Laser-driven proton beams for precise nanoparticle synthesis and cultural heritage diagnostics

M. Barberio[1], S. Giusepponi, S. Vallières, M. Scisció, S. Veltri, M. Celino, P. Antici

Outline

1)

Laser driven proton beams for precise nanoparticle synthesis

Motivation: Nanomaterials in medicine and nano-imaging

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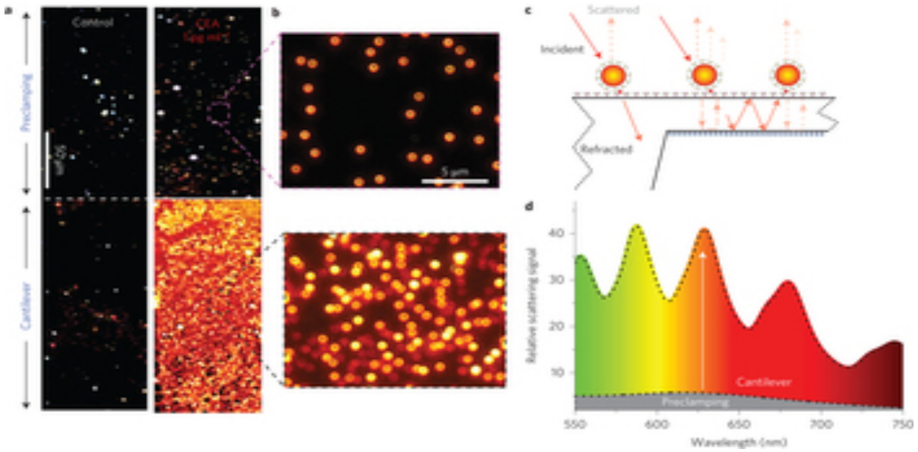
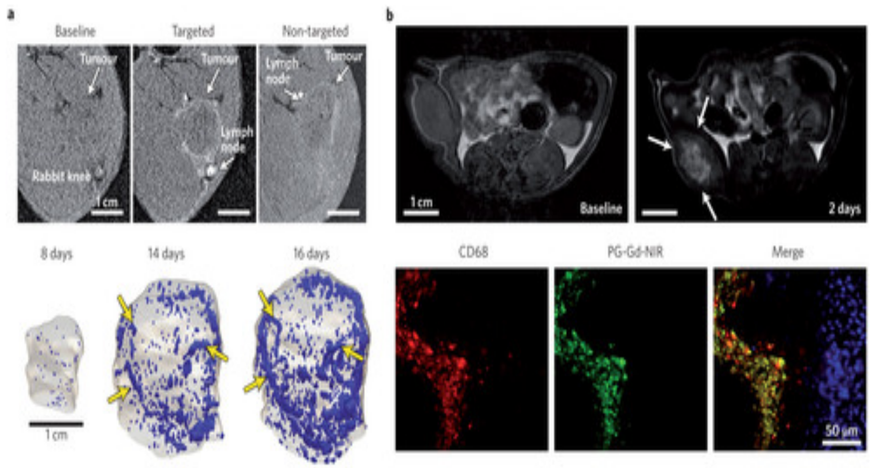
"Detection of cancer biomarkers in serum using a hybrid mechanical and optoplasmonic nanosensor", *Nature Nanotechnology* 9,1047–1053, (2014) - Gold nanoparticles act as biosensors in cancer cell detection



"A targeted approach to cancer imaging and therapy", *Nature Materials* 13,110–115 (2014)

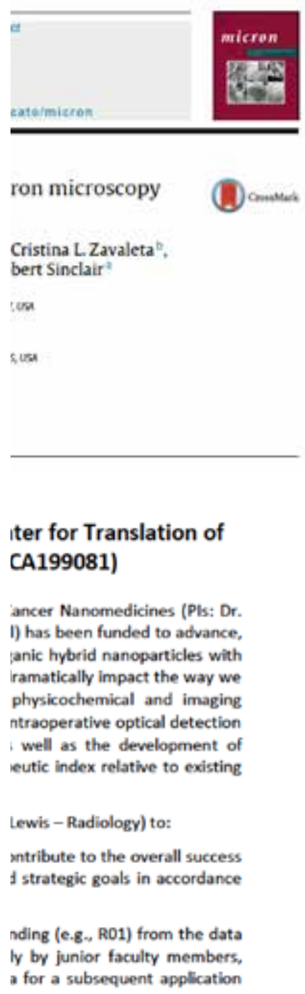
Nanoparticle-based imaging plays a crucial role in cancer diagnosis and treatment. Here, we discuss the modalities used for molecular imaging of the tumour microenvironment and image-guided interventions including drug delivery, surgery and ablation therapy.



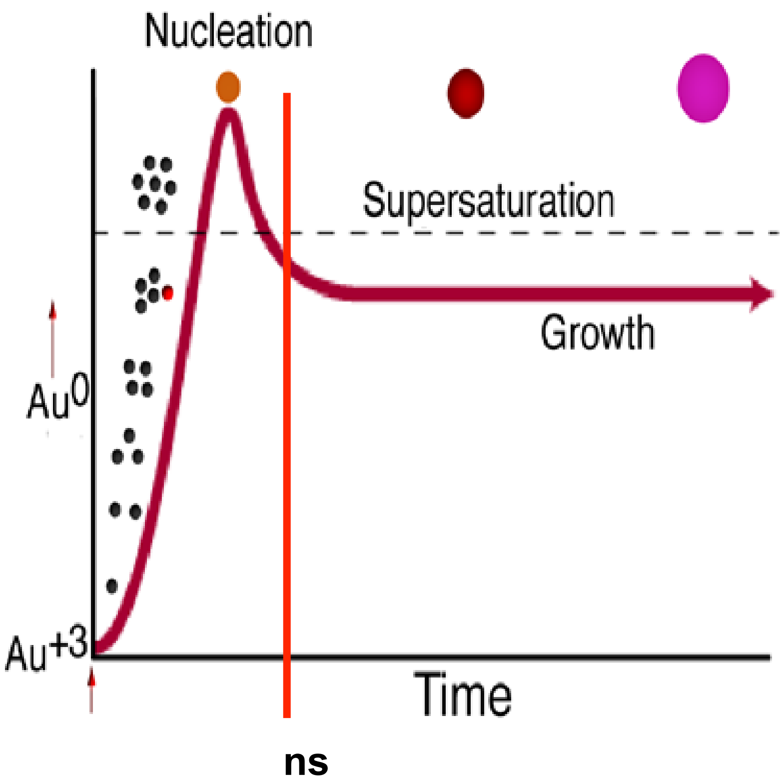
...and recently also there is a call for applications in Material Science...

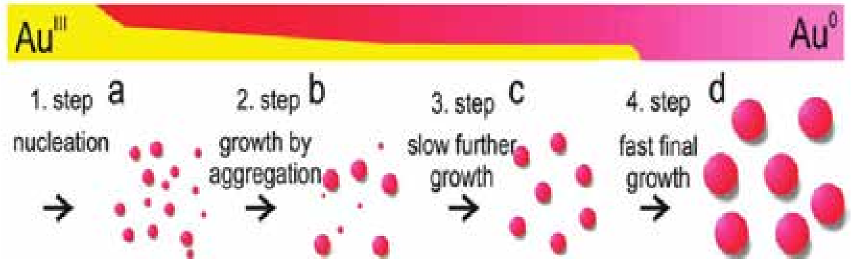
Palmer R. E., et al., Nanostructured surfaces from size-selected clusters, Nature Materials 2, 443 - 448 (2003) *"The precise definition of the size and density of nanoscale surface features has potential applications in many fields, such as the fabrication of semiconductor nanostructures and the immobilization and orientation of biological molecules.* ***Future challenges include the development of additional control over the shape (as well as the size)****…"*

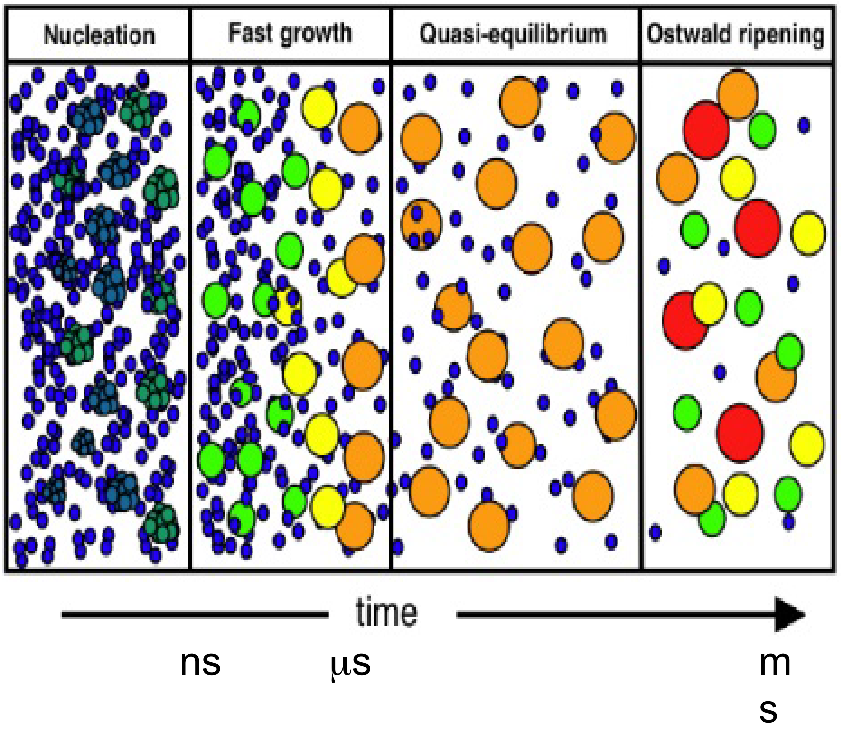
Guduru D., et al., Nanostructured material surfaces – preparation, effect on cellular behavior, and potential biomedical applications: A review, Int J Artif Organs (2011); 34 (10): 963-985 *"A surface-patterning method capable of high precision and accuracy over large areas is required to investigate topographic effects on cell behavior. Many lithographic methods, including EBL, nanosphere lithography (NSL) and microcontact printing (µCP) have been explored to mimicECM. Even so, many of these methods fall short due to their small-sized patterned areas, micron resolution limits, slow processing speeds, and high costs. A variety of chemical and solution-based methods, such as electrospinning and solvent casting, provide larger patterning areas of higher resolution,* ***however, they lack precise control over the feature dimensions and orientation****.*



Conventional NP growth



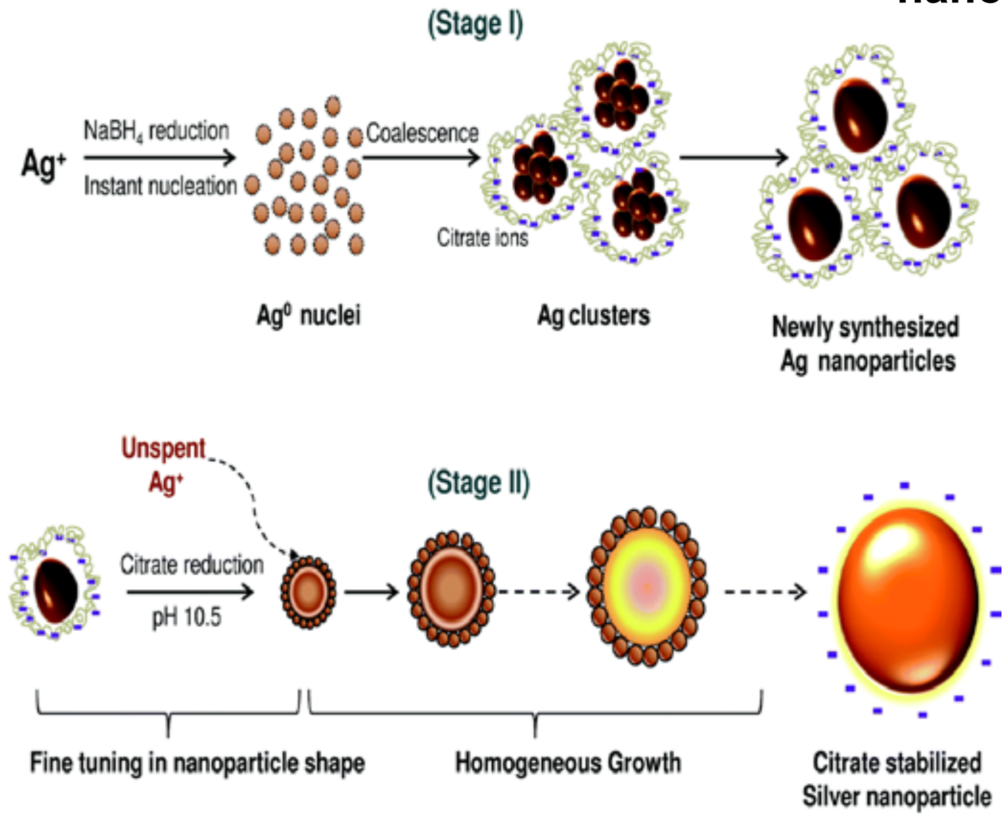


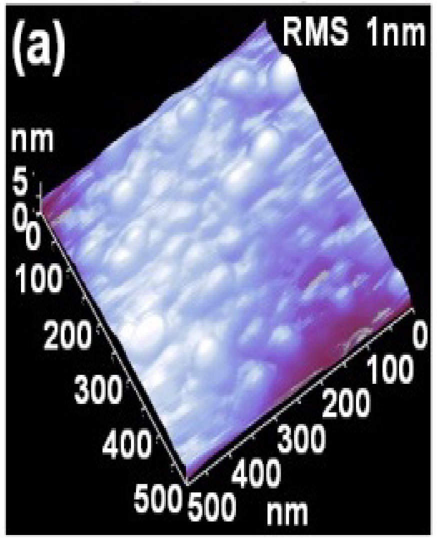


Methods for nanomaterials using Chemical methods

Chemical methods: the nucleation phase starts with a chemical reaction

Advantage: Good control of nucleation phase (time of reaction in the order of ns).
Problem: formation of a surfactant shell on the nanomaterials which strongly affect the nanoparticle's properties





Methods for nanomaterials using Physical methods

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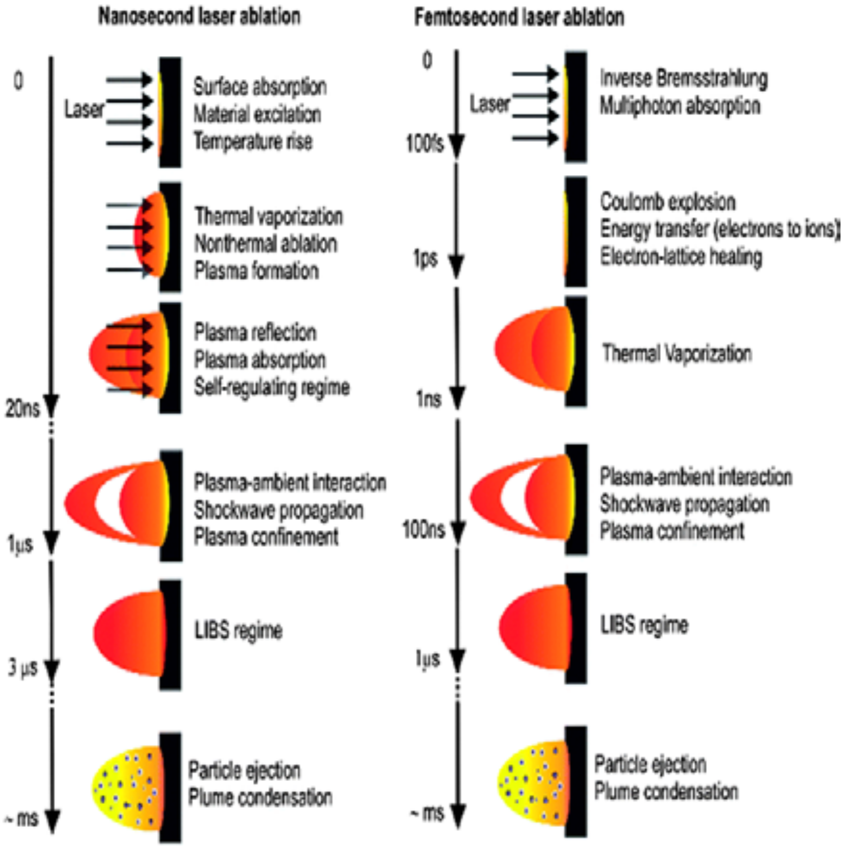
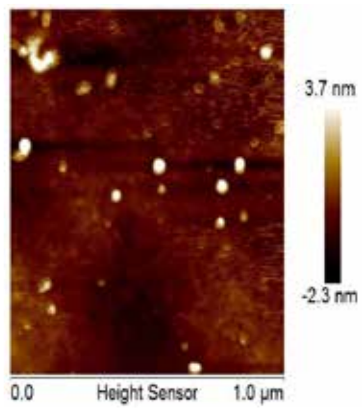
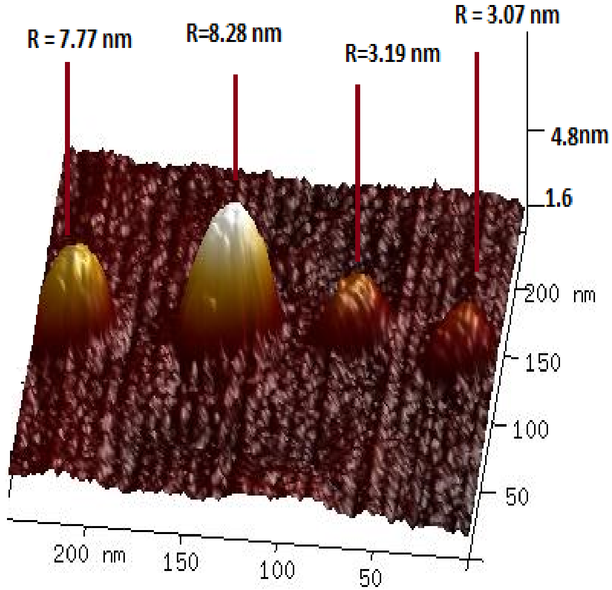
Physical methods (i.e. laser based): the nucleation phase starts with an evaporation of atoms from a surface

Advantage: Good control on the chemical composition of nanomaterials.

Problem: the control of particle dimensions and shape is difficult. The particle aggregation is slow with the consequent formation of big particles and aggregates







Principle of Laser-Driven Proton Ablation (LDPA) mechanism

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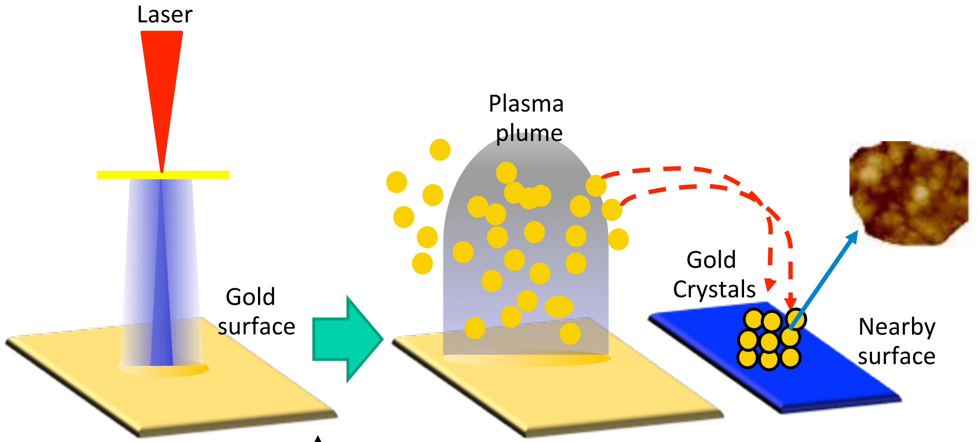
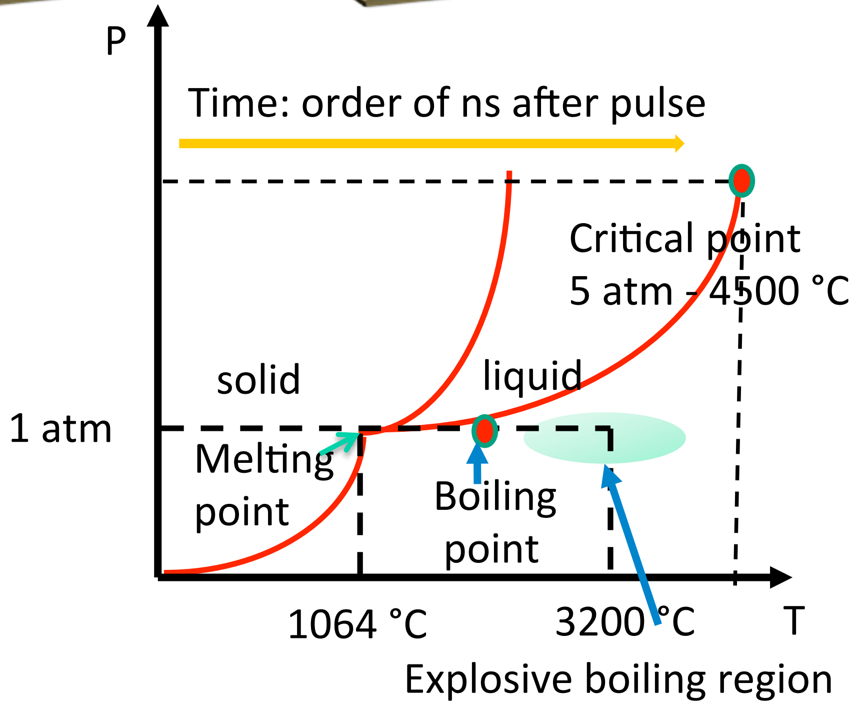




Proton irradiation

Temperature increases above boiling point in sub-ns timescale

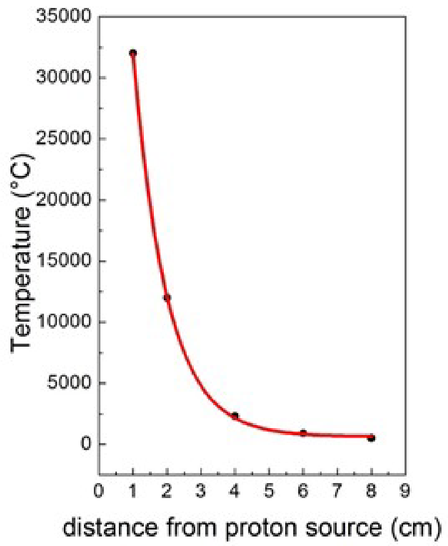
«Explosive boiling» with formation of plasma plume

Aggregation of particles in plasma plume and formation of supersonic jet

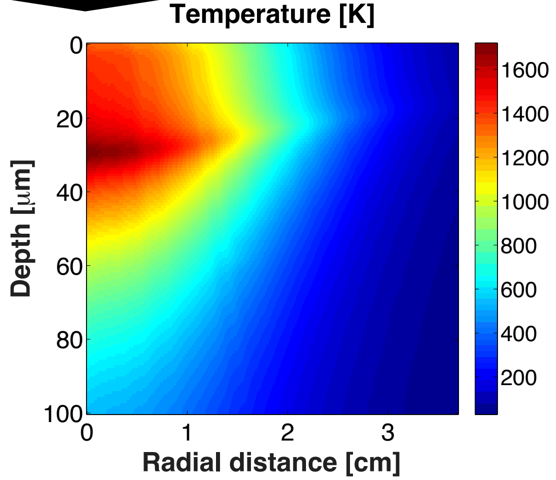
Deposition of particle on close surfaces

Energy Deposition code confirms Explosive Boiling conditions

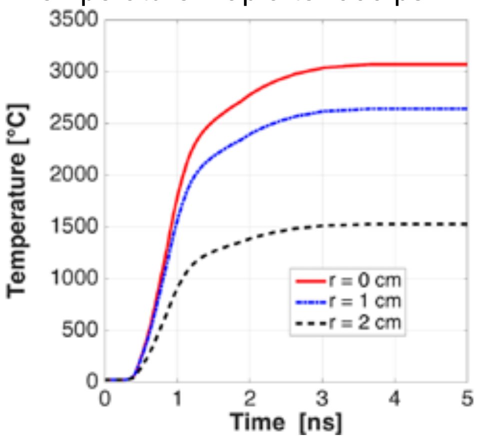
Chose the correct distance:



protons



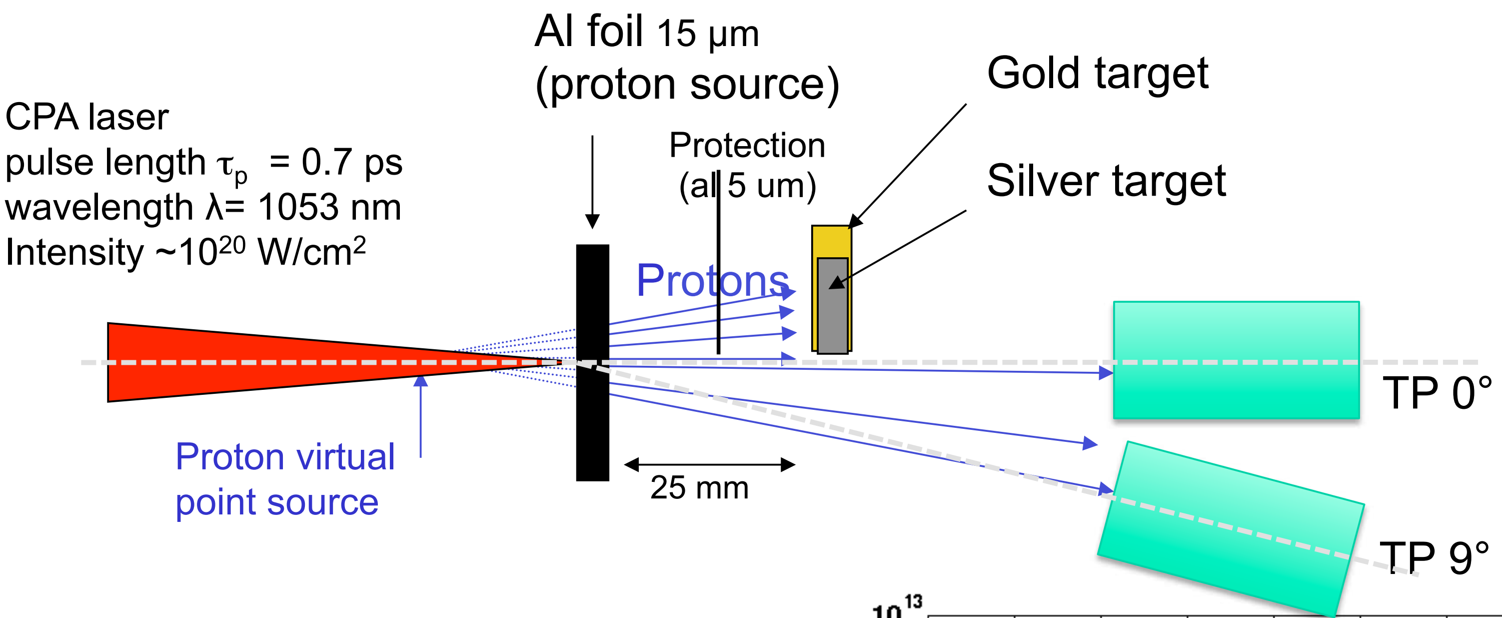
Temperature map after 900 ps

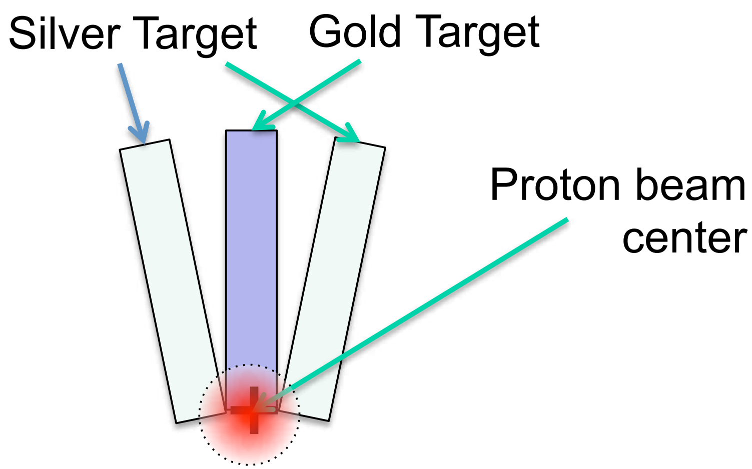


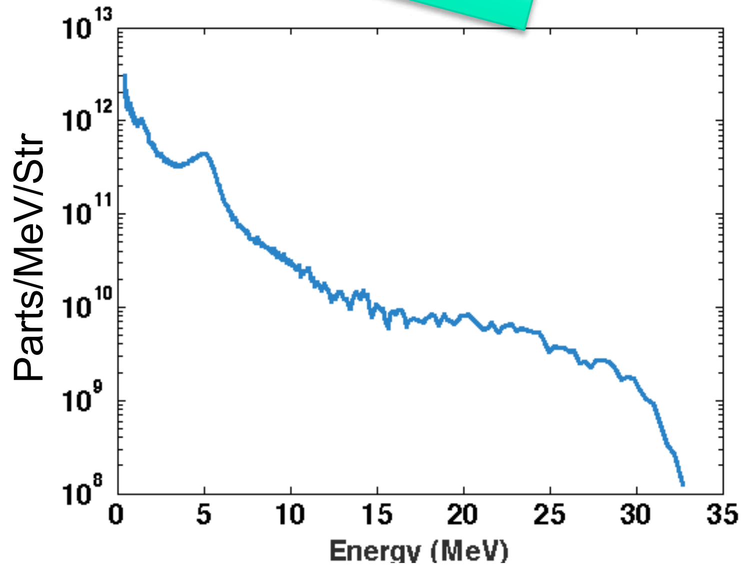
Temperature on the Gold target surface

Gold boiling point 2700 °C

First experimental verification on TITAN laser (LLNL, USA)

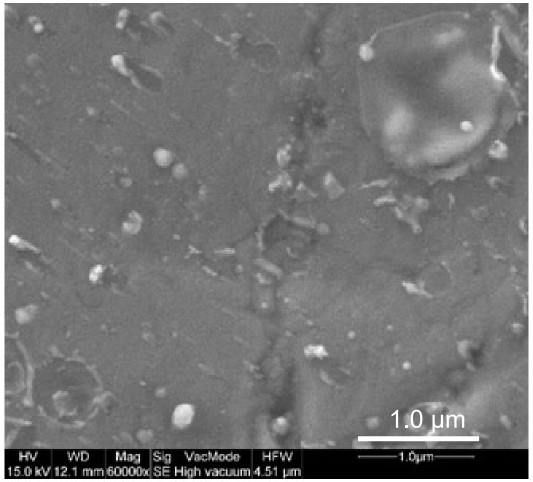






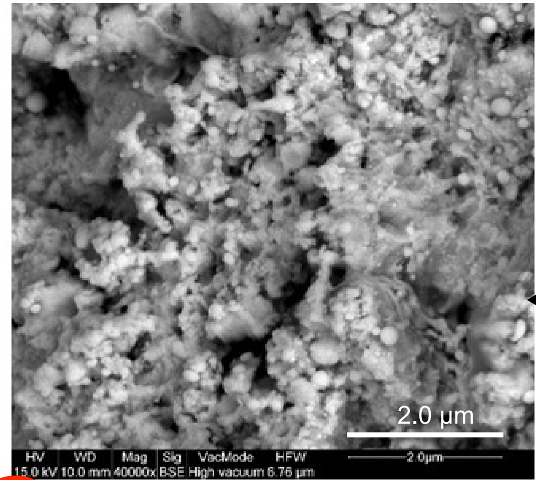
Explosive Boiling texture on the gold target

Gold Target before

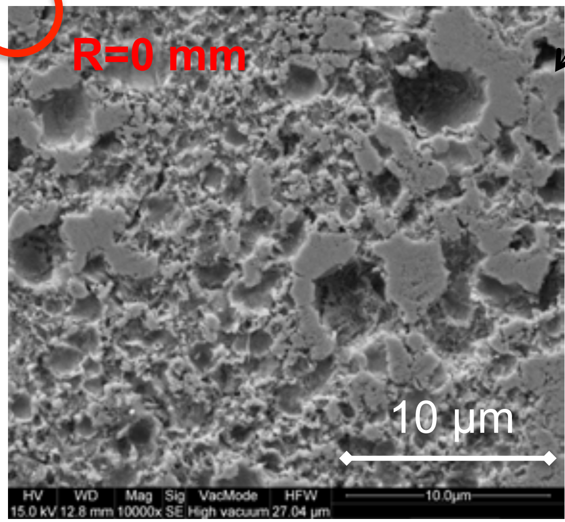


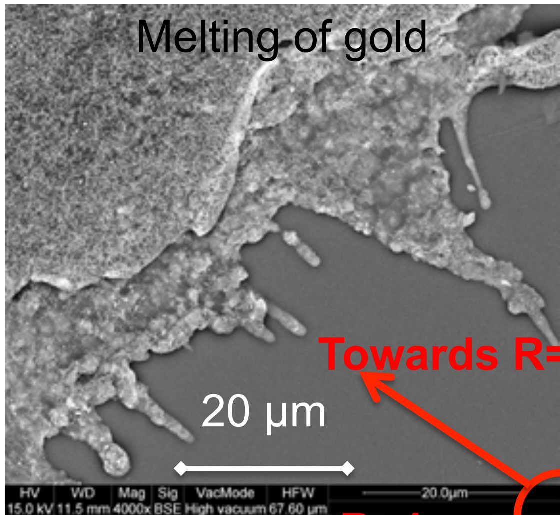
Gold Target after

R=~1 mm

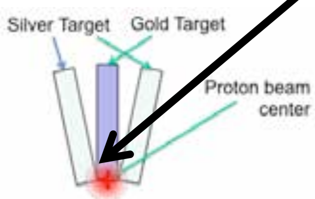


Massive erosion of gold
(highly porous and rough)
-> thermal shock



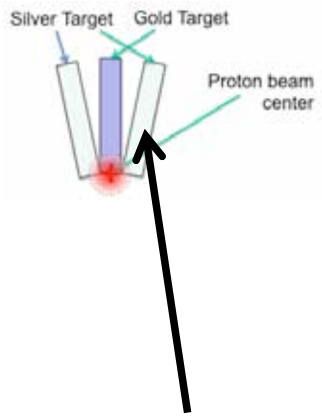


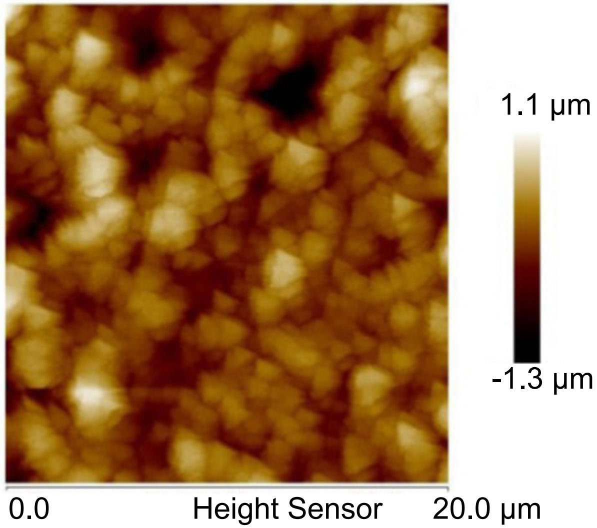
R=4 mm

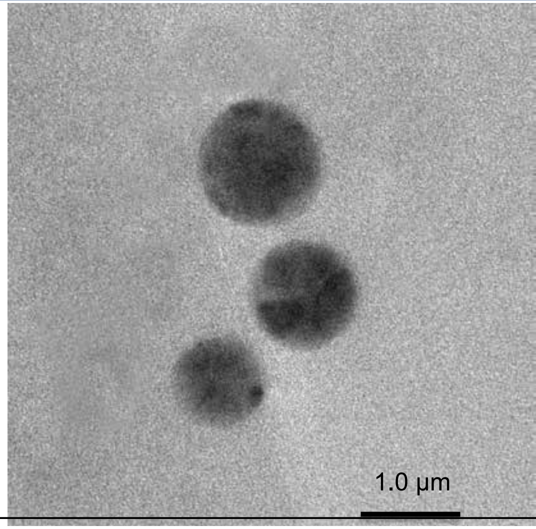


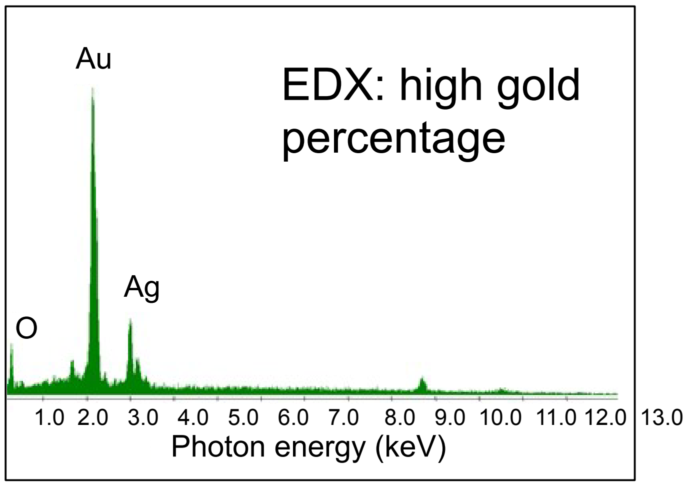
SEM images

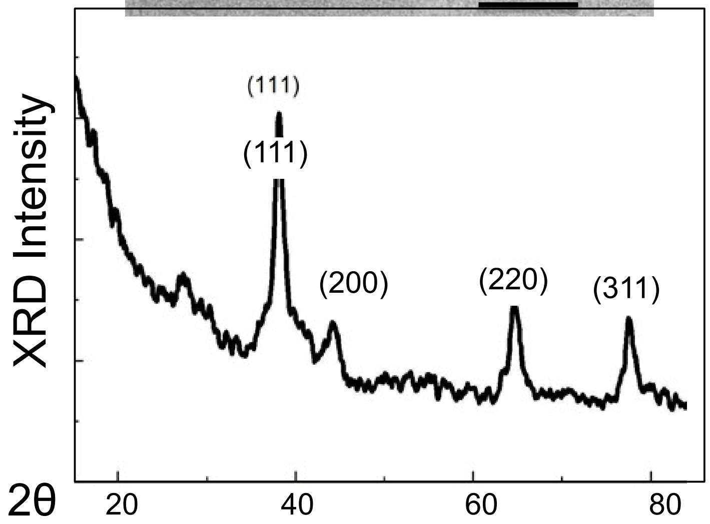
Nanoparticle production on the Silver target located nearby



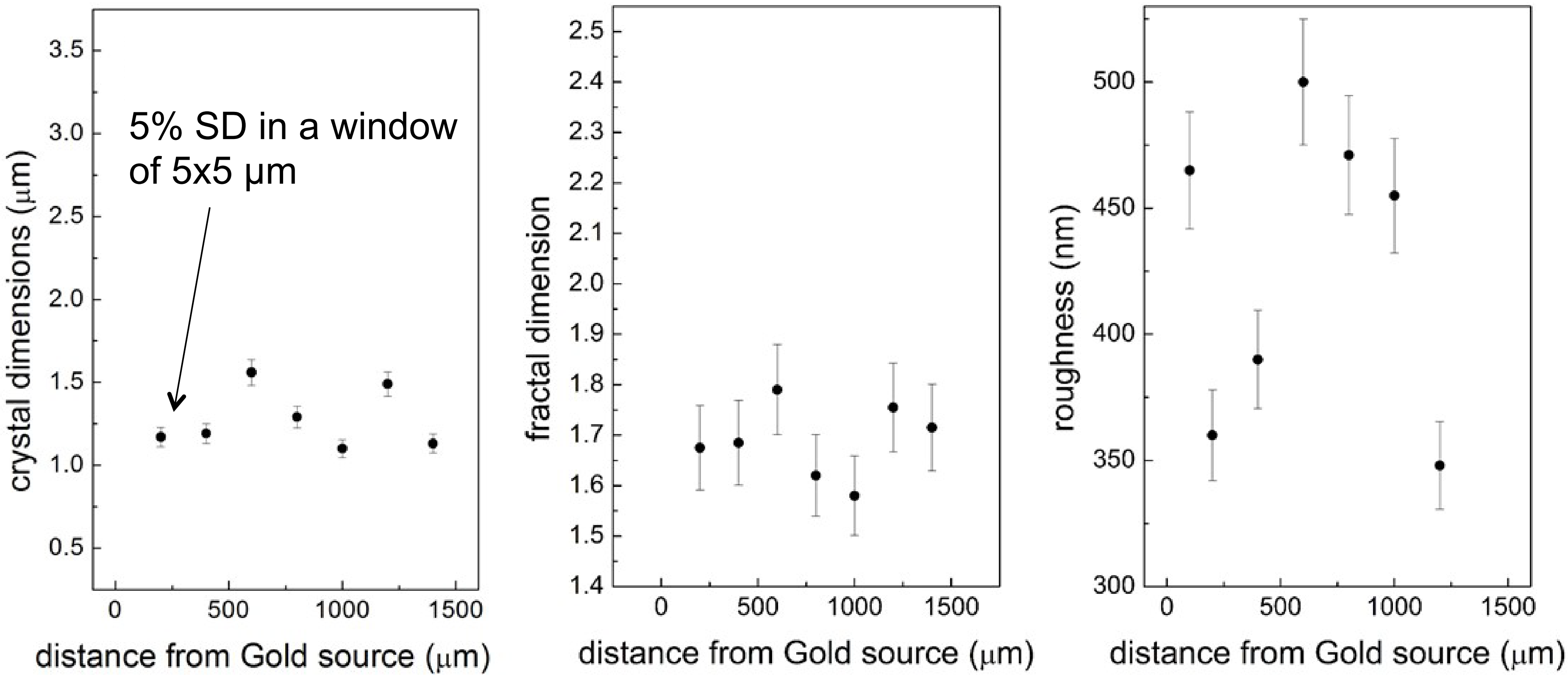








Statistics of the Gold Nanoparticles



M. Barberio, M. Sciscið, S. Vallières, S. Veltri, A. Morabito, P. Antici, Sci. Rep. 7, 12522 (2017)
P. Antici, M. Barberio, Patent Pending US 14448.128

Outline

2)

Laser driven proton beams for cultural heritage diagnostics

Context and motivation: chemistry and physics for Cultural Heritage (CH) analysis

"Accessibility and preservation of cultural heritage is needed for the vitality of engagement within and across European cultures by also considering the importance of cultural heritage as strong economic driver in a post-industrial economy and its contribution to sustainable economic growth."

- M. Barberio, …, P. Antici: **TiO_2 and SiO_2 nanoparticles film for cultural heritage: Conservation and consolidation of ceramic artifacts**
  Surface and Coatings Technology, 271, 174 (2015)
- M. Barberio,…, P. Antici: **AFM and Pulsed Laser Ablation Methods for Cultural Heritage: Application to Archeometric Analysis of Stone Artifacts**
  Appl. Physics A 0947, 8396 (2015)
- S. Veltri, …, P. Antici: **Synthesis and Characterization of thin-transparent nanostructured films for surface protection**
  Superlattices and Microstructures 101, 209 (2017)
- M. Barberio, …, P. Antici: **Pigment darkening as case study of In-Air Plasma Induced Luminescence**
  Sciences Advances (in press)

Current Challenge in CH:

Physics and Chemistry for Cultural Heritage: obtain a complete chemical/ morphological analysis of artifacts, preventing damage

Chemical analysis:
X-ray photoelectron *Spectroscopy* (**XPS**)
X-ray fluorescence **(XRF)**
Energy Dispersive *Spectroscopy* (**EDX**)
Photoluminescence
Particle induced X-Ray Emission **(PIXE)**

Morphological analysis:
SEM
AFM

What is Particle Induced X-ray Emission (PIXE)

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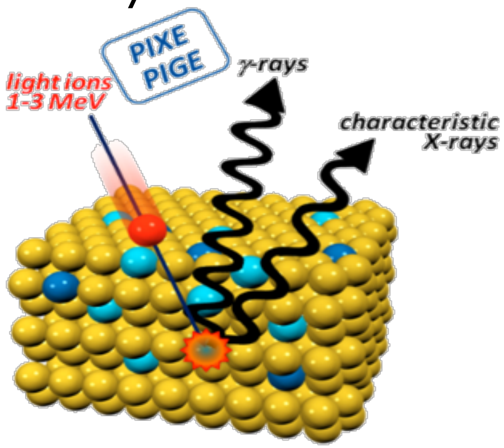
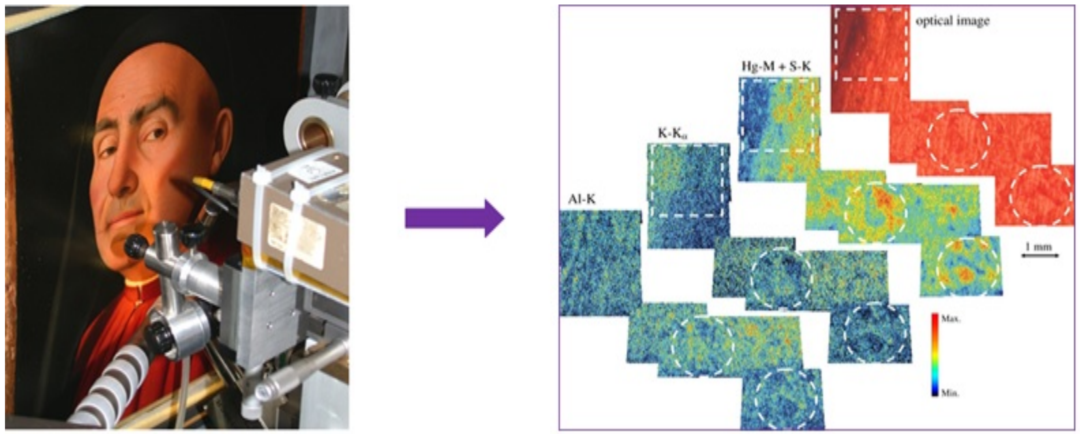
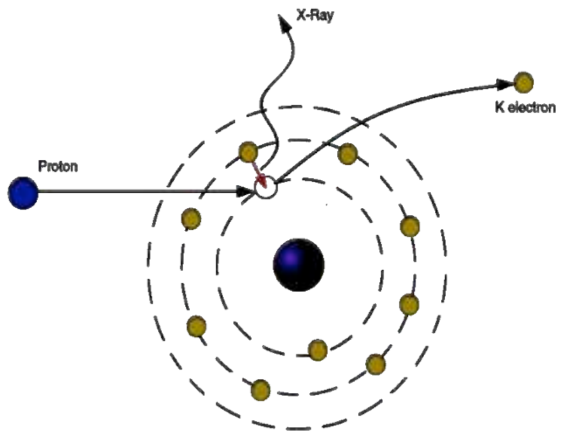
PIXE: proton beams stimulate the emission of X-rays (Gamma), which allows performing a chemical analysis of the material.

A technique used in the Cultural Heritage analysis, medicine, industry



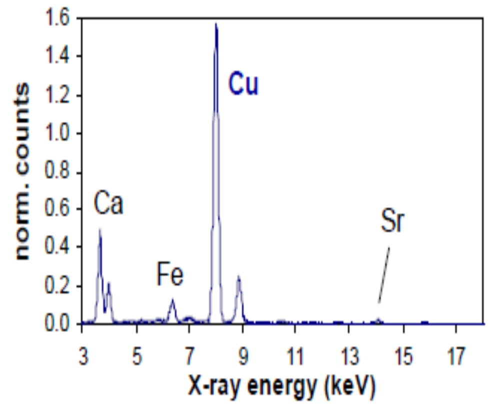


Example of PIXE to analyze the pigment's composition of The Trivulzio portrait by Antonello da Messina



K and L shell X-ray emission

- Advantage over X-Ray Fluorescence: detection of low Z elements and higher spatial precision
- Detection of elements up to 10 ppm
- Little invasive



Analysis performed at LABEC (Florence)

Current PIXE analysis with conventional accelerators

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20 minutes
TECHNOLOGIE
Le patrimoine du Louvre passe sous le faisceau de l'accélérateur de particules Aglaé P.14
GRAND PARIS
ARNAQUE
Un banquier jugé pour avoir escroqué une octogénaire P.2
PRÉSIDENCE LR
La candidate Florence Portelli
PAPY, C'ÉTAIT COMMENT LA RETRAITE?


14 Culture
PATRIMOINE L'unique accélérateur de particules consacré à l'étude des chefs-d'œuvre est au Louvre
L'art percé par le faisceau d'Aglaé
Provenance et composition
La machine permet d'établir la carte d'identité chimique des œuvres.
MORGANE SIRGUEY
DÉDICACE FNAC BERCY VILLAGE
SAMEDI 25 NOVEMBRE À 15H
Le trésor de Bavay passé au crible
Plus de 350 bronzes

	DR-N	
	Total	Pixel
Scan size	1 × 1 mm^2	
Pixel resolution		10 μm
Beam charge	18 μC	1.8 nC
Av. current	8 nA	
Acquisition time	35 min	210 ms
Matrix count rate	11 kc/s	
Trace count rate	69 kc/s	
Average counts		2500

A well established technique in many laboratories: AGLAE (France), LABEC (Florence), CENBG (Bordeaux) etc.

L. Pichon et al., Nucl. Instr. Methods Phys. Res. B 363, 48-54 (2015)

Laser-driven PIXE: we replace a conventional accelerator source with a laser plasma based

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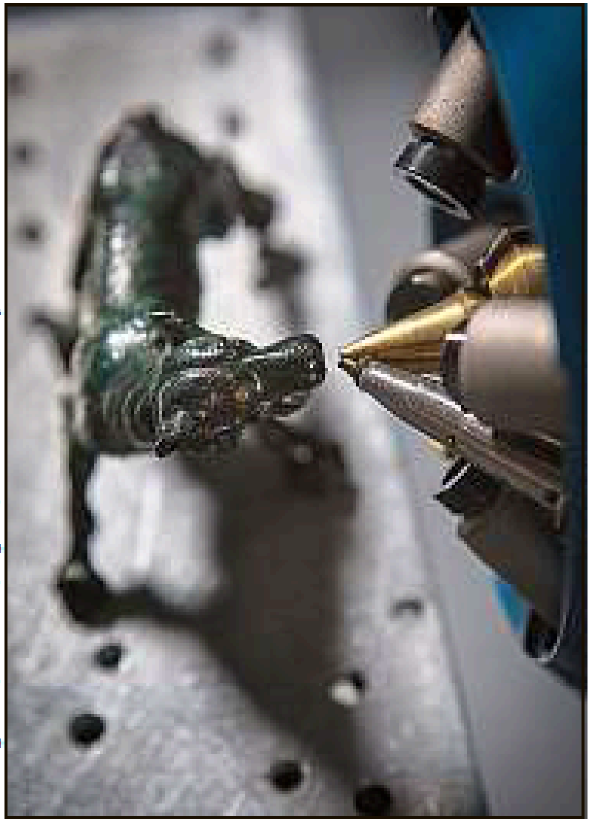
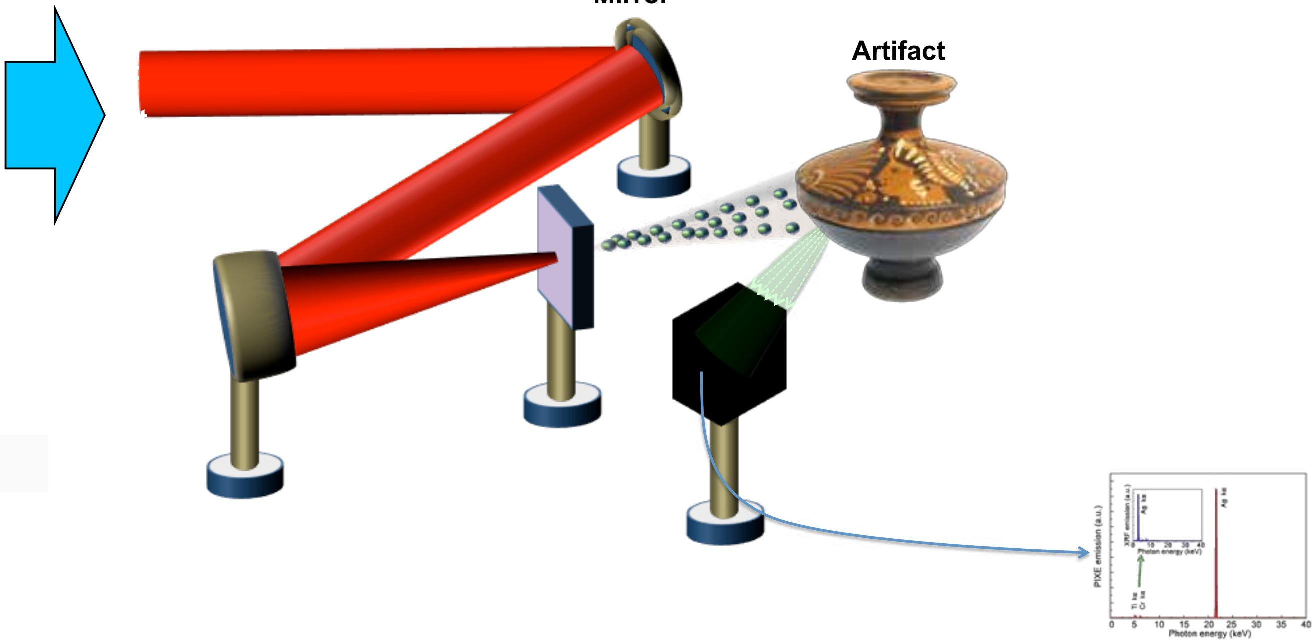
C. Hargoues/ C2RMF / Aglaé / CNRS Photothèque



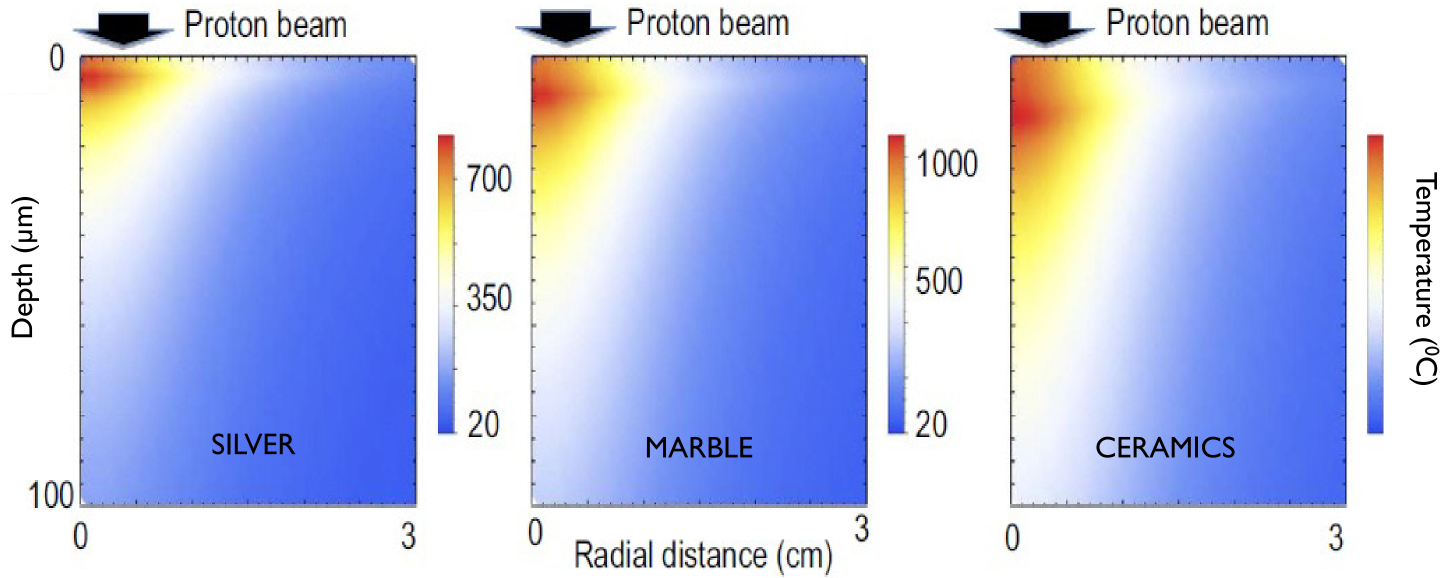
Une statuette en bronze du forum de Bavay analysée avec Aglaé.



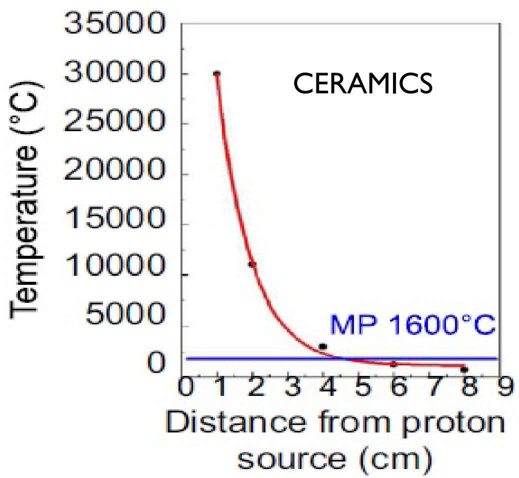
Numerical simulations to verify material sample heating (don't melt it!)

Typical CH materials:

Silver/Gold/
Bronze
Marble
Ceramics
Paper

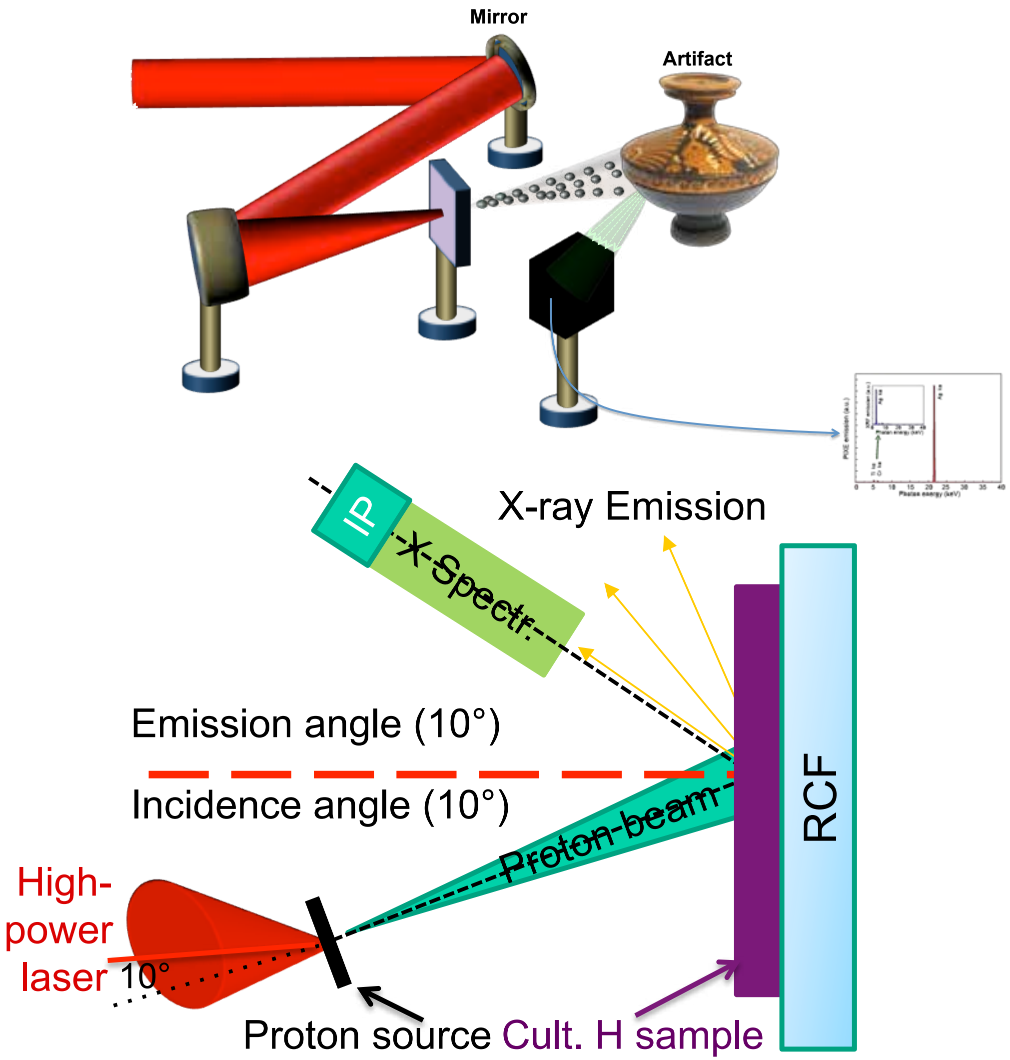


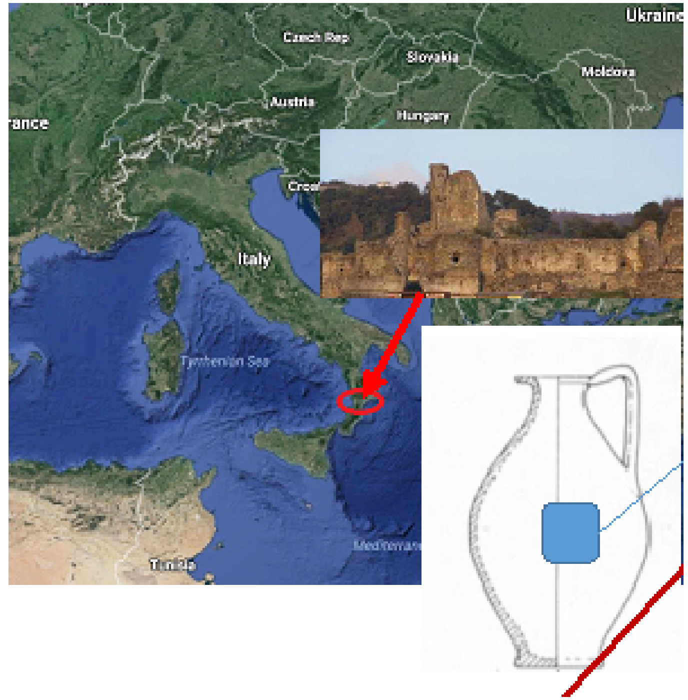
Energy deposition code simulating the TITAN spectrum (distance=6 cm)



- Temperature below meting point
- "Safe" distance in the >5 cm range
- Bunch duration in the ns range, cooling phase in the ns range

Experimental setup: first test the damage on the most sensitive material in the CH





Ceramics artifact from AD 1650 (archeological situ of Nicastro), provided by the Ministry of Culture in collabration with the Regione Calabria

Investigation of damage induced in a ceramic sample

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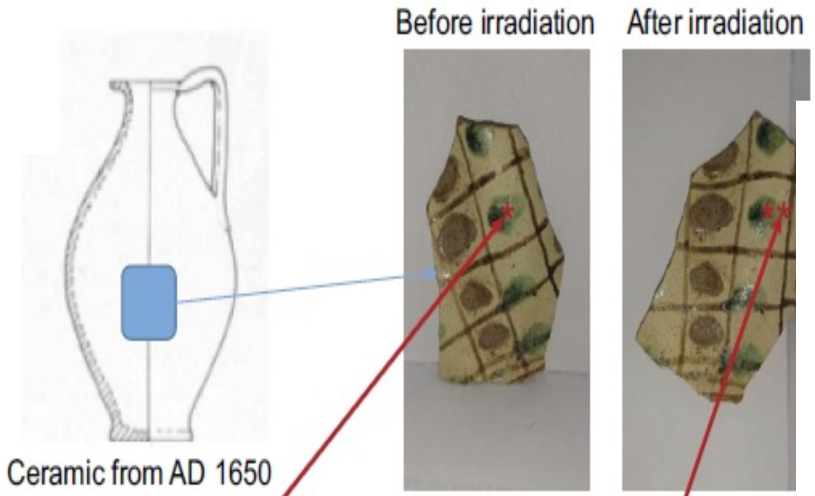
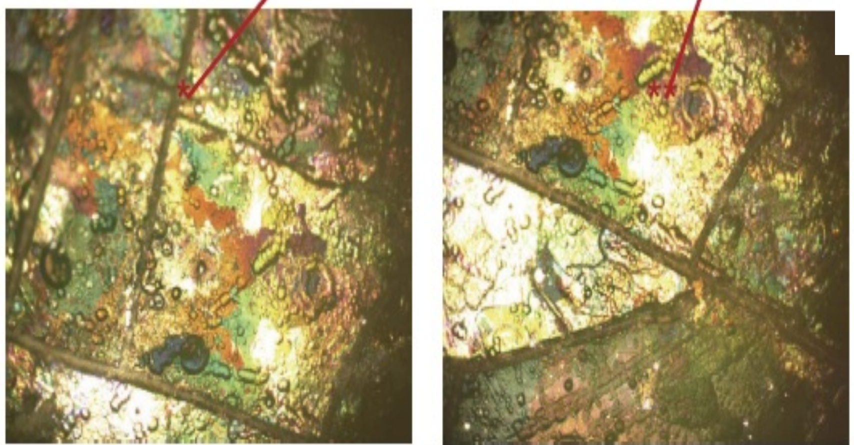
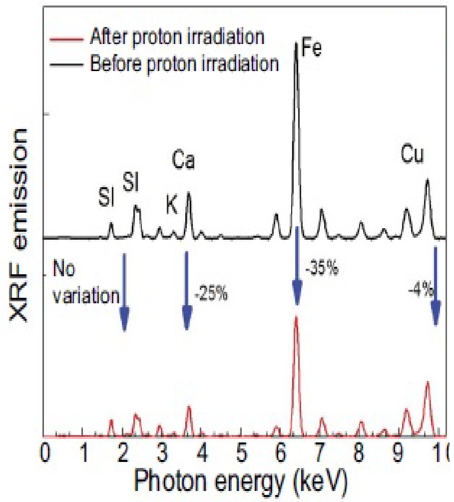


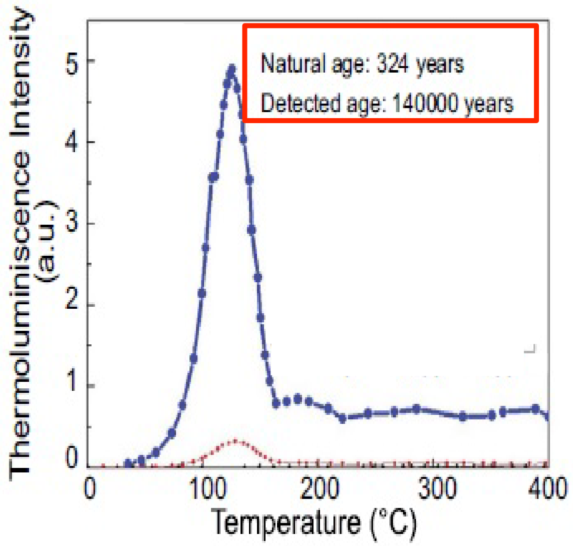
Morphological analysis: (100x mag.)



No aesthetical change after irradiation

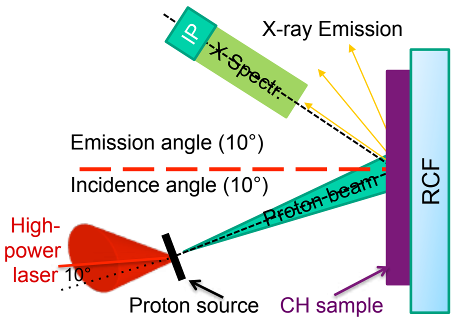
Chemical analysis: Conventional XRF spectroscopy and Thermoluminescence

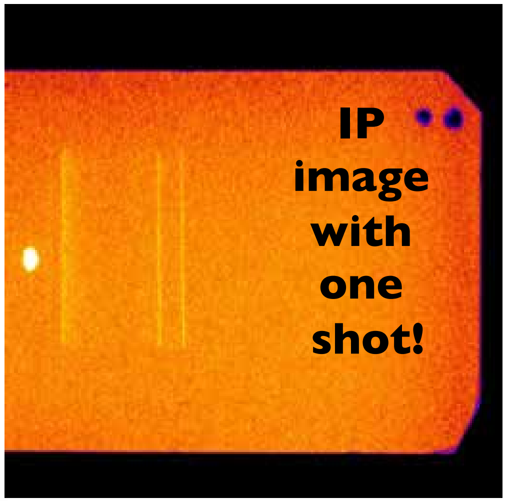




- XRF: no chemical modifications of the irradiated sample
- Absorbed dose alters the thermoluminescence of the materials: the age of the sample is artificially increased

X-ray spectrum analysis of a Silver sample with Bragg-spectrometer

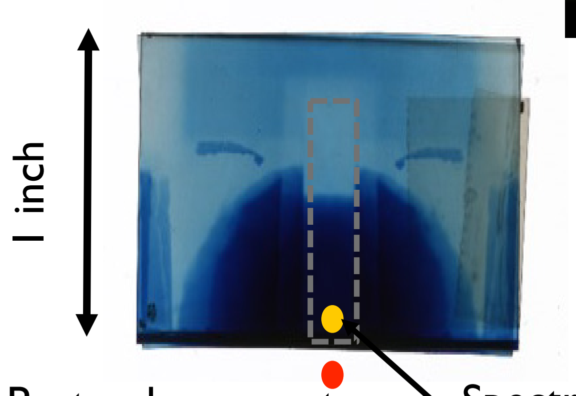


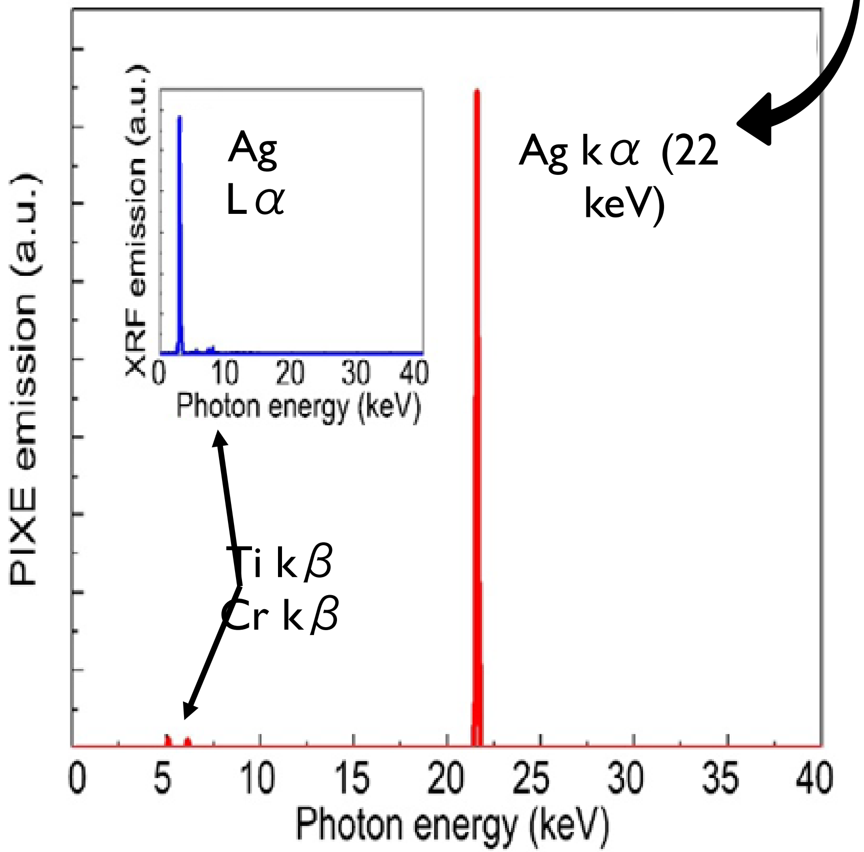


Bragg's law

$$\lambda = 2d \sin \frac{\vartheta}{2}$$

$$E = \frac{hcR}{2dx}$$

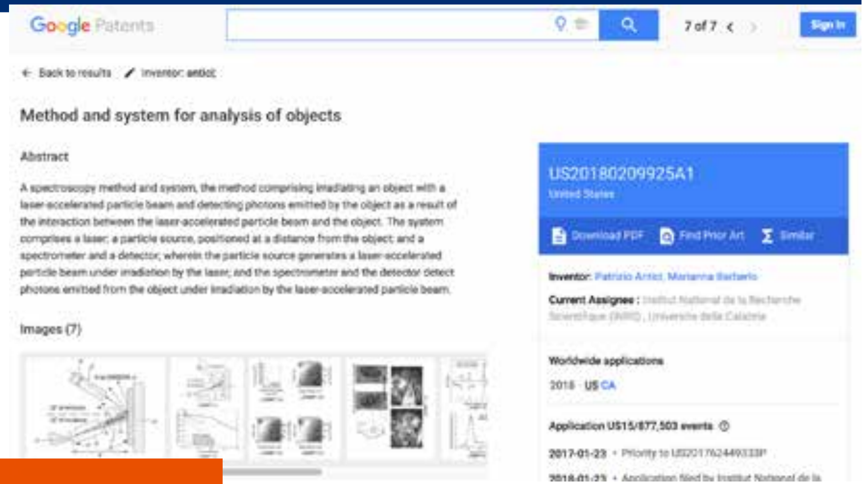


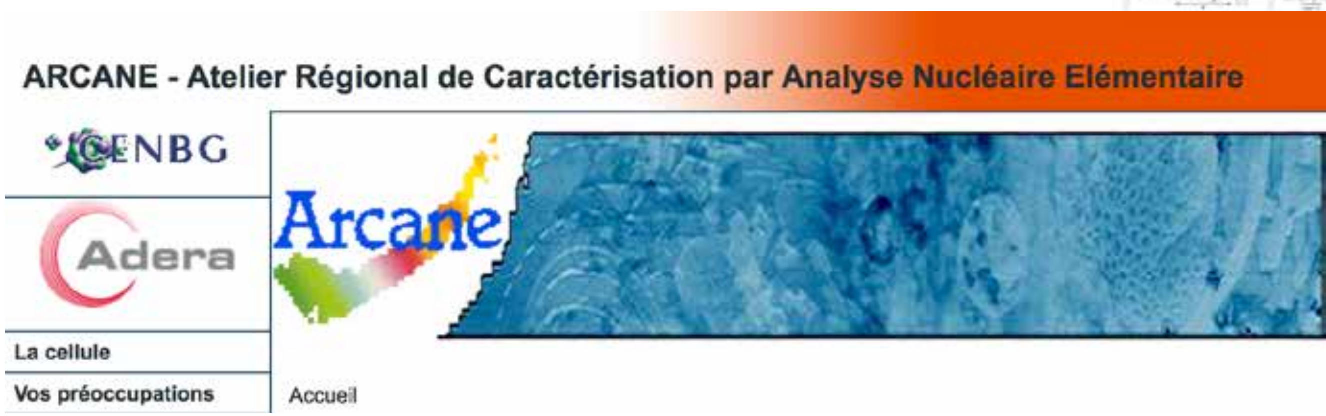


- Chemical composition of the sample successfully retrieved (Ag 97%, impurities of Ti, Cr and Cu)

Our new client: ARCANE@CENBG/Louvre & Biomed

M. Barberio, S. Veltri, M. Sciscià and P. Antici, Sci. Rep. 7, 40415 (2017)





- Laser-driven proton acceleration produces routinely energies 1-11 MeV, is this useful - can you do volumetric testing ?
- What materials can be analyzed ?
- Can you go beyond 10 ppm ? How quick is the analysis ?

We are not alone !

ELI Beamlines and its new project: Non-Destructive Laser-driven Heritage Testing

Institute of Physics of the Academy of Sciences of the Czech Republic / E
a new project called Non-Destructive Methods of Heritage Testing , fina
(Pól rüstu ČR, the City of Prague). Researchers from ELI-Beamlines work
Acceleration by Laser, and currently commissioning the ELIMAIA user be
the project by the Institute of Nuclear Physics in Řež and Istituto Nazion
Nazionali del Sud (INFN LNS) in Catania. INFN ranks among the best in
and restoration of monuments, but with a conventional particle acceler
protons will be worldwide unique for the combination of the planned a

Intense Laser Irradiation Laboratory

National Institute of Optics – National Council of Research

CNR INO
CONSIGLIO NAZIONALE DELLE RICERCHE
ISTITUTO NAZIONALE DI OTTICA

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Laser-PIXE

Laser driven Particle Induced X-ray Emission:
source development and X-ray spectral/spatial analysis

The PIXE (Particle Induced X-ray Emission) is a multi-elemental, quantitative, highly s
ments with concentrations in parts-per-million), rapid, non-invasive and non-destructive
to determine the composition of surface layers of a sample. The technique is based on s
characteristic X radiation emitted by each atom following irradiation with proton/ion bea
protons with MeV energies (2-3 MeV typically) that are currently produced with particle a
dimensions (and costs) represent a limit for the application of the same technique outside o

ENSURE

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Exploring the New Science and engineering unveiled by Ultraintense ultrashort Radiation interaction with mattEr

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PEOPLE

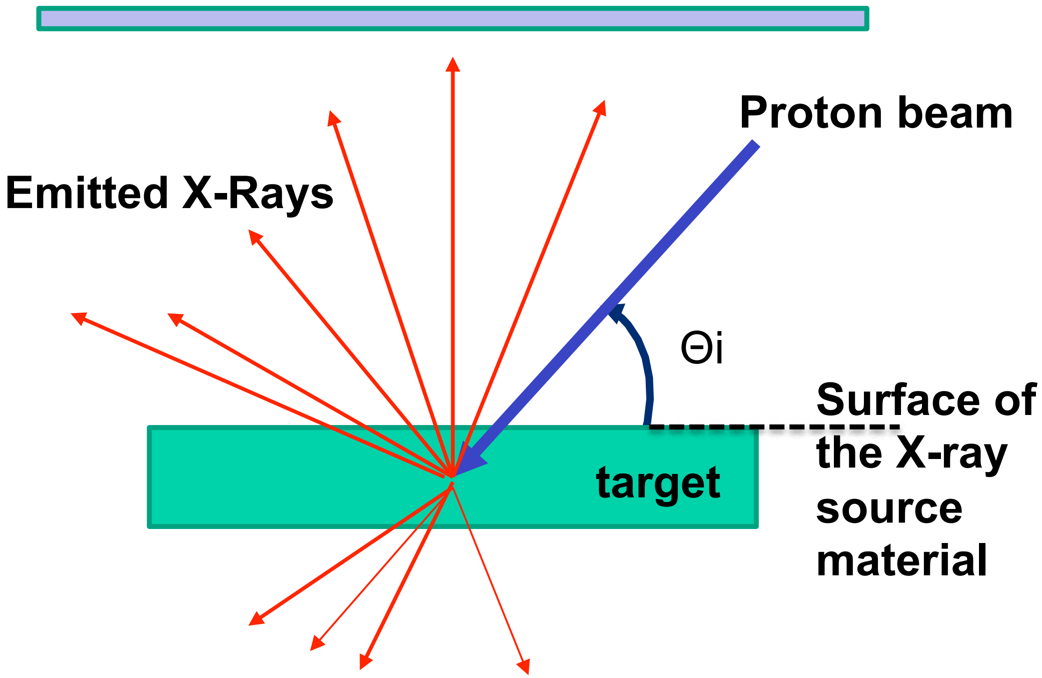
CORE TEAM

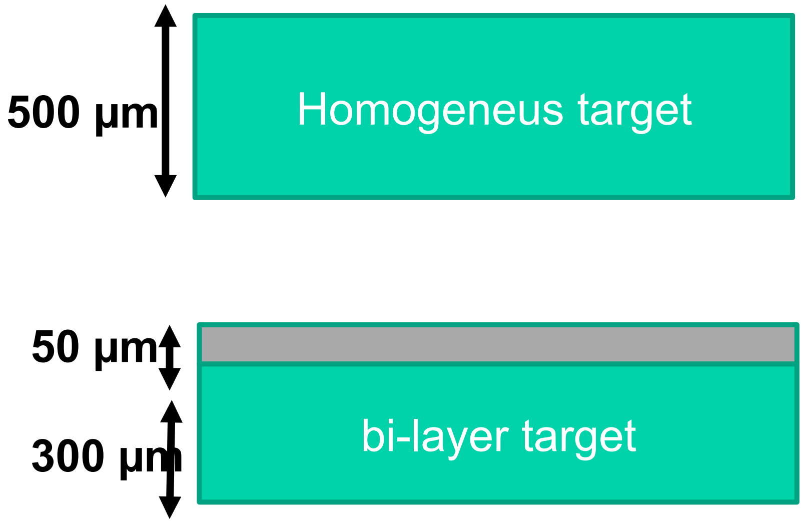
PRINCIPAL INVESTIGATOR

Matteo Passoni

Optimization of laser-PIXE using GEANT4

CCD DETECTOR (4 cm x 4 cm x 250 µm)
(solid angle ~0.81 str)

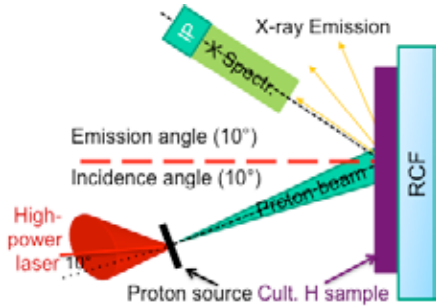




GEANT4 with the package G4ParticleGun, cylindrical symmetry, number of particles between 10^4 and 10^{13}. The proton beam diameter = 1 µm (typical applications)

M. Pia, et al, PIXE simulations with Geant, *IEEE Trans. Nucl. Sci.*, **56**, 3614 (2009)

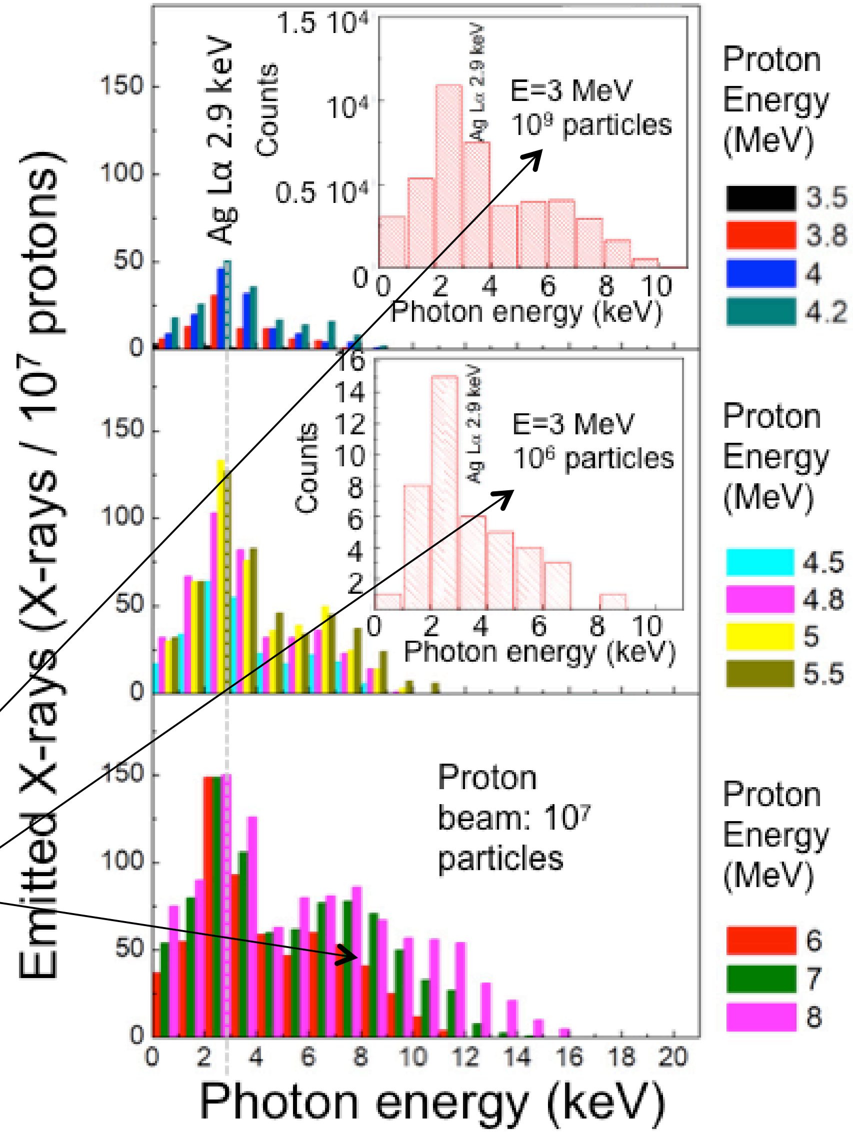
Reproducing the experimental results on a silver sample



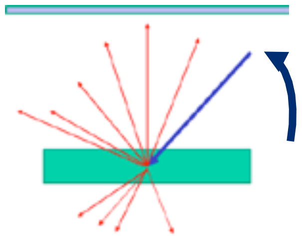
Efficience 10^{-4} photons/proton
About 10^{13} protons/MeV/str @ 3 MeV
->10^{-9} photons / str - close to experimental value

Very high Bremsstrahlung
Energies above 5 MeV of little interest

Ratio is independent from particle number

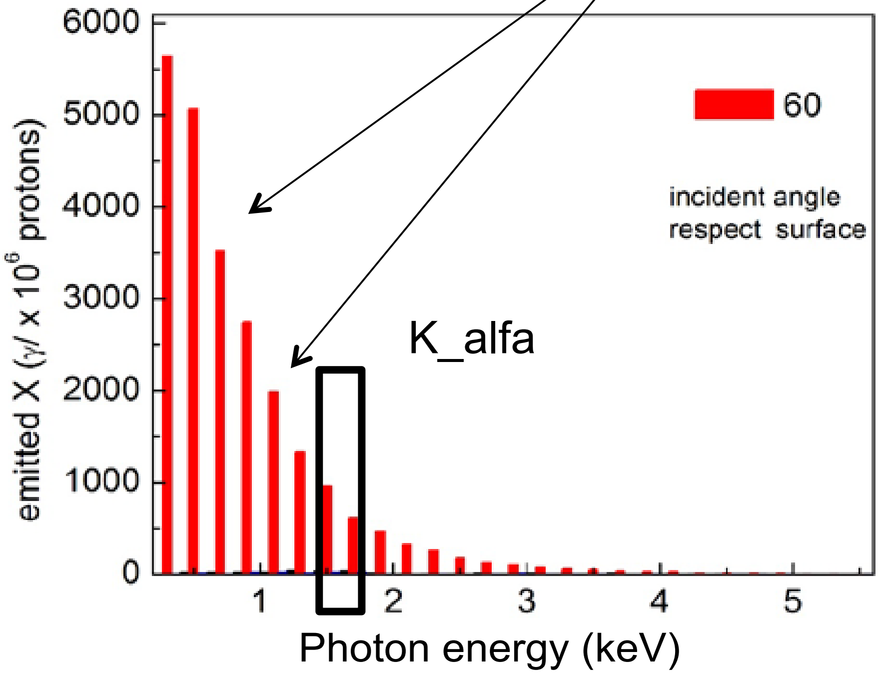


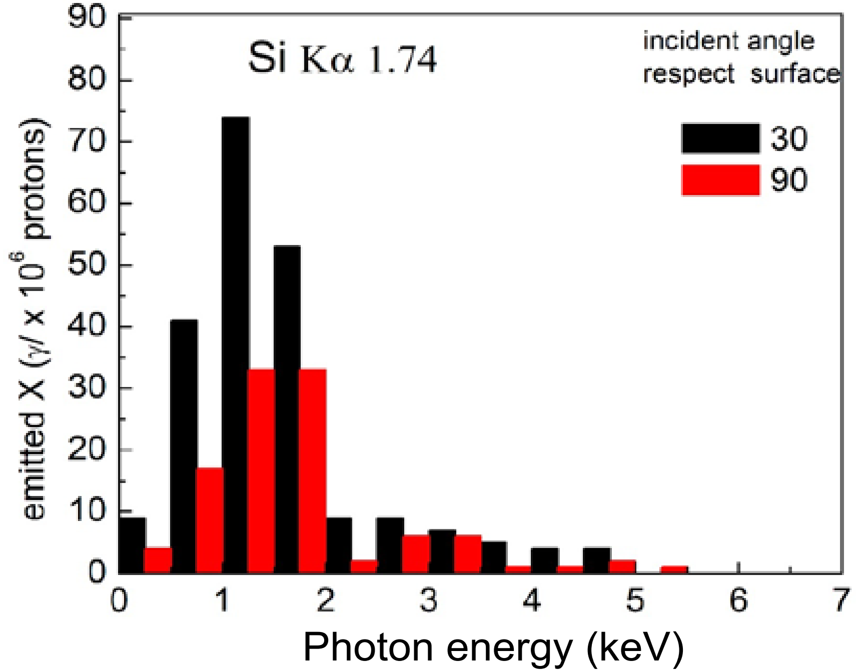
Optimization of the incident angle: 60 degrees incident angle favors Bremsstrahlung for 3 MeV incident protons on SiO_2



Optimized angle for the detection of the K_alfa is around 30°

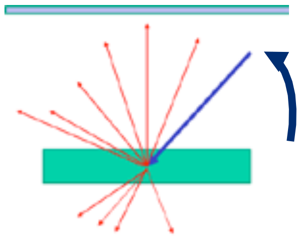
Significant Bremsstrahlung

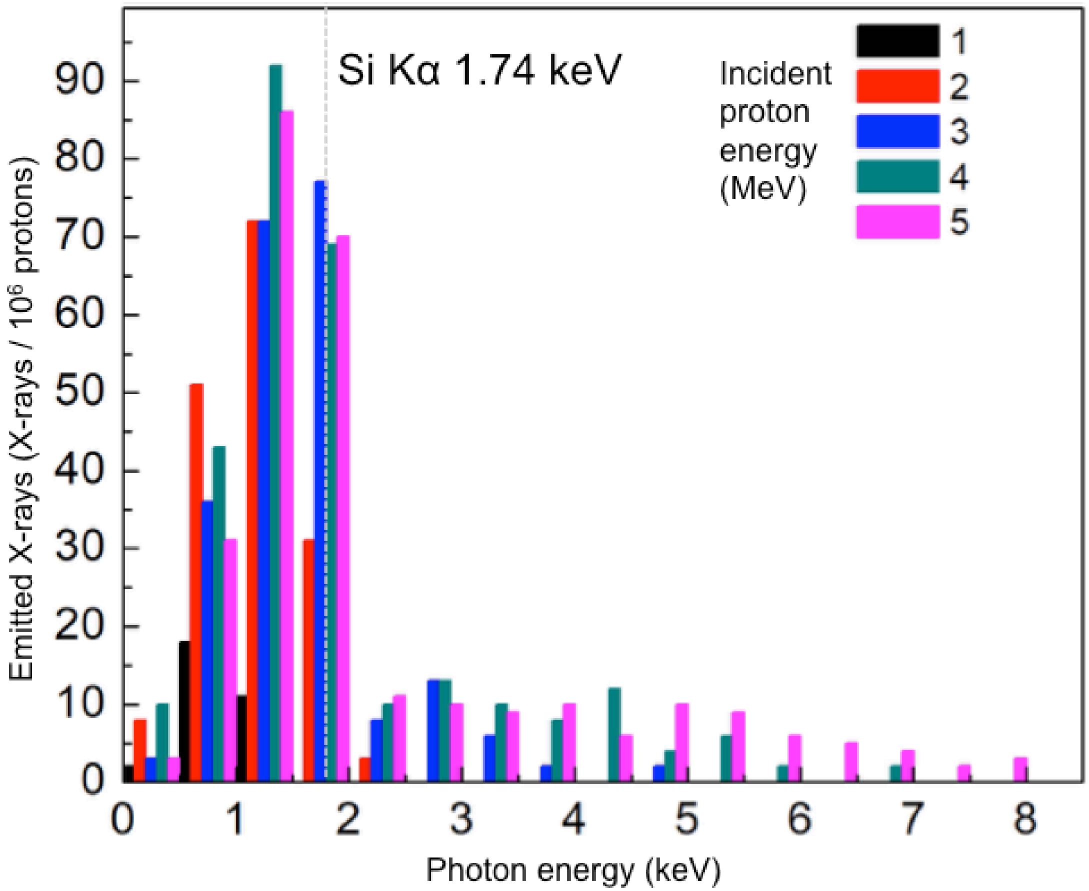




One of the most analyzed materials: Ceramics (SiO_2), Mean energy 3 MeV, 10^6 protons, 3 cm distance

30° incident angle better also for other incident energies

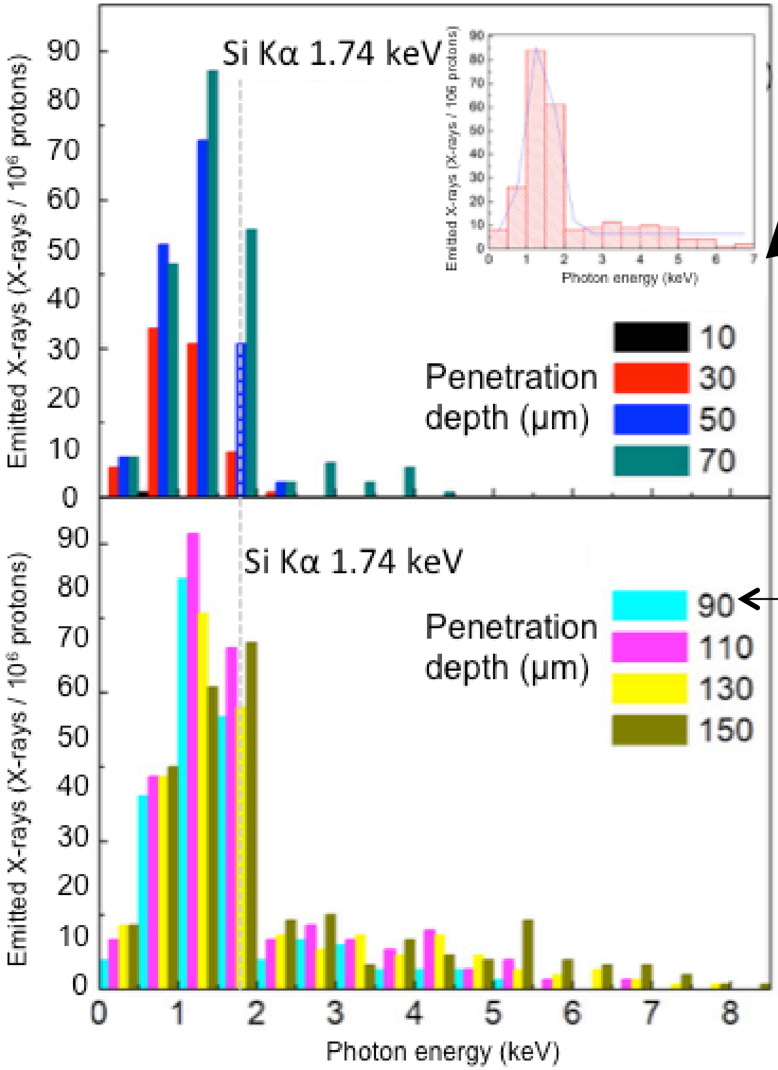




In the following we chose 30° as best incident angle !

One of the most analyzed materials: Ceramics (SiO_2), different mean energies, 10^6 protons, 3 cm distance

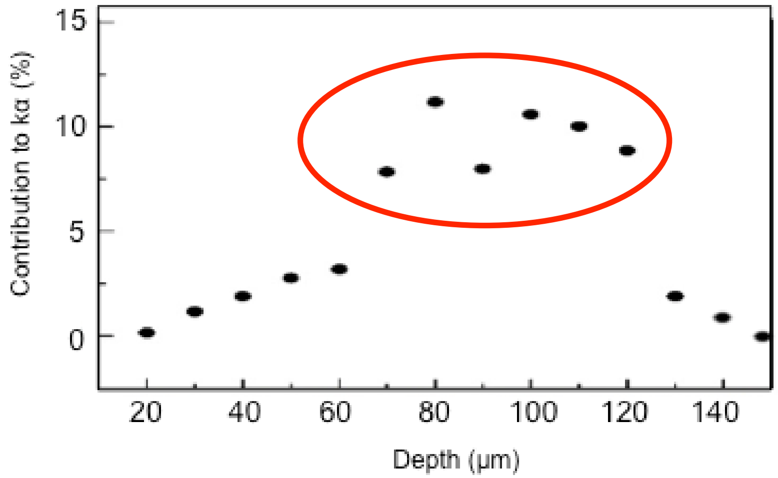
Contribution of the different depth to the total X-ray production



Bin size = 0.5 keV

3 MeV

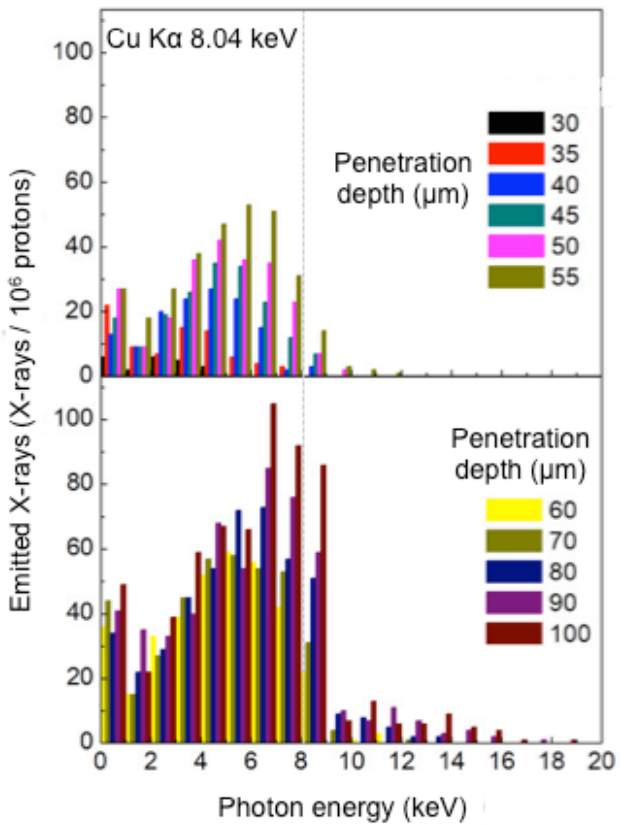
Bin size = 0.02 keV

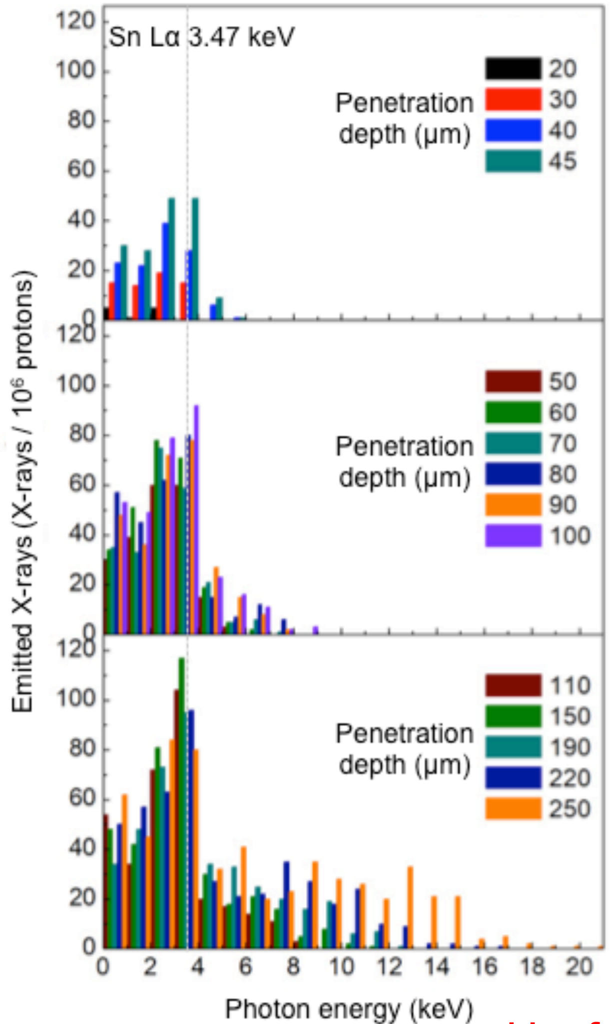


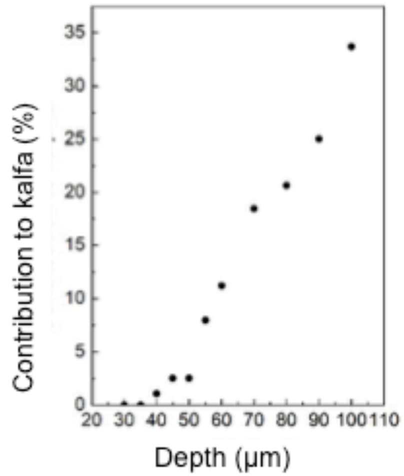
Useful region: ~2.5 – 4 MeV

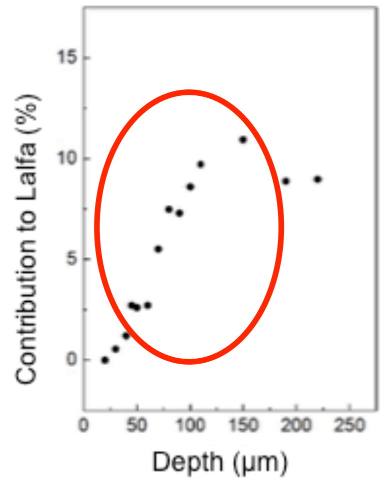
One of the most analyzed materials: Ceramics (SiO_2)

We verify this on a bronze alloy (material which consists of 90 % Cu and 10 % Sn)









Useful region: <~ 6 MeV

How does this compare with other Multi-layer targets ??

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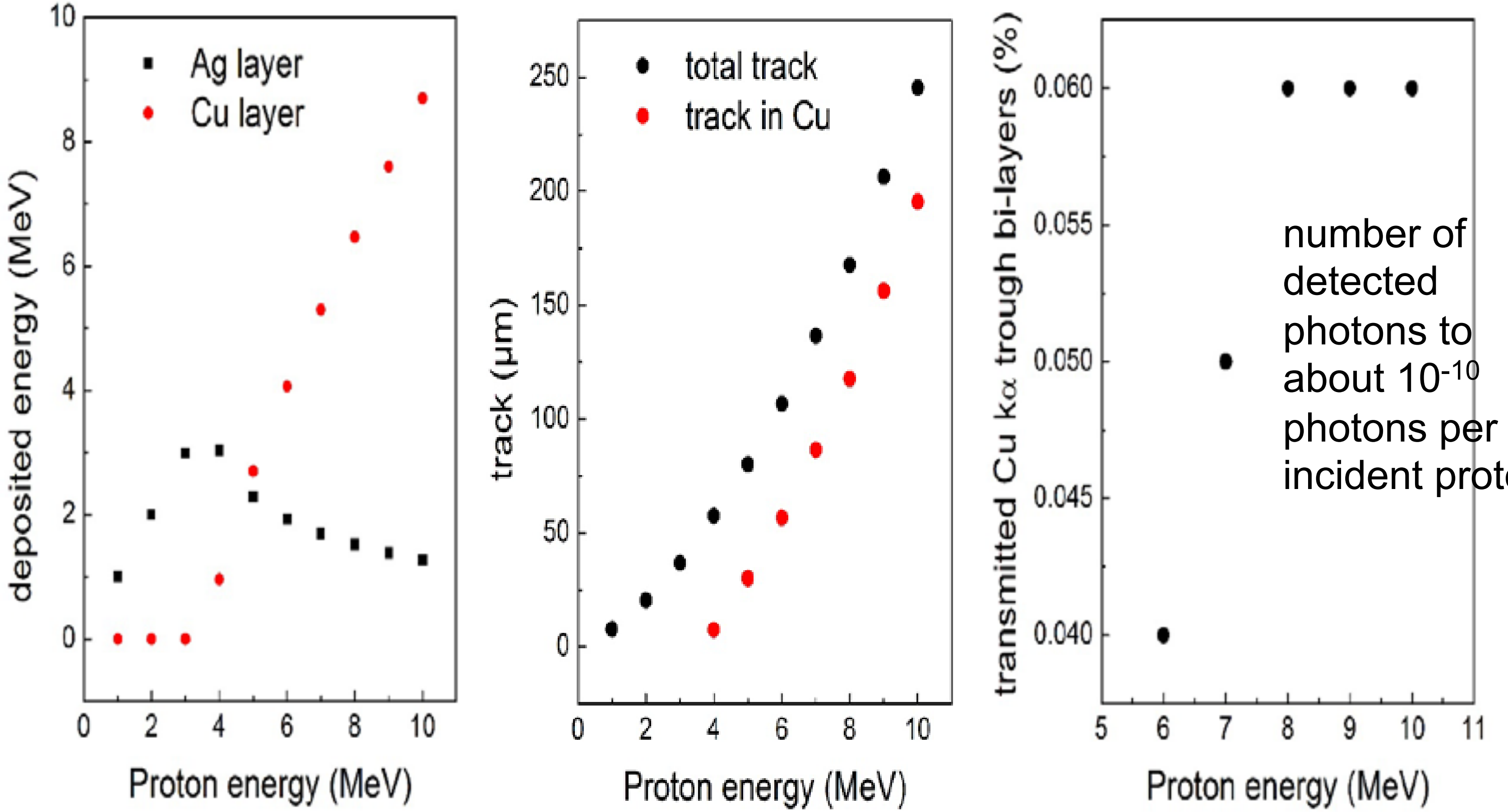
Ag(50μm)//Copper(300μm) multilayer sample



number of detected photons to about 10^{-10} photons per incident proton

For other multilayer targets ?

- For example Pb//SiO_2 or Ti//SiO_2) the transmittance of photons generated in the SiO_2 layer is extremely low (close to zero) due to the very low energy of the SiO_2 K_alfa photons.

- Multilayers consisting of Pigments//Ceramic and Pigments//Canvas, simulated as a PbO layer on SiO_2 (or $CaCO_3$ for the canvas), or $Ti0_2$ layer on SiO_2 is the same.

Laser-PIXE very difficult for multilayers with $K\alpha$ < 6 keV…

M. Barberio et al., Submitted

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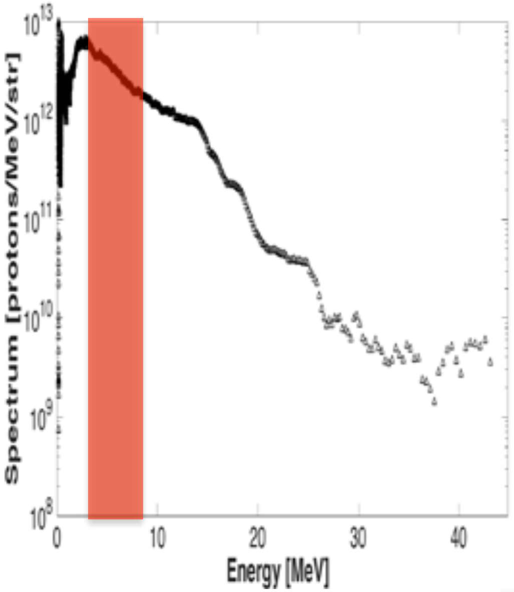
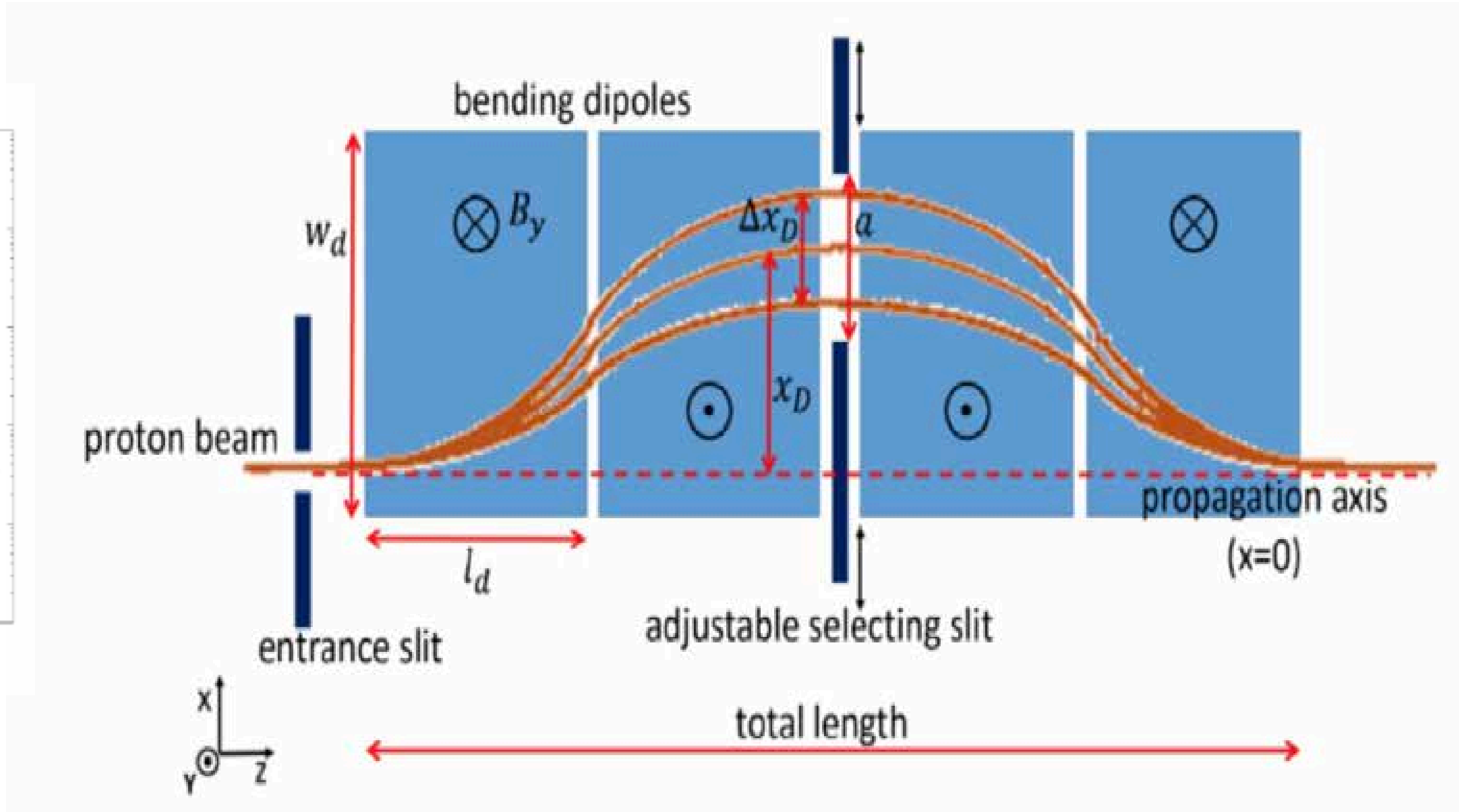
Laser-PIXE very difficult for K_alfas < 6 keV…

Adding layer-by-layer analysis





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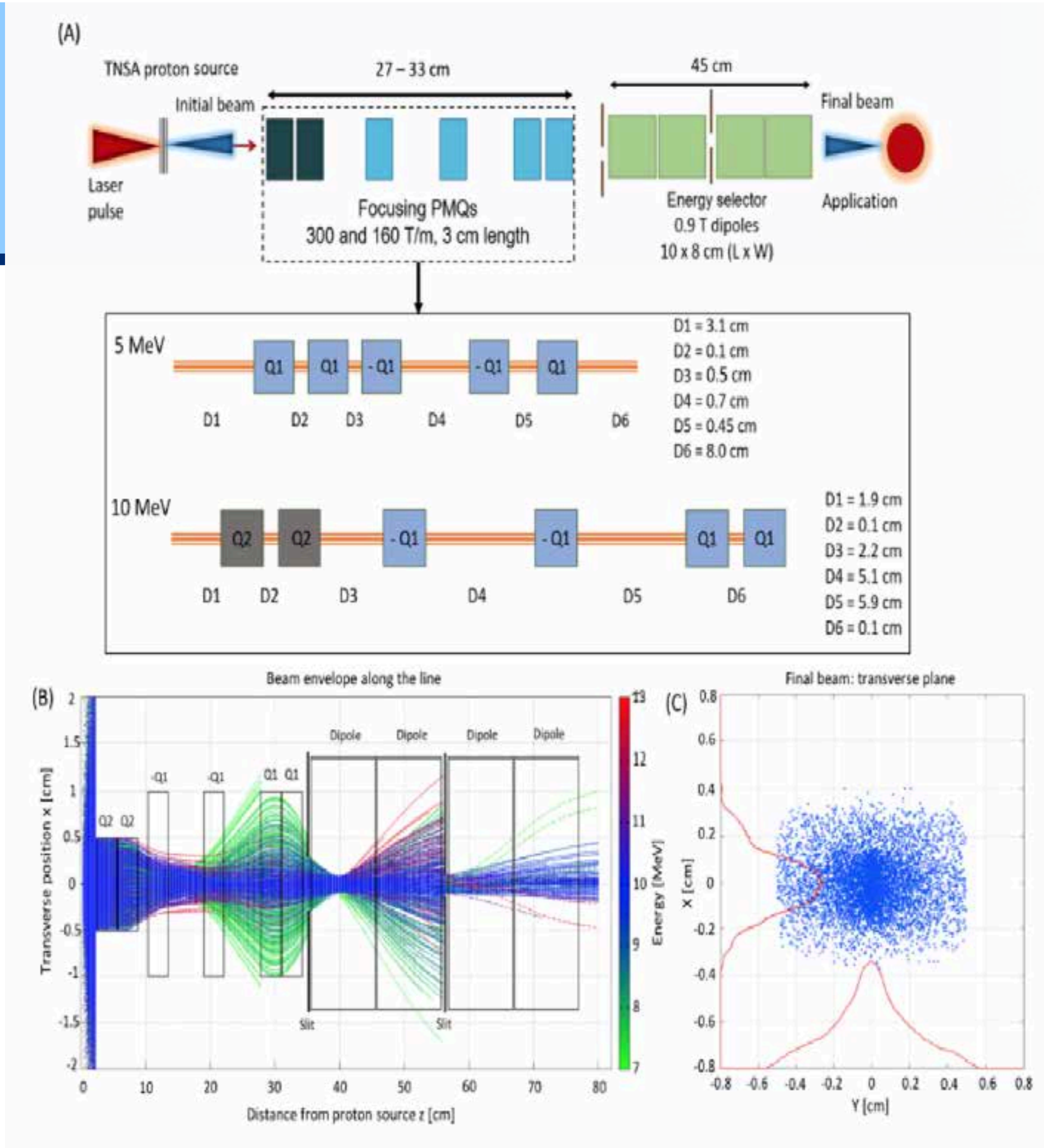
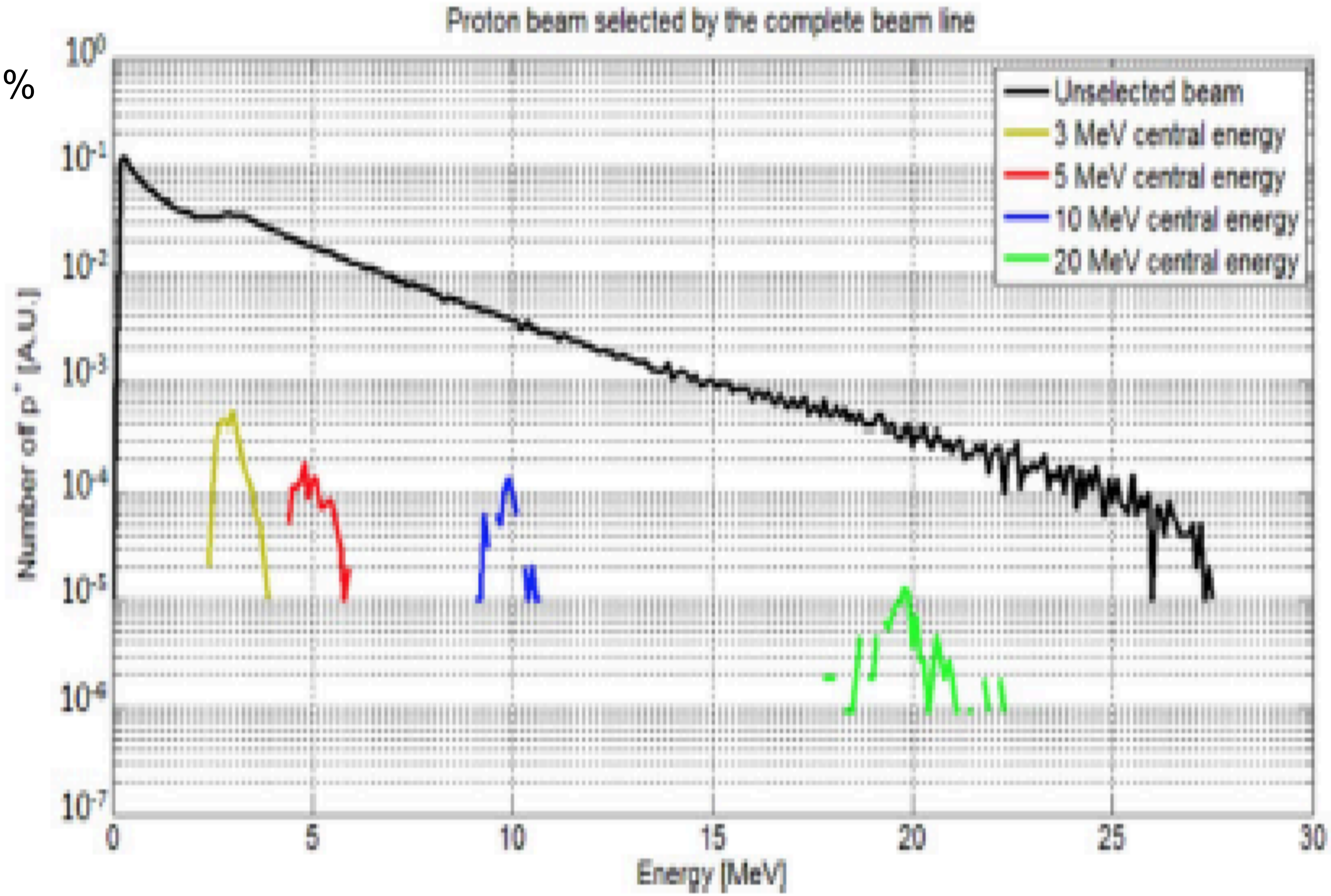
Potential beamline design optimizing protons around 5 & 10 MeV with energy spread of 10 %.

Q1 and Q2: 160 T/m and 300 T/m



Efficiency of an Energy Selector ?

About 1 %



Application to CH

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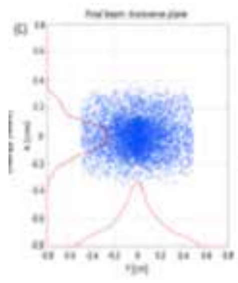
- Novel Ti:Saphire laser systems operate at multi-Hz repetition rate and generate proton bunches in the nC range (~10^{11} protons/MeV for a mean energy of 3 MeV).
- Choosing an energy spread of FWHM 20 % (3 MeV ±30 keV) leads to a penetration depth of about 36 ± 6 μm into a Silver sample
- With an overall transmission efficiency of η = 1.2%, a final beam charge of ~7.2 10^8 protons/bunch, i.e. ~ 0.12 nC/bunch.
- At repetition rate of 10 Hz allows irradiating the sample with a proton flux of 1.2 nC/s, scanning an area of about 10 mm^2


SCIENTIFIC REPORTS

OPEN Design and optimization of a compact laser-driven proton beamline

M. Scisciò, M. Migliorati, L. Palumbo & P. Antici

M. Scisciò et al., Sci. Rep. 8, 6299 (2018)



Thanks to the entire teams

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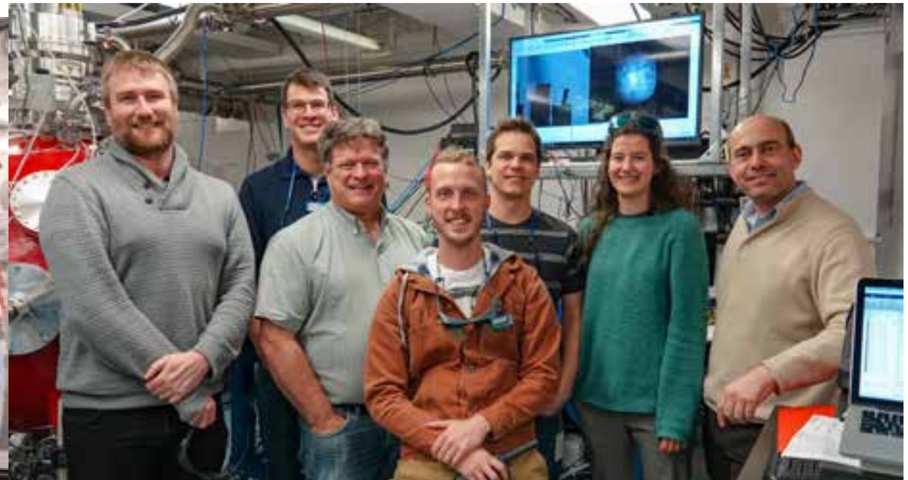

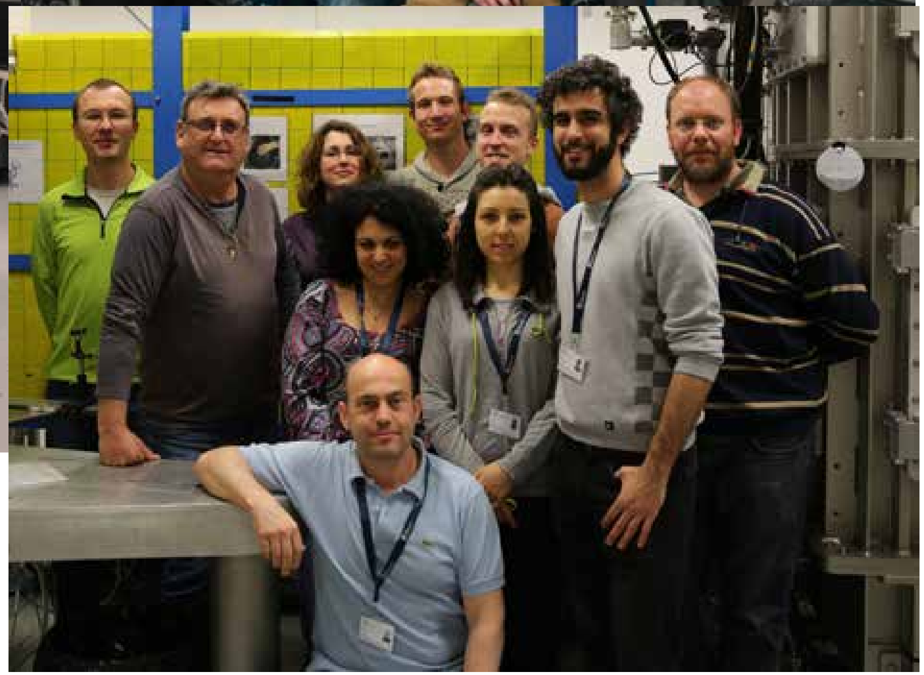

Interested ?
…collaborate or join us !

Thank you for your attention !