EMIR&A user's meeting 2025

ANR-21-ESRE-0050

Development of mitigation strategies for safer IBA of fragile heritage artefacts

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[3] Fondation des Sciences du Patrimoine, Paris, France

NewAGLAE

external microbeam beam

AGLAE : 2 MV 6-SDH-2 Pelletron tandem accelerator

outline

1 *Challenges*

- preserve heritage objects
- complex / unknown composition
- unpredictable behavior

2 *PIXXL*

- special accelerator beamline
- new scanning protocols

3 *MONIONS*

- real-time monitoring of target
- chemical, optical & thermal effects

1

Strategies to mitigate modifications

International Atomic Energy Agency

- working groups (TMs, Paris 2015, Amsterdam 2017)
- Round-robin program on painting materials

European heritage science networks

- Joint Research Activities in IPERION european programs

Keypoints

- **identify** sensitive materials
- **understand** and predict beam-induced modifications
- **establish** safe radiation exposure limits
- **lower as much as possible** exposure (ALARA principle)
- **optimize** beam conditions and detection
- **develop** processing of low statistics (frugal) spectra
- **monitor** the object under the beam

L. Bertrand, S. Schöder, I. Joosten, S. M. Webb, M. Thoury, T. Calligaro, E. Anheim, A. Simon, Practical advances towards safer analysis of heritage samples and Objects, Trends in Analytical Chemistry 164 (2023) 117078

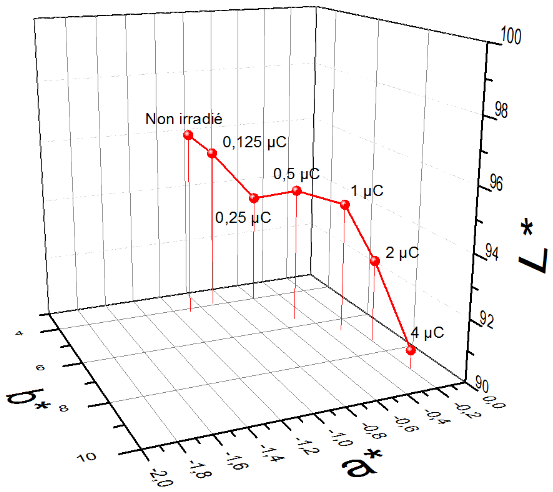
Identify sensitive materials

lead white paint

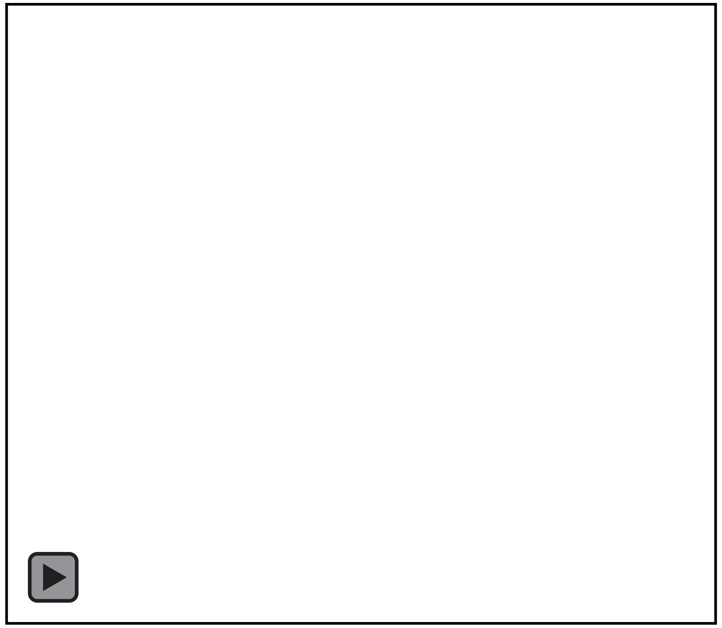
lead carbonate + organic binder

Darkening

Colour vs beam dose



T. Calligaro, V. Gonzalez, L. Pichon,
PIXE analysis of historical paintings: Is the gain worth the risk? NIM B363 (2015) 135–143



Old painting 80 µC/cm^2 3-MeV ^{1}H

Lead white + oil binder 40 µC/cm^2

18th c. painting 40 µC/cm^2

understand beam effects : absorbed dose

absorbed dose of radiation per unit of mass

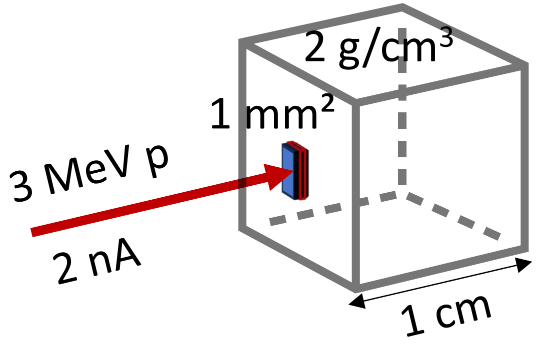
D ≡ dE/dm in **Gray (Gy) [J/kg]**

D varies with:

- fluence Φ (μC/cm^2)
- ion type (p, d, He)
- incident energy E
- target composition
- depth z

Louis Harold Gray 1905-1965

D for standard PIXE conditions



t = 200 s
Q = 0.4 μC
z = 0.1 mm
v = 10^{-4} cm^3

Φ = 40 μC/cm²

E = qV = 0.4 μC × 3 MV = 1.2 J
m = 0.2 mg = 2.10^{-7} kg

D ≈ E / m = 6 MGy

Calculate dose along beam trajectory

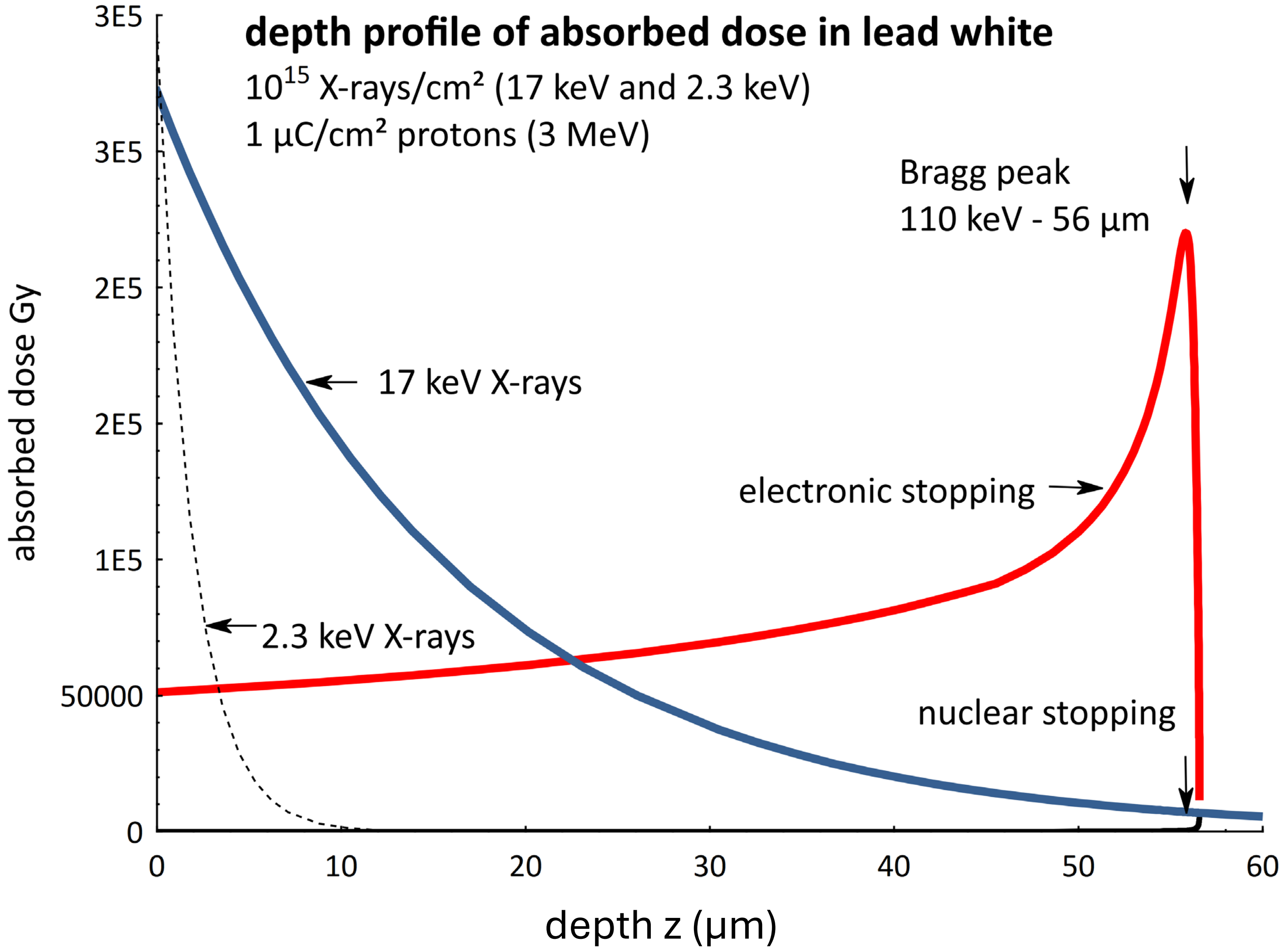
Absorbed dose D at depth z

$$\mathbf{D}\,(z) = 10^6\ \mathbf{S}/\rho\,(z) \times \mathbf{\Phi}$$

Φ [μC/cm^2]
stopping power S/ρ [MeV/mg/cm^2]

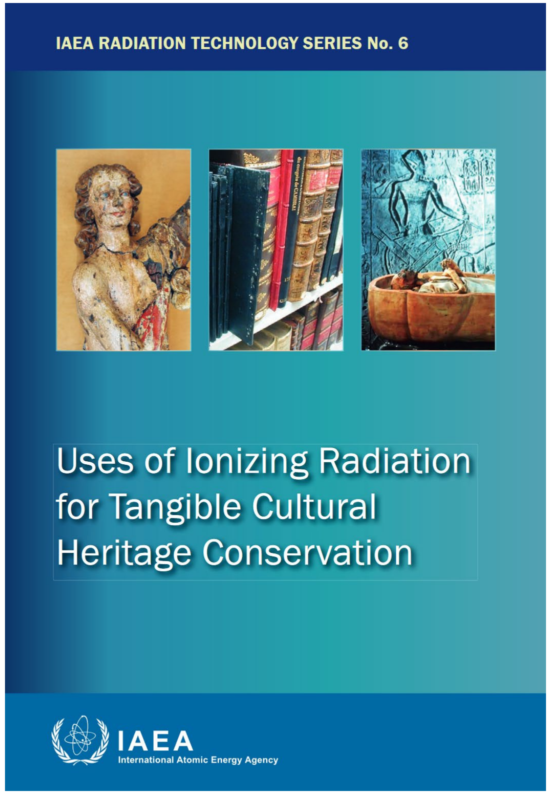
SRIM calculations

James F. Ziegler 1935-2024



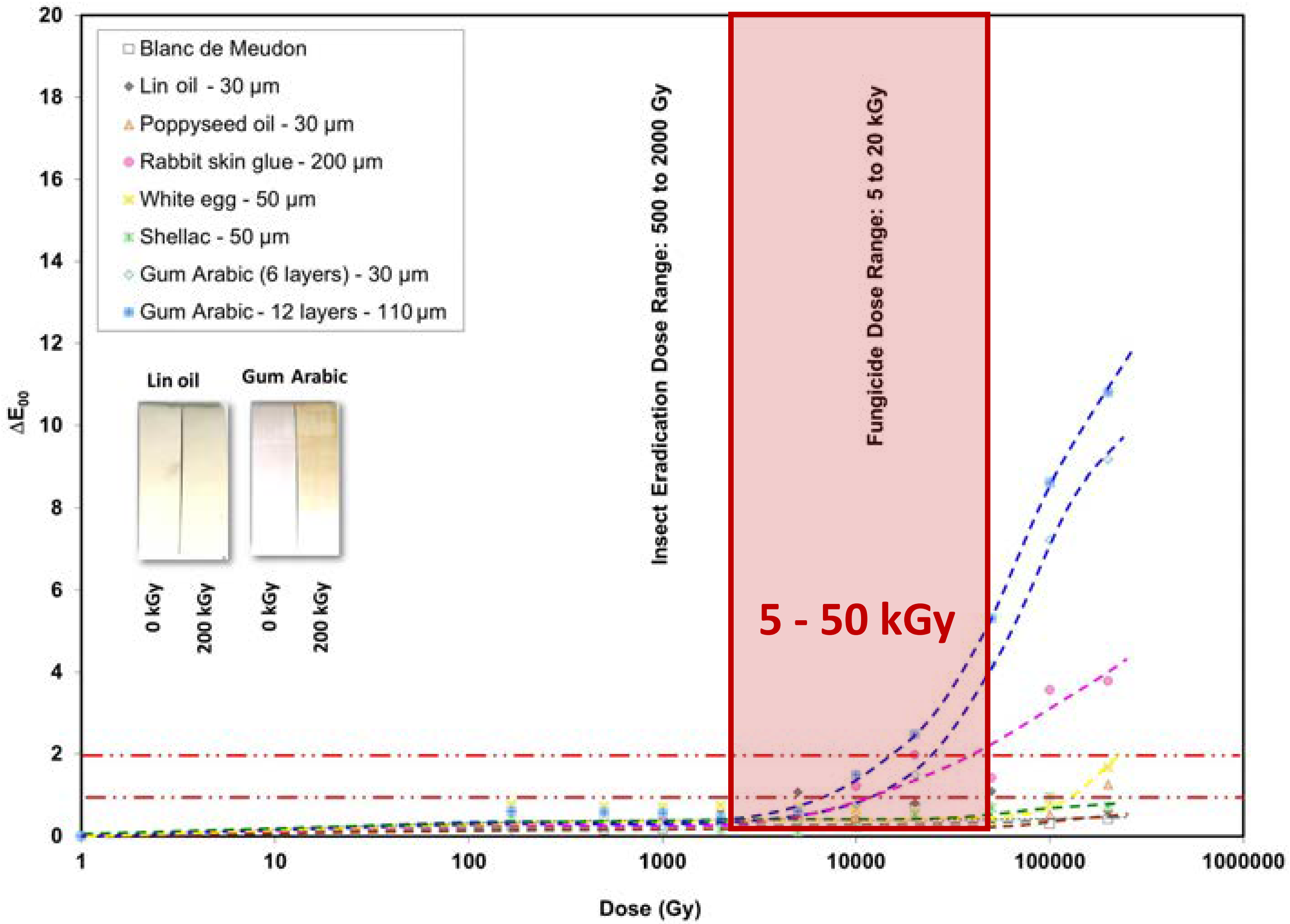
Safe Gy treshold : γ-ray treatment for heritage preservation

determined for fragile materials
case of paintings



www.iaea.org/publications/10937 *(2017)*

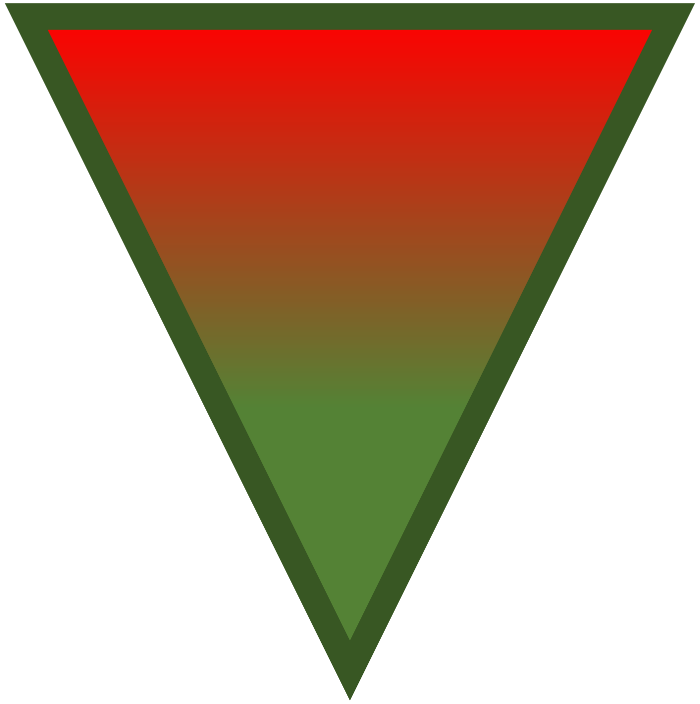
visual modifications (ΔE CIE La*b*) versus D



New paradigm for safer PIXE analysis

drastic Gray decay

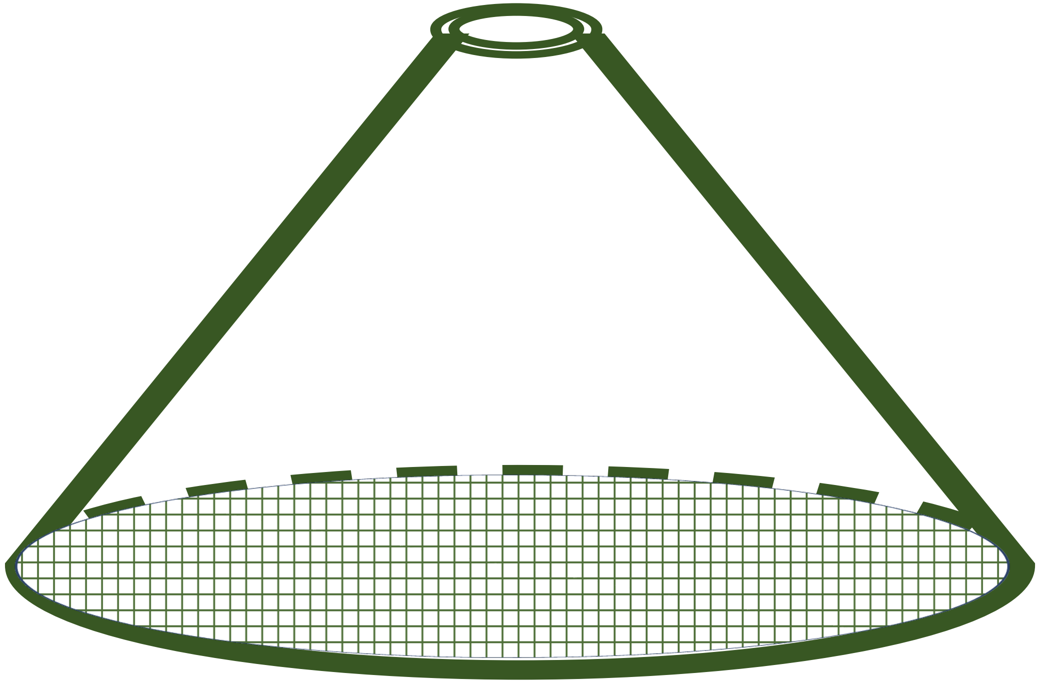
5 MGy



50 kGy

Φ downscale

40 μC/cm²



0.4 μC/cm²

PIXE XL

Particle Induced X-rays for Xtra Large objects

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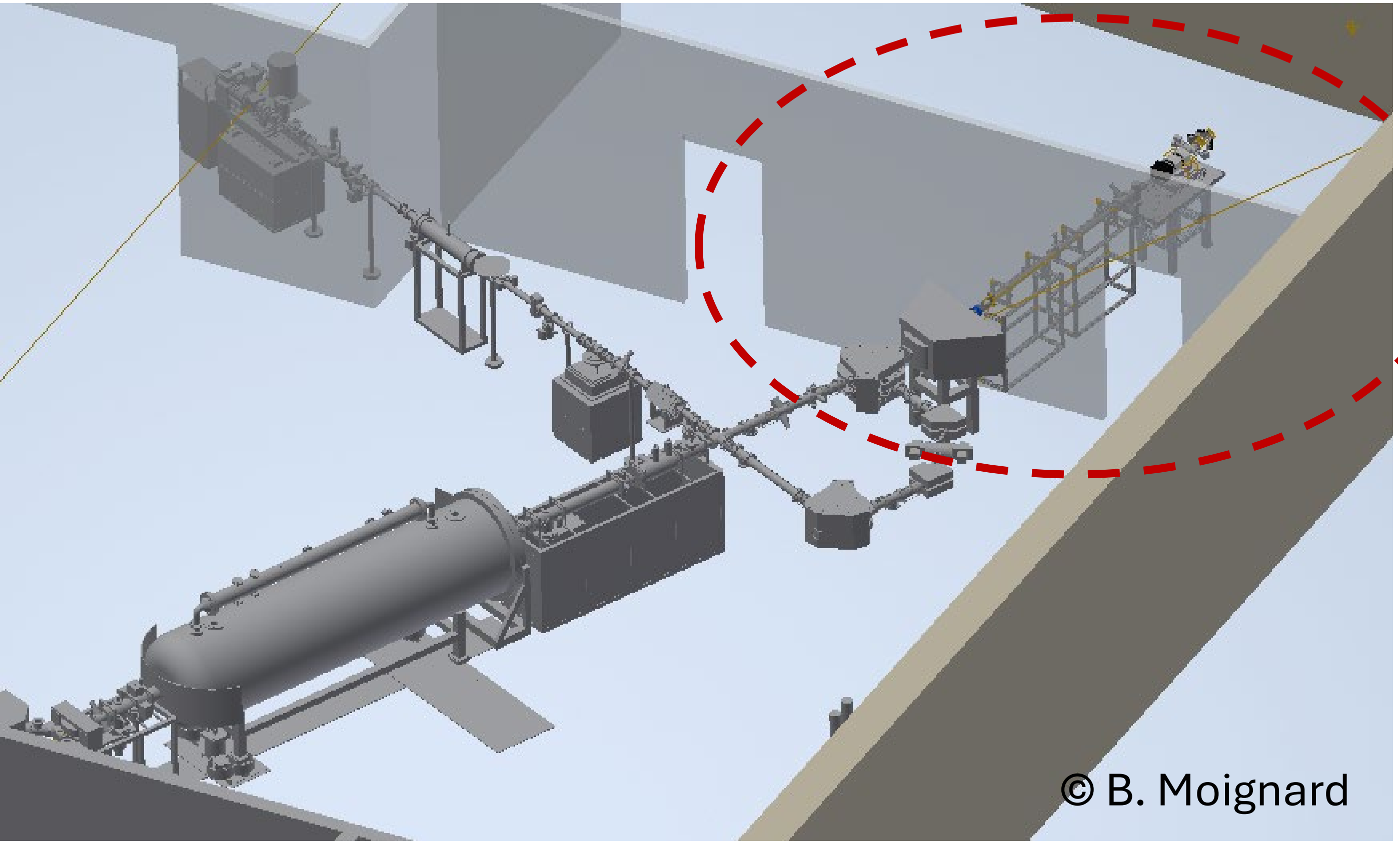
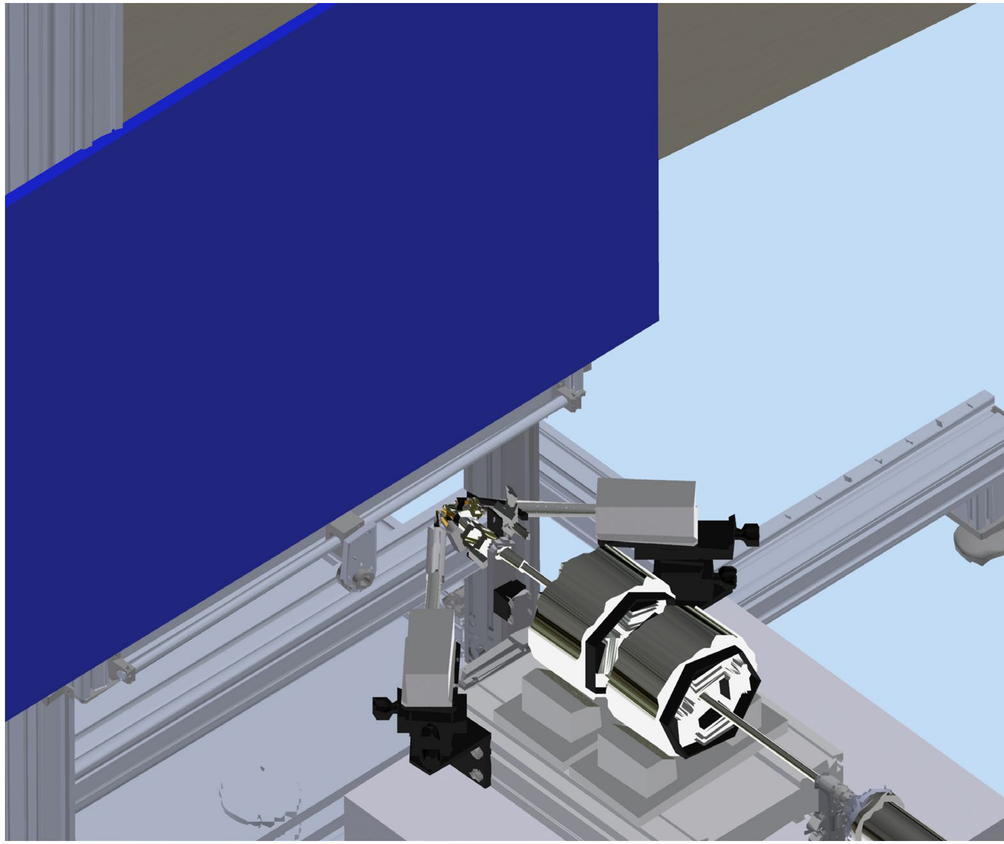
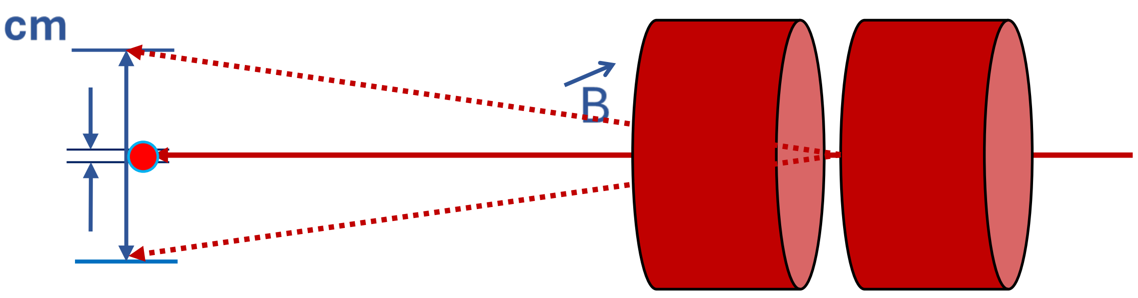




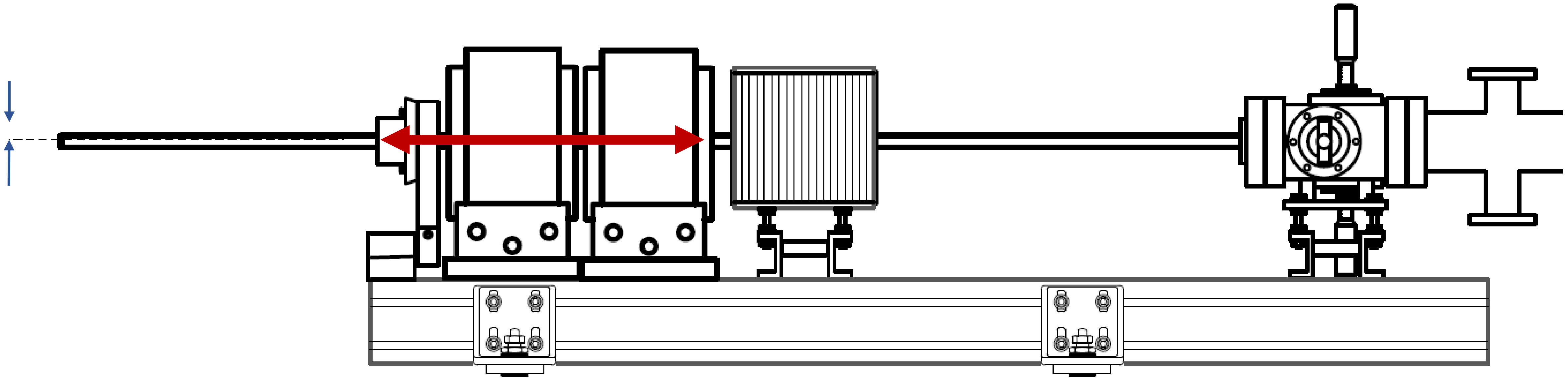
PIXE imaging of large surfaces: a paradigm shift

- dramatically upscale map size: mm → meter
- 200-µm beam ∅ → display all visible details
- keep exposure within safe limits → avoid modification
- record 25 M spectra in a single a scan
- acquire flash spectra (2 ms) → 12 hours total time
- maximize detection (Ω=1 Sr) → every X-ray matters
- process frugal spectra (few k counts)
- exploit huge number of low stat spectra using hyperspectral techniques

wide field beam optics



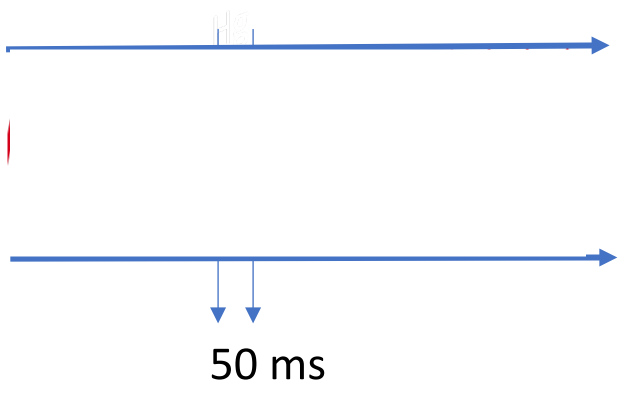
- Q-pole doublet
- long working distance (15 -> 50 cm)
- large beam scan (1.5 mm -> 10 mm)
- autofocus for non-flat objects (sliding doublet)

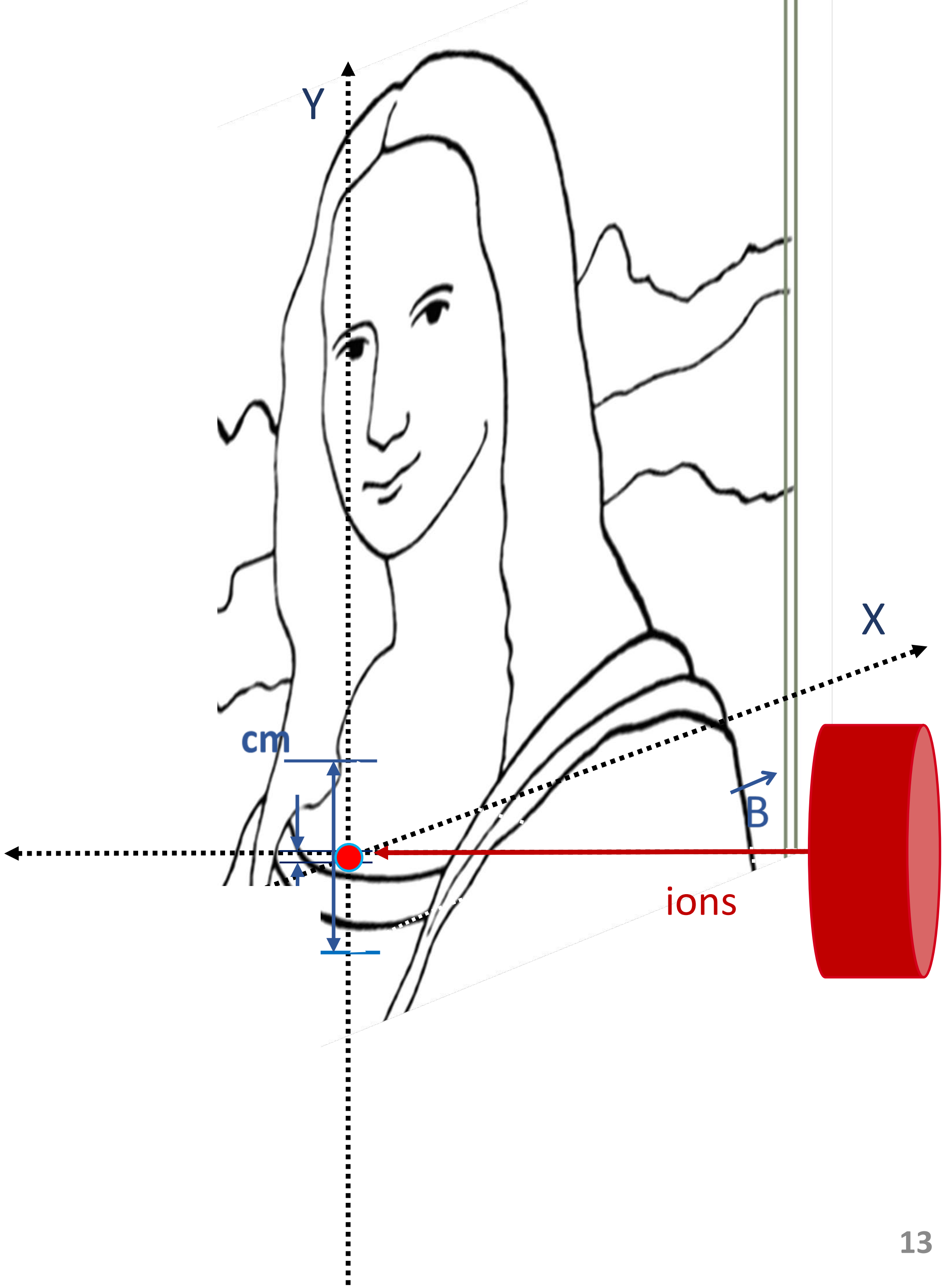


novel push-broom scanning

electromagnetic deflection

mechanical raster scanning





constant-speed scanning → on-the-flight acquisition
X, Y positions not required → X, Y = f(time)

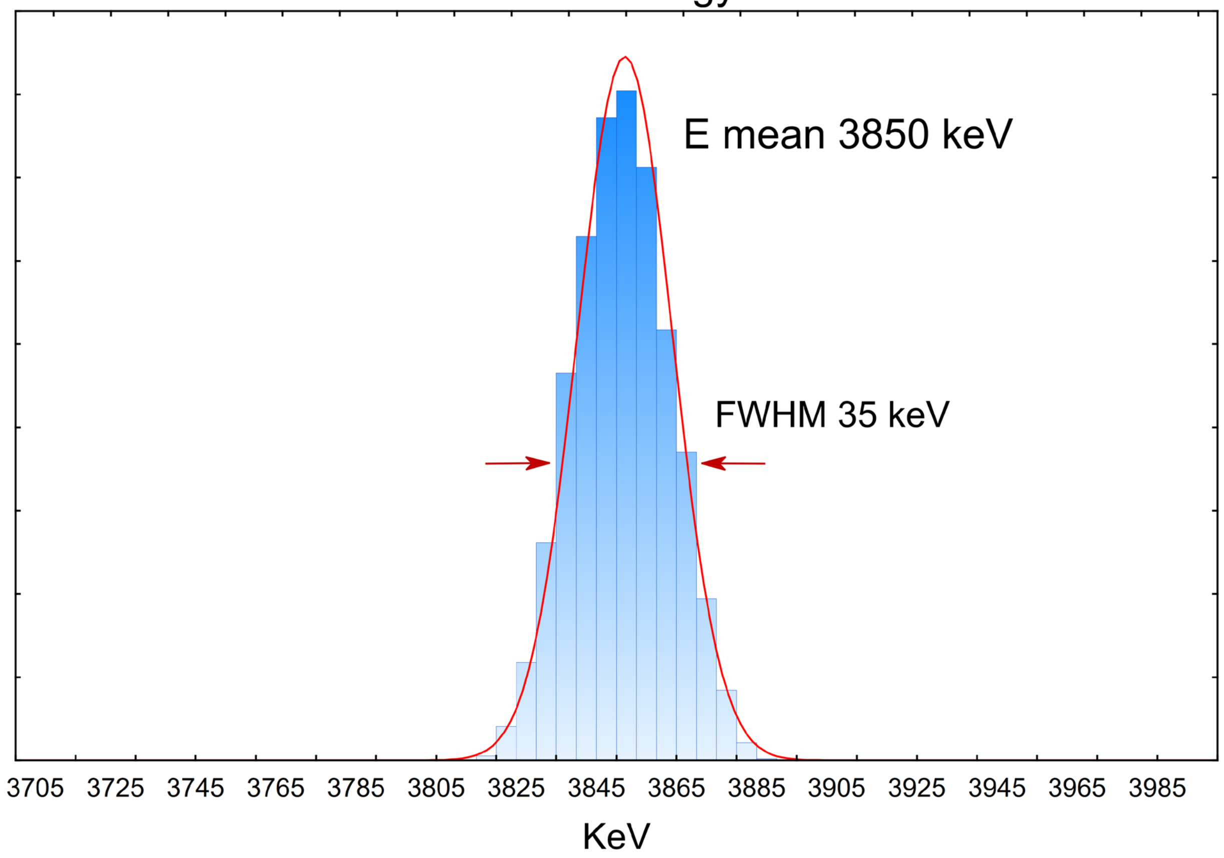
Folded beam trajectory → slow object movements

2 PIXXL specifications

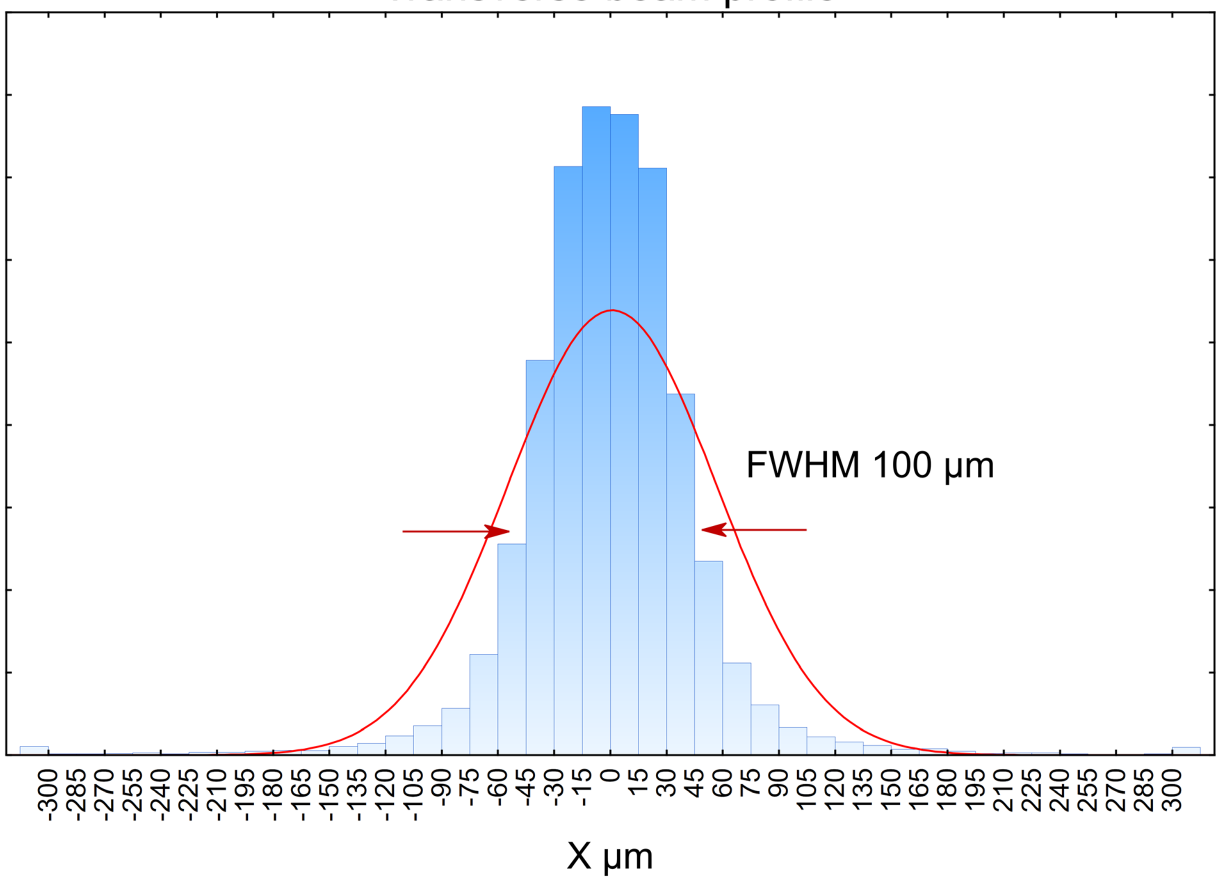
Ion type PIXE method	ion	energy	intensity	Φ
	^{1}H	**4 MeV**	**20-60 nA**	**0.1-0.3 μC/cm²**
	beam shape	count rate	dwell time	spectrum stats
	10 mm V × 0.2 mm H	**10^6 counts/s**	**0.002 sec**	**2000 counts**
Object specs	size	weight	scanned area	**movement speed**
	2 x 2 m²	**50 kg**	**1 m x 1 m**	**2 mm/s**
Mapping specs	resolution	spot size	total time	data volume
	5000 × 5000 = 25 M	**0.2 mm × 0.2 mm**	**12 h**	**10-100 Go**

beam quality after 15-mm air path
^{1}H 4 MeV – SRIM

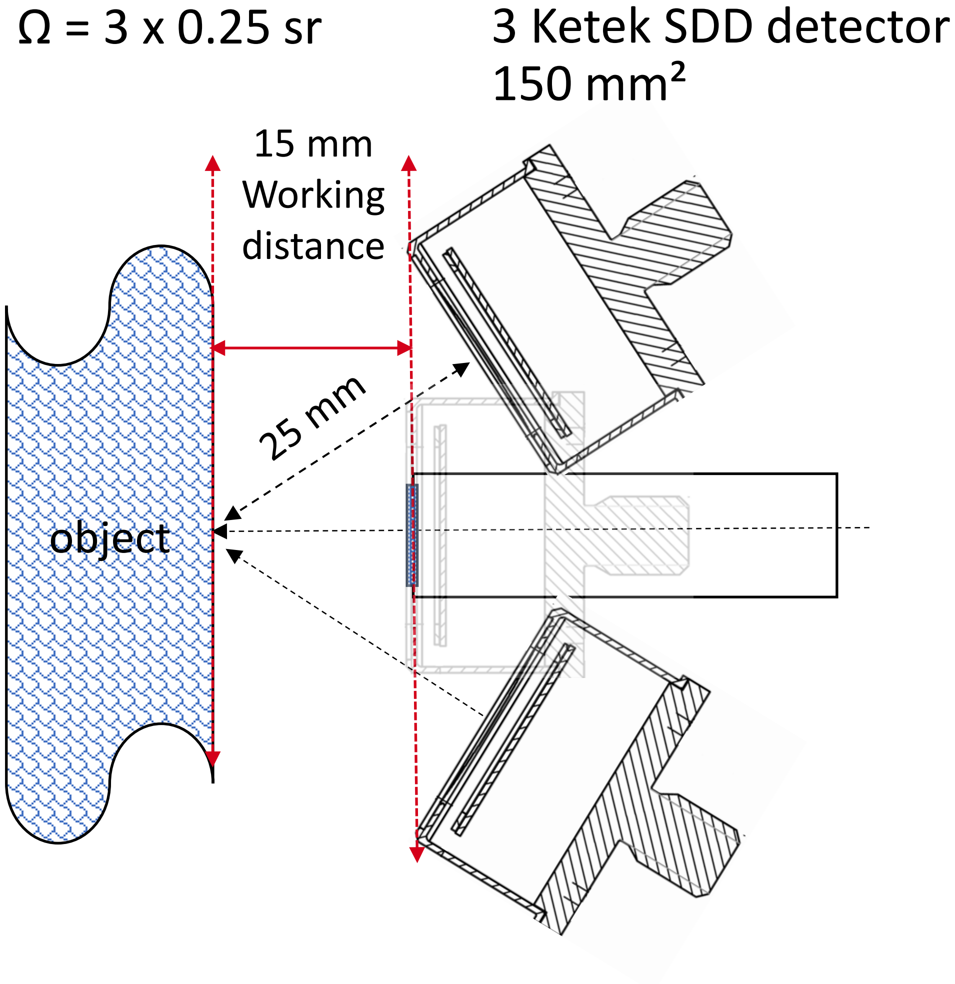
Beam energy



Transverse beam profile



improve efficiency detection



improve DAQ throughput

- 4 M counts/sec input for 3 detectors
- 85 ns dead time / event
- 4 detector channels
- 1 ms spectrum accumulation
- direct HDF5 format

Proof of principle
18th c. oil painting

4-MeV ^{1}H

3 x SDD 50 mm^2

125 μm Be absorbers

2400 x 2200 pixels

100 μm pixel size

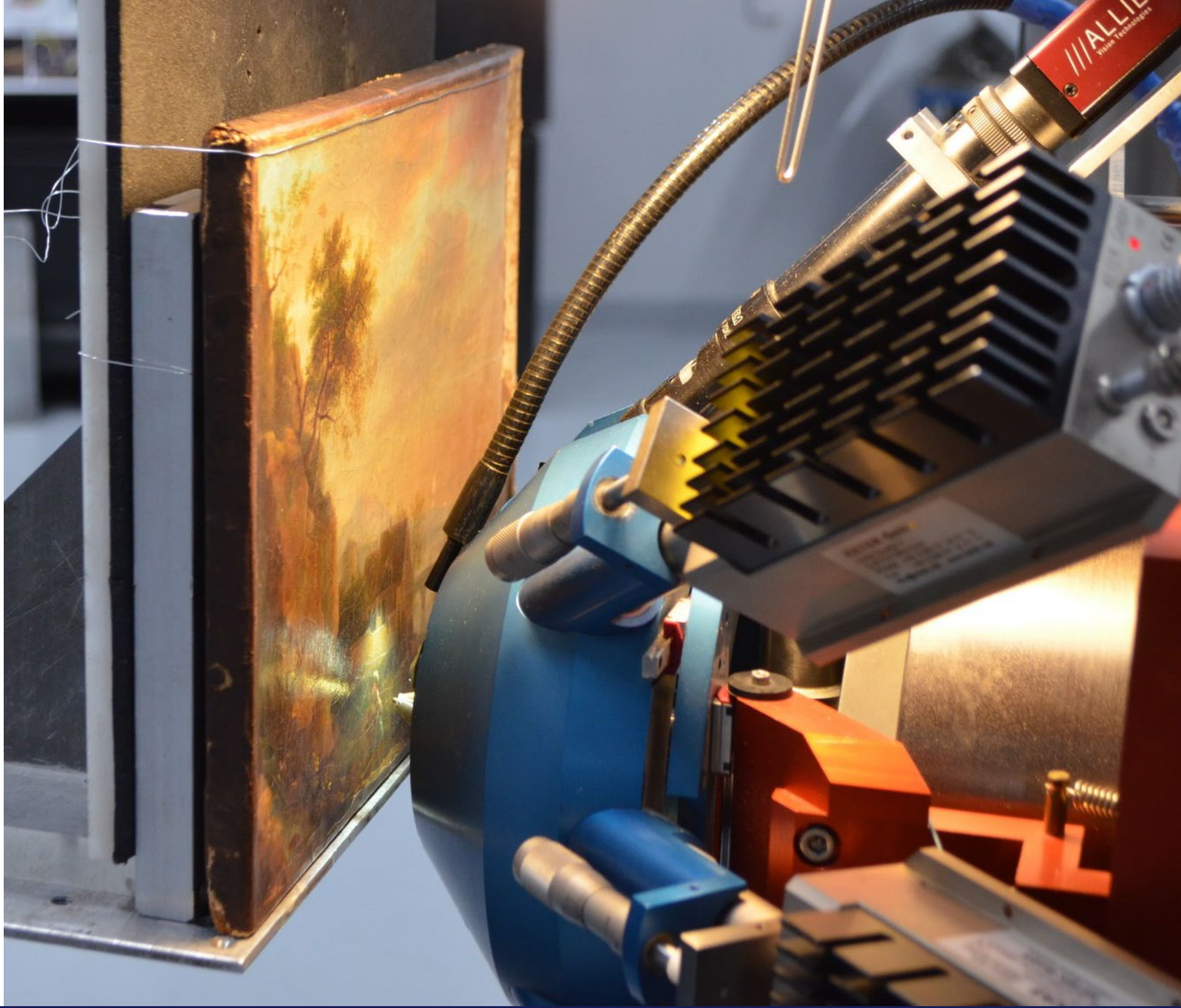
6 ms / pixel, 15 h total

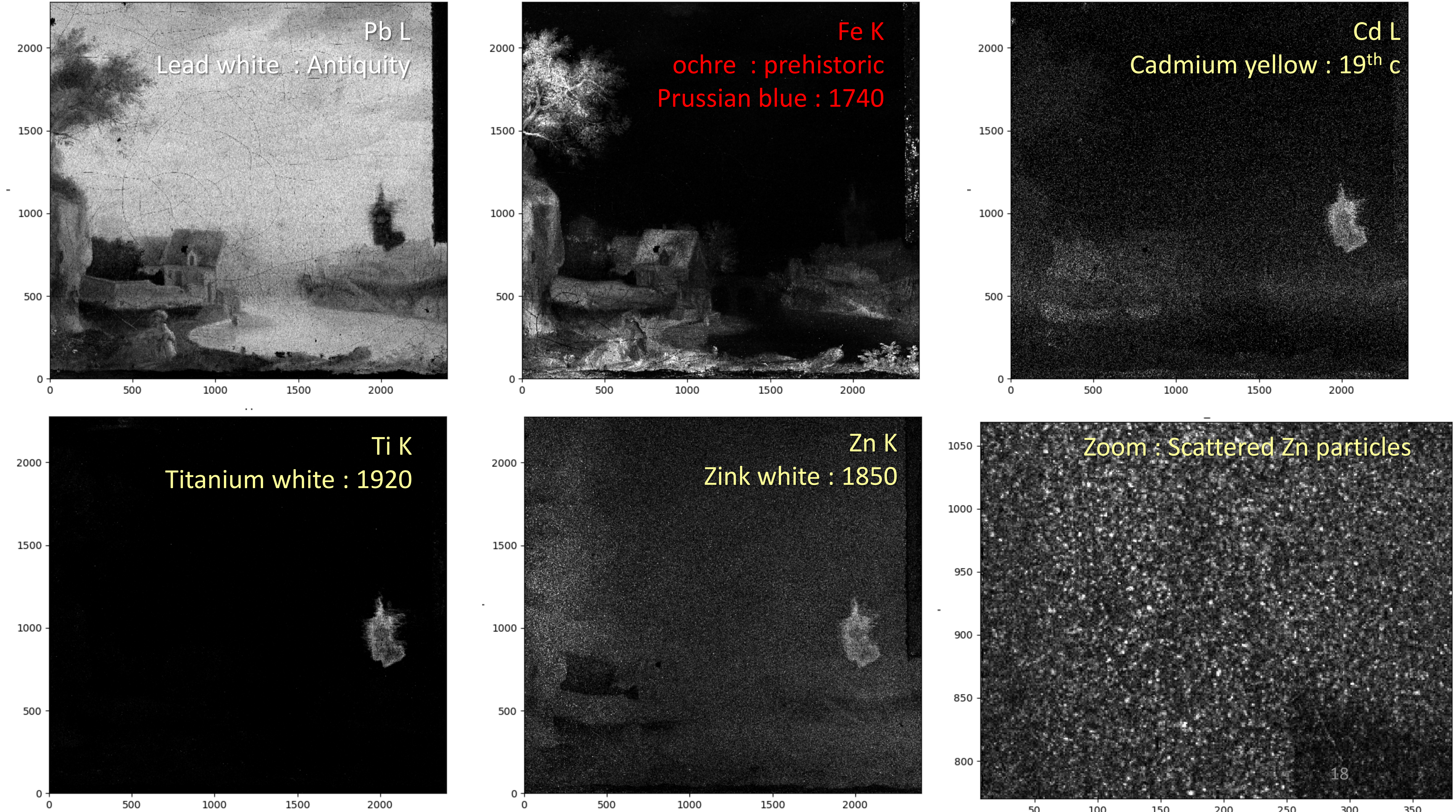
200 μC total - 0.4 μC/cm^2

100000 c/s

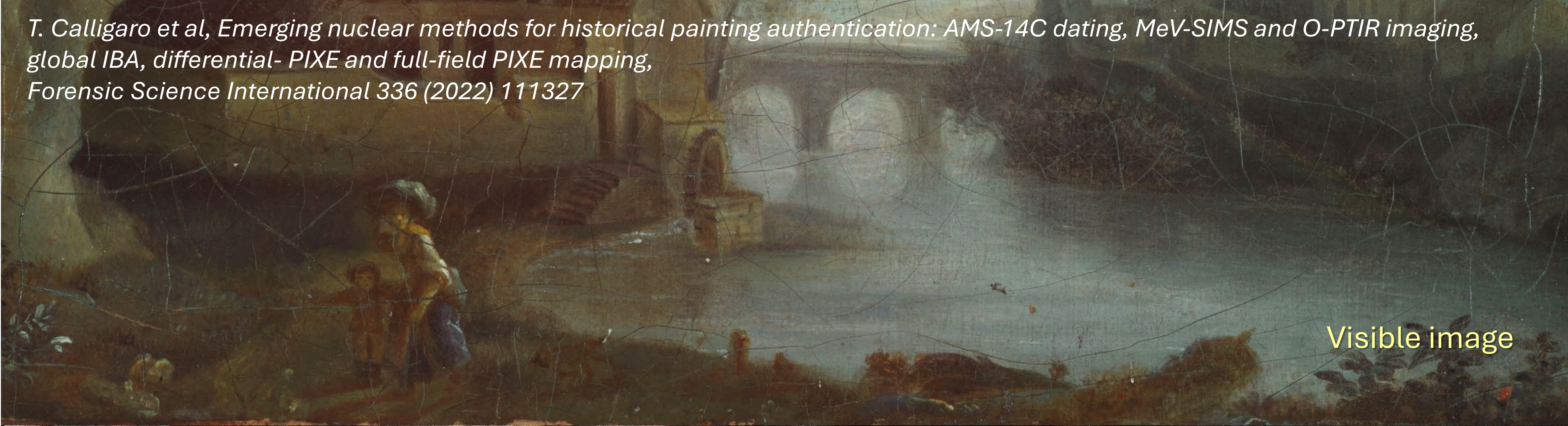
1 kcounts /pixel

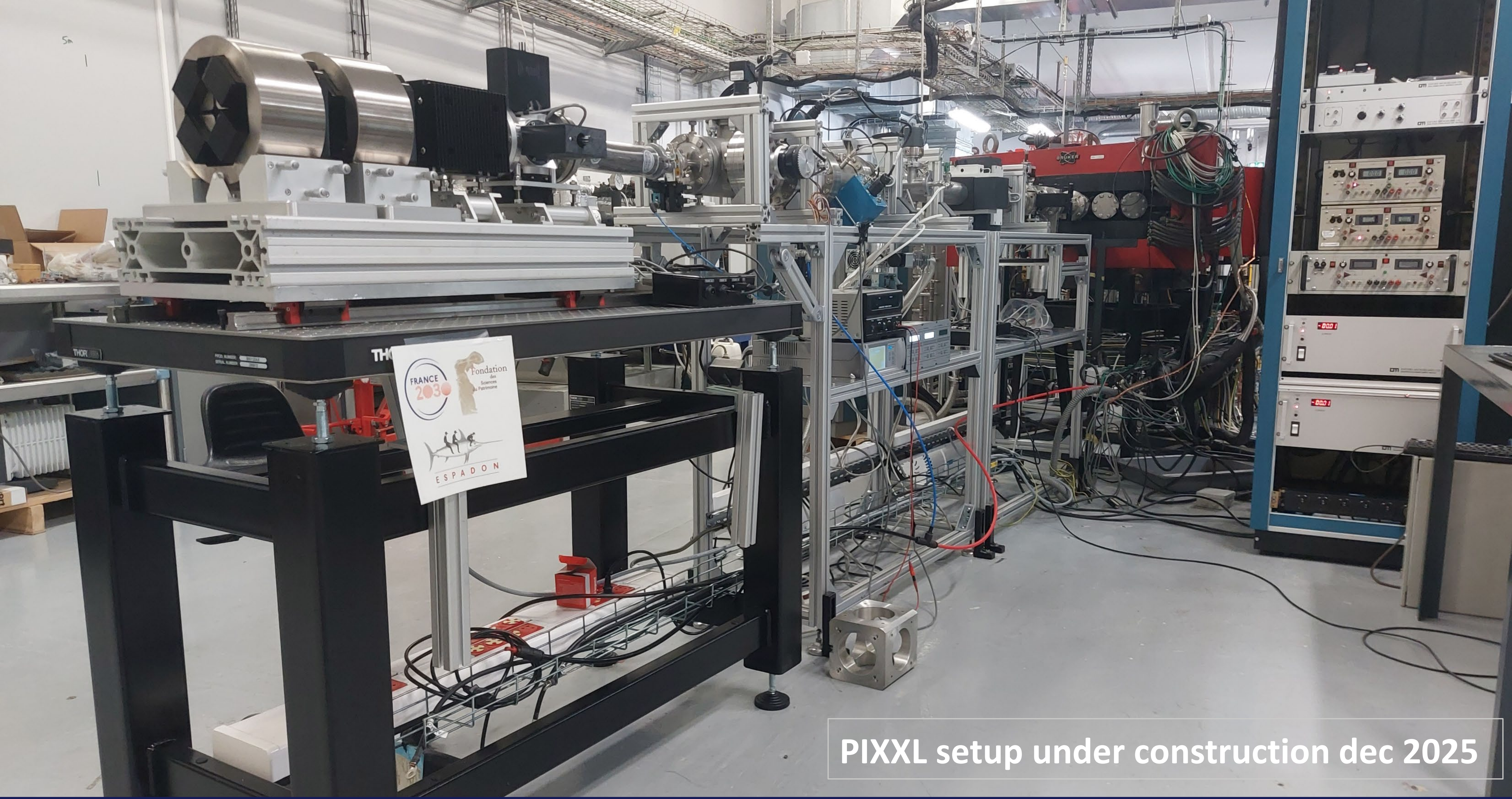
20 Gb total (EDF format)




Pb L
Lead white : Antiquity
Fe K
ochre : prehistoric
Prussian blue : 1740
Cd L
Cadmium yellow : 19th c
Ti K
Titanium white : 1920
Zn K
Zink white : 1850
Zoom : Scattered Zn particles

T. Calligaro et al, Emerging nuclear methods for historical painting authentication: AMS-14C dating, MeV-SIMS and O-PTIR imaging, global IBA, differential- PIXE and full-field PIXE mapping,
Forensic Science International 336 (2022) 111327




FRANCE 2030
Fondation des Sciences du Patrimoine
ESPADON
PIXXL setup under construction dec 2025

3

MONIONS

MONitoring of sensitive targets under **IONS**

New strategies for non-invasive IBA of heritage goods

MONIONS strategies

- Anticipate modifications : live control of D
- Monitor in real-time macroscopic target properties:

real time
< 1 sec

Optical

- ☑ reflectance & luminescence
- ☐ n, transparency, gloss, etc

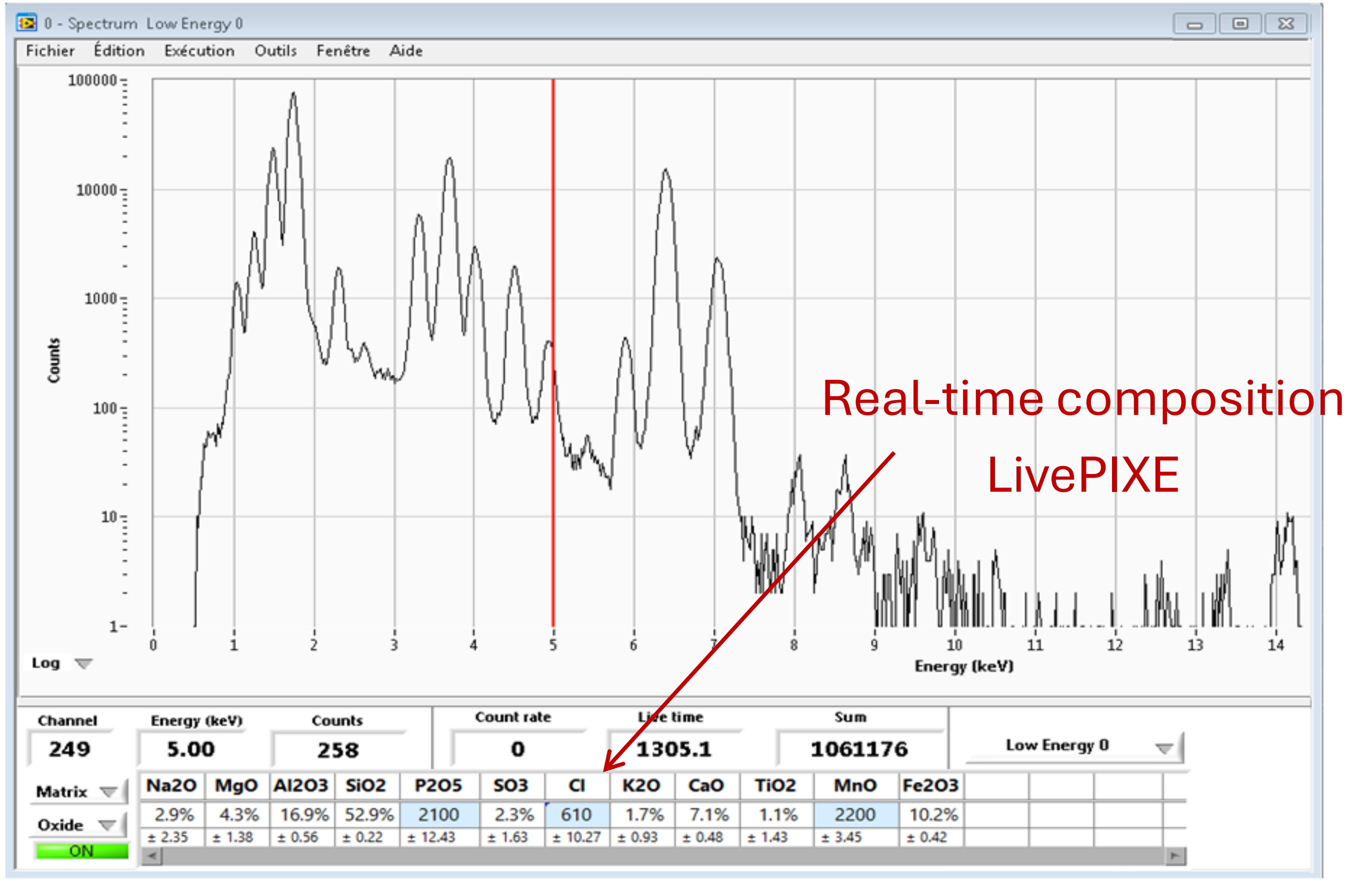
Chemical and molecular

- ☐ disorder using FTIR, HP-LC, ESR, Raman, XRD, etc
- ☑ gas release from fragile compounds, radical formation

Physical

- ☐ surface shape, thickness
- ☐ mechanical properties: hardness, tensile strength, etc.
- ☑ T° increase

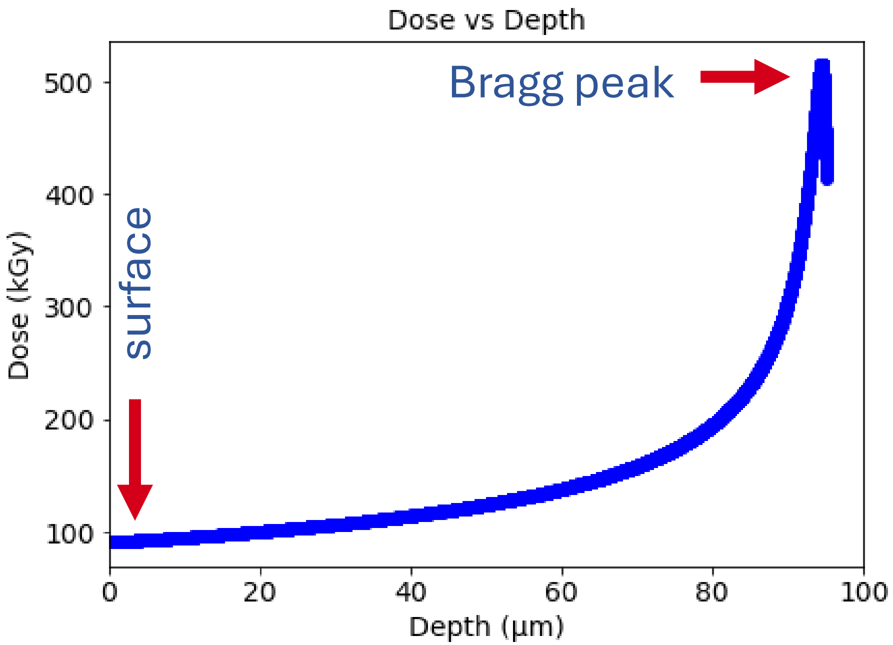
Live calculation of D



Real-time composition

LivePIXE

L. Pichon, Q. Lemasson, D. Bachiller-Perea, C. Pacheco, Advances in dynamic and batch processing of PIXE spectra, NIM B565 (2025) 165734

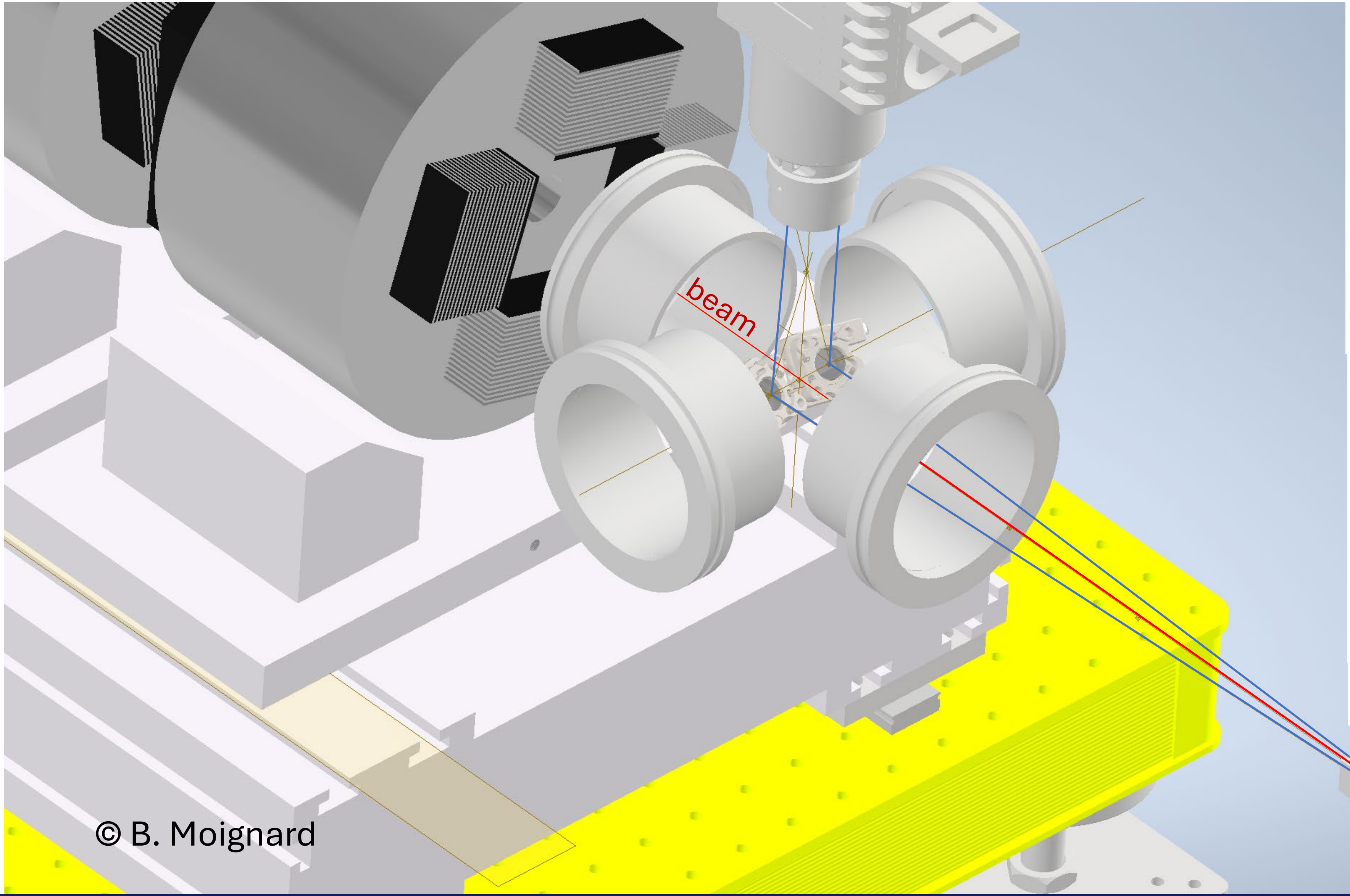


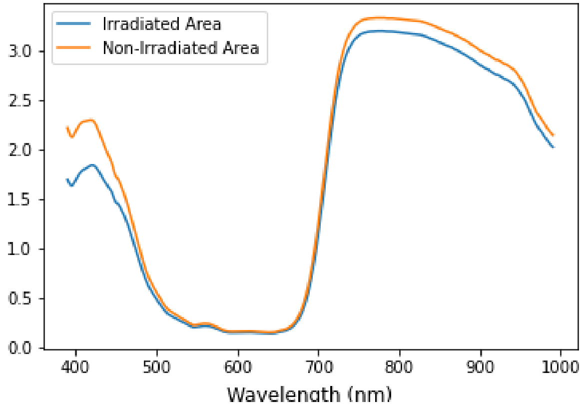
J. A. Maxwell, GUPIX package code, 1988
J.F. Zeigler, U. Littmark & H. Biersack
The Stopping & Ranges of Ions in Matter, 1985

Φ from experimental settings

$$\mathbf{D}(z) = 10^6\ \mathbf{S/\rho}(z) \times \Phi$$

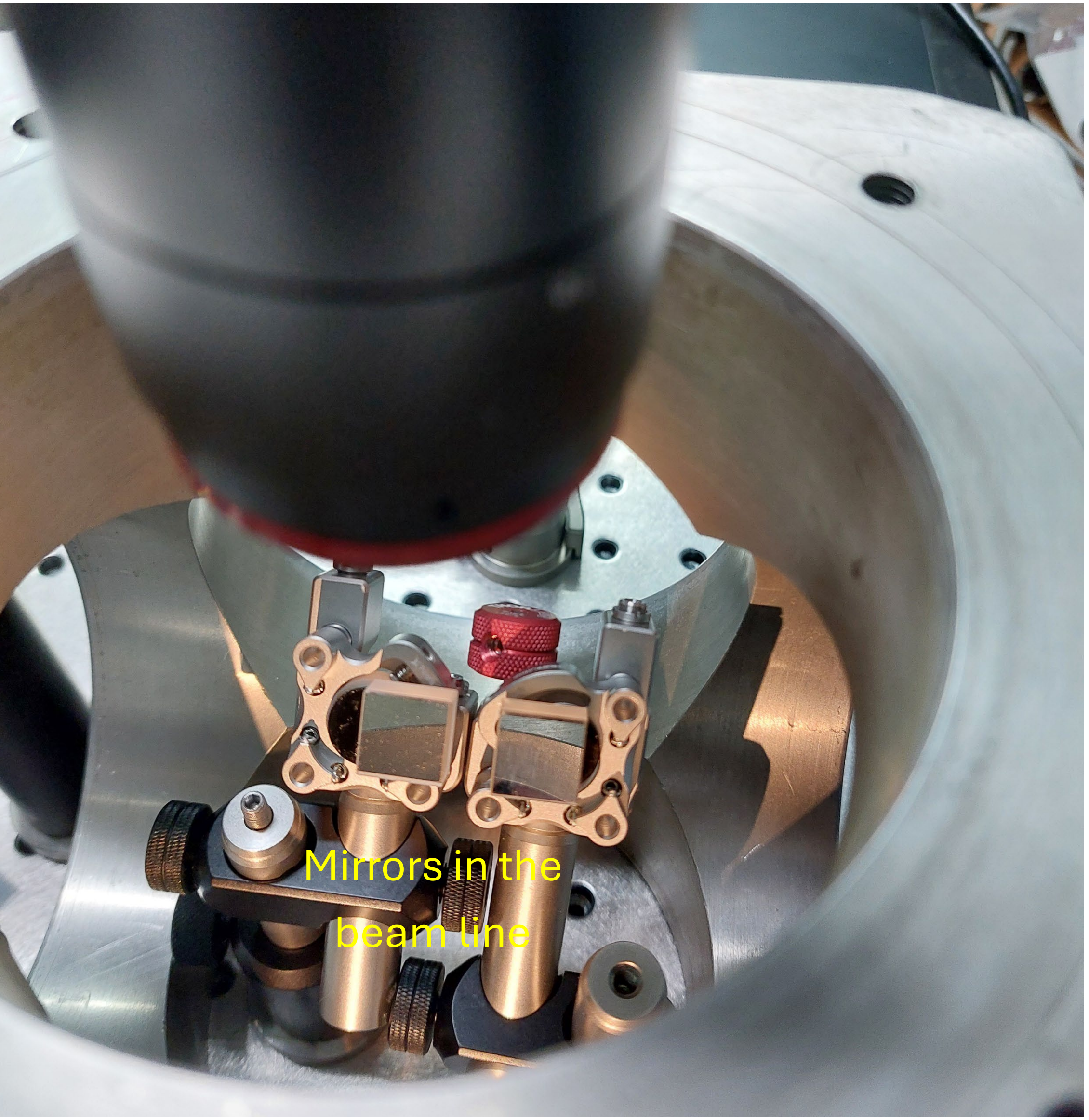
reflectance monitor : hyperspectral camera in the beamline axis

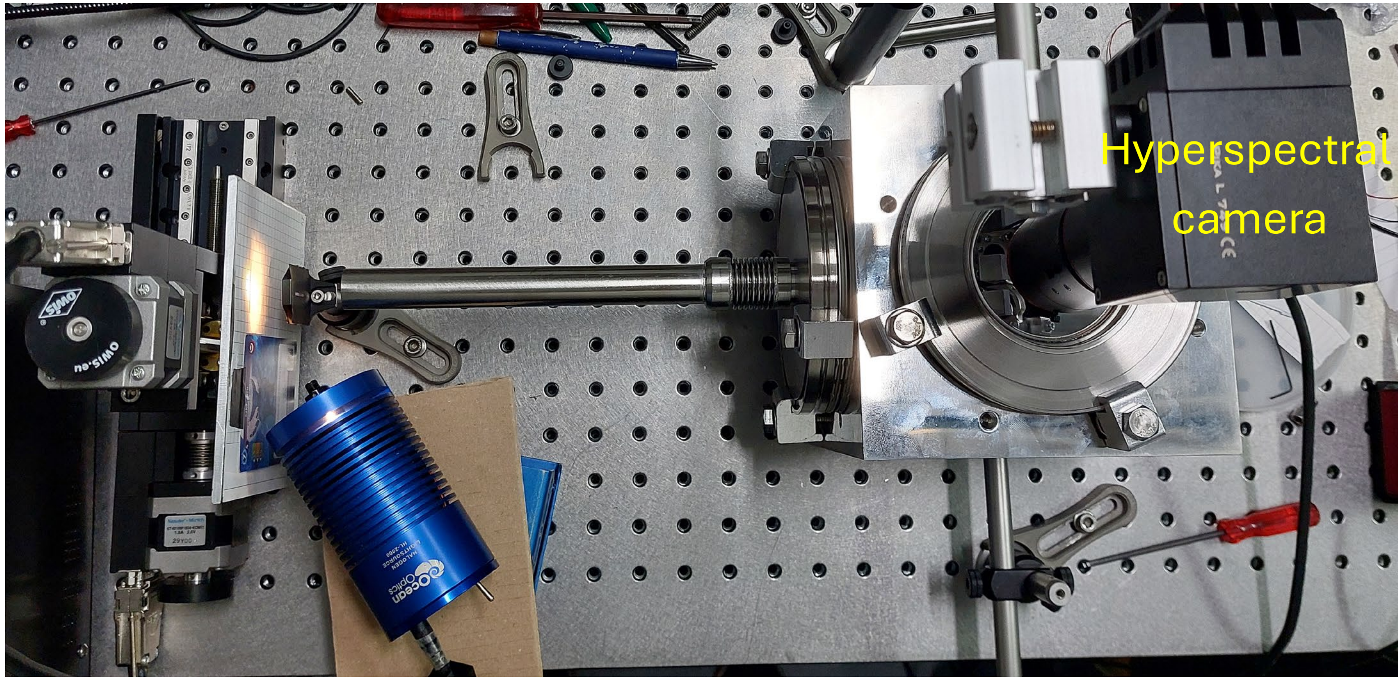




Reflectance before/after

simultaneous reflectance monitor before / after irradiation



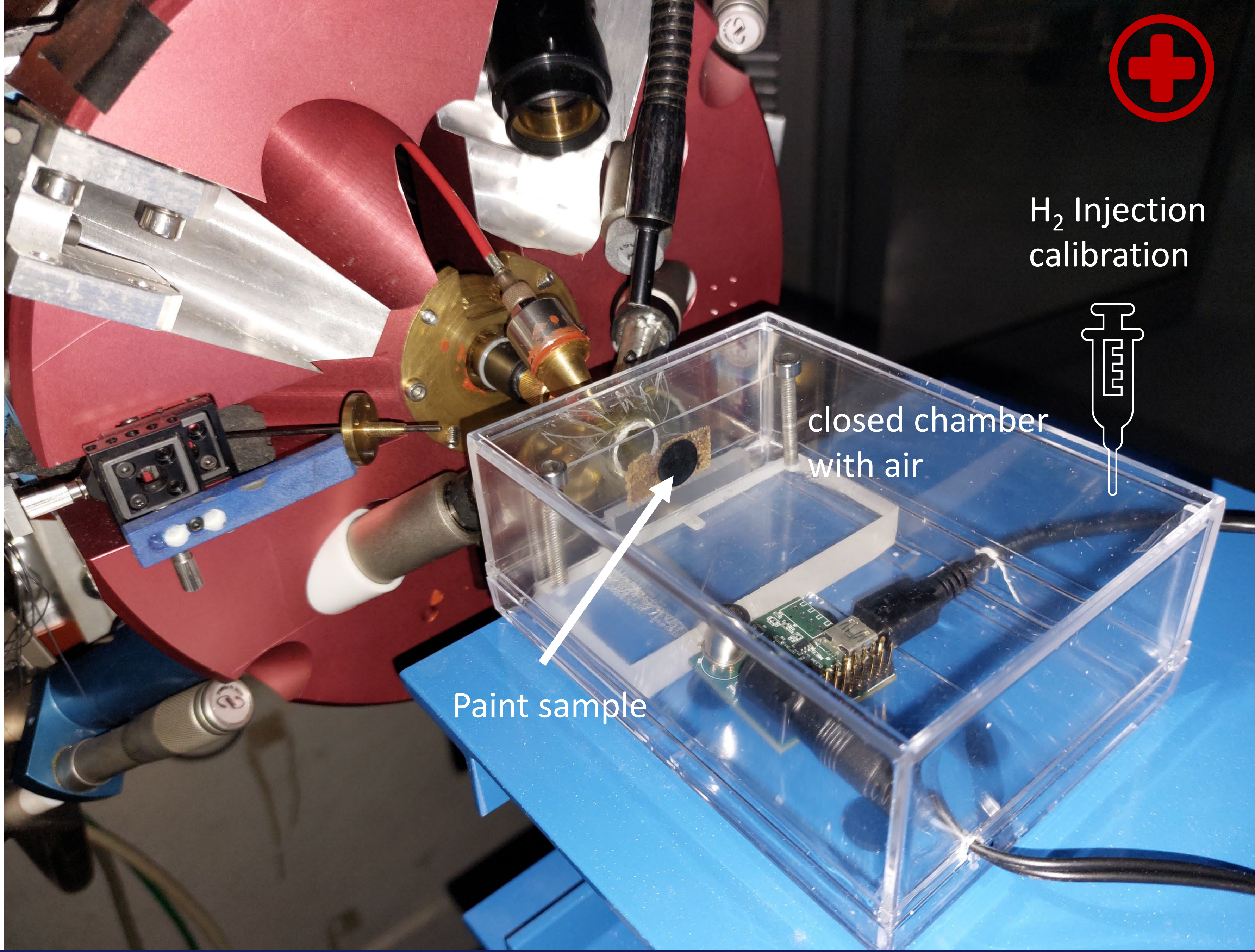


H_2 release: detection in air

chemioresistive sensor

SGAS701

sensitivity 10 ppm vol H_2 in air

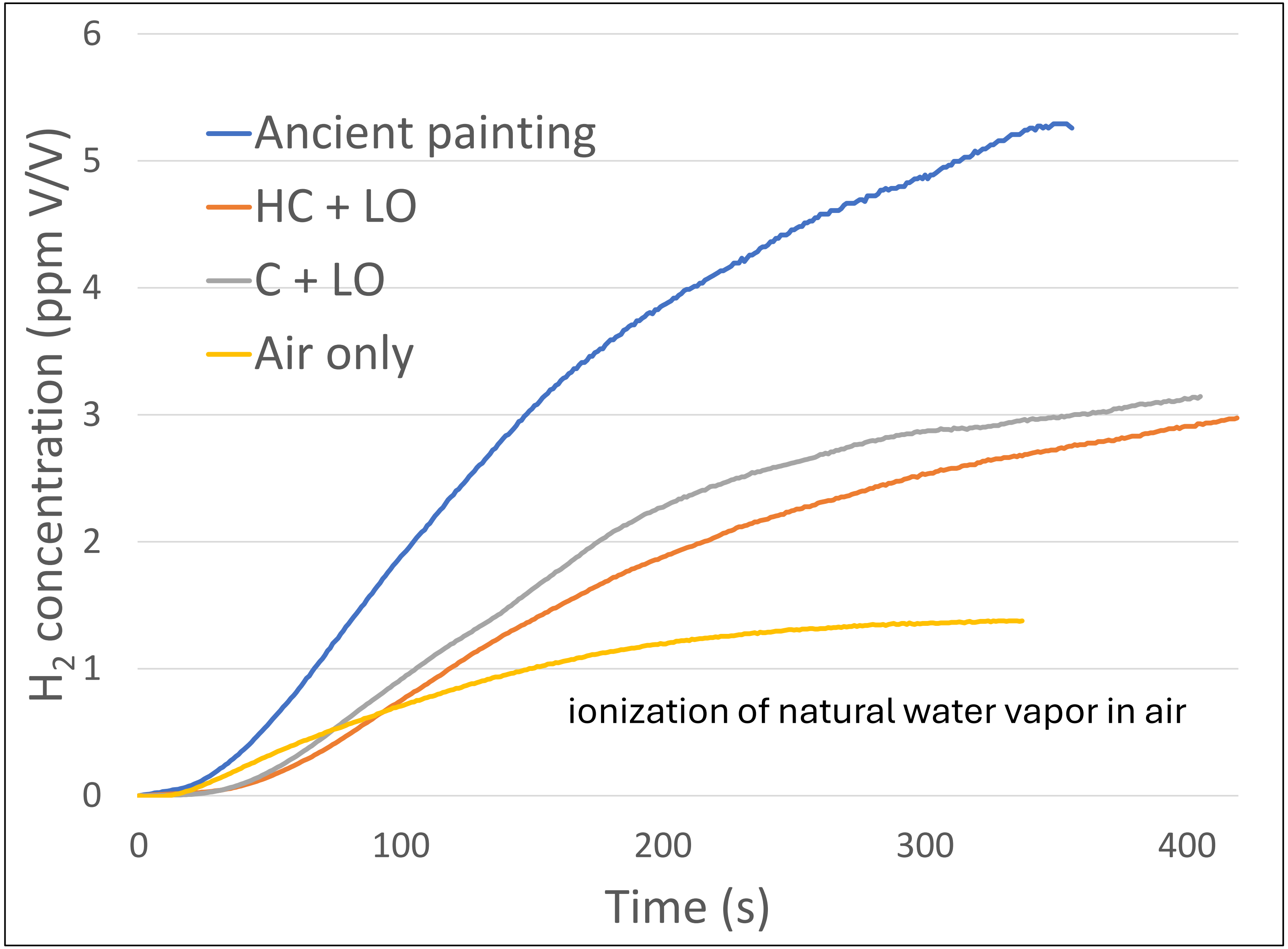


H_2 release by paint layers in air

several µl H_2

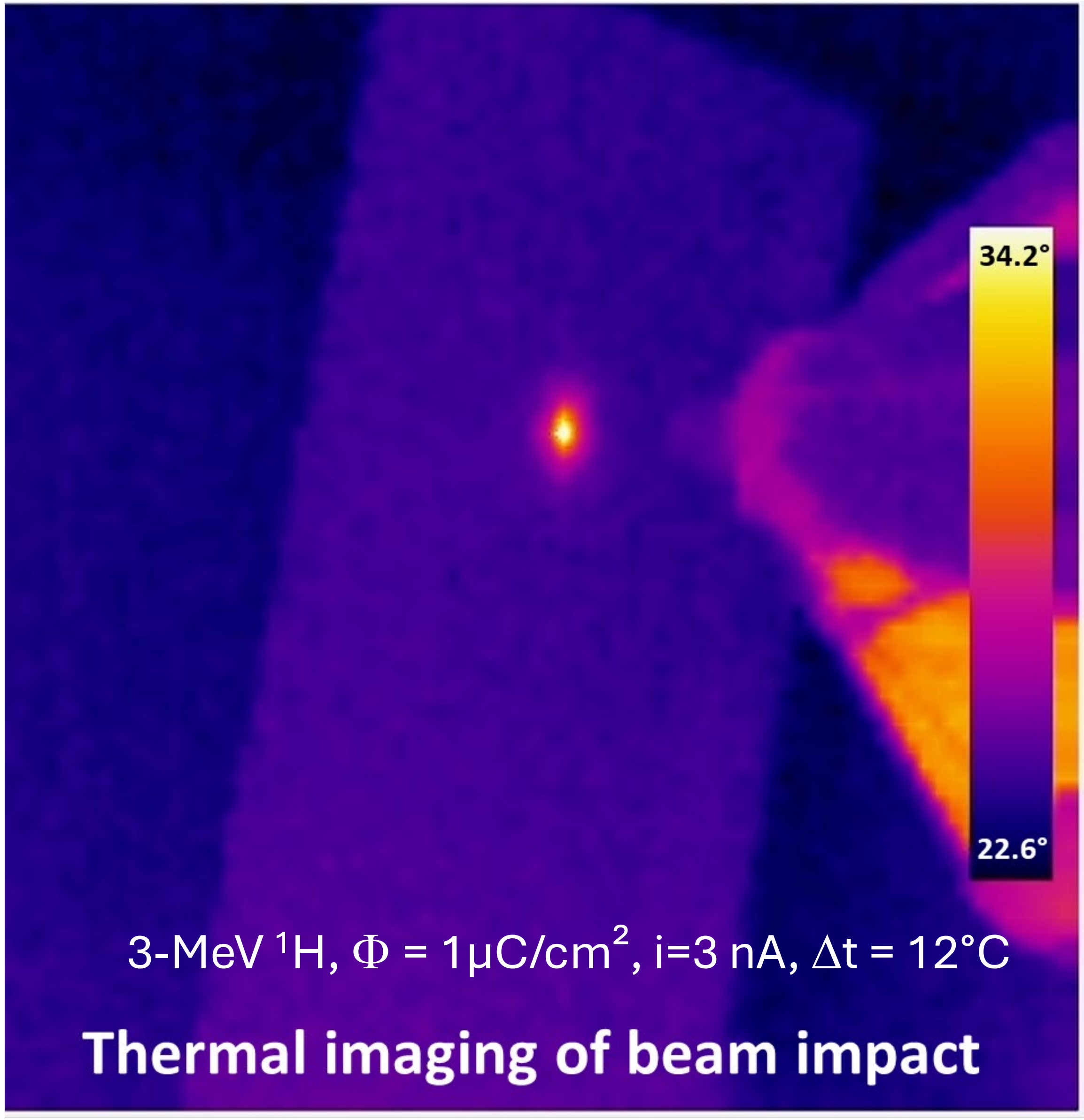
dH_2/dt → proxy of modification

3-MeV ^{1}H
2 µC, 5 nA
4 mm x 4 mm
$\Phi = 12.5\ \mu C/cm^2$
D = 0.6 MGy @surface
T= 25° C, HR 50%

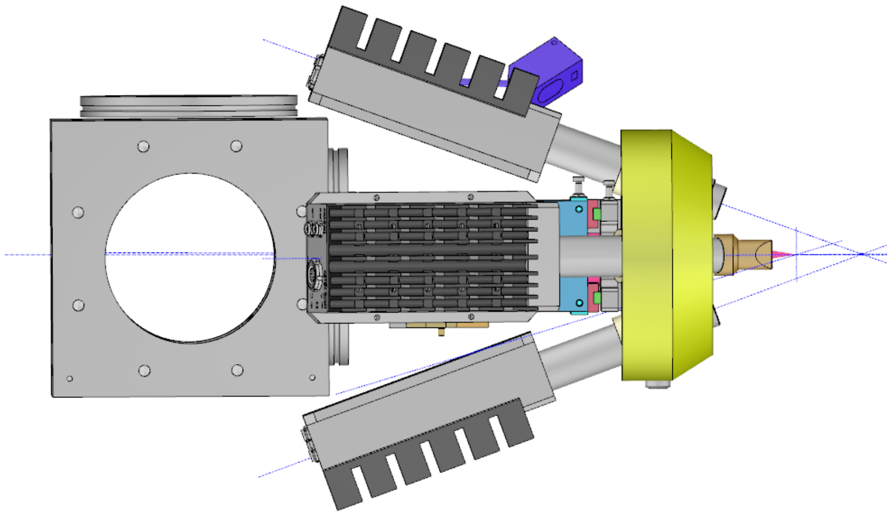


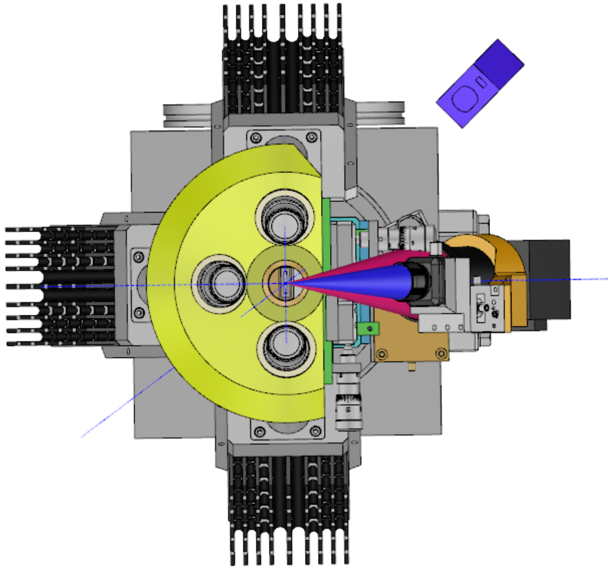
thermal monitoring of beam impact

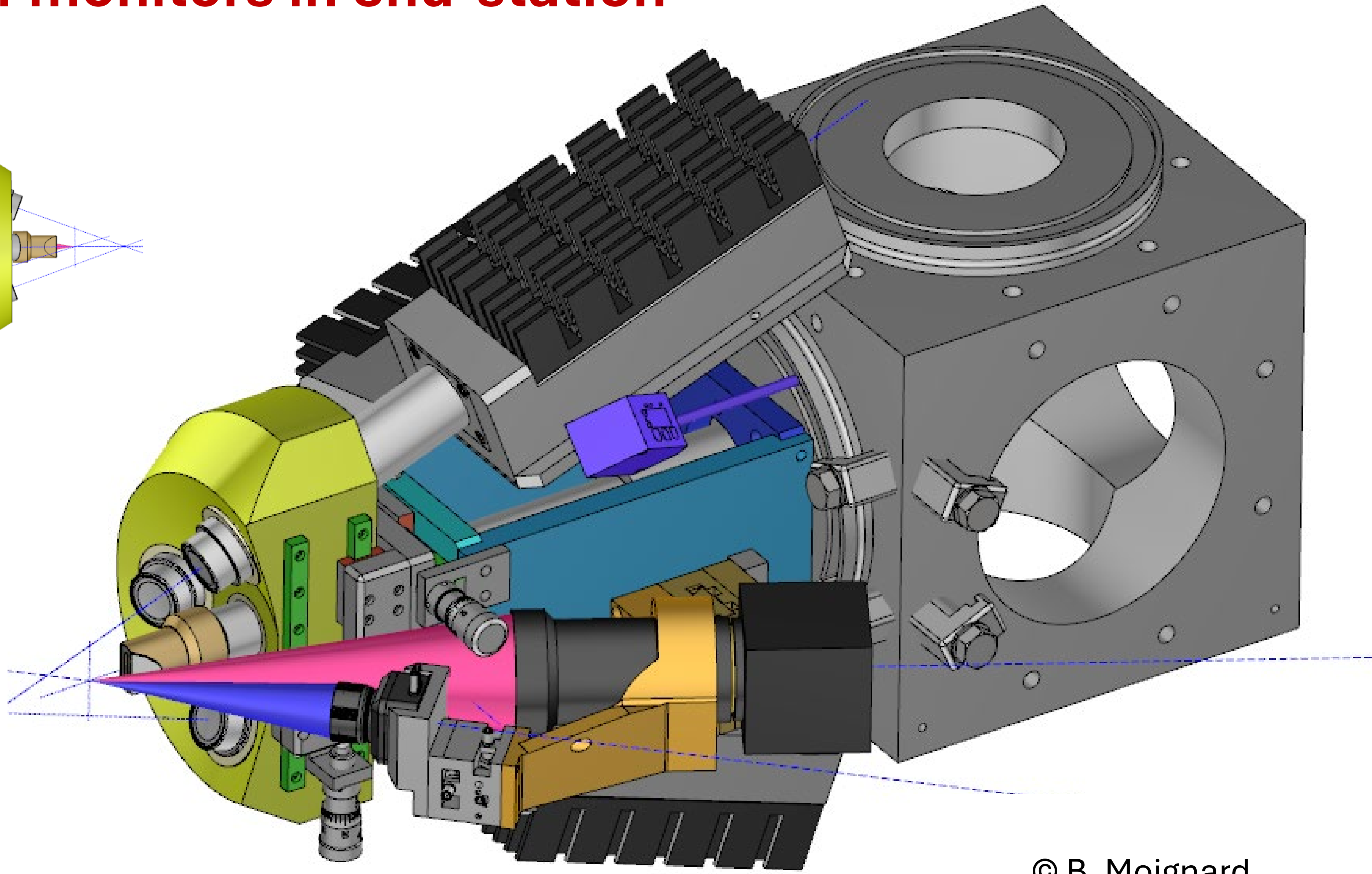
FLIR camera
λ = 8 – 14 µm
-20° - +350°C
accuracy ±2%
sensitivity : 0.1°C



Integration of monitors in end-station







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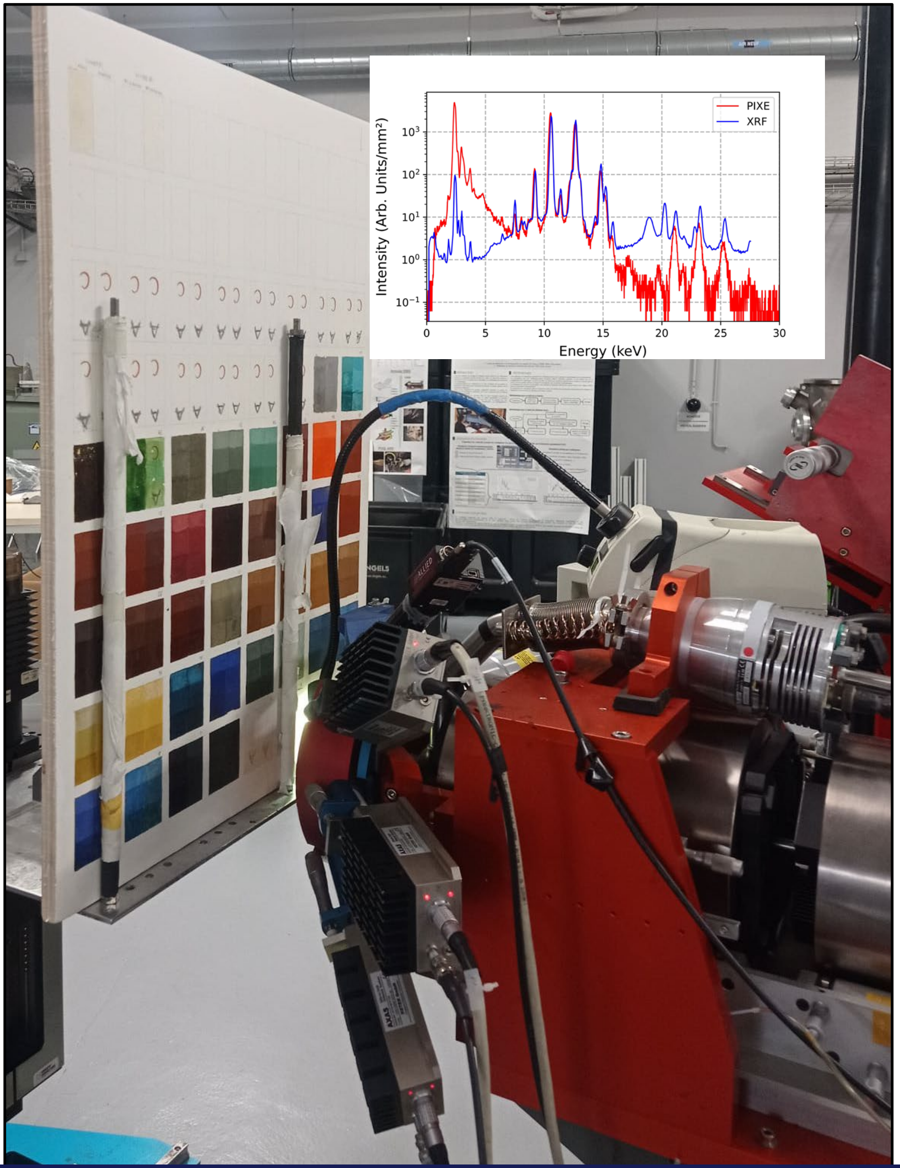
Testing MONIONS protocols on reference painting panels

collaboration with the *Venaria Reale*, Torino, Italy

1st reference database on painting panels



PIXE/IBIL mapping

^{1}H 3-MeV

Φ = 0.4 μC/cm^2

i ~ 1.1 nA

4 × 4 mm^2

t = 60 s

Effects

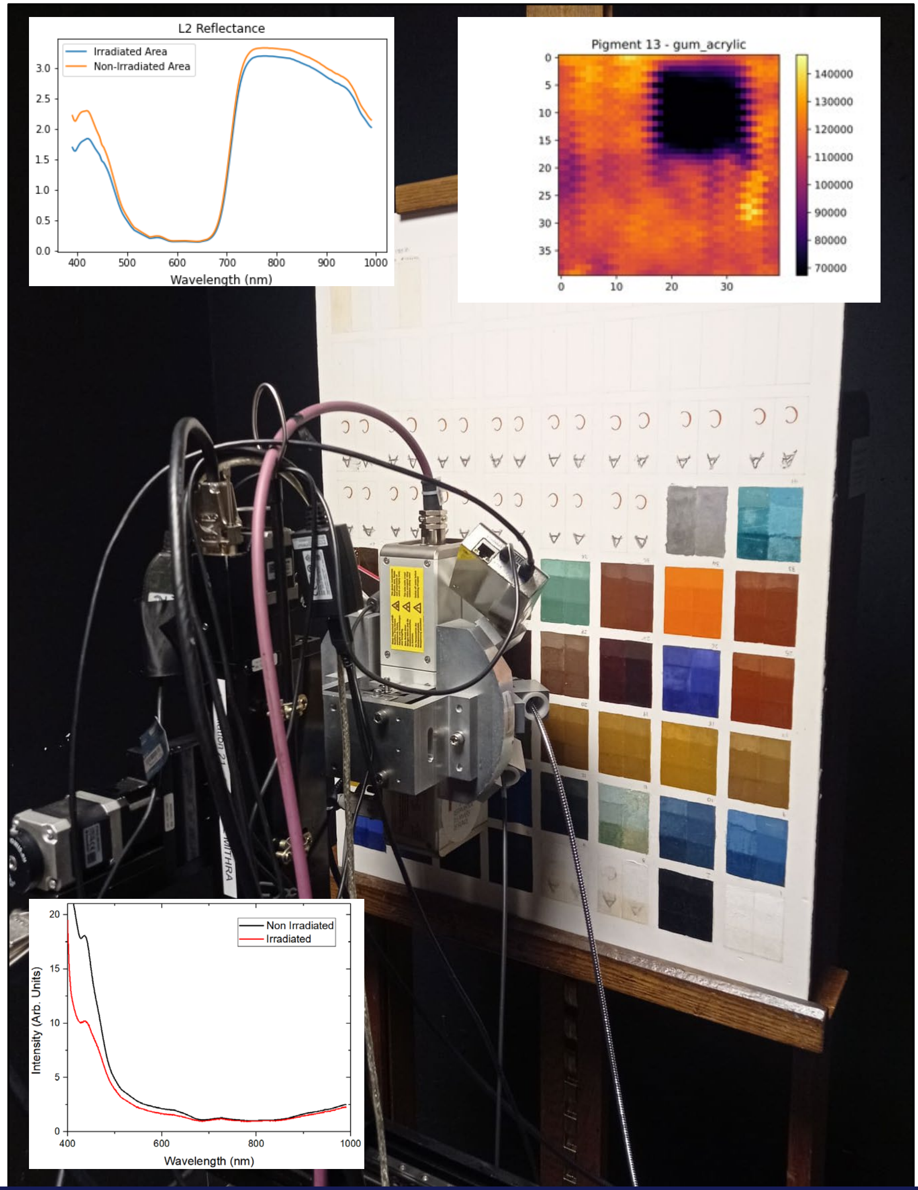
Reflectance VNIR 400-1000 nm

SWIR 1200-2400 nm

Photoluminescence @365 nm

1 × 1 cm^2

t = 128 s



Achievements

Pathways for safer ion beam analysis of heritage objects

Development of instrumental setup integrating monitors

Oulook (among other things)

systematics on modifications induced in fragile materials

deeper understanding of mechanisms – comparison with simulation

evolution of modifications with time

gain advices from EMIR&A community

Acknowledgements

Institutions

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Collaborating laboratories


Thank you for listening
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PAVILLON