Nuclear Reaction Analysis (NRA) & Proton-Induced Gamma-ray Emission (PIGE)

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Outline

- Ion Beam Analysis
- Theoretical background
- Particle Induced Gamma – ray Emission
- Nuclear Reaction Analysis
- Conclusions

Pros / Cons

- They are generally least destructive and are suitable for use with delicate materials.
- They are to a certain extent multielementary and produce high-accuracy quantitative results.
- They require little or no preparation of the sample with the result that a specimen (like an artifact) could be directly analyzed.
- Only very small quantities (mg) of sample are needed.
- They permit the analysis of a very small portion of the sample by reducing the diameter of the ion beam to less than 0.5 mm.

- **Some damage cannot be avoided (thermal, carbon buildup etc.)!**
- **A VdG type of accelerator is required.**
- **In most of the cases the experiments are carried out in vacuum chambers.**
- **Several experimental issues need to be addressed, thus a minimum knowledge of nuclear physics (experimental and theoretical) is mandatory.**
- **No direct information about the chemical environment can be produced.**
- **The analysis concerns only a few microns below the surface of the samples.**
- **In most of the cases, a combination of techniques is required to solve a problem, and this implies time consuming experiments!**

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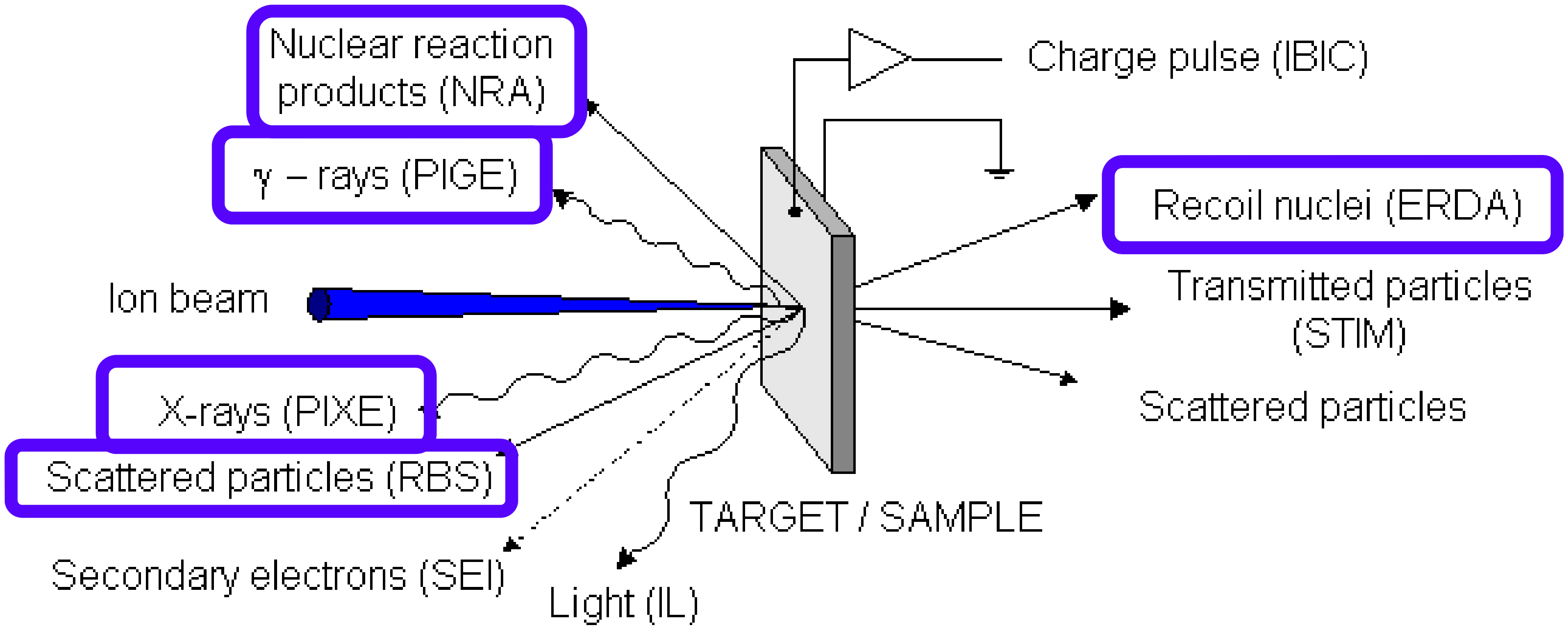
Ion Beam Analysis

Ion Beam Analysis (IBA) is based on the **interaction**, at both the atomic and the nuclear level, between **accelerated charged particles** and the bombarded material. When a charged particle moving at high speed strikes a material, it interacts with the electrons and nuclei of the material atoms, slows down and possibly deviates from its initial trajectory. This can lead to the **emission of particles or radiation** whose energy is characteristic of the **elements** which constitute the **sample material**



Theoretical Background I

Nuclear Reaction:

The interaction between two nuclei which results in the emission of nuclei and/or gamma rays.

Cross Section:

The probability of a nuclear reaction to occur

$$\sigma = \frac{N_{det}}{\Omega \cdot N_{inc} \cdot N_{tar}}$$

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Theoretical Background II

Scattering:

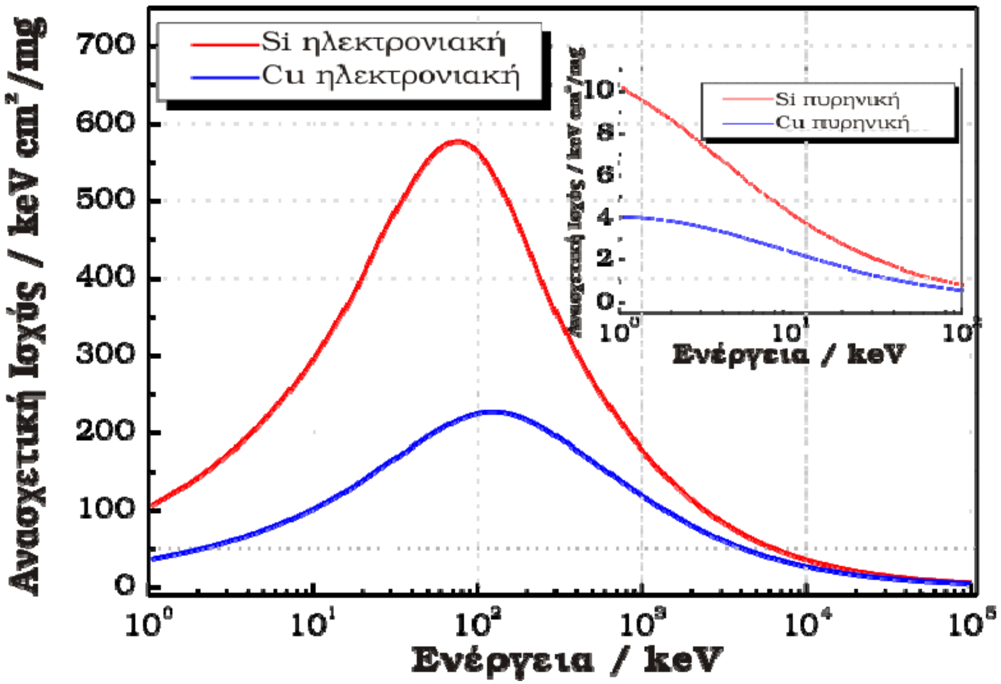
When a charged particle impinges on a material, it interacts with the electrons and the nuclei of the material. The result of the interaction is the loss of energy and the change of trajectory of the initial ion.

Energy Straggling:

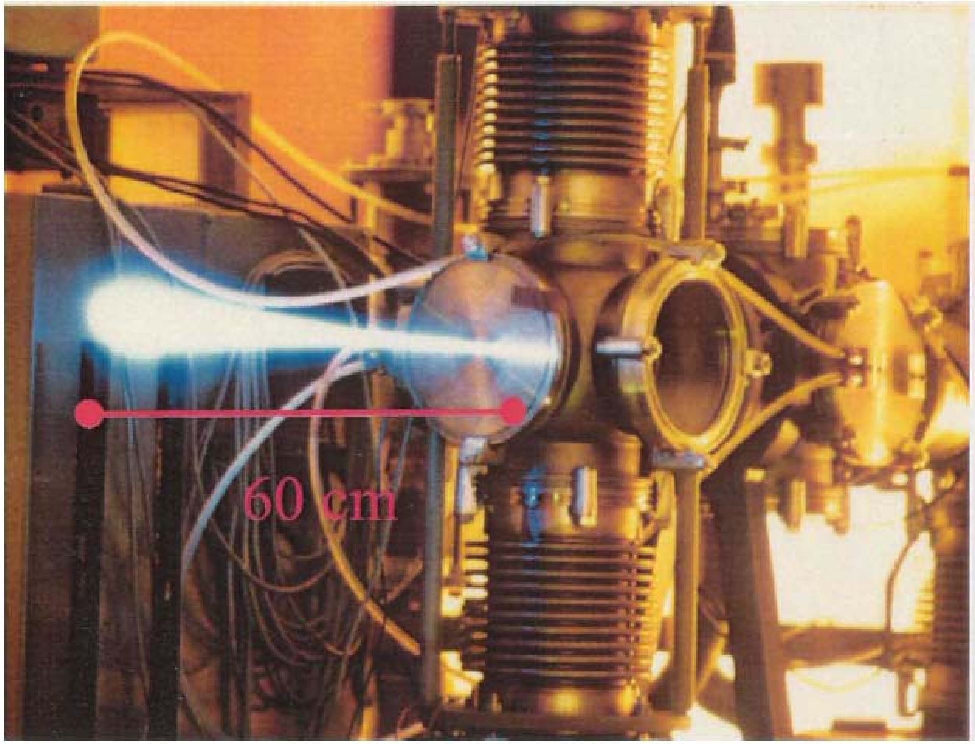
Loss of kinetic energy per length unit

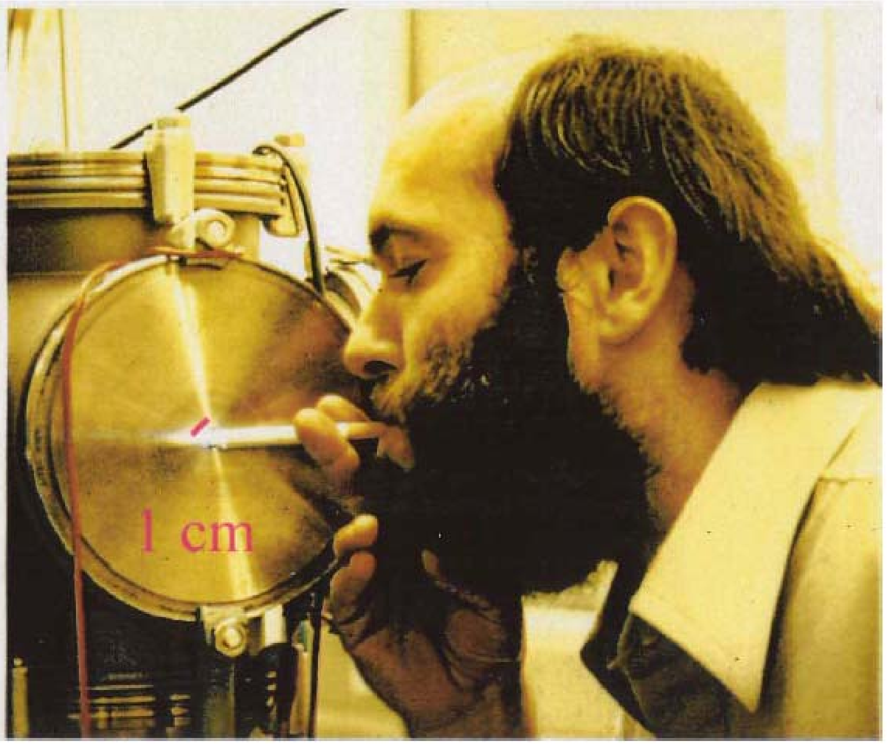
Inelastic collisions with the electrons and the nuclei

$$S(E) = S_e(E) + S_n(E)$$



Theoretical Background II





7 MeV beam energy
R≈60 cm

0.3 MeV beam energy
R≈1cm

Depth Profiling

YES

- Rutherford Backscattering Spectroscopy (RBS)
- Nuclear Backscattering Spectroscopy (NBS)
- Elastic Recoil Detection Analysis (ERDA)
- Nuclear Reaction Analysis (NRA)

- **Particle Induced γ –Ray Emission (PIGE)**

NO

- **Charged Particle Activation Analysis (CPAA)**
- **Particle Induced X-Ray Emission (PIXE)**
- **Neutron Activation Analysis (NAA)**
- **Secondary Ion Mass Spectroscopy (SIMS)**

Sample Size Selection

There are three possibilities

Under Vacuum

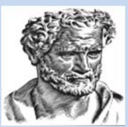
Small samples (1 to 10 cm)
Can withstand vacuum (no wood)
Preferably good electrical conductivity
Greater accuracy

External Beam

No size limitation
No vacuum conditions
Flow of He
Limited accuracy

Microbem

Small samples (less than 1 cm)
Elemental mapping possibilities



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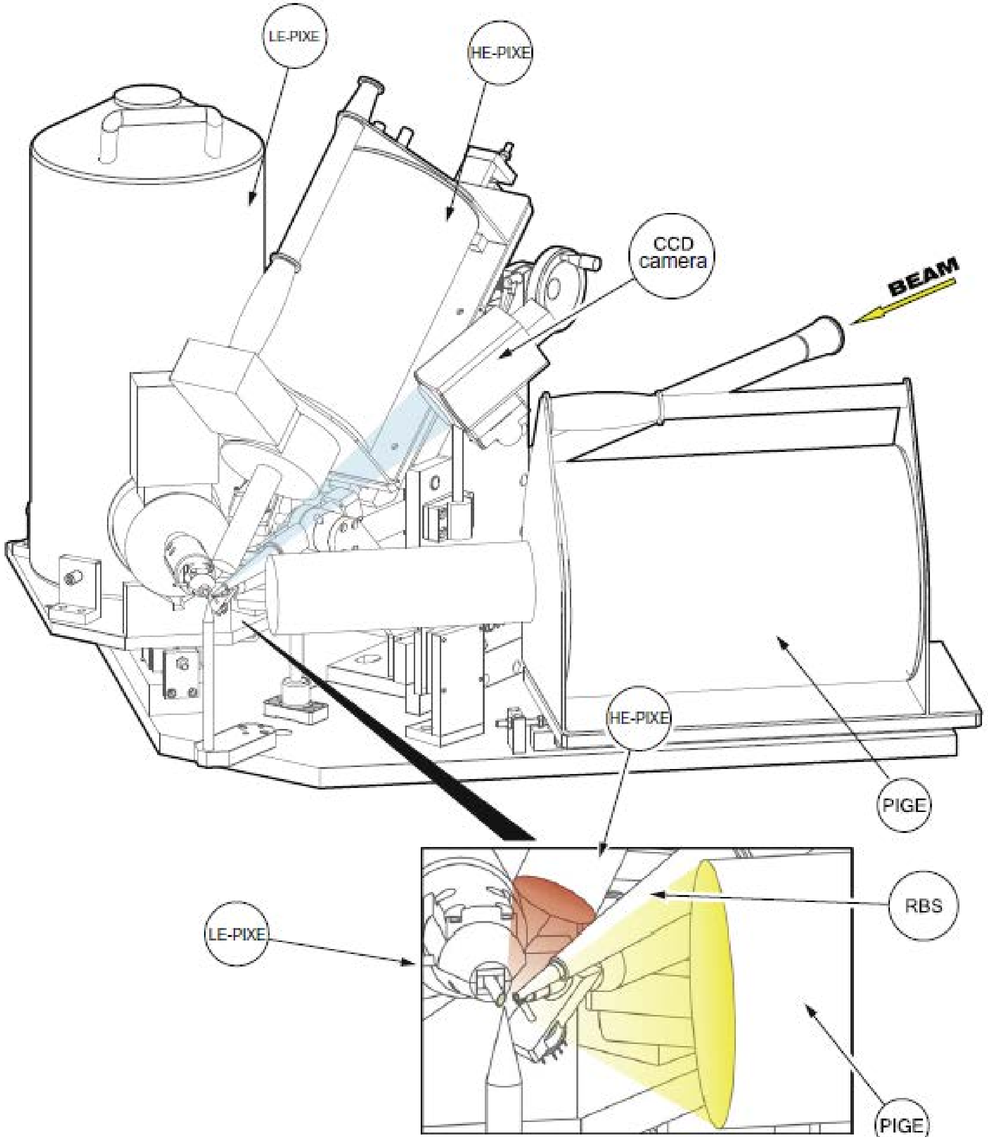
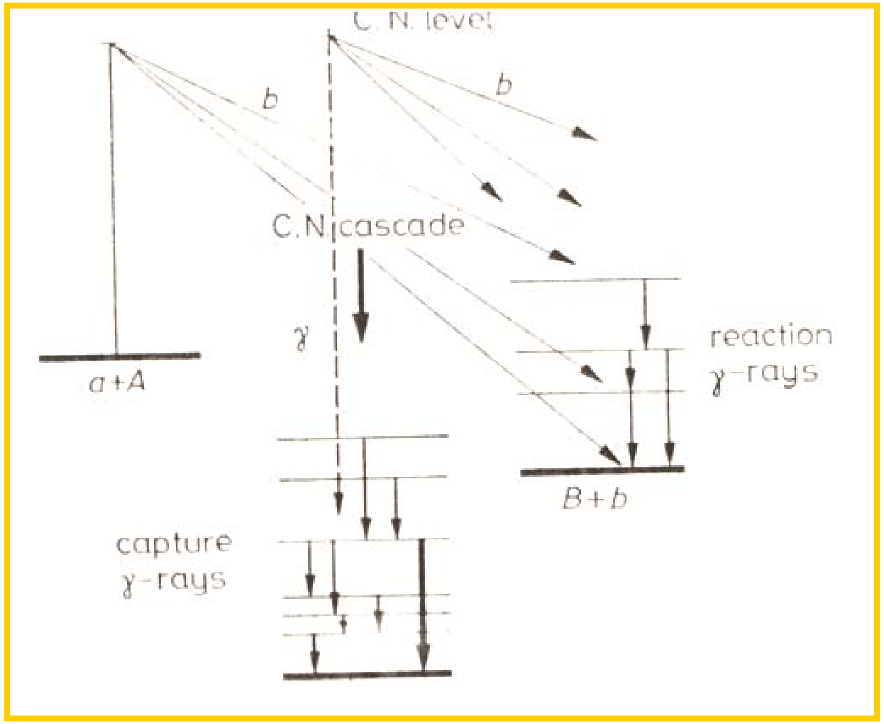
External Beam Setup



Particle Induced Gamma ray Emission

Detection of the gamma rays from the produced nuclei.
They are characteristic of the produced nuclei thus of the initial one



In most cases it is combined with PIXE for the detection of light elements
e.x. Sodium (440 keV)
Boron (2125 keV)
Berillium
Fluorine (197 keV)

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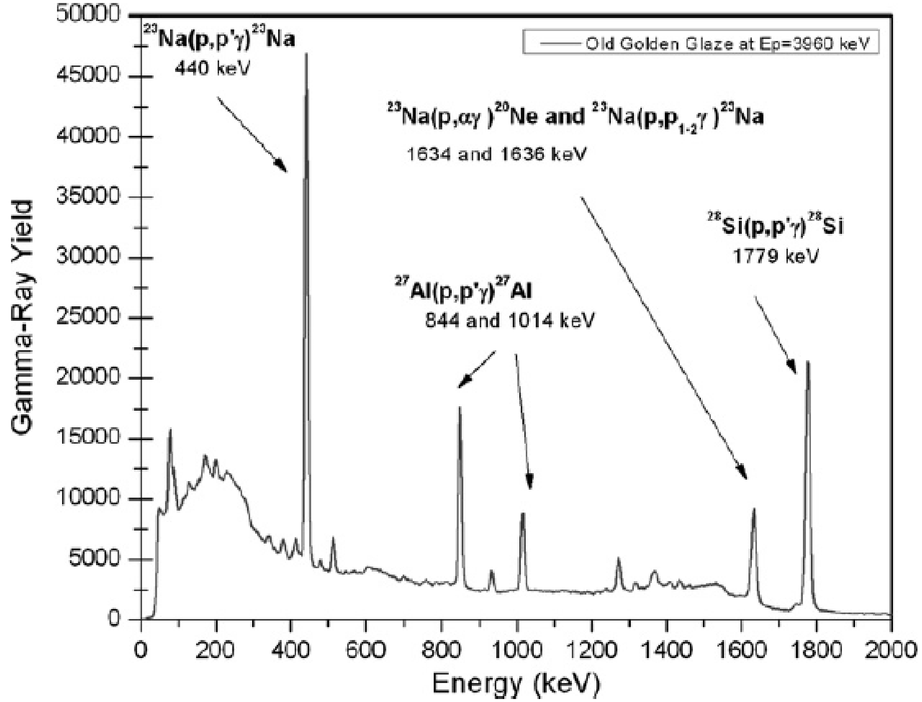
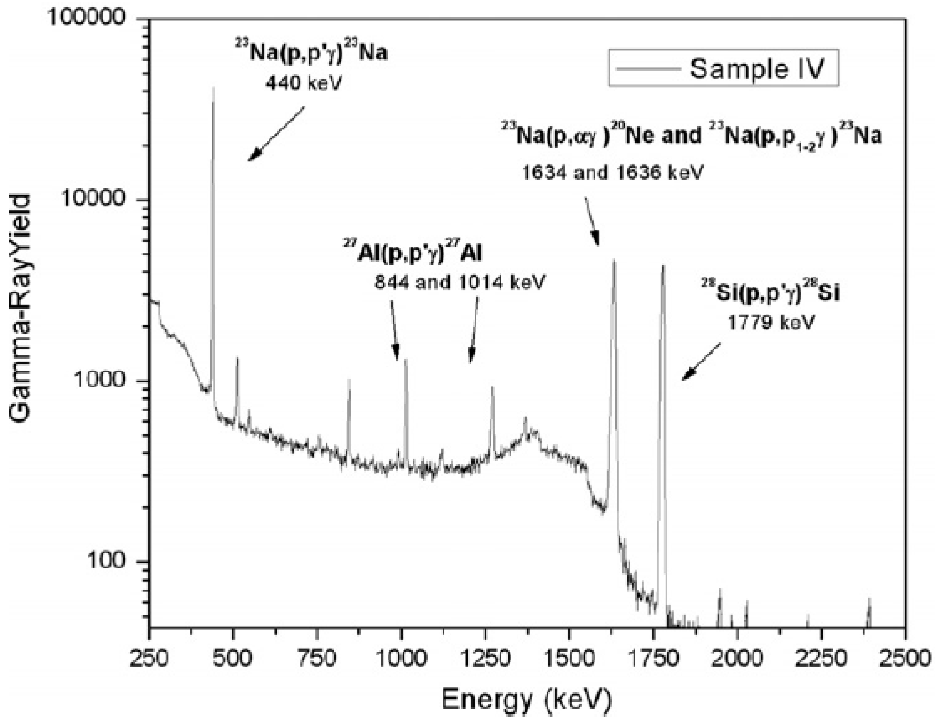
Particle Induced Gamma ray Emission

Golden glazes analysis by PIGE and PIXE techniques

M. Fonseca et al. NIMB 269 (2011) 3060





Analysis of Indian pigment gallstones

T.R. Rautray et al. NIMB 255 (2007) 409

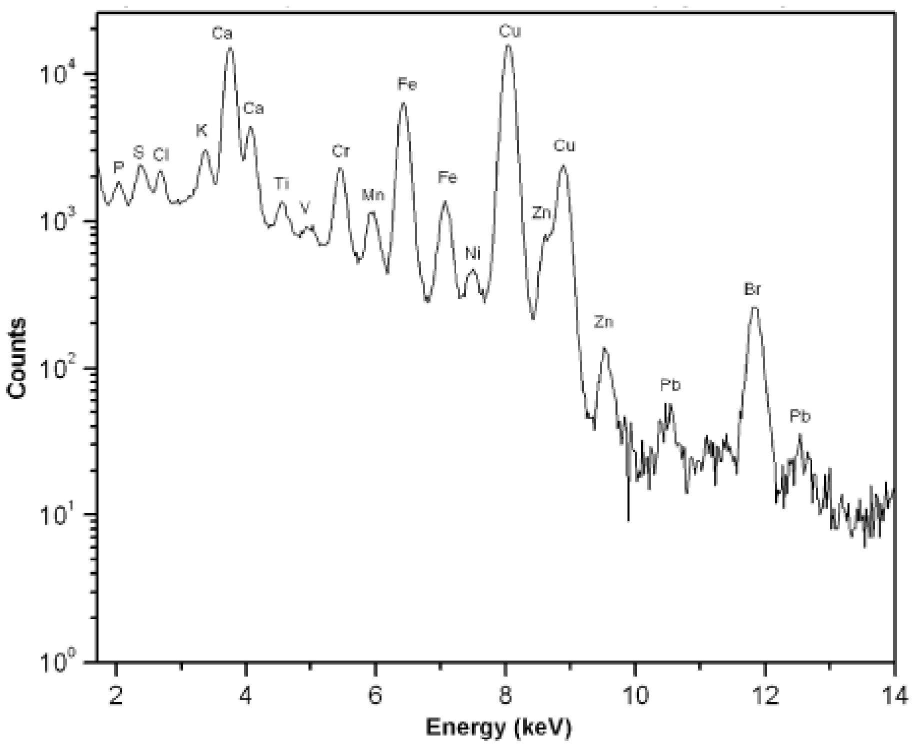
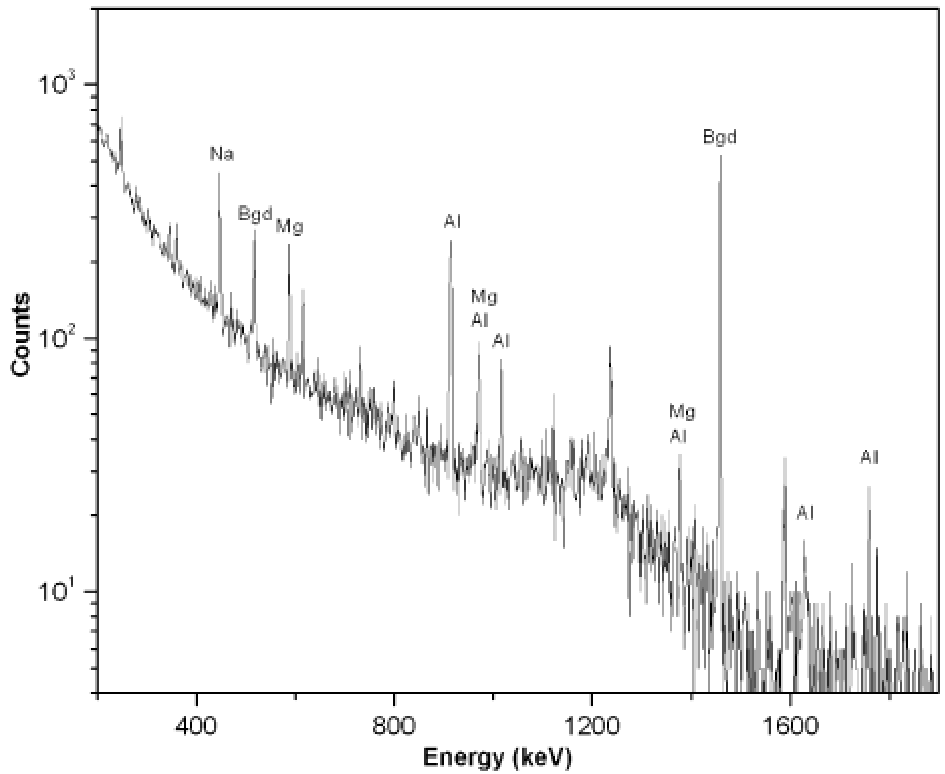
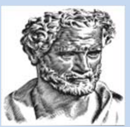


Fig. 2. PIXE spectrum of a south indian pigment gallstone.





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Particle Induced Gamma ray Emission

Advantages of scanning-mode ion beam analysis for the study of Cultural Heritage *N. Grassi et al. NIMB 256 (2007) 712–718*

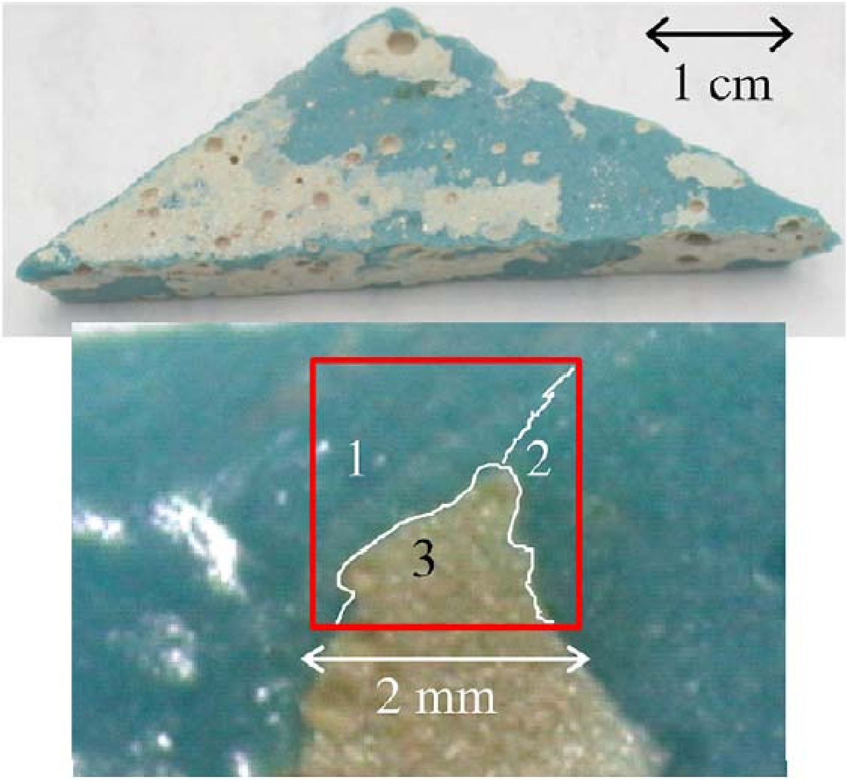
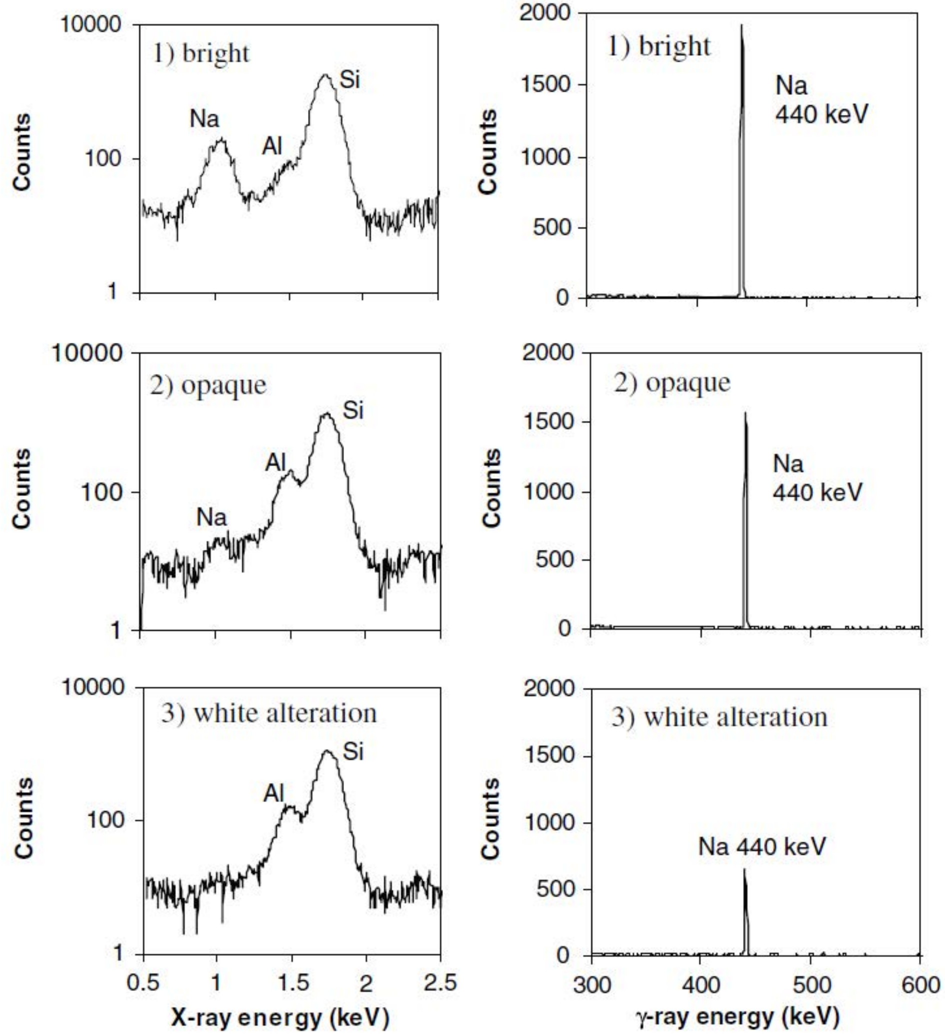


Fig. 4. Photograph of the glass *tessera* under study (top) and detail of the region analysed (bottom). Within the frame, showing the scanned area, three different zones can be distinguished: (1) bright, freshly broken; (2) opaque, with colour still visible and (3) white alteration.



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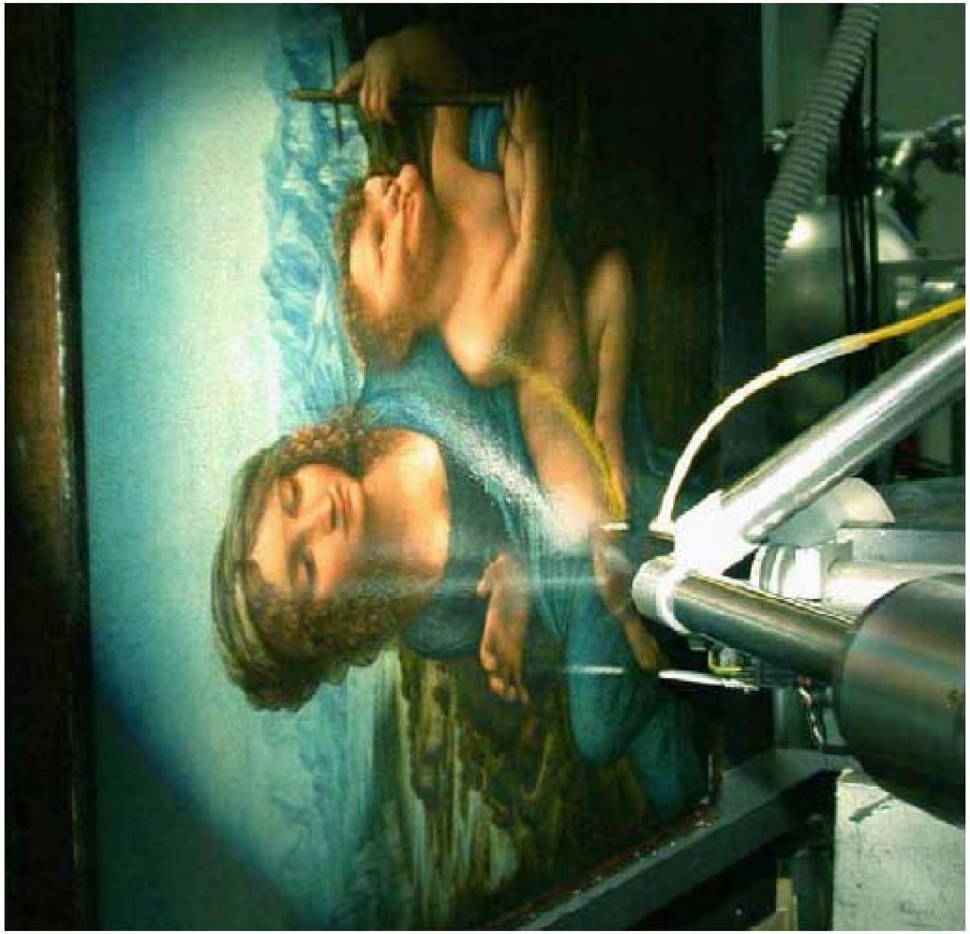
Particle Induced Gamma ray Emission

Identification of lapis-lazuli pigments in paint by PIGE measurements

N. Grassi et al. NIMB 219–220 (2004) 48



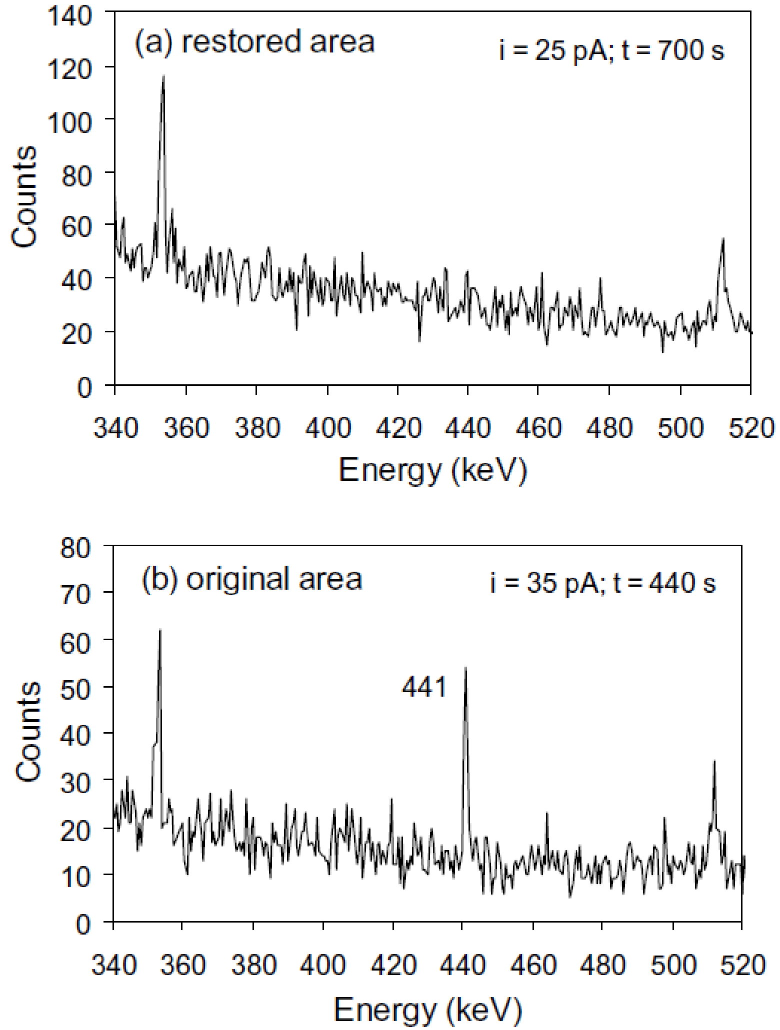


Fig. 5. PIGE spectra obtained irradiating two blue areas in the “Madonna dei fusi”: (a) a restored area; (b) an original area; here, the presence of lapis-lazuli in the paint layer is pointed out by the 441 keV γ-ray peak.

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Detection Apparatus

Beams used

- **Protons from 0.5 to 3 MeV** — **Probe larger depths**
- **Heavier ions (12C, 160) 10 to 20 MeV** — **Probe only surface layers**
Higher mass resolution
Higher depth resolution

Most commonly used detectors are Surface Barrier Detectors (SSB)

- **Various thicknesses (μm) and apertures (mm^2)**
- **They work only under high vacuum**
- **Can detect the energy of the particle (resolution ~ 15 keV)**
- **Can't detect the mass of the particle**

Sample considerations

- **Small size (few cm)**
- **Capable of being under vacuum (no wood e.t.c.)**
- **Preferable good electrical conductivity**

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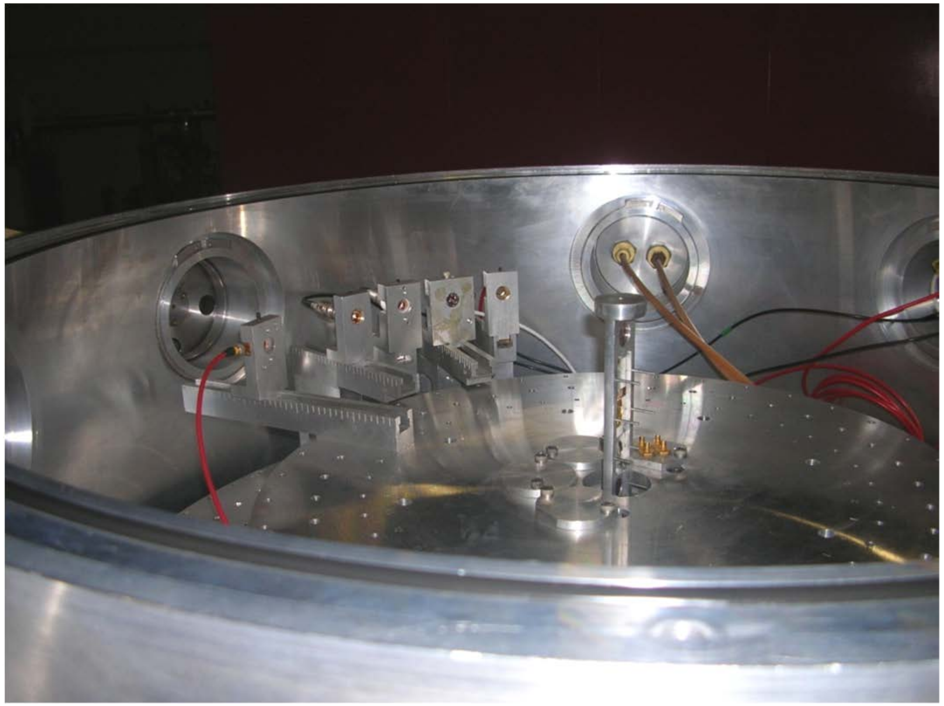
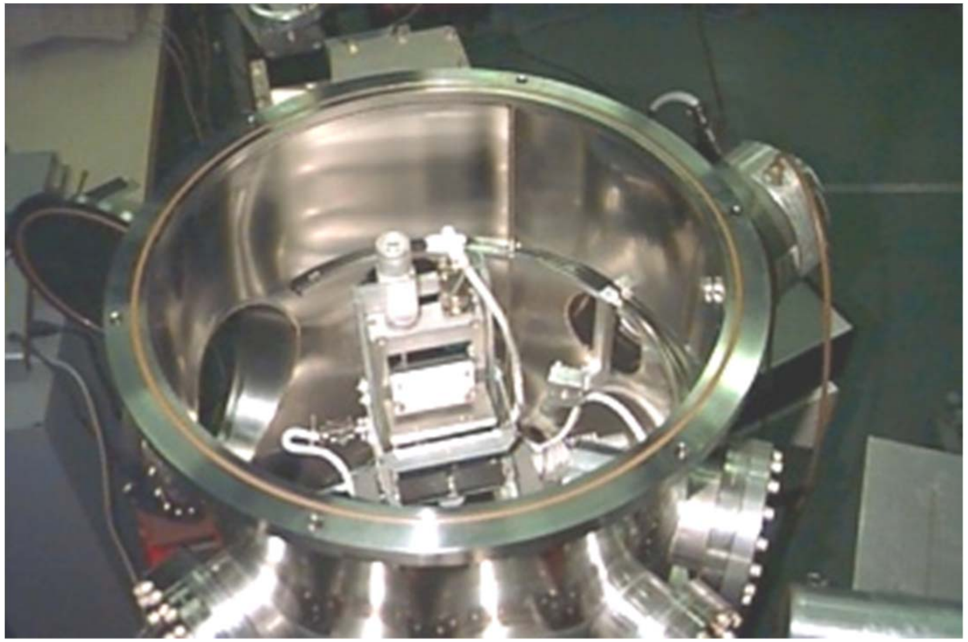
Experimental Setup





Motor driven goniometer
Great angular accuracy (0.01 deg.)
Up to 4 targets
Water cooling available

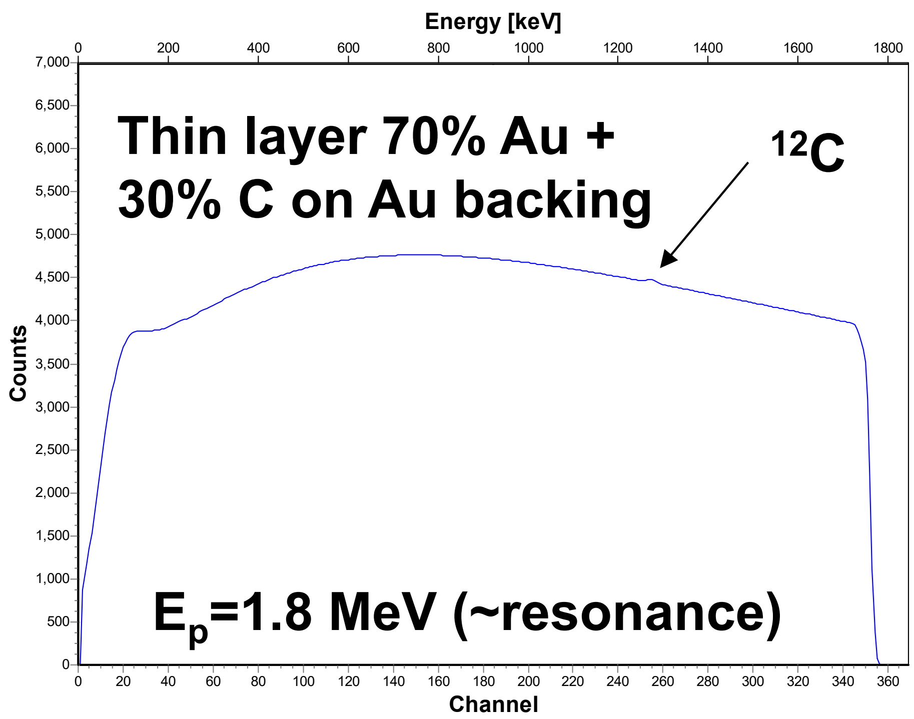
Motor driven goniometer
Suitable for channeling studies
4 – axis target movement
Place for PIGE detector

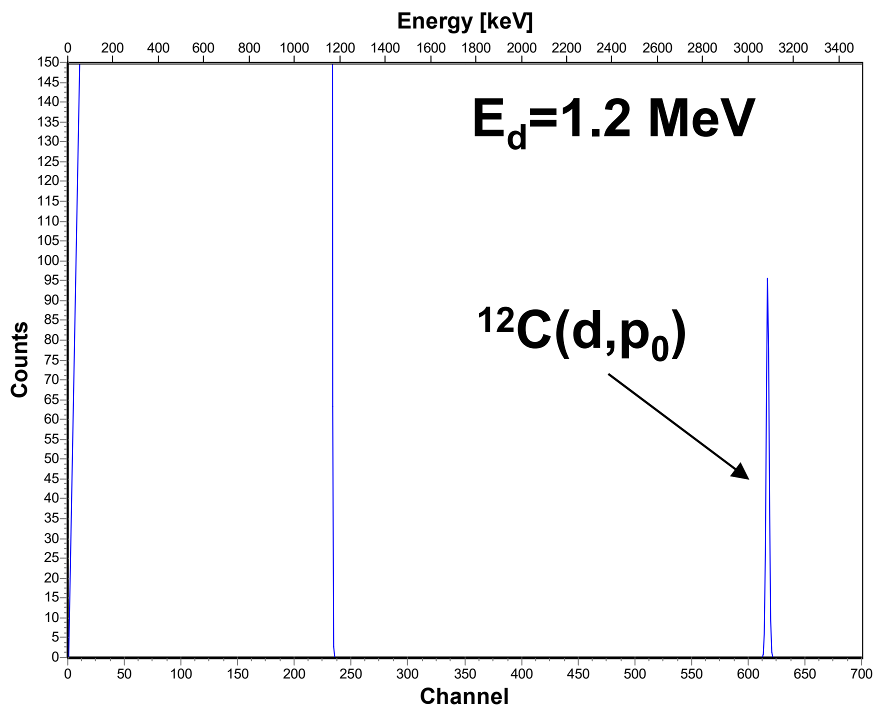
Nuclear Reaction Analysis

Use of nuclear reactions, (d,p), (d,α), (p,α), (α,p) etc.

Usually with high enough Q-values

e.g. The '**carbon problem**': RBS is weak, EBS can be applied only in certain cases (no other light elements present, no high-Z matrix, very case-specific measurements):





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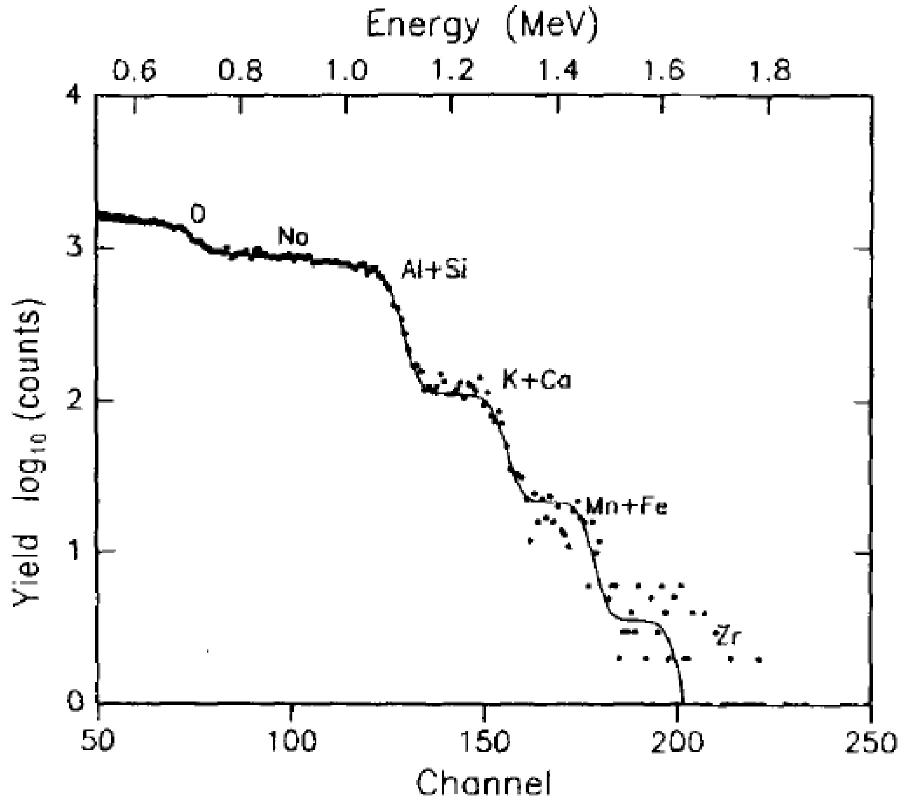
Examples

Analysis of Mexican obsidians by IBA techniques

G. Murillo et al. NIMB B 136-I 38 (1998) 888



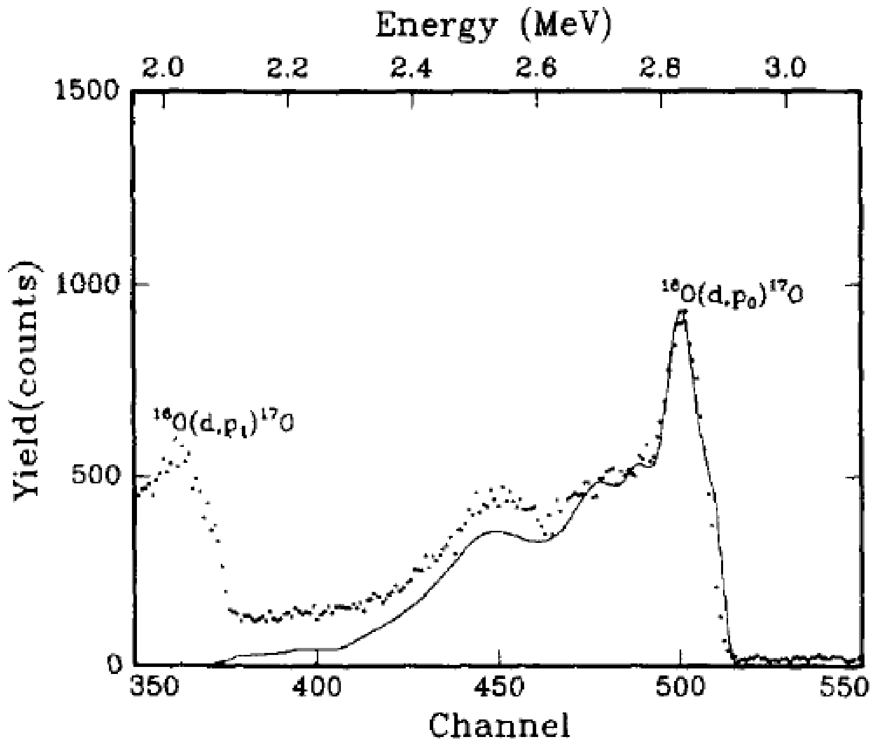
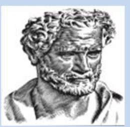


Fig. 4. Typical ion energy spectrum of an obsidian sample, used



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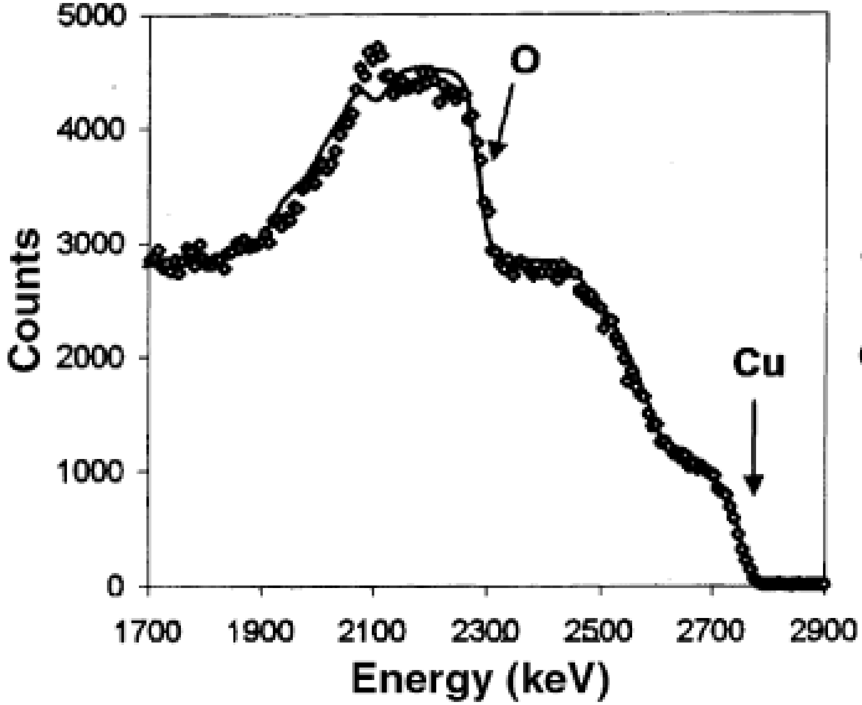
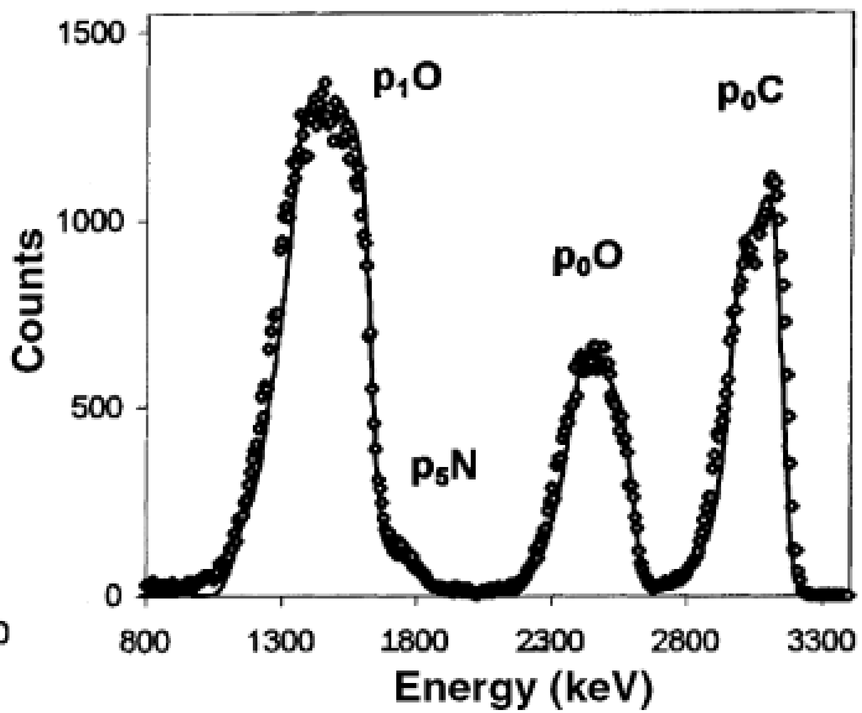
Examples

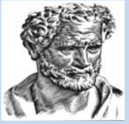
RBS and NRA with external beams for archaeometric applications

E. Ioannidou al. NIMB B 161±163 (2000) 730±736

Examination of patina layers on ancient steel







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Resonant PIGE

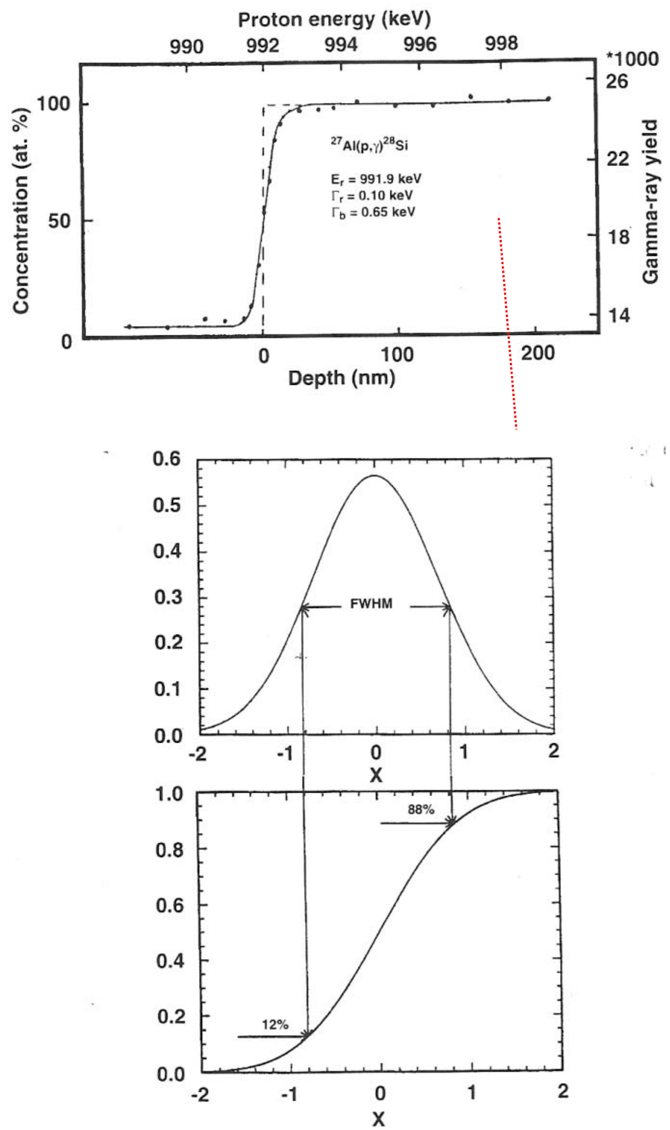
Reactions between particle and γ - rays

Use of the resonance phenomenon

Necessary to have a **STRONG** resonance and at the same time **NARROW** because this determines the depth resolution

Scanning of the sample by increasing the ion beam’s energy

The resonance **propagates** into the sample providing thus information about the depth profiling



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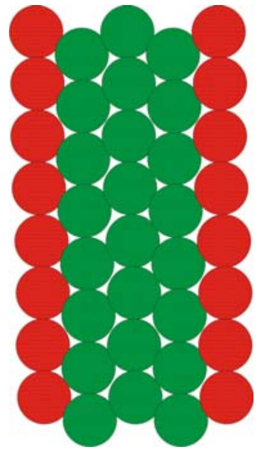
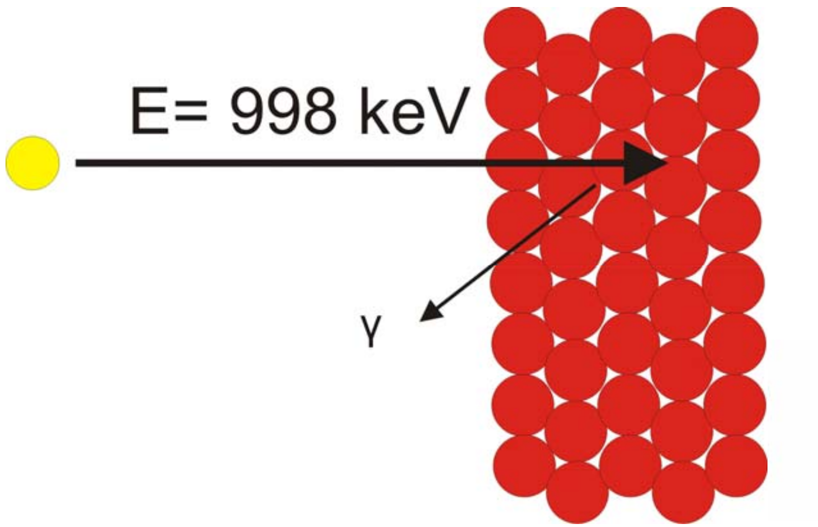
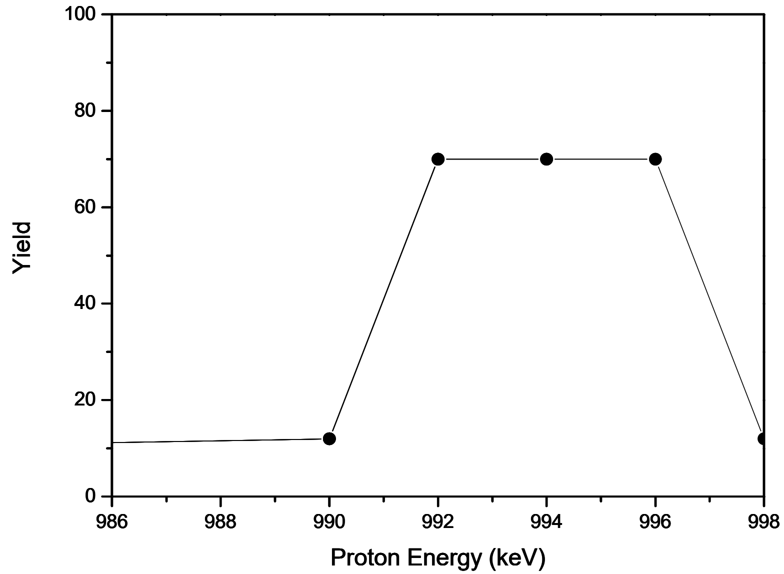
Resonant PIGE

Example: Resonance $^{27}Al(p,\gamma)^{28}Si$ E_p=992 keV







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Non-destructive evaluation of glass corrosion states

M. Mader et al. NIMB 136 I38 (1998) 863-868

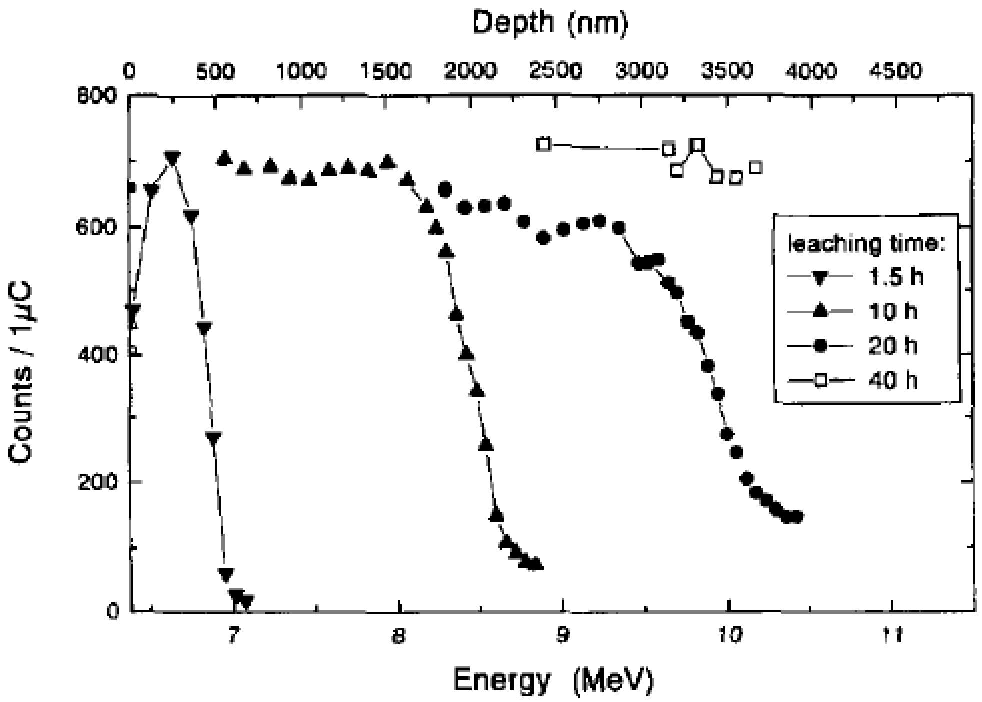

Fig. 4. Hydrogen depth profiles from M3 glasses treated in 0.1 n HCl for different times: 1.5, 10, 20, and 40 h. The $^{1}H(^{15}N,\alpha\gamma)^{12}C$ reaction was used around the 6.38 MeV resonance.

Resonant PIGE

PROS

- ✓ Ideal for depth profiling of hydrogen, fluorine and aluminum (EOA~1-10 ppm)
- ✓ Satisfactory results for carbon, nitrogen, oxygen, magnesium and silicon
- ✓ Quantification is made with the use of standards

CONS

- ➲ HPGe are expensive, fragile and they need cooling
- ➲ Time consuming measurements
- ➲ Suitable for only one element
- ➲ If the sample is thick, there is the possibility of resonance overlapping
- ➲ Prior knowledge of detector's efficiency is obligatory

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Destructiveness

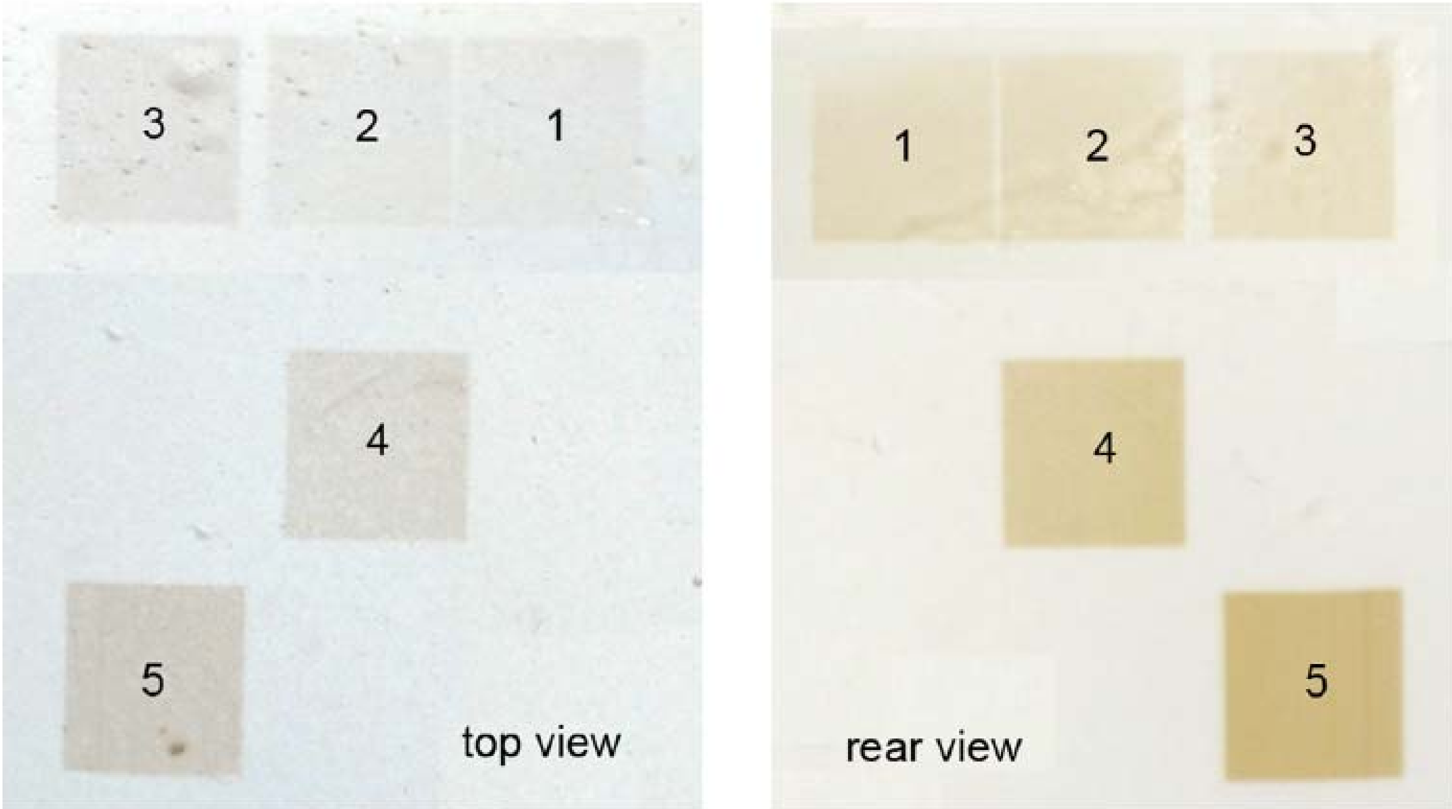
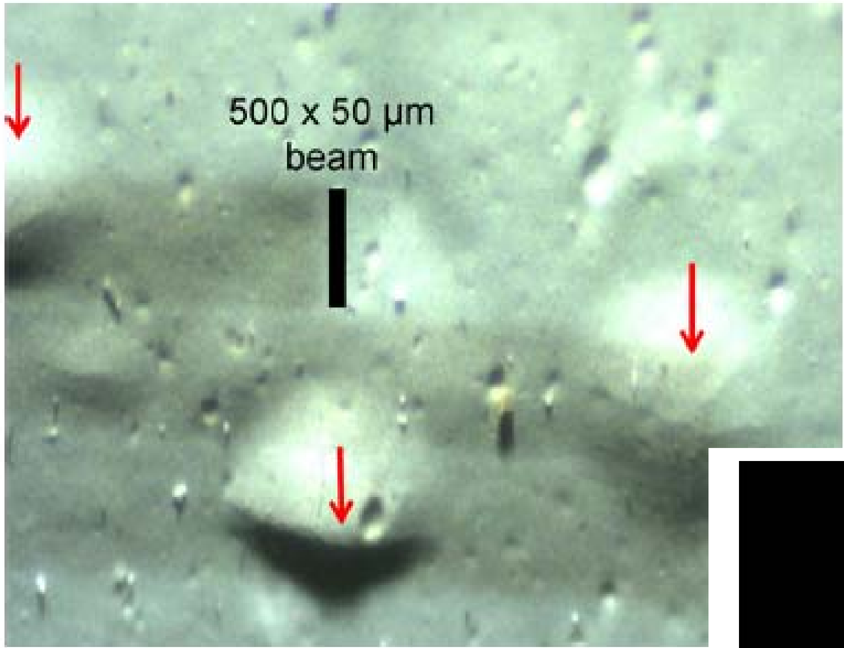
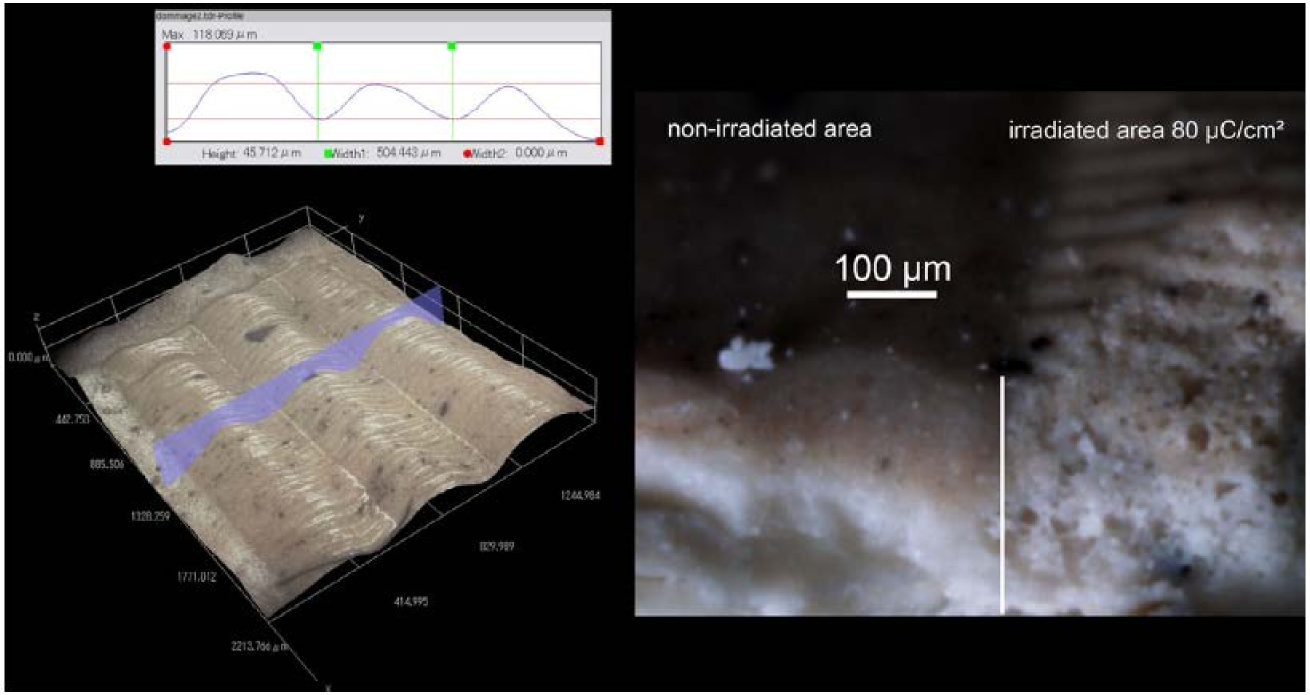


Fig. 3. Front and rear view of the model paint layer irradiated with various values of low fluences from 2.5 to 40 $\mu C/cm^2$, as in referred in Table 2. Damage starts to appears at 2.5 $\mu C/cm^2$, and is more marked on the rear view through the glass slide, where the end of the range can be observed.

Destructiveness





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Destructiveness

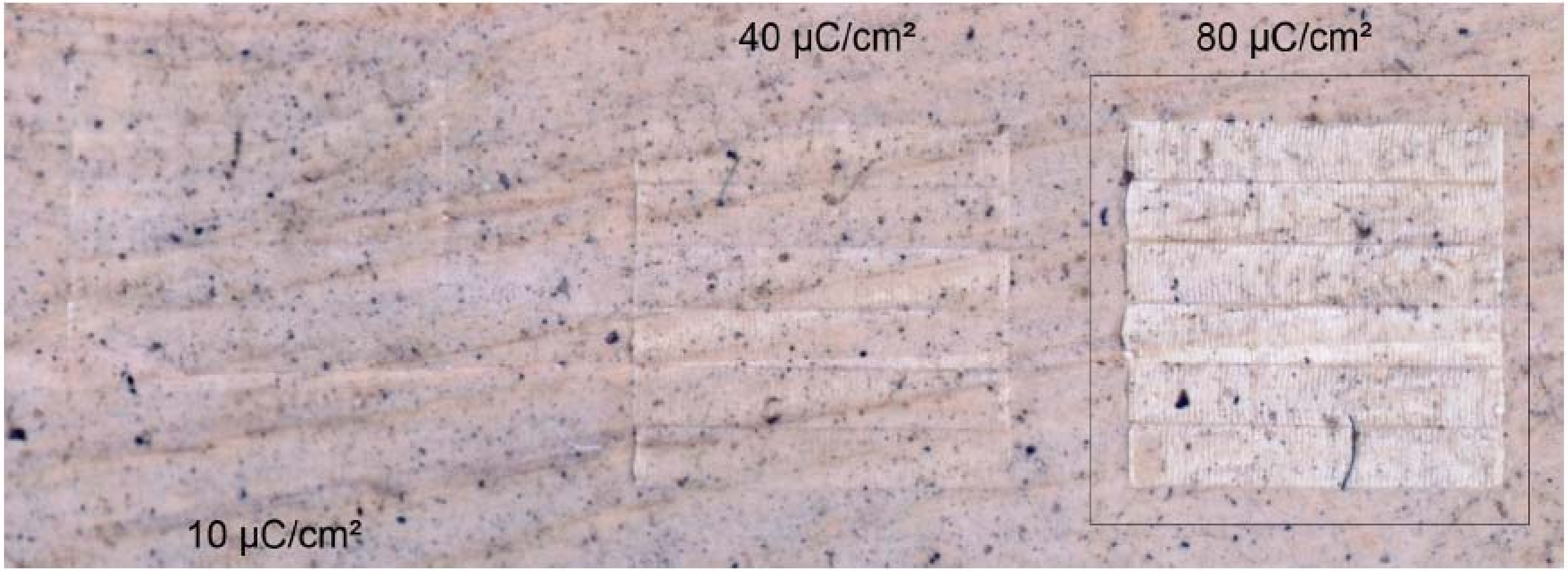

Fig. 6. Photomicrograph of the 3 × 3 mm^2 painting areas irradiated at fluences of 10, 40 and 80 $\mu C/cm^2$, left to right). On the two leftmost areas white stripes following the beam scanning can be noted

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Synopsis

Main Applications

- Material Science
- Archaeometry and Cultural Heritage
- Earth and Environmental Sciences
- Biological Sciences
- Nuclear Safety and Radioprotection
- Fundamental Nuclear and Atomic Physics
- Forensic

IBA

Relevant Techniques	Detection Capability
Rutherford Backscattering Spectrometry (RBS)	Z>1
Elastic Recoil Detection (ERD)	Z<17 (typically including H)
Particle Induced X-ray Emission (PIXE)	Z>11
Particle Induced Gamma-ray Emission (PIGE)	Z<17 (Li, B,F,Na, Mg, Al, Si)
Nuclear Reaction Analysis (NRA)	Z<17 (often for C, N, O and isotope detection)
Scanning Transmission Ion Microscopy (STIM)	Sample density
Ionoluminescence (IL)	Defects and Sample Structure

Conclusions

Summary

1. Rutherford backscattering (RBS) is ideal for depth-profiling heavy elements on lighter substrates.
2. Elastic recoil detection analysis (ERDA) is excellent for depth-profiling very light elements in thin films.
3. Nuclear reaction analysis (NRA), is excellent for high resolution depth-profiling of specific isotopes.

Present Situation

1. A lot of work is being done in PIGE and NRA.
2. Micro-beams and measurements in air (Louvre) have enhanced IBA capabilities.

Future Perspectives

1. New techniques are always evolving (e.g. HR-RBS).
2. PIGE analytical algorithms?
3. CAN WE SOLVE ALL THE PROBLEMS??? NO (BUT MANY YES…)

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