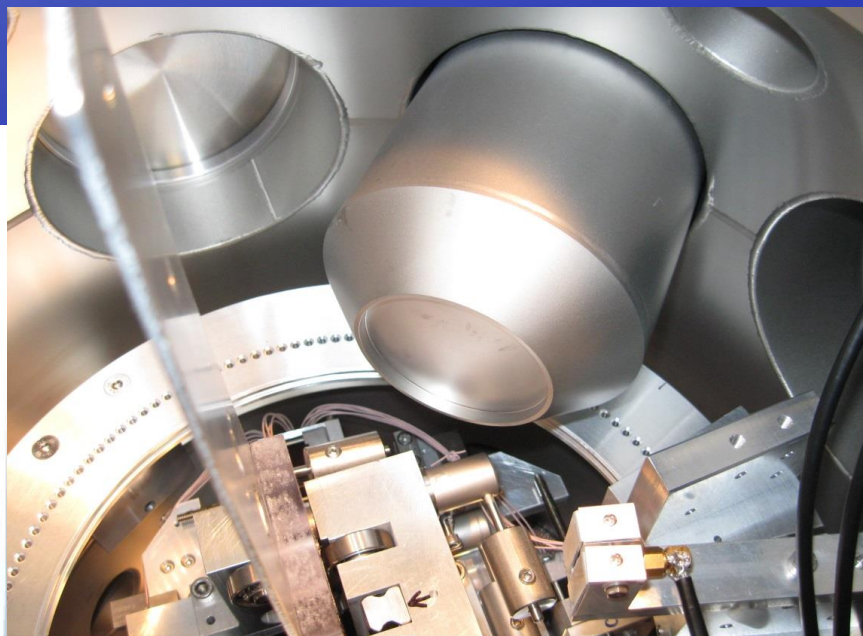
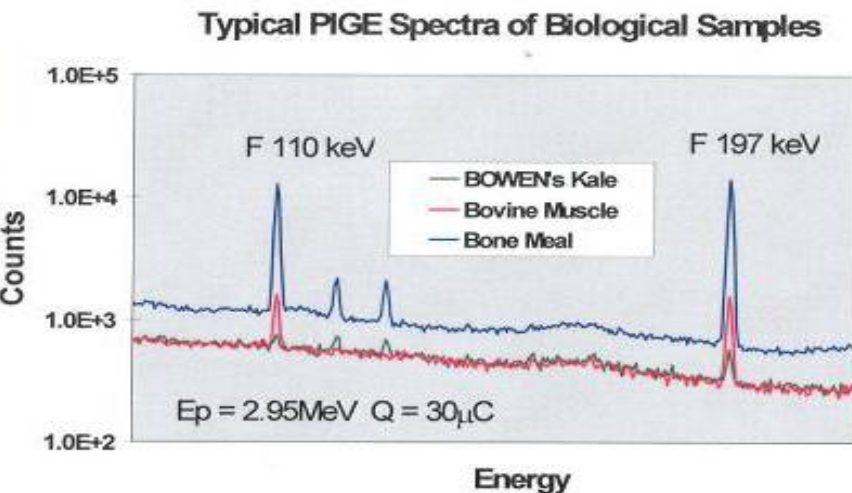
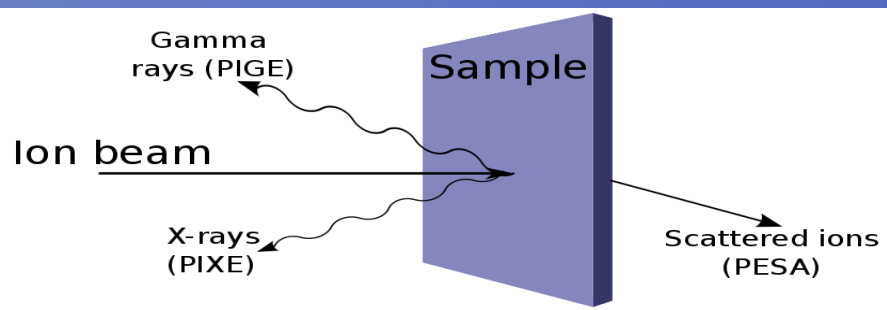


Experimental chamber for simultaneous analyses
by PIXE, RBS, PESA and PIGE



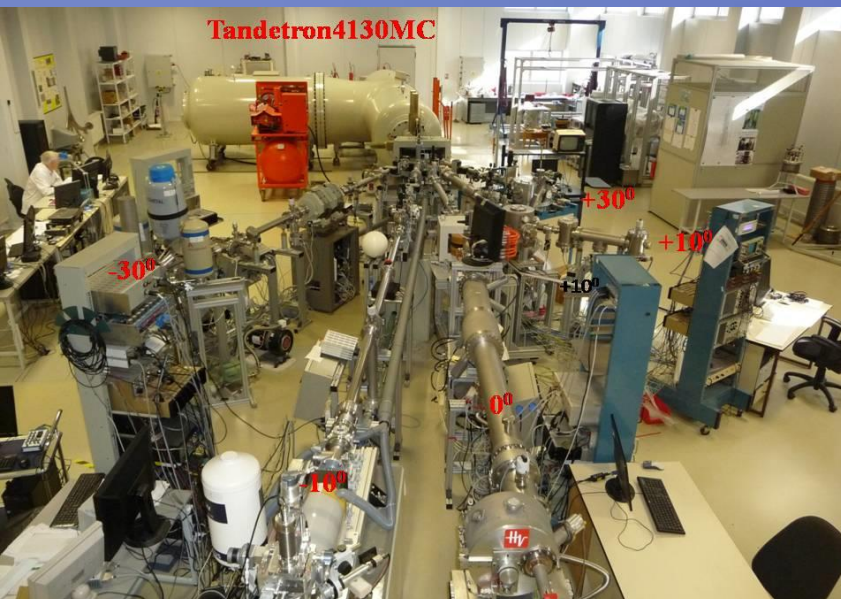
PIGE INSTRUMENTATION

- PIGE (particle-induced gamma-ray emission) is a versatile non-destructive analytical and depth profiling technique based on the (p, γ) reaction. The energy and intensity of the γ -ray lines indicate the elements that are present and their amounts, respectively.
- For protons with energies from 1 to 3 MeV, the best sensitivities are found for Li, B, F, Na, and Al. The highest cross sections are for light isotopes ($A < 30$), which can be determined with a sensitivity of $10 \mu\text{g} \cdot \text{g}^{-1}$ or less.



ION MICROBEAM

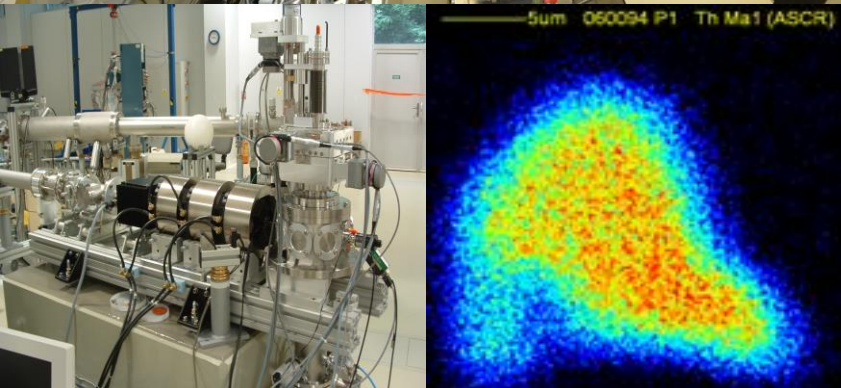
Particle-Induced X-ray emission spectroscopy (PIXE) - The energy of a peak in the X-ray spectrum is specific for a particular element, and its intensity is proportional to the elemental concentration. PIXE has a very low detection limit, down to several ppm in the standard practice.



Ion microbeam - the ion beam from the accelerator passes through a lens (a combination of magnetic quadrupoles with alternated polarities) focusing the high energy ions.

The samples are irradiated with an ion beam focused to a spot that can be as small as few hundreds of nm in diameter and standard IBA techniques are used to characterize the irradiated object.

By raster-scanning the beam over the sample surface, 2D or 3D distribution of elements can be determined with nm depth resolution and lateral resolution limited by the size of the beam spot



EXTERNAL ION BEAM

Some archaeological artefacts cannot be placed in a vacuum chamber because of their large size or the presence of volatile components.

Such samples can be analysed using an **external ion beam**.

The beam is extracted from the evacuated beam line into air through a thin window.

The target is encircled by an array of detectors; normally there are at least two X-ray detectors, with a thin window detector for soft X-rays and a detector with a large solid angle but equipped with an additional absorber for hard X-rays.

The external microbeam set-up can be improved to be versatile and allows all IBA techniques to be used individually or in combination, namely PIXE–PIGE–RBS with protons, PIXE–PIGE–NRA with deuterons, PIXE–RBS with He^+ ions.

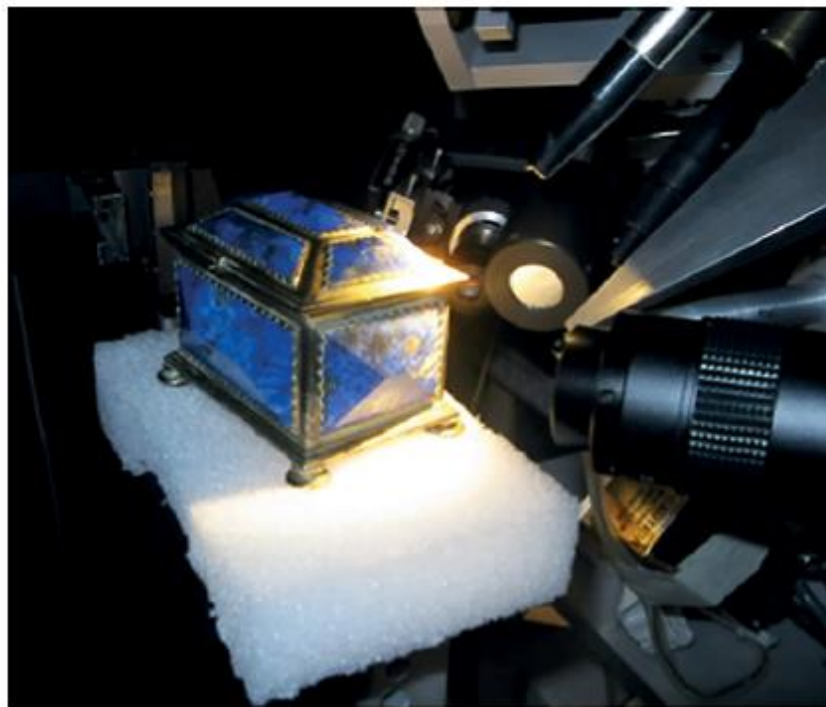


Figure 2.14: An artwork from the *Collezione Medicea* photographed during an external microbeam IBA analysis carried out at INFN-LABEC, Florence.

NEUTRON BEAM ANALYSIS

- Analytical neutron techniques require sources with constant and sufficiently high neutron fluxes, and suitable neutron energies.
- For detailed studies scientists require much more intensive neutron fields which may be produced in nuclear reactors.
- A constant and intense neutron flux is produced by a controlled and sustained chain reaction in the reactor. Many of them (e.g. those in Garching, Delft, Saclay, Budapest and Řež near Prague) still provide very reliable and effective neutron fields for research.
- The most common questions addressed by neutron beam analysis are the provenance of objects or the workshop or technique used in its production.

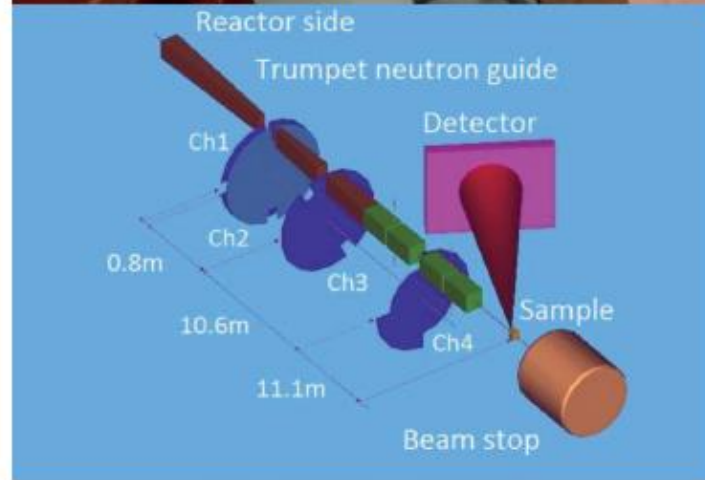


Figure 3.3: Photograph and Schematic illustration of the neutron TOF instrument set-up showing the real neutron guide together with its shielding components.

Budapest Research Center, Hungary

Cold neutron prompt gamma activation analysis – a non-destructive method for characterisation of high silica content chipped stone tools and raw materials, *Z. Kasztovszky, K. T. Biró, A. Markó, V. Dobosi*, *Archaeometry* 50 (2008) 12-29,

NEUTRON BEAM ANALYSIS

The bulk elemental composition of objects can be determined from the detection of characteristic γ -rays produced in (n, γ) reactions Neutron Activation Analysis (NAA).

NAA thanks to its high potential for accuracy and well defined theoretical background.

Investigation of elastic or inelastic scattering patterns can provide information on the atomic, molecular or nano-scale structural properties including their crystalline or amorphous morphology, phase composition, mechanical strains, impurities, *etc.* Using neutron techniques, 2D or 3D images of objects can be reconstructed in a manner similar to X-ray radiography or tomography – neutron diffraction techniques.

Neutron diffraction or elastic neutron scattering is the application of neutron scattering to the determination of the atomic or magnetic structure of a material.

The *Yellow Submarine* SANS spectrometer, operating at the cold neutron source of the Budapest Research Reactor covers a study range of material inhomogeneities from 50 Å to 1500 Å.

How Can Neutrons Contribute to Cultural Heritage Research?,
Zs. Kasztovszky, L. Rosta,
Neutron News 23:1, 2012, 25-28,

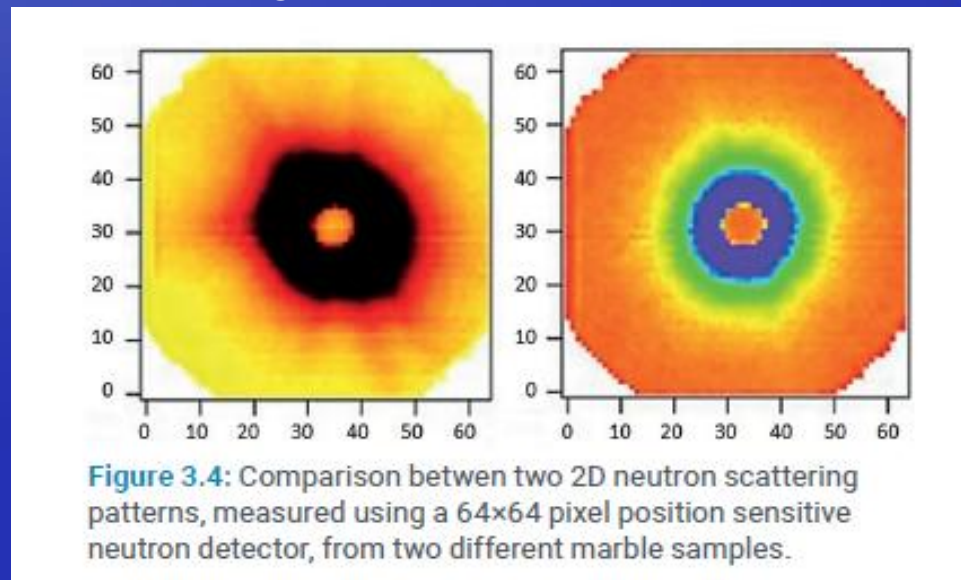


Figure 3.4: Comparison between two 2D neutron scattering patterns, measured using a 64×64 pixel position sensitive neutron detector, from two different marble samples.

- Neutron activation analysis is a multi-elemental analytical technique used for qualitative and quantitative analysis of major, minor, and trace elements. Samples weighing, typically in the range from sub-mg to g, are irradiated with neutrons and the newly formed radioisotopes are created, mostly via the (n, γ) nuclear reaction with thermal neutrons (neutron radiative capture).
- The radioactive decay of newly formed radionuclides is often accompanied by the emission of characteristic γ -rays. In general, the lower the neutron energy, the higher the probability of the neutron radioactive captures. Detection limits are primarily determined by neutron capture cross-sections, i.e. the probability of the (n, γ) reaction, neutron flux, abundance of the target isotope and the measured characteristics of the emitted radiation.
- NAA can detect up to 74 elements depending on the experimental procedure, with minimum detection limits ranging from 10^{-7} to 10^{-12} g/g depending on the element and matrix composition.
- The NAA technique requires a small sample to be taken from the object analysed, but the size of the sample is usually so small that damage to the object is minimised.

DISCOVERING NEW INFORMATION FROM HISTORICAL ARTEFACTS

USING ELECTROMAGNETIC RADIATION
AND CHARGED PARTICLES AS A PROBE

FEATURES

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✦ ² Department of Physics, Faculty of Science, J.E. Purkinje University, Ceske Mladeze 8, 400 96 Usti nad Labem, Czech Republic

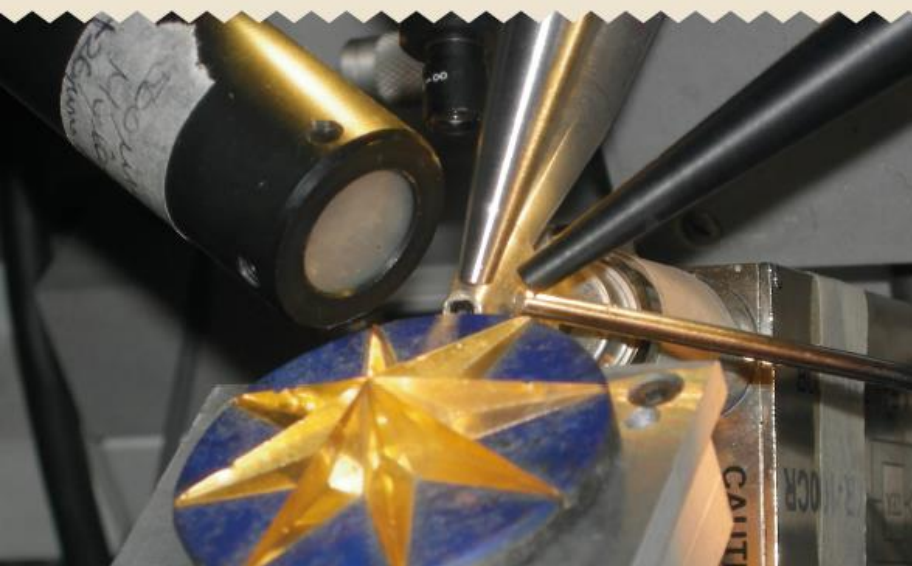
✦ ³ Faculty of Mathematics and Physics, University of Ljubljana, Jadranska 19, SI-1000 Ljubljana, Slovenia

✦ ⁴ Jožef Stefan Institute, Jamova 39, POB 3000, SI-1000 Ljubljana, Slovenia

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Archaeological artefacts included in cultural heritage contribute to the knowledge of our roots, which may help us to learn about our future. Although the connection between ancient times and nuclear technology seems farfetched, this paper will try to show how nuclear radiation of various kinds can be irreplaceable in the elemental composition analysis of an archaeological find.



A. Macková, Ž. Šmit, L. Giuntini, Discovering new information from historical artefacts using electromagnetic radiation and charged particles as a probe. *Europhysics News*. 47, 3 (2016), p. 17-20

A. Macková, J. Kučera, J. Kameník, V. Havránek & K. Kranda, Ion and Neutron Beams Discover New Facts from History, *Nuclear Physics News*. 27, 4 (2017) 12-17

CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

- The example of metal analysis demonstrates the identification of the gilding technique. The objects studied come from the Late Antiquity, which favoured gilded silver or bronze jewellery with inlaid garnets.
- The methods applied were differential PIXE and RBS with in-air proton beam. Differential PIXE is based on the sequential measurement in the same spot such that protons reach different target depths.
- The results of the de-convolution procedure are concentration profiles, which can reach up few tens of microns below the target surface.

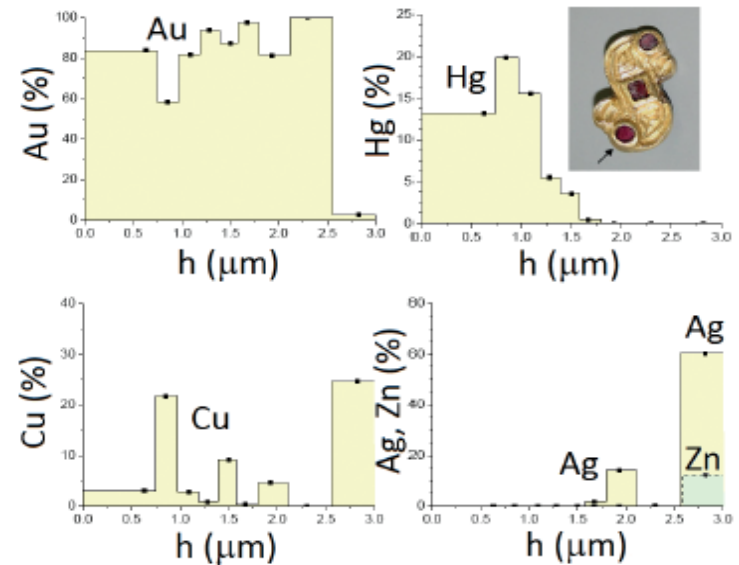
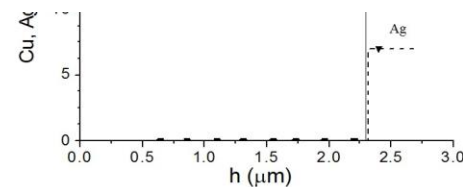


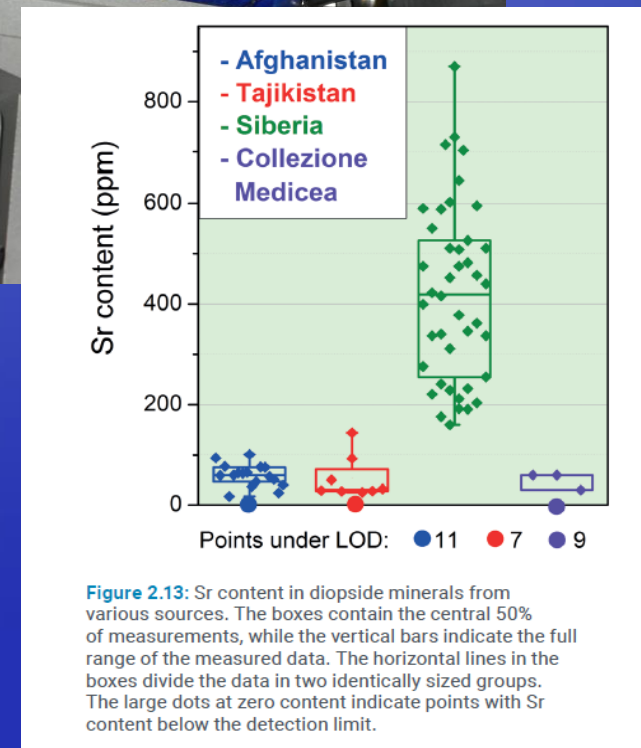
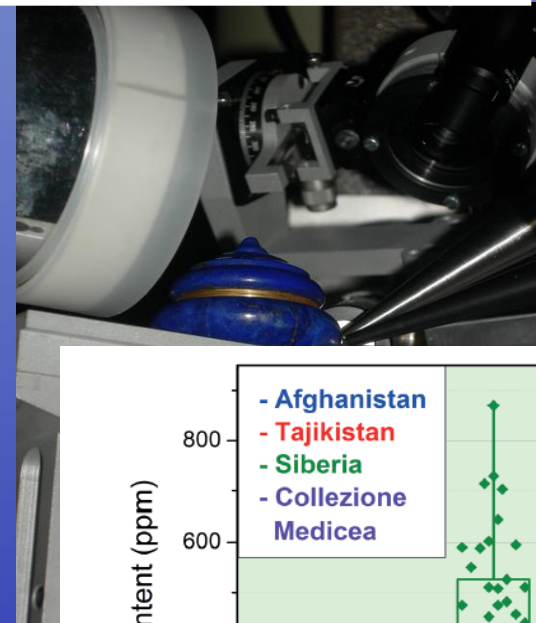
Figure 2.6: Au, Hg, Cu, Ag and Zn concentration profiles of the gilded layer on a Late Antique brooch obtained by differential PIXE; h is the distance from the surface [33].



Plating of archaeological metallic objects – studies by differential PIXE, *Ž. Šmit, J. Istenič, T. Knific*, Nucl. Instrum. Meth. Phys. Res. B 266 (2008) 2333
External beamline setup for plated target investigation, *D. Jezeršek, Ž. Šmit, P. Pelicon*, Nucl. Instrum. Meth. Phys. Res. B 268 (2010) 2006-2009,

CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

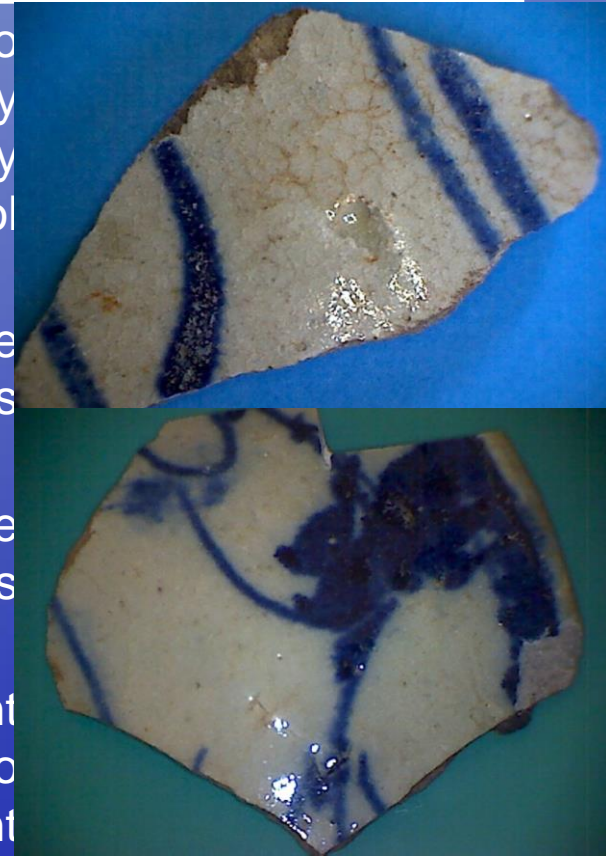
- Lapis lazuli is a semi-precious blue stone widely used for different purposes since the antiquity, but, at present, there are still some lacking pieces of information about both its trade in ancient times.
- External proton microprobe was used as the external beam allows for non-invasive, multitechnique (PIXE and PIGE) study of objects of almost any shape and dimension.
- For the provenance discrimination the study focused on markers, such as for example the presence or absence of the trace elements in the stone of a specific mineral phase.
- After this study, some of the markers found on rocks have been successfully used to characterise six precious artworks of the “Collezione Medicea” in Florence.



A. Re, A. Lo Giudice, D. Angelici, . Calusi, L. Giuntini, M. Massi, G. Pratesi, Lapis lazuli provenance study by means of micro-PIXE, Nucl Instr Meth B. 269(20) (2011) 2373-2377.

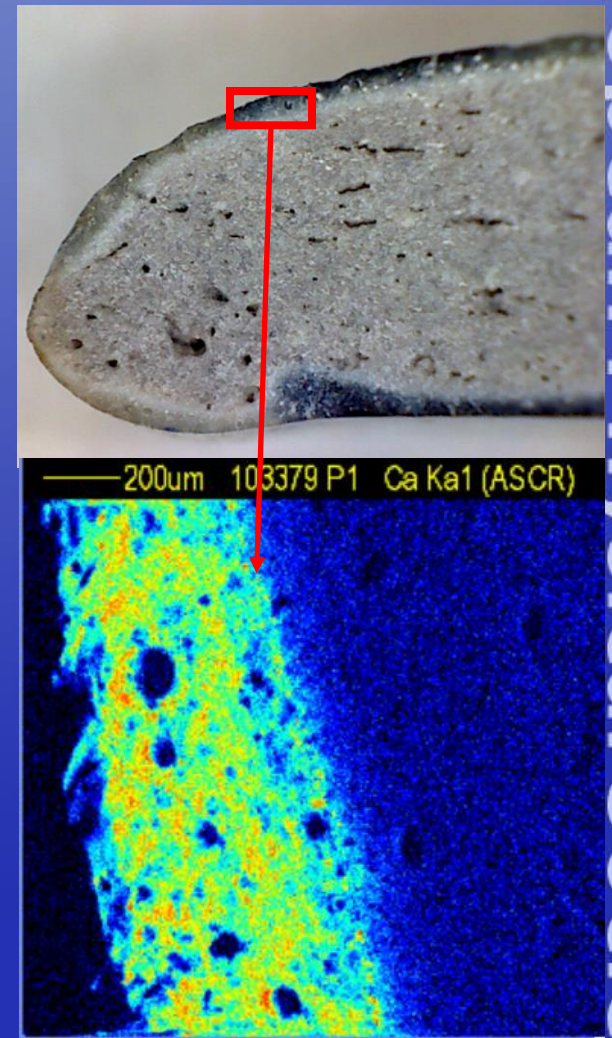
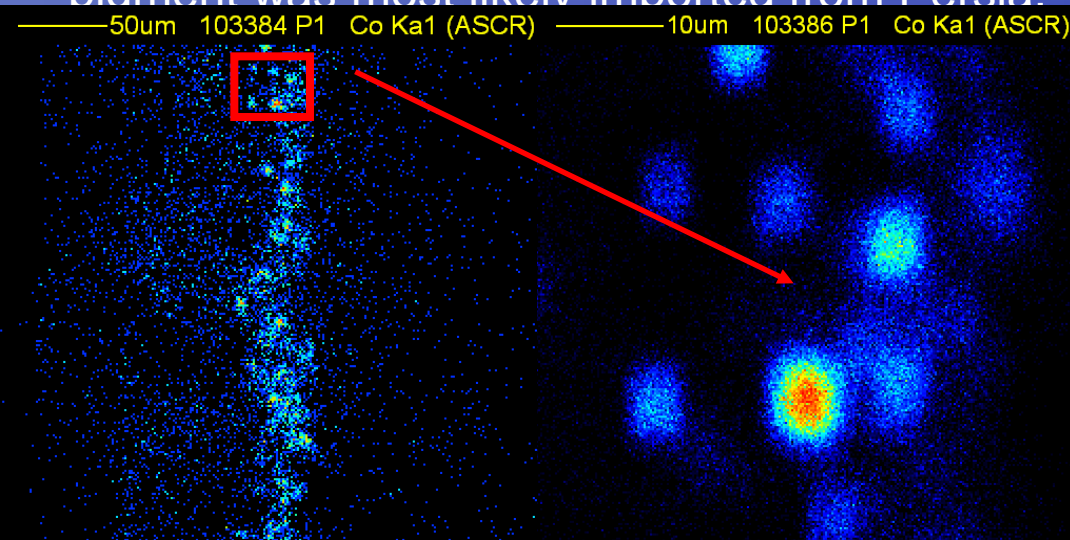
CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

- The purpose of the investigation was to determine the possible origin of Chinese pottery sherds, presumably dating to the Ming dynasty found in excavated material from an ancient pool at the Royal palace grounds of Angkor Thom.
- It was our intention to separately analyse the composition of the glaze and the painted sections containing cobalt.
- Furthermore, we attempted to find the possible origin of the kilns in China where the sherds found were actually manufactured.
- We compared the characteristic trace element content in the sherds body determined by macro PIXE and the composition of the cobalt pigment inclusions determined by μ -PIXE with reported measurements of elemental composition of ancient Chinese porcelain produced at various kiln locations in China.



CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

- From the μ -PIXE analysis the maps of individual elements were constructed from their emission spectra showing a space-resolved concentration of each particular element.
- Microbeam measurement clearly distinguished the glaze on the shards, as calcium content is much higher in the glaze. From the comprehensive elemental analysis of about 20 elements in the glaze, body and cobalt pigment, it appears, that the pigment was most likely imported from Persia.



K. Kranda, V. Havránek, V. Peřina, P. Malínský, On the Composition of Cobalt Pigment of Chinese Ming Pottery Found In Angkor, TECHNART 2011 – Berlin, April 26 - 29, 2011

CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

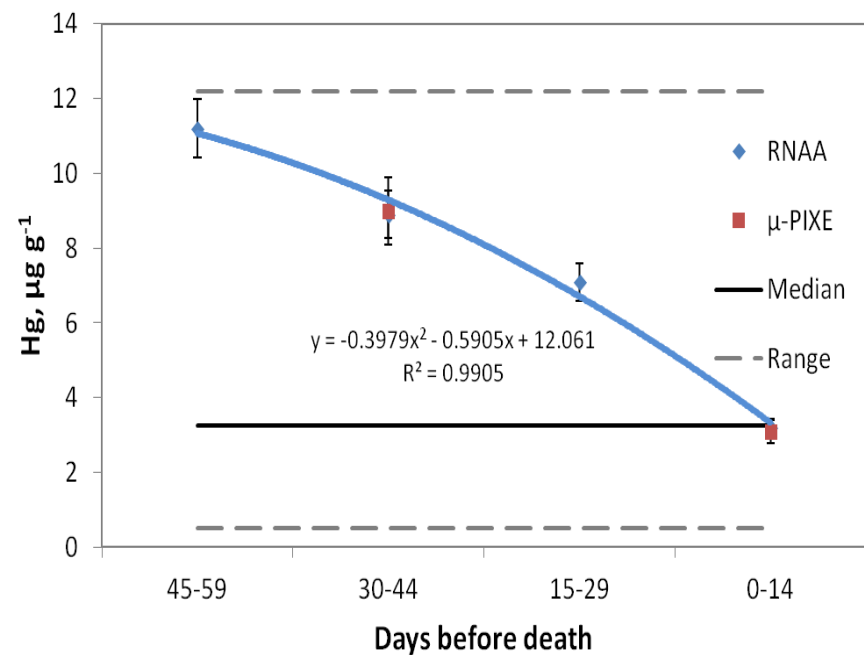
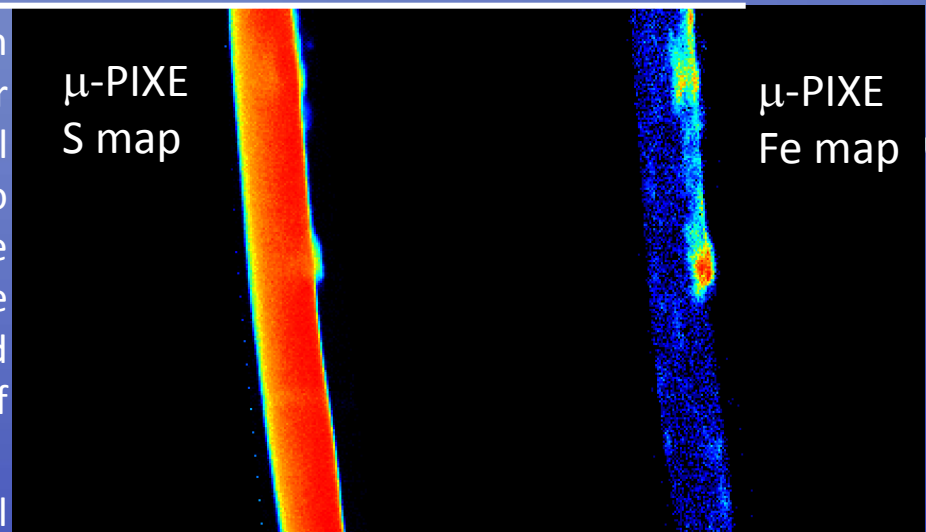
Was He Murdered Or Was He Not?— Determination of Mercury in the Remains of Tycho Brahe

- World-renowned Renaissance astronomer Tycho Brahe died on 24 October 1601, after 11 days of sudden illness. To test the murder hypothesis, Brahe's grave in Prague was reopened in 2010 and samples of his bones, hair, teeth and the textiles were collected and analysed.
- For NAA, hairs with identifiable roots were cut into ~ 5 mm long sections. The sectioned hair samples from 20–25 individual hairs weighing 200–300 μg were sealed in pre-cleaned high-purity quartz ampoules and irradiated in the LVR-15 nuclear reactor in Řež (operated by Research Centre Řež, Ltd.) at a thermal neutron fluence rate of $3 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ for 20 h.
- The extract was measured with high-resolution gamma-spectrometry. Unsectioned hair samples were also analyzed by $\mu\text{-PIXE}$, using a Tandetron 4130 MC accelerator with a 2.6 MeV proton beam focused to a diameter of 1.5 μm .



CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

- Multiple scans were performed over 500 μm sections of hair at a 0.1 nA beam current for 1–3 h. The concentrations and the spatial distributions of the other elements are also important, as these may provide some information on a possible hair surface contamination, hair ageing process and reveal the health and professional status of particular person.
- The element map of Fe shows external contamination of the hair specimen analyzed, demonstrating that $\mu\text{-PIXE}$ could distinguish between the elements present on the hair surface and those homogeneously distributed in the hair matrix, like S.
- An excellent agreement between the NAA and $\mu\text{-PIXE}$ results for one hair sample Tycho Brahe. The Hg concentration decline along the hair length indicates that Brahe was not exposed to any excessive Hg doses shortly before his death.
- Analysis of Brahe's bones revealed no long-term exposure to Hg (no chronic poisoning).



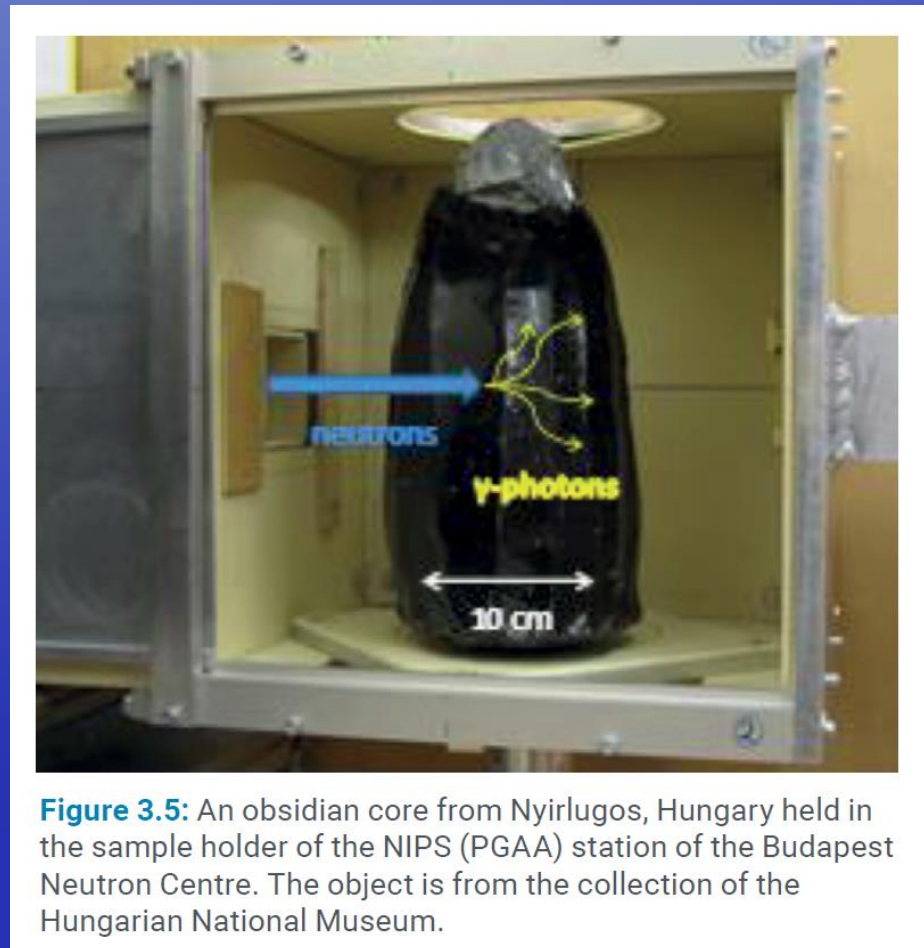
CULTURAL HERITAGE CHARACTERIZATION - EXAMPLES

In prehistoric times people knew where good quality materials could be quarried to make everyday tools. If we can analyse the composition of tools and *fingerprint* chemical components characteristic of the material's provenance, this can enormously help archaeologists to reconstruct prehistoric trade and migration routes.

The task is to determine the provenance of obsidian – a volcanic glass which was popular as a raw material from the early Palaeolithic period.

Besides trace elements of Rb, Nb, Yb, *etc.* that can be measured by destructive INAA, B and Cl, which are both easy to measure non-destructively by PGAA, were found to be *fingerprints* as well.

A border zone between the distribution areas of the so-called Carpathian and Lipari obsidians has been identified. This border falls in the inland part of modern Croatia.



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Conclusions

- The application of atomic and nuclear techniques to the study of archaeological objects gives the historian or archaeologist material information that can help them to understand life during ancient times. This knowledge tests the authenticity and provenance of artefacts and helps prepare restorations. These objectives are common to a very large community of people working in the field of archaeometry, the “application of science to art and archaeology”.
- For this research a multi-disciplinary approach is essential, bringing together physicists, chemists, archaeologists, numismatists, historians, geologists and conservators from different laboratories, institutions and museums.
- This topical paper, brought to you by the Nuclear Physics Division of the European Physics Society, shows the public, and the professional community, how important nuclear techniques have become in the study of cultural heritage, its characterisation and preservation.
- This publication records the very fruitful collaboration of many scientists from different fields focused on the same goal – to enlarge and deepen our knowledge of cultural heritage to keep it safe for future generations.

Thank you