PAUL SCHERRER INSTITUT PSI

Wir schaffen Wissen – heute für morgen

Paul Scherrer Institut, Switzerland

Eberhard H. Lehmann
and members of the
Neutron Imaging & Activation Group, Spallation Neutron Source Division

Contribution to the knowledge about cultural heritage objects by means of neutron and X-ray investigations

Italy
is the country
with the highest density
of
UNESCO
cultural heritage sites
(and objects for investigations?)

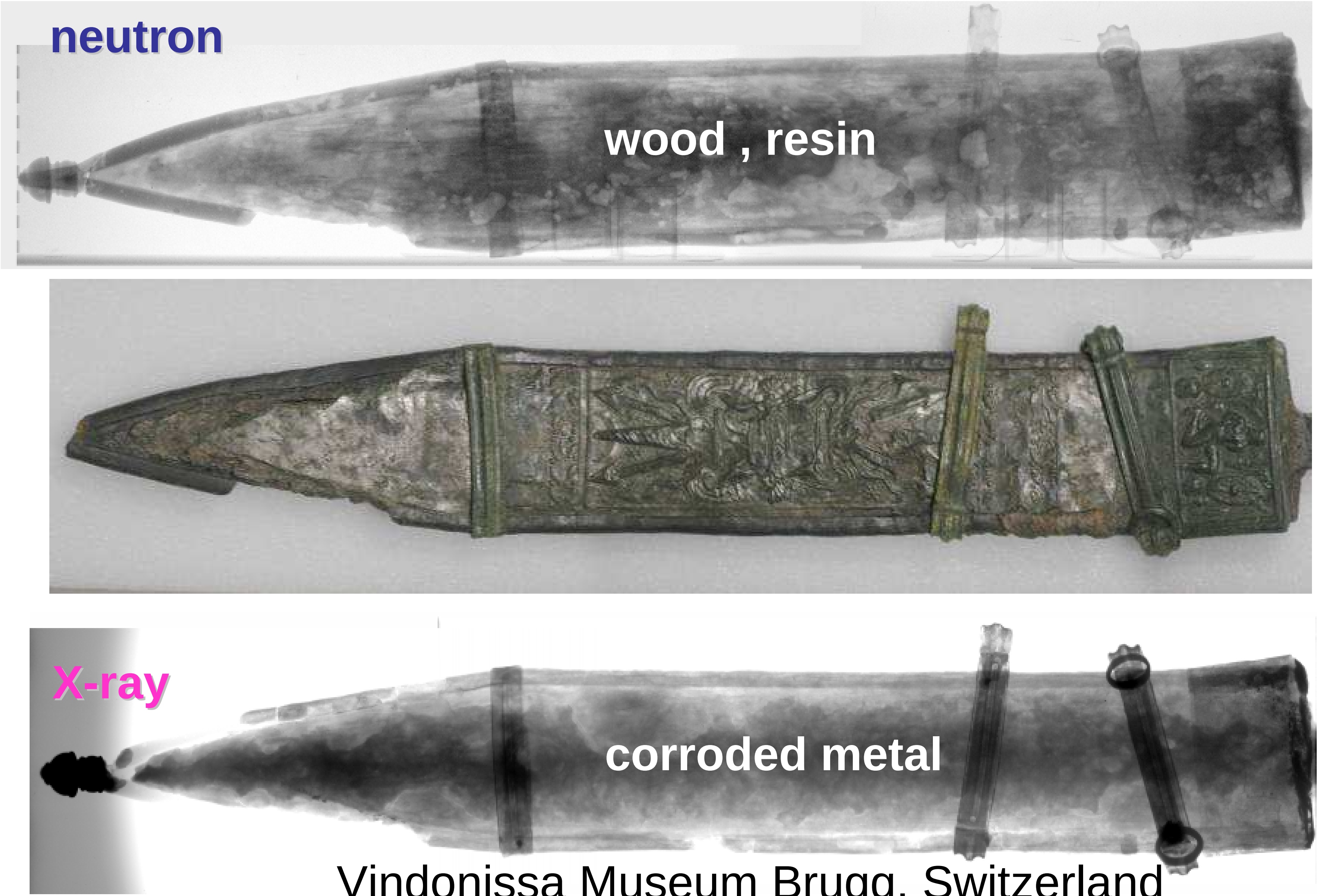
OUTLINE

1. Introduction: how it started …
2. Neutrons vs. X-rays
3. Facilities at Paul Scherrer Institut, Switzerland
4. Options in neutron imaging today
5. Successful studies of relevant objects
6. The relation to archaeology and museums experts
7. Neutron Imaging & Cultural Heritage in Italy?
8. Further trends and activities
9. Conclusions

VINDONISSA – a Roman military camp in northern CH

Roman sword

Gladius type

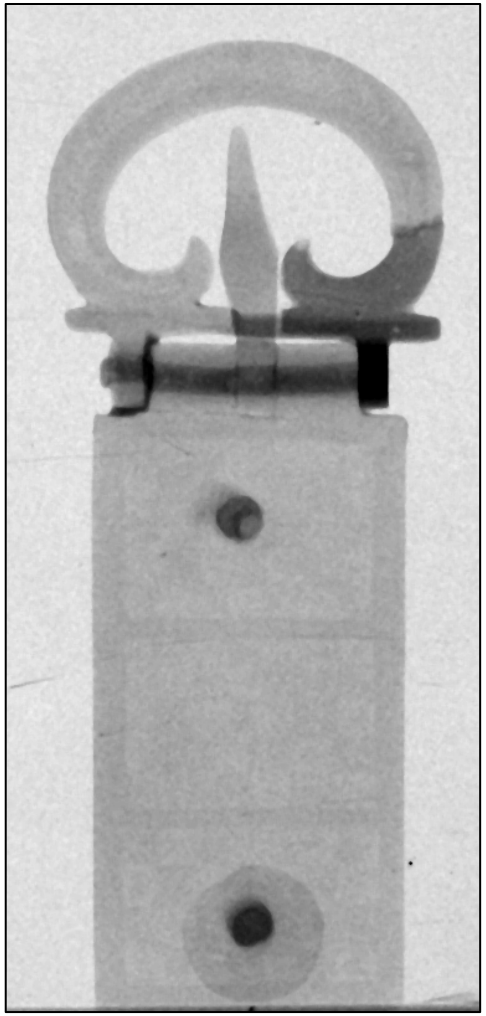


Vindonissa Museum Brugg, Switzerland

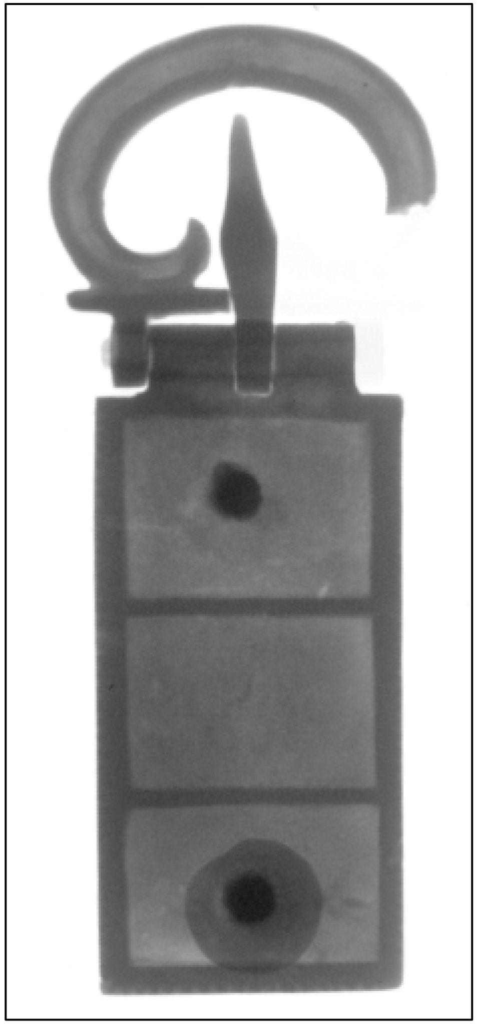
Roman belt buckle – „Millefiori“ technique

Photo: front view

Photo: back view



neutron

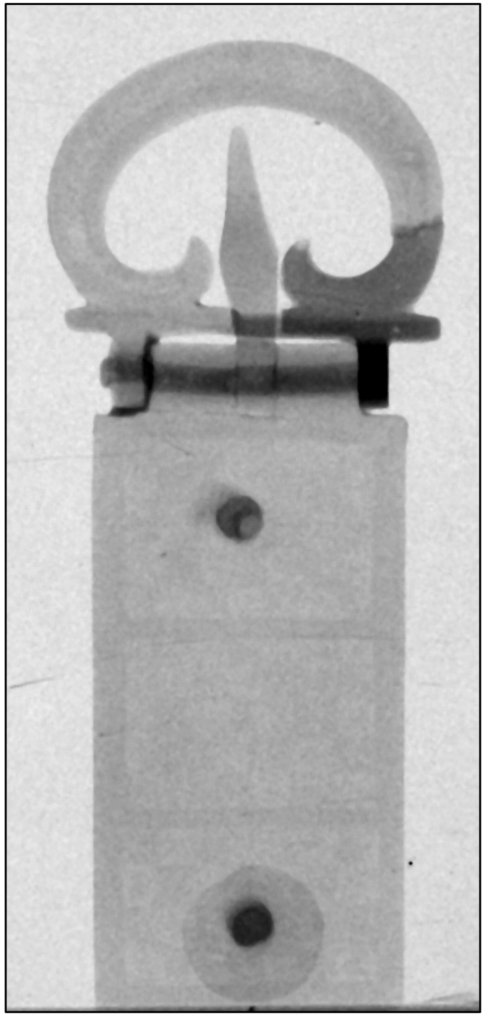


X-ray

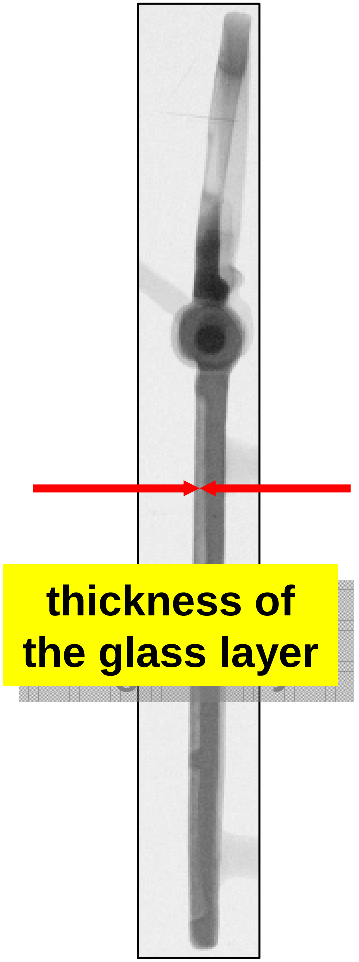
Roman belt buckle – „Millefiori“ technique

Photo: front view

Photo: back view



neutron



neutron, side view

Introduction

•Dealing with unique objects from cultural heritage requires **non-invasive** and **non-destructive** techniques

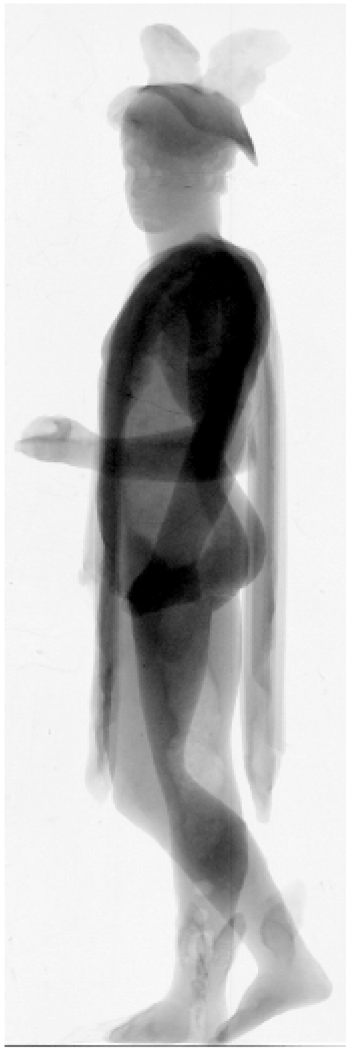
•transmission radiation methods can provide information about
-*material distributions*
-*composition*
-*structure*
-*manufacturing*
-*corrosion*
-*repairs*
-*provenance*

•**X-ray techniques** are common, but limited in the case of metals in penetration power
•**neutron imaging** helps to overcome these limitations

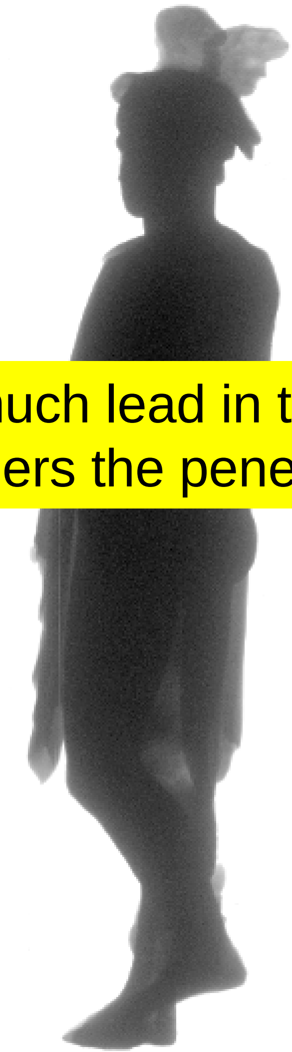
MERKUR from THALWIL, Switzerland

30 cm

photo



neutron



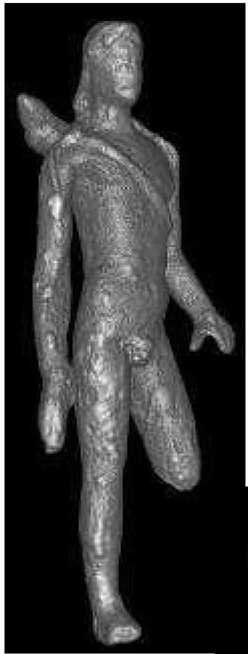
X-ray

too much lead in the alloy
hinders the penetration

2004:first study
tomography with
limited performence
512*512*512 voxels

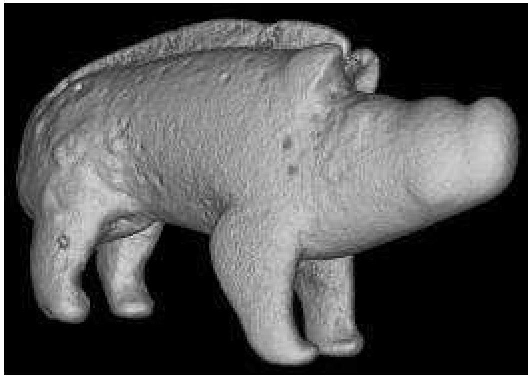
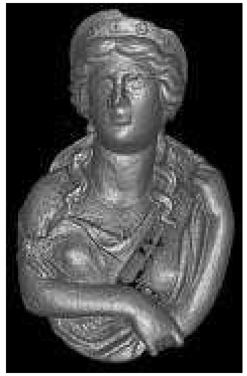
Mercure
from
Uster

Catalogue for Roman bronze sculptures located in Switzerland (examples)




about 200 pieces



studies with neutron tomography and radiography will become part of the 4th volume

FIRST CONCLUSIONS

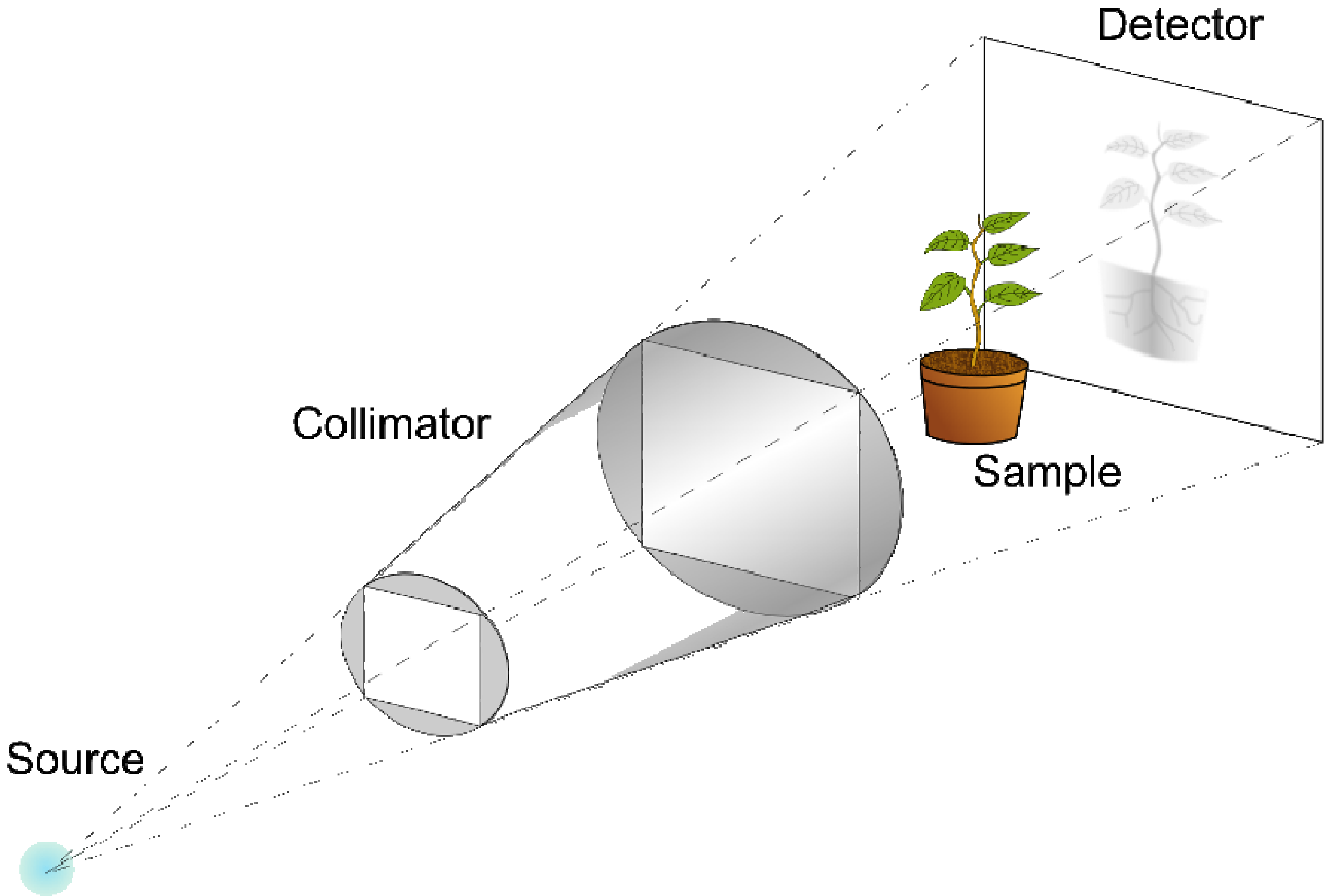
1. Neutron imaging provides another option for non-invasive studies of cultural heritage objects

2. The performance with respect to spatial resolution, exposure time are in the same order compared to standard X-ray imaging

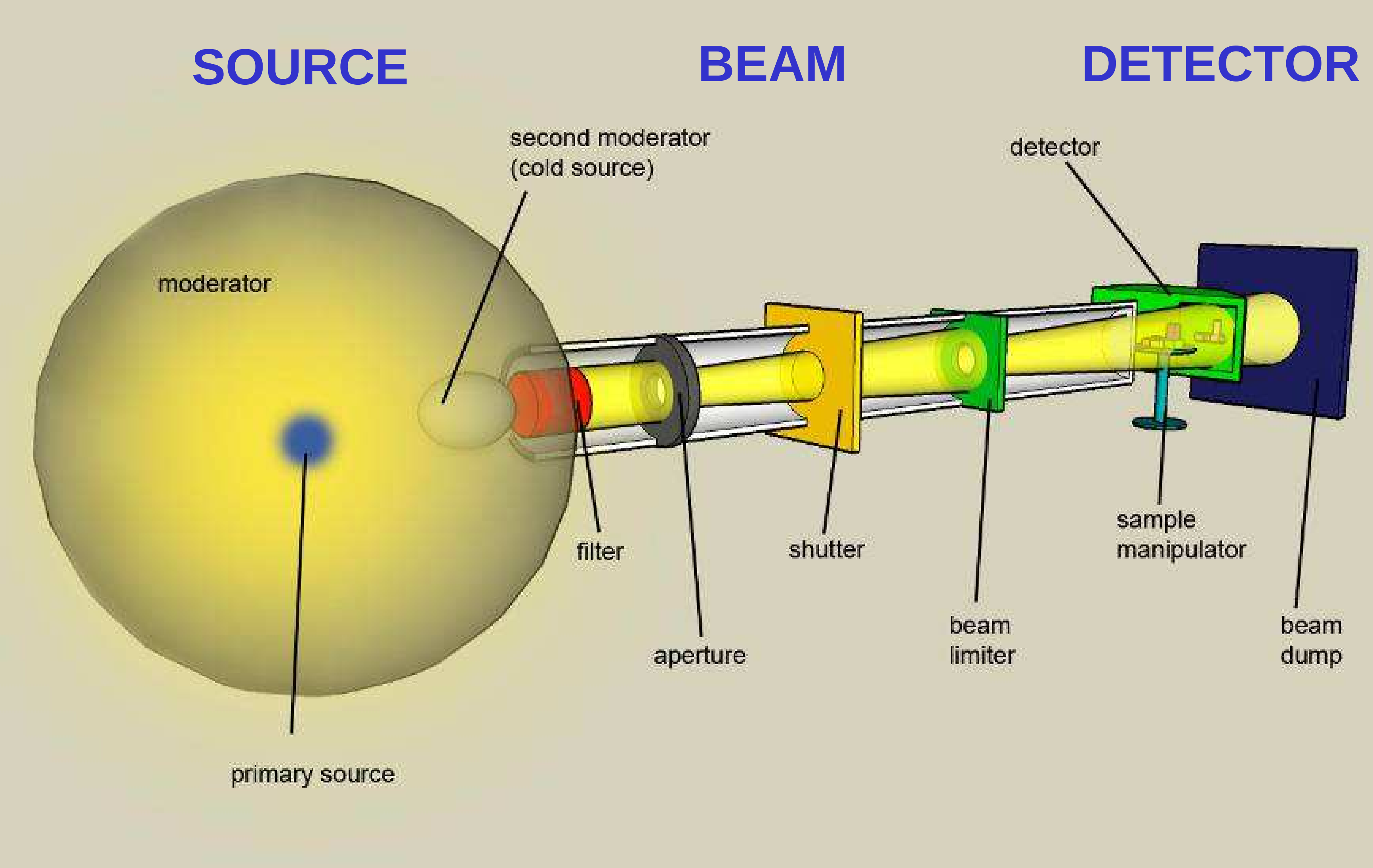
3. The high sensitivity for hydrogenous materials (resin, water, wax, glue, …) and the higher penatrability for most metals can be considered as new options

4. Advanced methods (tomography, real-time imaging, phase-contrast) are also available with neutrons

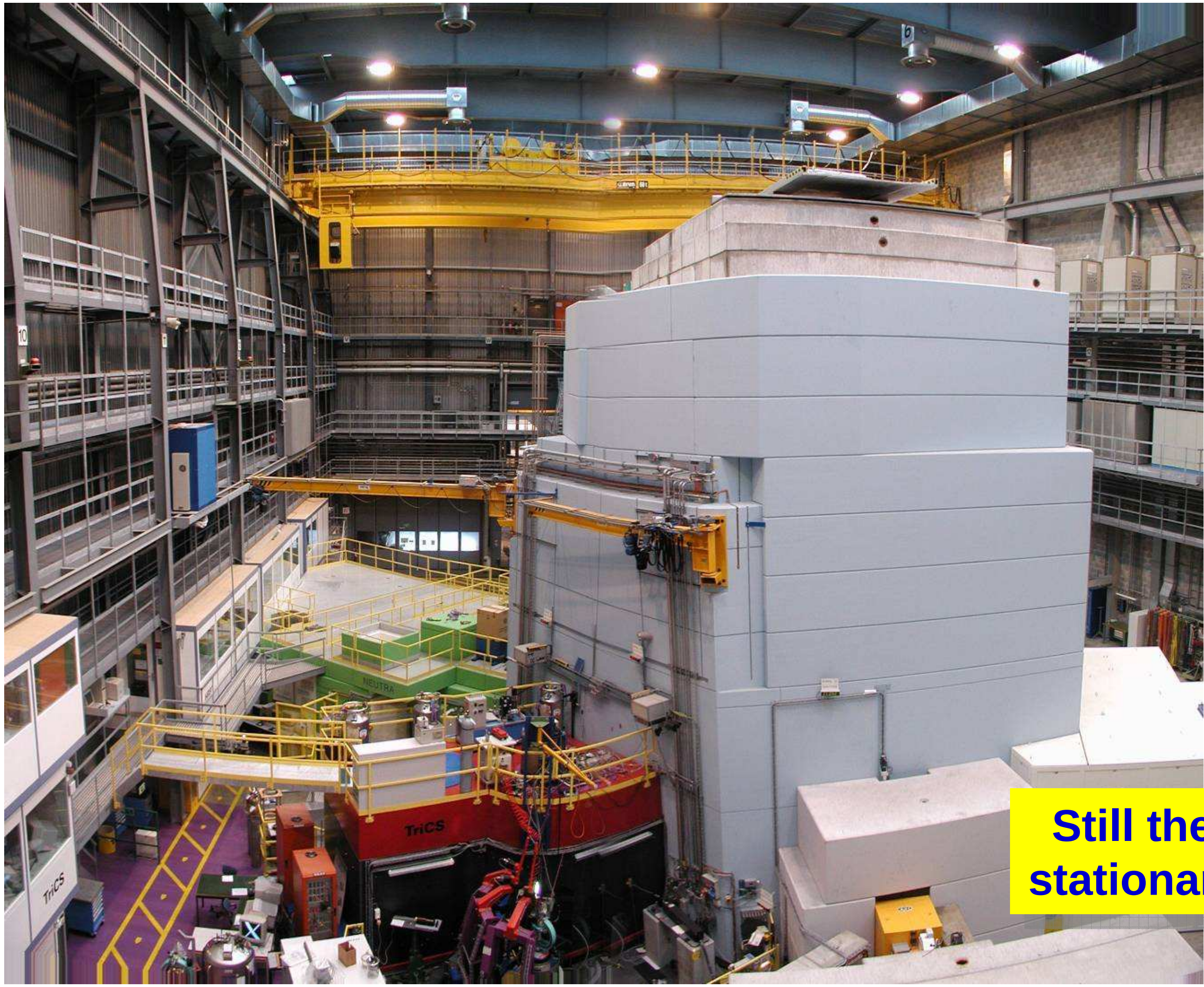
NEUTRON IMAGING (very simplified scheme)



Neutron Imaging – Setup TODAY



Spallation neutron source SINQ @ PSI



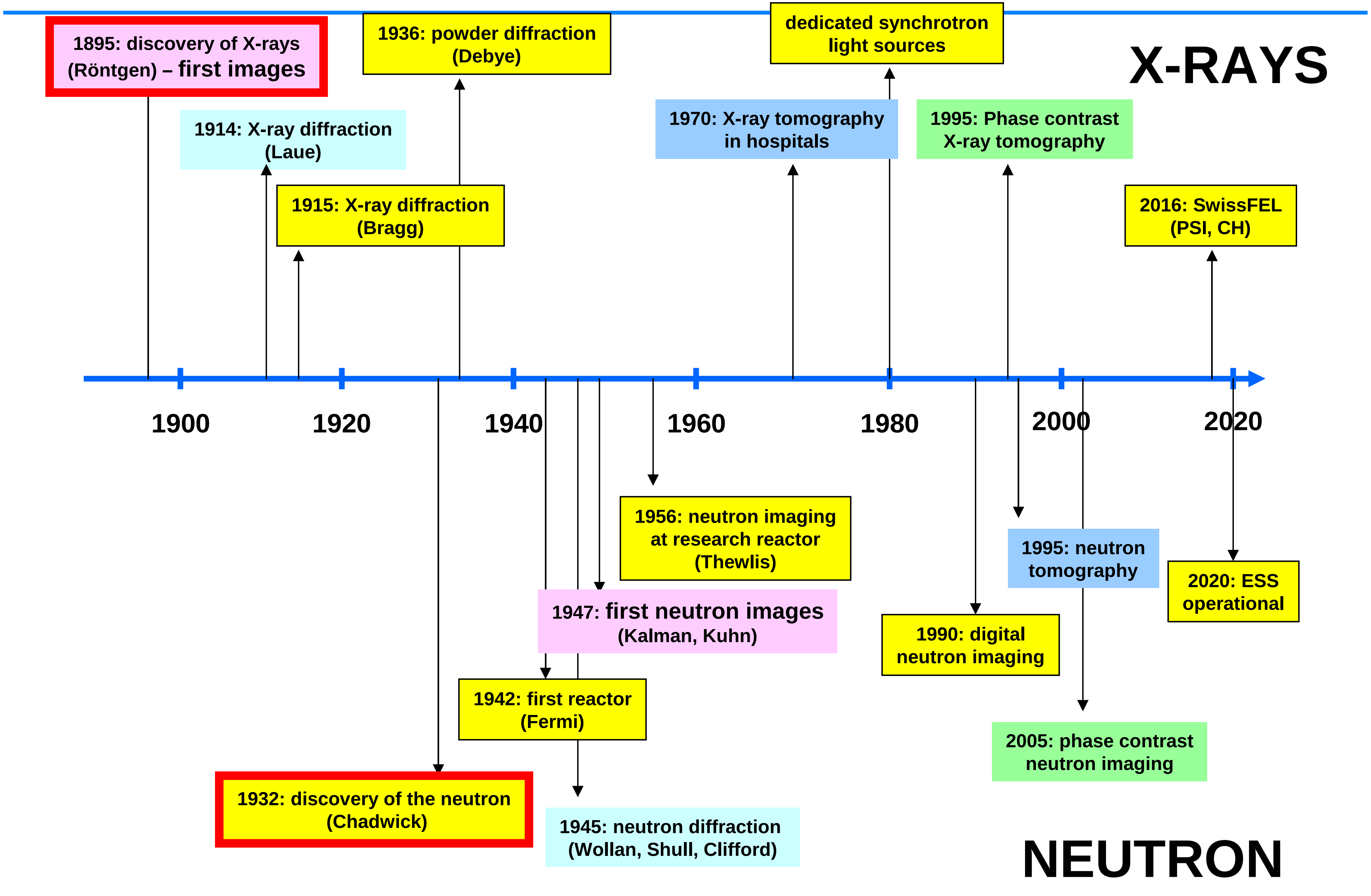
- In operation since 1997
- Driven by 590 MeV protons on a Pb target
- Intensity about 1.4 mA, corresponding to 1MW thermal power
- Installations for research with thermal and cold neutrons

Still the world's strongest stationary spallation source

Neutron Imaging TODAY: Definition

- Dedicated beam line at a (most) powerful neutron source → intensity
- Well defined thermal or cold spectrum ←→ Quantification
- Best possible beam collimation (L/D>100) ←→ high resolution
- Reasonable large field-of-view (diameter > 10 cm) - homogenous
- DIGITAL IMAGING DETECTION SYSTEM →**tomography enabled**
- Experimental infrastructure (remote control of processes, radiation protection, access control, …)
- Prepared for user access

Neutrons vs. X-rays
competition - and complement

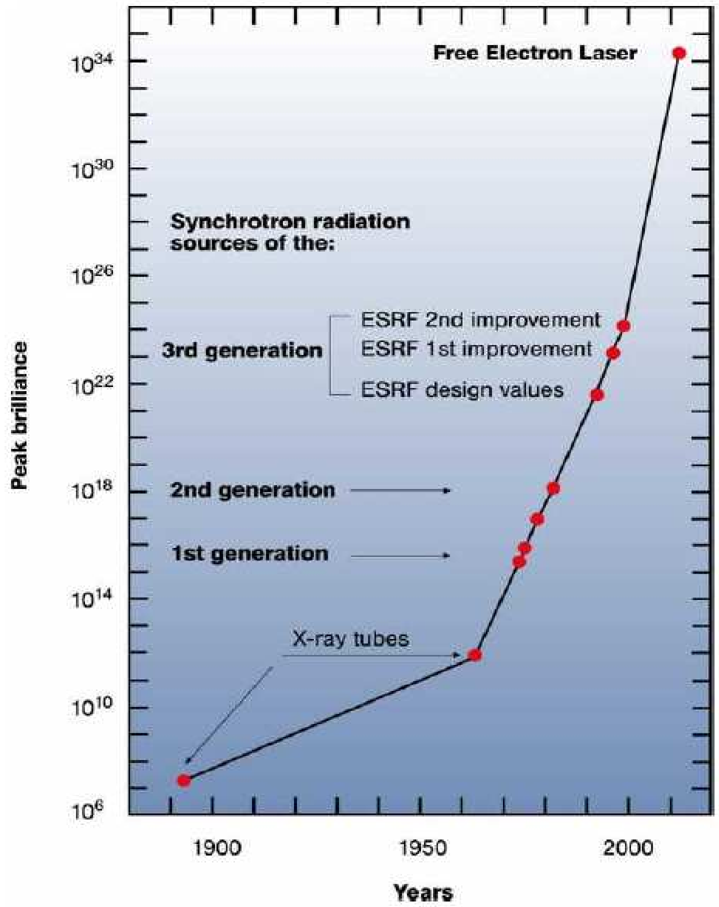

1895: discovery of X-rays (Röntgen) – first images
1936: powder diffraction (Debye)
dedicated synchrotron light sources
X-RAYS
1914: X-ray diffraction (Laue)
1970: X-ray tomography in hospitals
1995: Phase contrast X-ray tomography
1915: X-ray diffraction (Bragg)
2016: SwissFEL (PSI, CH)
1900
1920
1940
1960
1980
2000
2020
1956: neutron imaging at research reactor (Thewlis)
1995: neutron tomography
2020: ESS operational
1947: first neutron images (Kalman, Kuhn)
1990: digital neutron imaging
1942: first reactor (Fermi)
2005: phase contrast neutron imaging
1932: discovery of the neutron (Chadwick)
1945: neutron diffraction (Wollan, Shull, Clifford)
NEUTRON

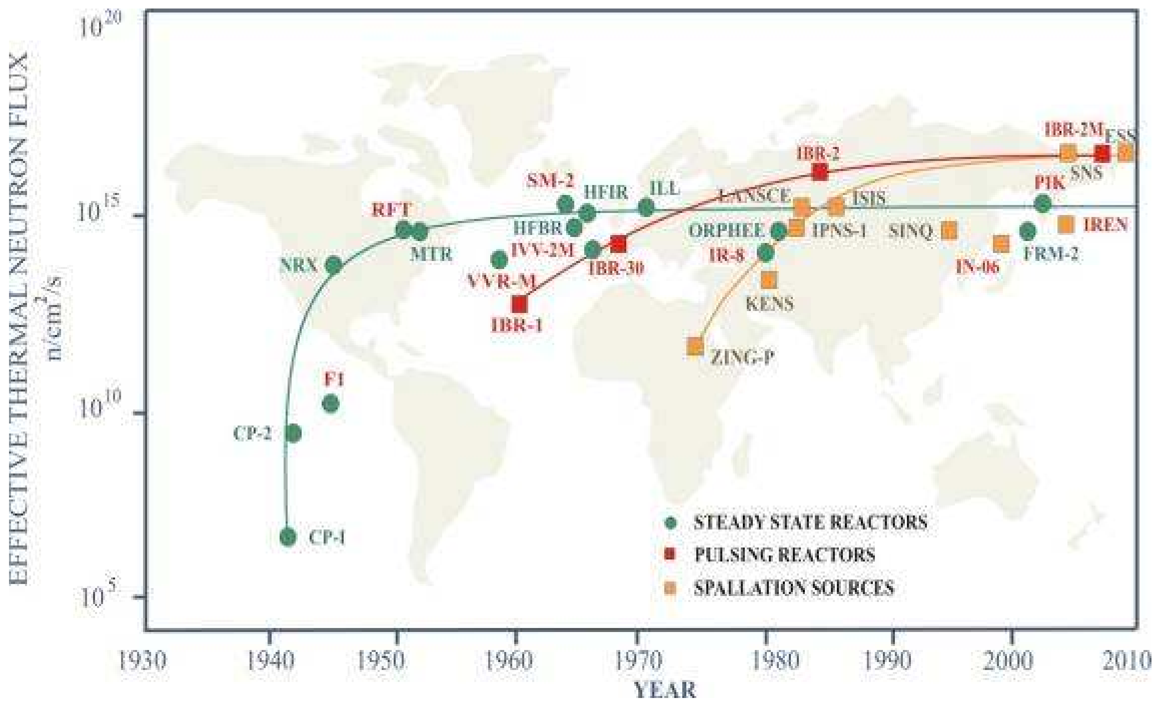
X-rays vs. neutrons

- ***free neutrons*** were discovered **37 years** after the X-rays were found
- ***neutron imaging*** started **50 years** after first X-ray images were made
- ***neutron diffraction*** comes **30 years** later than X-ray diffraction
- ***neutron tomography*** comes **25 years** later than X-ray tomography in hospitals
- ***phase contrast imaging*** with neutrons comes **10 years** later than with X-rays
- ***neutron imaging*** is **now** a competitive and complementary method compared to the X-ray techniques

→ **BUT: there are upcoming further gaps …** e.g. coherence

“Brilliance” of synchrotron and neutron sources





neutron sources are intensity limited!

source: Anatoly M. Balagurov

Frank Laboratory of Neutron Physics, JINR, Dubna, Russia

NEUTRON BEAMS

strong neutron sources: reactors (1 MW – 50 MW);
spallation neutron sources ~ 1MW
(but more efficient in n yields)

source intensity: max. flux: 10^{15} cm^{-2} s^{-1} (fast → thermal)

beam intensity: **10^{7} cm^{-2} s^{-1} to 10^{10} cm^{-2} s^{-1}** of thermal or cold neutrons (white beam – Maxwellian)

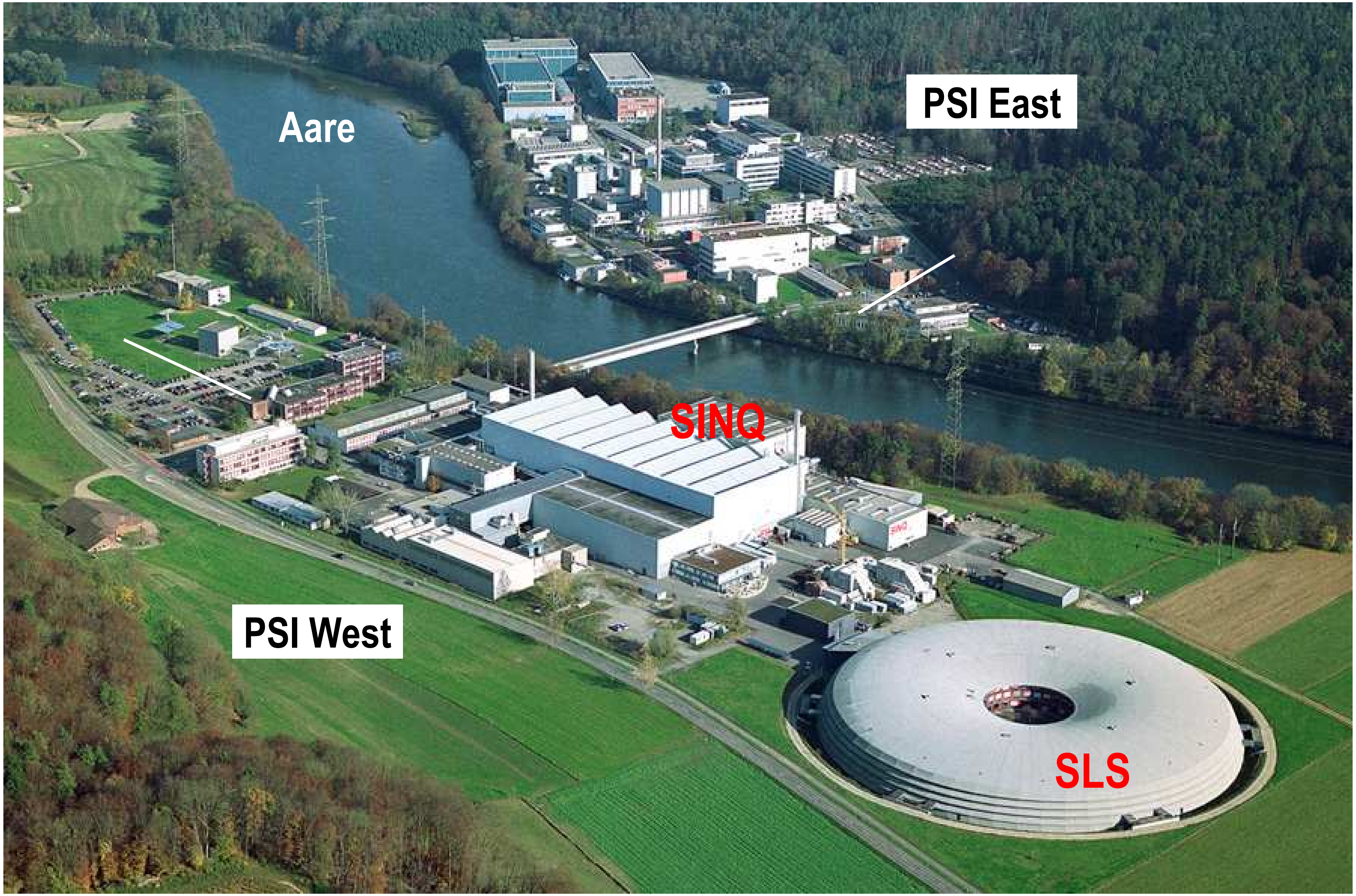
collimation: by sorting out the divergent part

coherence: limited and not (yet) an issue for research

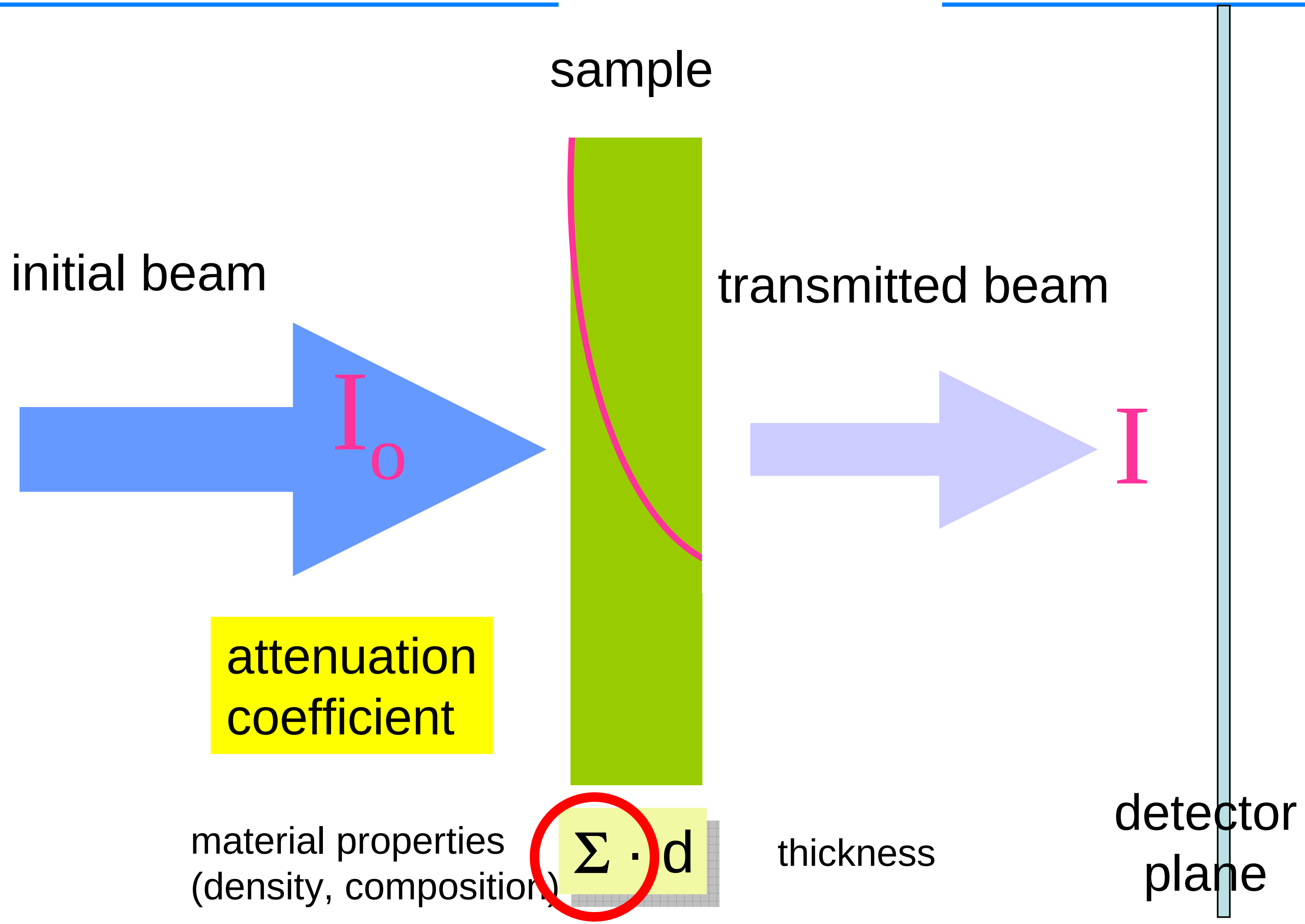
for comparison: brilliance SLS: 10^{18} photons/s/mm²/mrad²/0.1%BW

FUTURE: because no/few new reactor based sources are built,
better usage of existing facilities needed

Paul Scherrer Institut and its large scale facilities

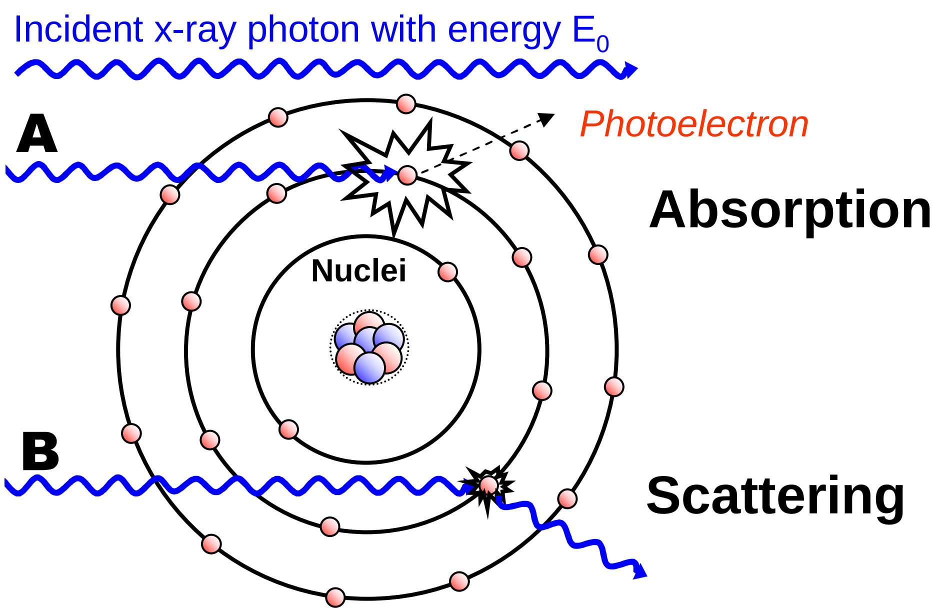


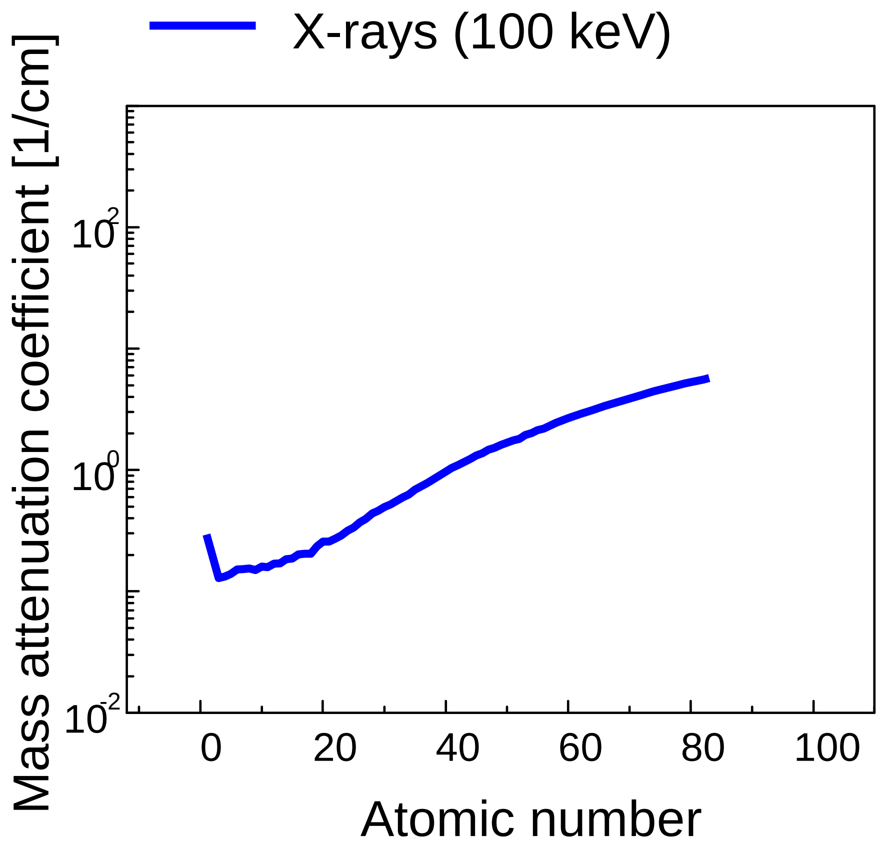
Attenuation in transmission mode



X-rays vs. Neutronen

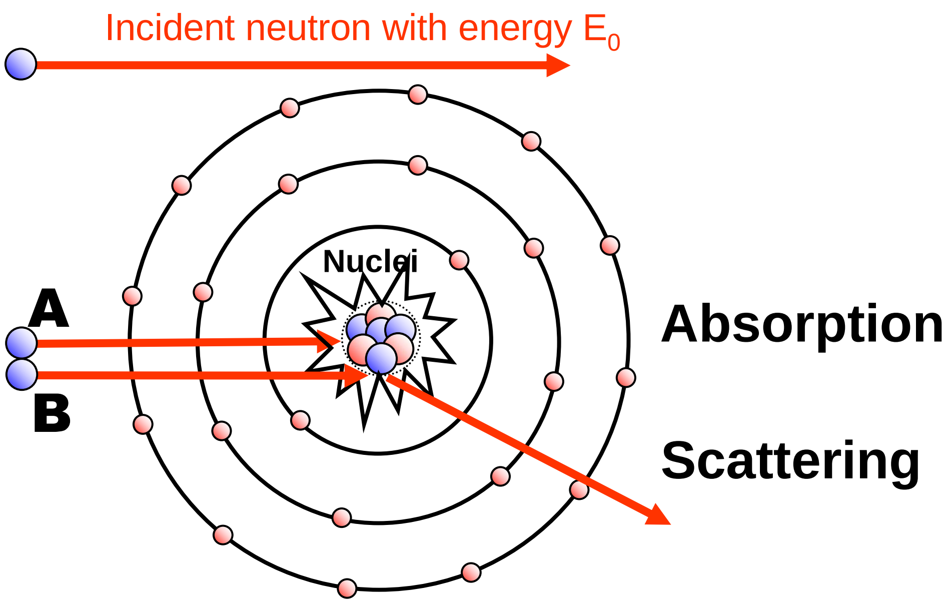
X-Rays

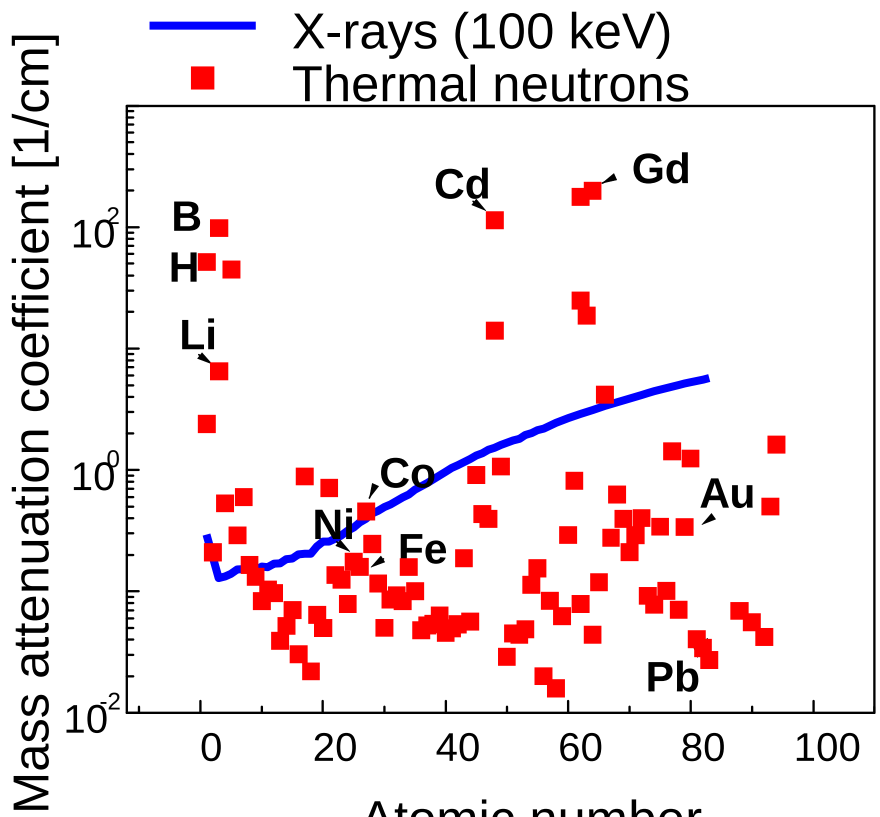




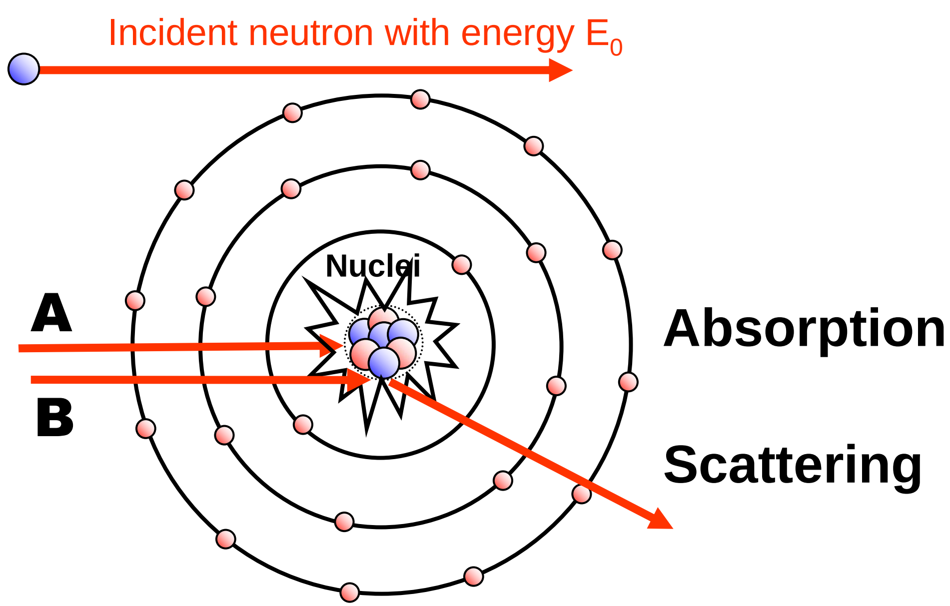
X-rays vs. Neutronen

Neutrons

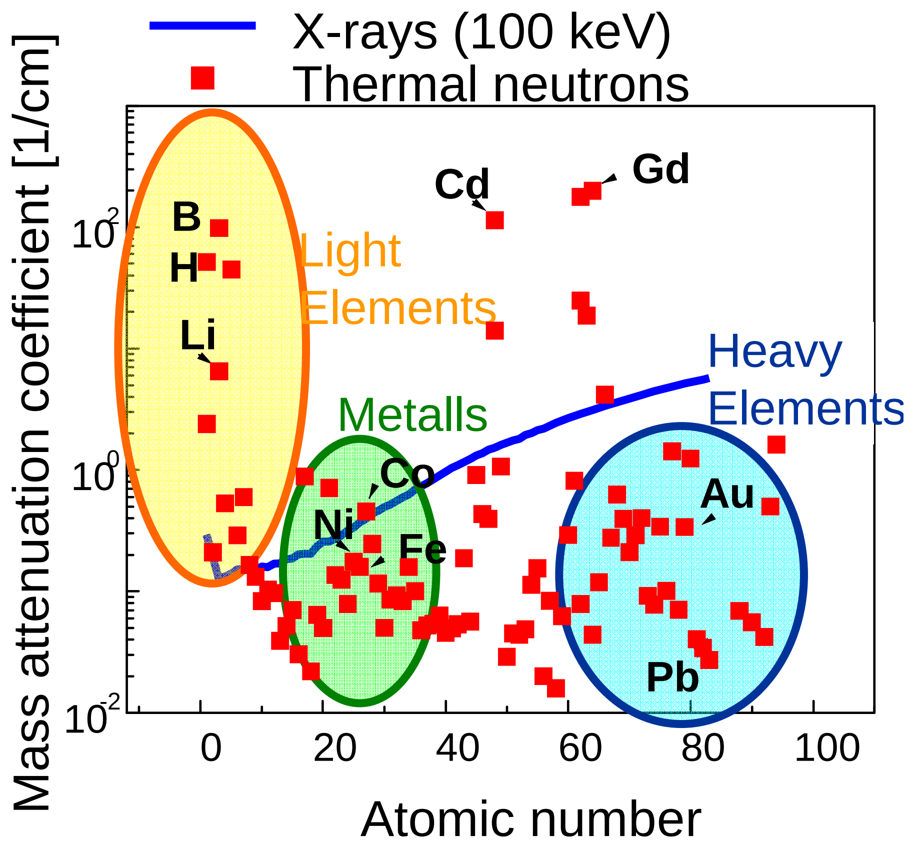




Neutrons



Incident neutron with energy E_0
Nuclei
A
B
Absorption
Scattering



X-rays (100 keV)
Thermal neutrons
Mass attenuation coefficient [1/cm]
10^2
10^0
10^{-2}
0
20
40
60
80
100
Atomic number
B
H
Li
Light Elements
Cd
Gd
Metalls
Co
Ni
Fe
Heavy Elements
Au
Pb

Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	H 0.02																	He 0.02
2	Li 0.06	Be 0.22											B 0.28	C 0.27	N 0.11	O 0.16	F 0.14	Ne 0.17
3	Na 0.13	Mg 0.24											Al 0.38	Si 0.33	P 0.25	S 0.30	Cl 0.23	Ar 0.20
4	K 0.14	Ca 0.26	Sc 0.48	Ti 0.73	V 1.04	Cr 1.29	Mn 1.32	Fe 1.57	Co 1.78	Ni 1.96	Cu 1.97	Zn 1.64	Ga 1.42	Ge 1.33	As 1.50	Se 1.23	Br 0.90	Kr 0.73
5	Rb 0.47	Sr 0.86	Y 1.61	Zr 2.47	Nb 3.43	Mo 4.29	Tc 5.06	Ru 5.71	Rh 6.08	Pd 6.13	Ag 5.67	Cd 4.84	In 4.31	Sn 3.98	Sb 4.28	Te 4.06	I 3.45	Xe 2.53
6	Cs 1.47	Ba 2.73		Hf 19.70	Ta 25.47	W 30.49	Re [illegible]	Os 37.92	Ir 39.01	Pt 38.61	Au 35.94	Hg [illegible]	Tl 23.23	Pb 22.81	Bi 20.28	Po 20.22	At -	Rn 9.77
7	Fr -	Ra 11.80		Rf -	Db -	Sg -	Bh -	Hs -	Mt -	Ds -	Rg -	Uub -	Uut -	Uuq -	Uup -	Uuh -	Uus -	Uuo -

Lanthanides	La 5.04	Ce 5.79	Pr 6.23	Nd 6.46	Pm 7.33	Sm 7.68	Eu 5.66	Gd 8.69	Tb 9.46	Dy 10.17	Ho 10.17	Er 11.70	Tm 12.49	Yb 9.32	Lu 14.07
Actinides	Ac 24.47	Th 28.95	Pa 39.65	U 49.08	Np -	Pu -	Am -	Cm -	Bk -	Cf -	Es -	Fm -	Md -	No -	Lr -

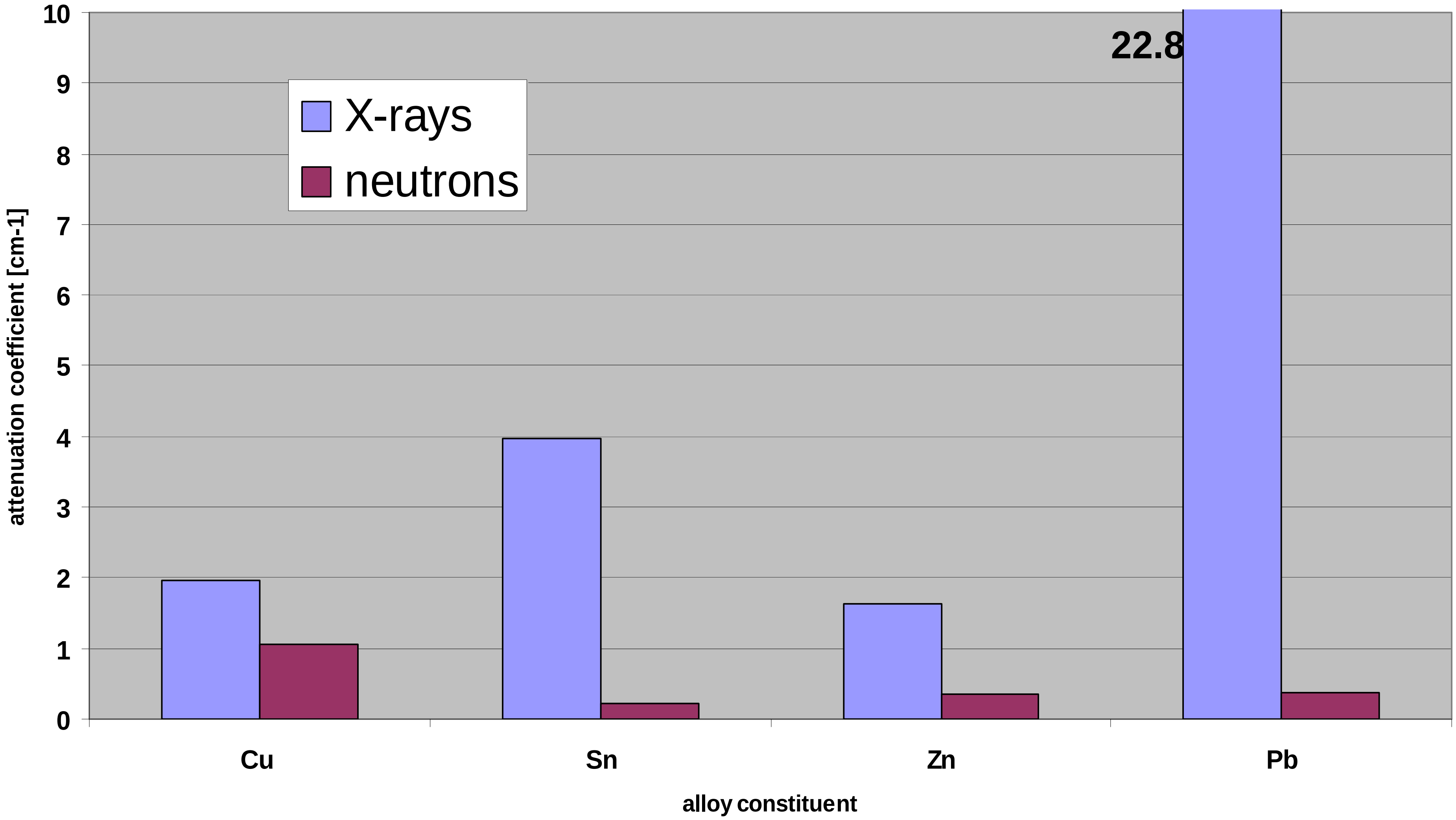
X-ray

Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	H 3.44																	He 0.02
2	Li 3.30	Be 0.79											B 101.6	C 0.56	N 0.43	O 0.17	F 0.20	Ne 0.10
3	Na 0.09	Mg 0.15											Al 0.1	Si 0.11	P 0.12	S 0.06	Cl 1.33	Ar 0.03
4	K 0.06	Ca 0.08	Sc 2.00	Ti 0.60	V 0.72	Cr 0.54	Mn 1.21	Fe 1.19	Co 3.92	Ni 2.05	Cu 1.07	Zn 0.35	Ga 0.49	Ge 0.47	As 0.67	Se 0.73	Br 0.24	Kr 0.61
5	Rb 0.08	Sr 0.14	Y 0.27	Zr 0.29	Nb 0.40	Mo 0.52	Tc 1.76	Ru 0.58	Rh 10.88	Pd 0.78	Ag 4.04	Cd 115.1	In 7.58	Sn 0.21	Sb 0.30	Te 0.25	I 0.23	Xe 0.43
6	Cs 0.29	Ba 0.07		Hf 4.99	Ta 1.49	W 1.47	Re 6.85	Os 2.24	Ir 30.46	Pt 1.46	Au 6.23	Hg 16.21	Tl 0.47	Pb 0.38	Bi 0.27	Po -	At -	Rn -
7	Fr -	Ra 0.34		Rf -	Db -	Sg -	Bh -	Hs -	Mt -	Ds -	Rg -	Uub -	Uut -	Uuq -	Uup -	Uuh -	Uus -	Uuo -

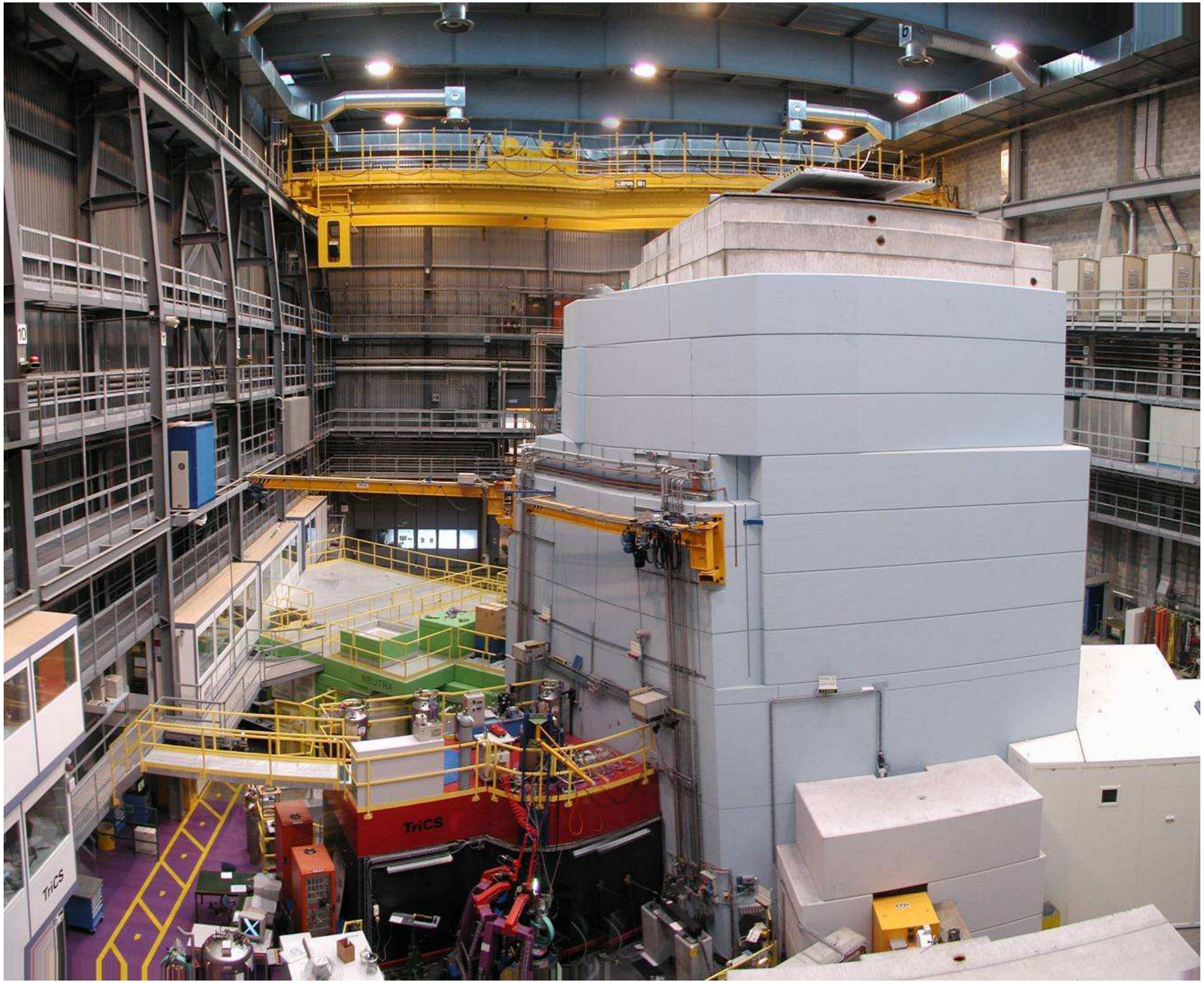
Lanthanides	La 0.52	Ce 0.14	Pr 0.41	Nd 1.87	Pm 5.72	Sm 171.47	Eu 94.58	Gd 1479.0	Tb 0.93	Dy 32.42	Ho 2.25	Er 5.48	Tm 3.53	Yb 1.40	Lu 2.75
Actinides	Ac -	Th 0.59	Pa 8.46	U 0.82	Np 9.80	Pu 50.20	Am 2.86	Cm -	Bk -	Cf -	Es -	Fm -	Md -	No -	Lr -

thermal neutrons

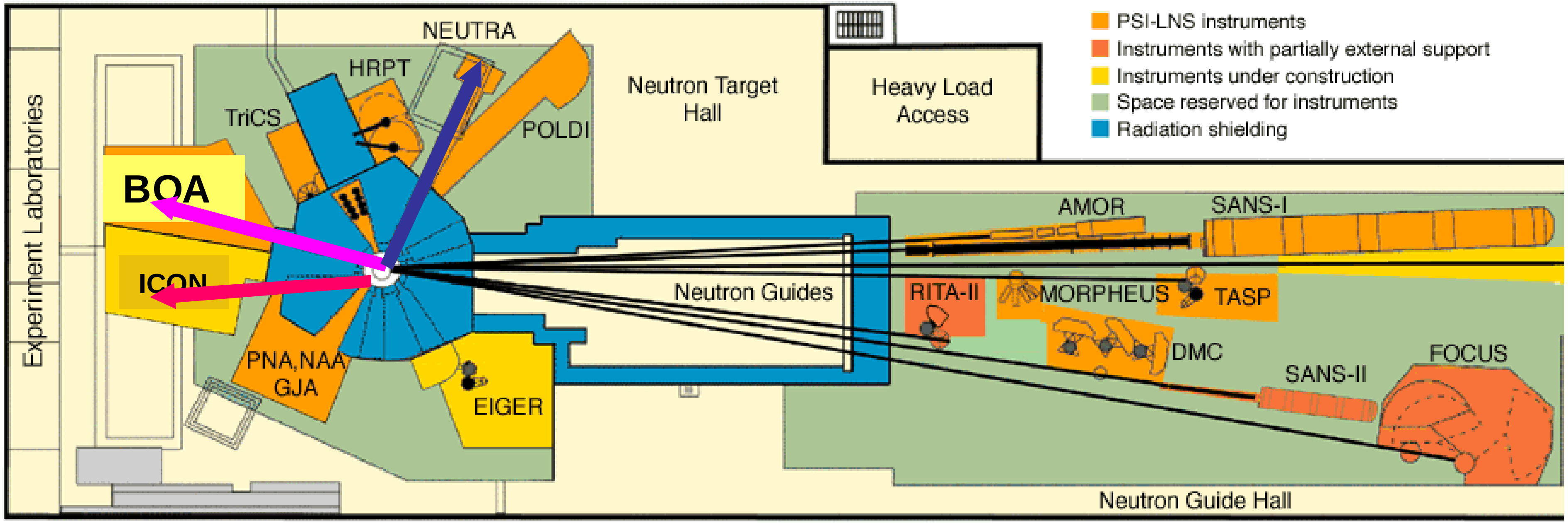
Attenauation properties of bronze for X-rays and neutrons



Spallation neutron source SINQ @ PSI

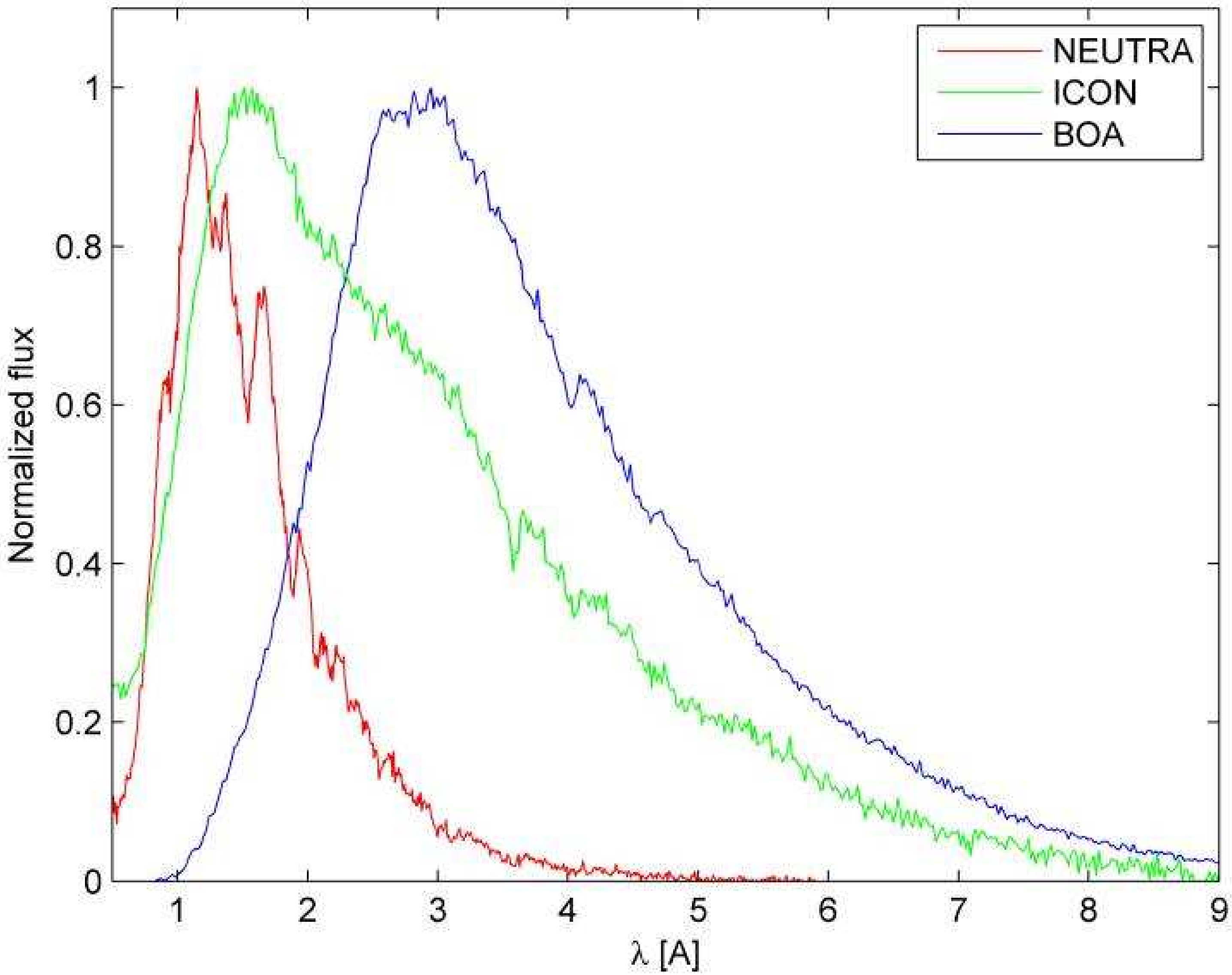


SINQ – Layout, Imaging Beam Lines



NEUTRA:	thermal neutrons
ICON:	cold neutrons
BOA:	very cold (polarized) neutrons

Spectra at PSI/SINQ Neutron Imaging beamlines



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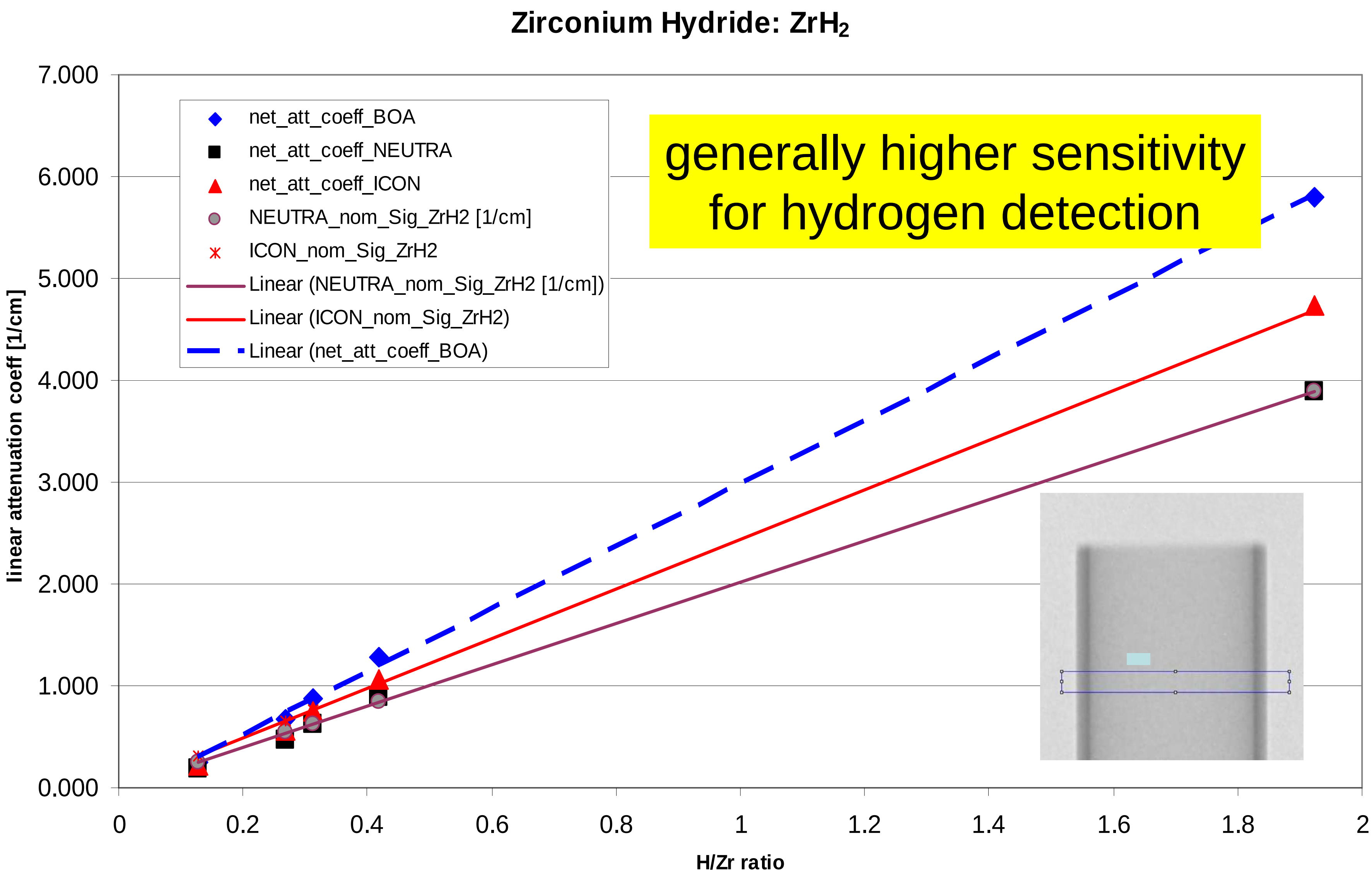
Attenuation coefficients in dependence on hydrogen content



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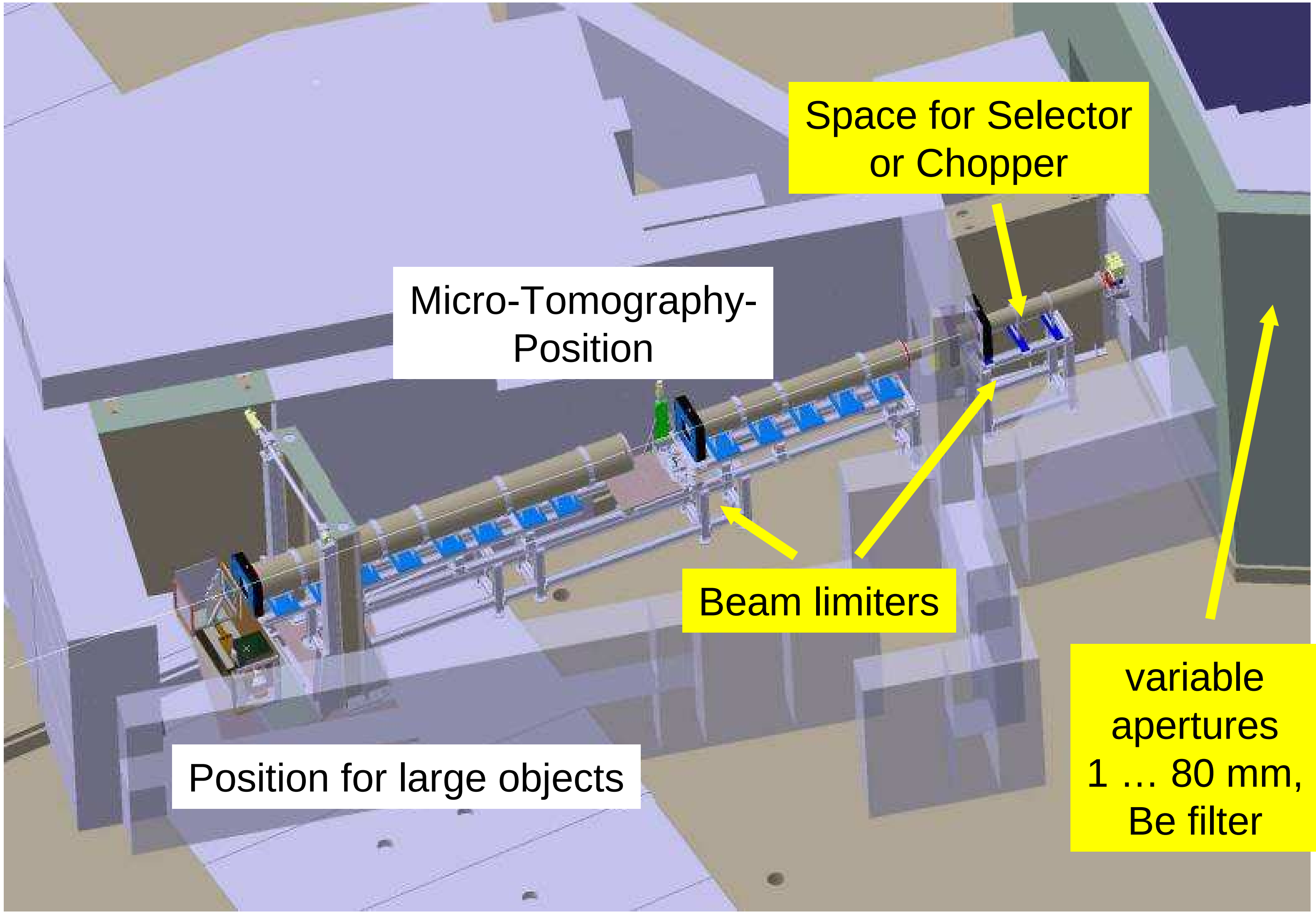
ICON-beam line @ SINQ



Methods in neutron imaging

current base line:

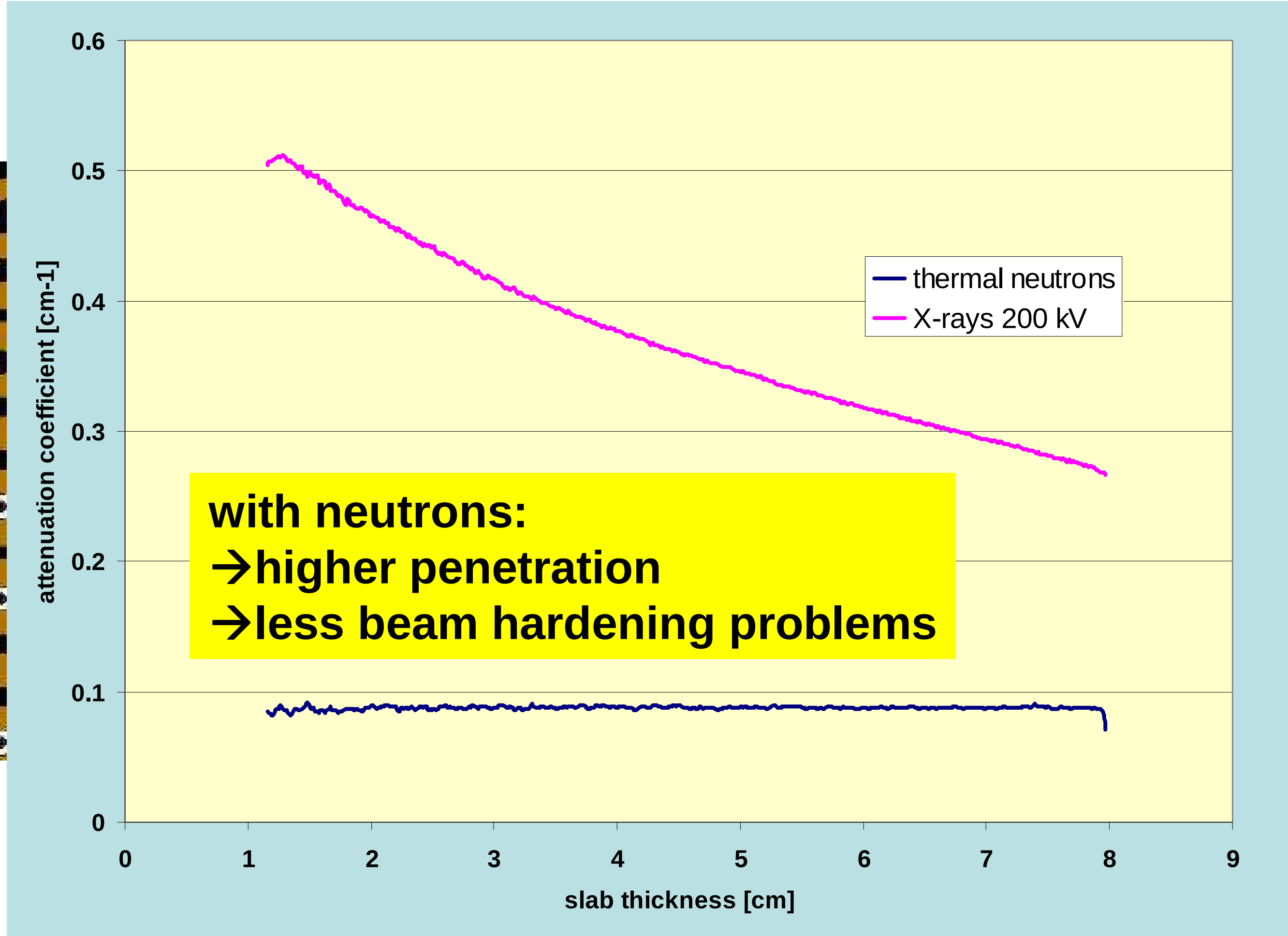
•digital
•2D and 3D
•with white cold or thermal beams
•on macro (40 cm Ø) and micro scales (13 µm pixel size)

new approaches:

•energy selection (selection devices, TOF)
•time-dependence (sequential or stroboscopic)
•diffractive imaging
•neutron interferometry (phase and “dark-field” imaging)
•edge enhancement by neutron refraction
•data fusion (e.g. to X-ray imaging)
•resonance imaging with epithermal neutrons
•polarized neutron imaging

Quantification in tomography

Cylinder head of a combustion engine (25 cm Al transmission length)

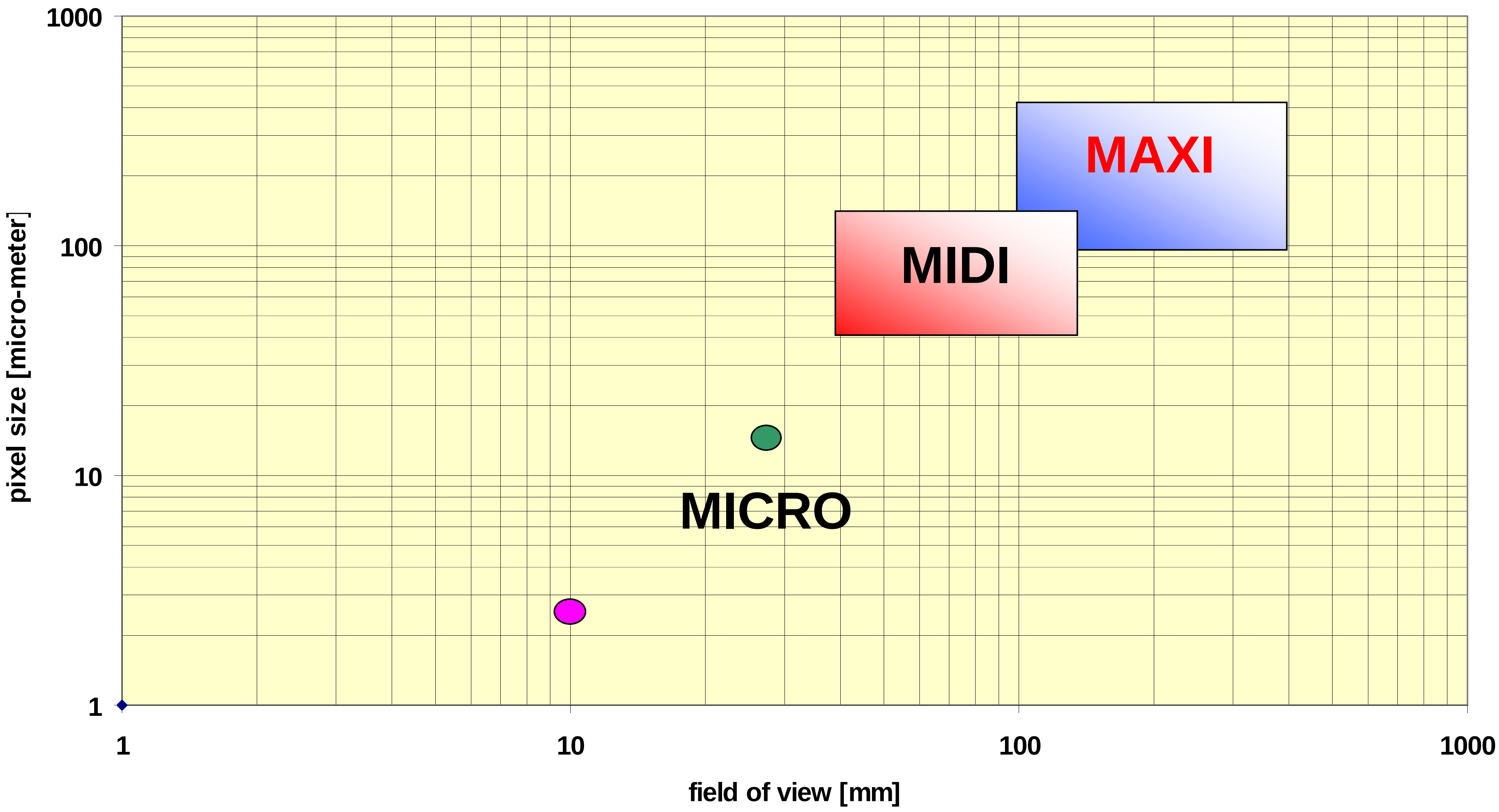


Trends in Neutron Beams

- colder neutrons
- energy selectivity → pulsed sources, higher E - resolution
- polarized neutrons
- highest possible intensities
- improvements in spatial resolution

→ **We are waiting for ESS in Lund/Sweden**

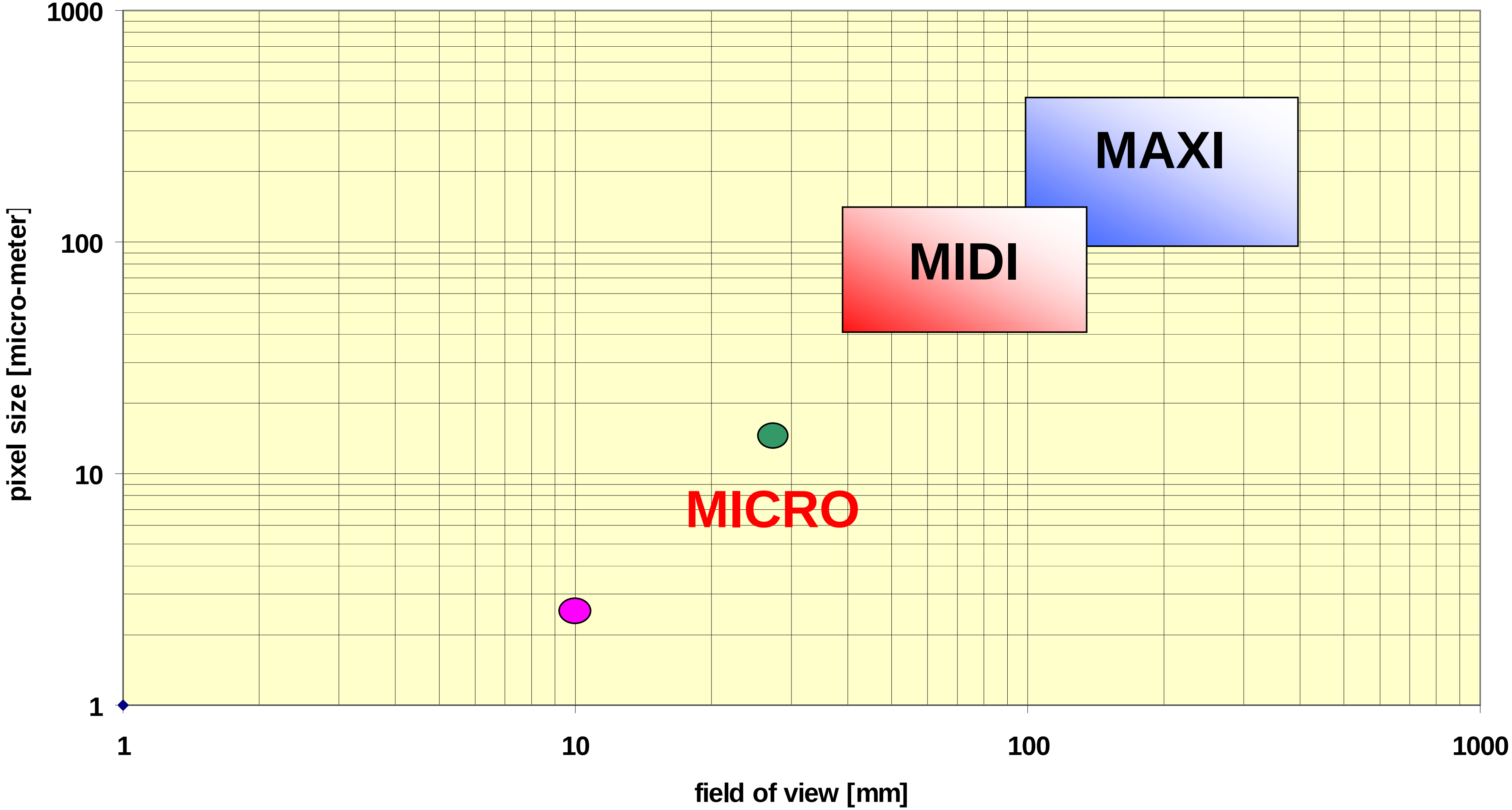
Working areas in neutron tomography



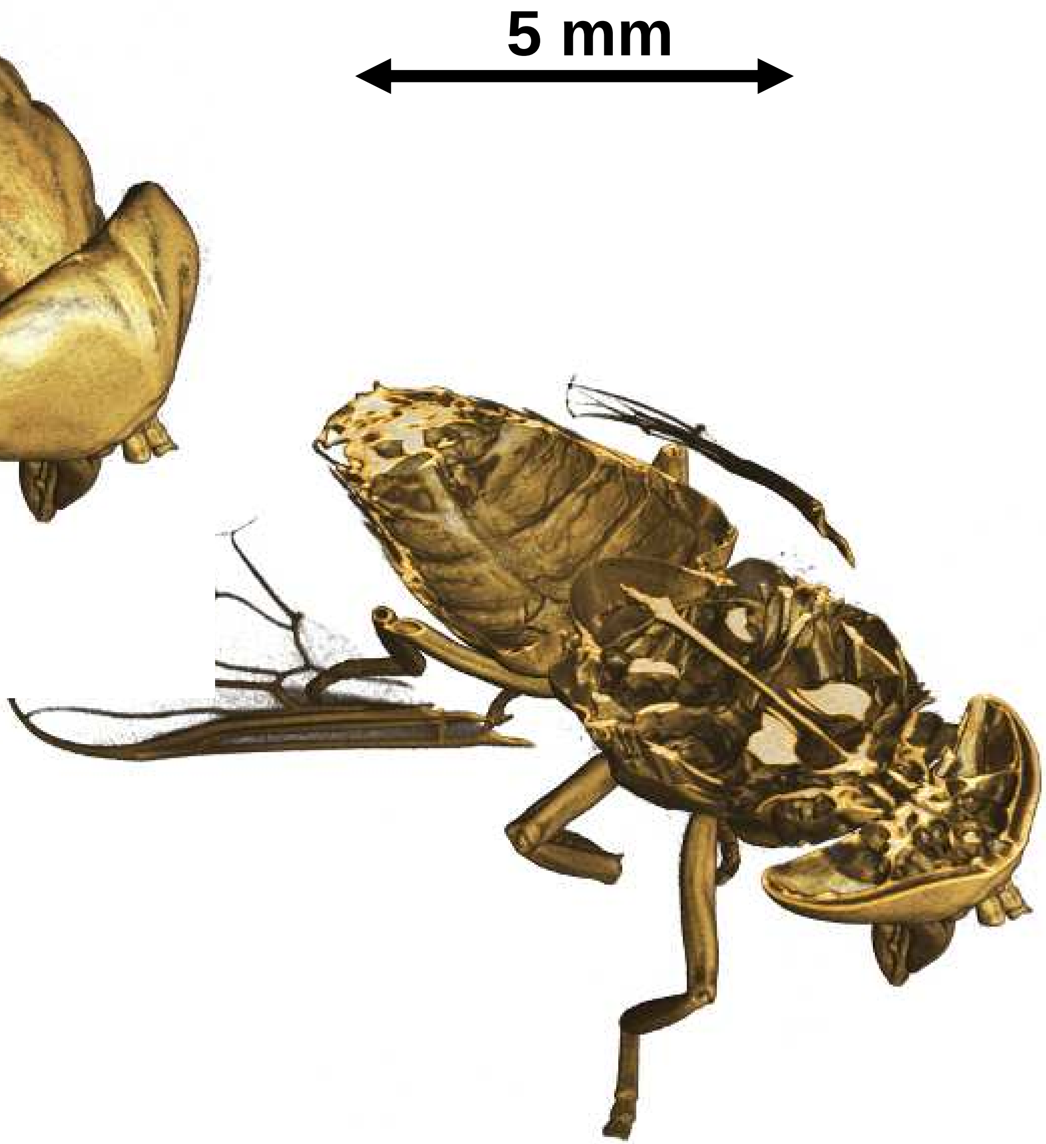
Tomo result: virtual 3D volume close to reality

10 cm

Working areas in neutron tomography

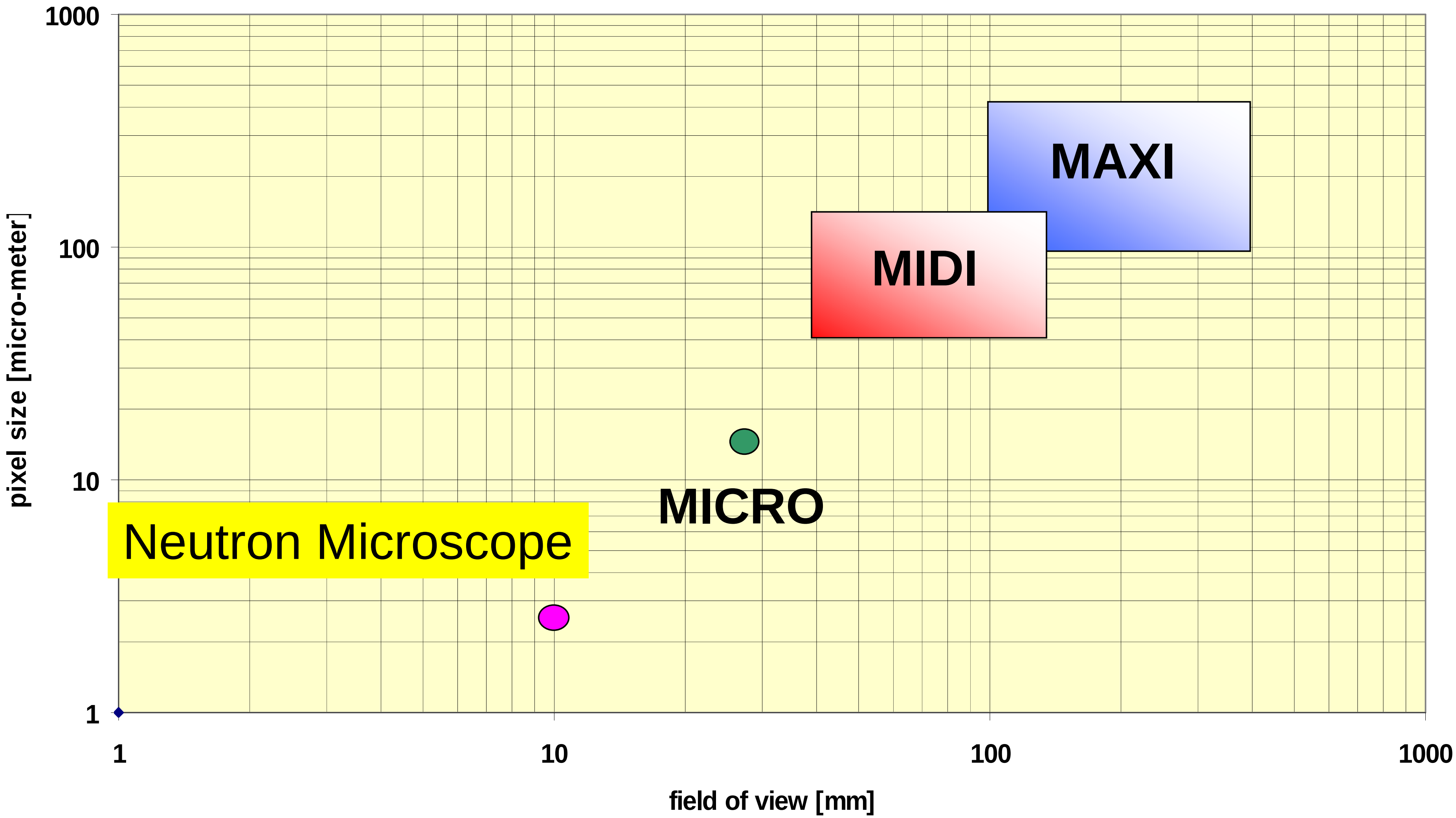


Tomography Result: Virtual Reality



Micro-Tomography
with cold neutrons

Demand to higher spatial resolution

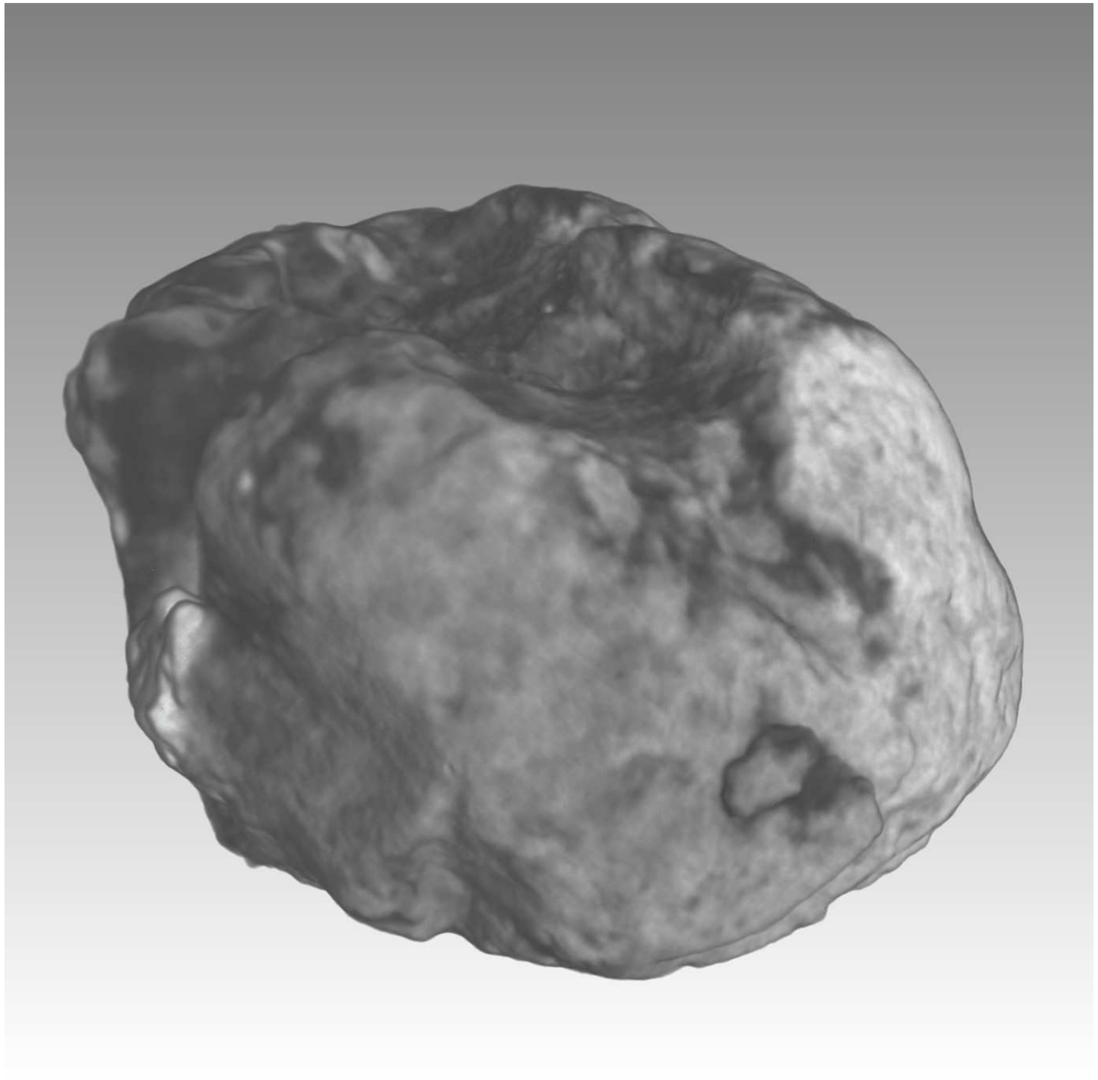


Examples for successful studies with neutrons (and X-rays)

Mary Rose – English war ship † 1545, * 500 dead ***, recovered 1982**

Canon balls from „Mary Rose“

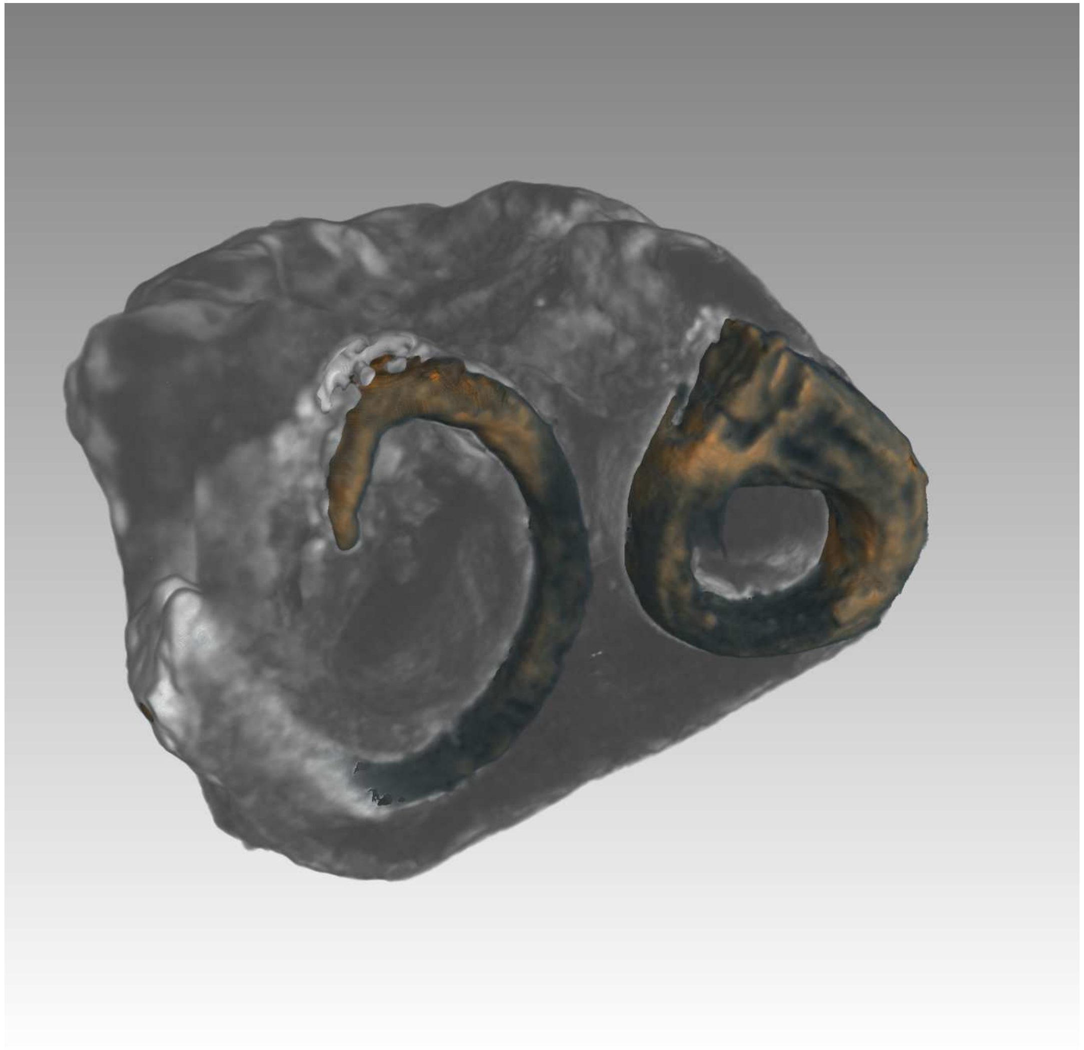
canon ball – iron core embedded in lead



impossible to perform with X-rays

Origin: England in the Middle Age → Cultural Heritage

canon ball – iron core embedded in lead



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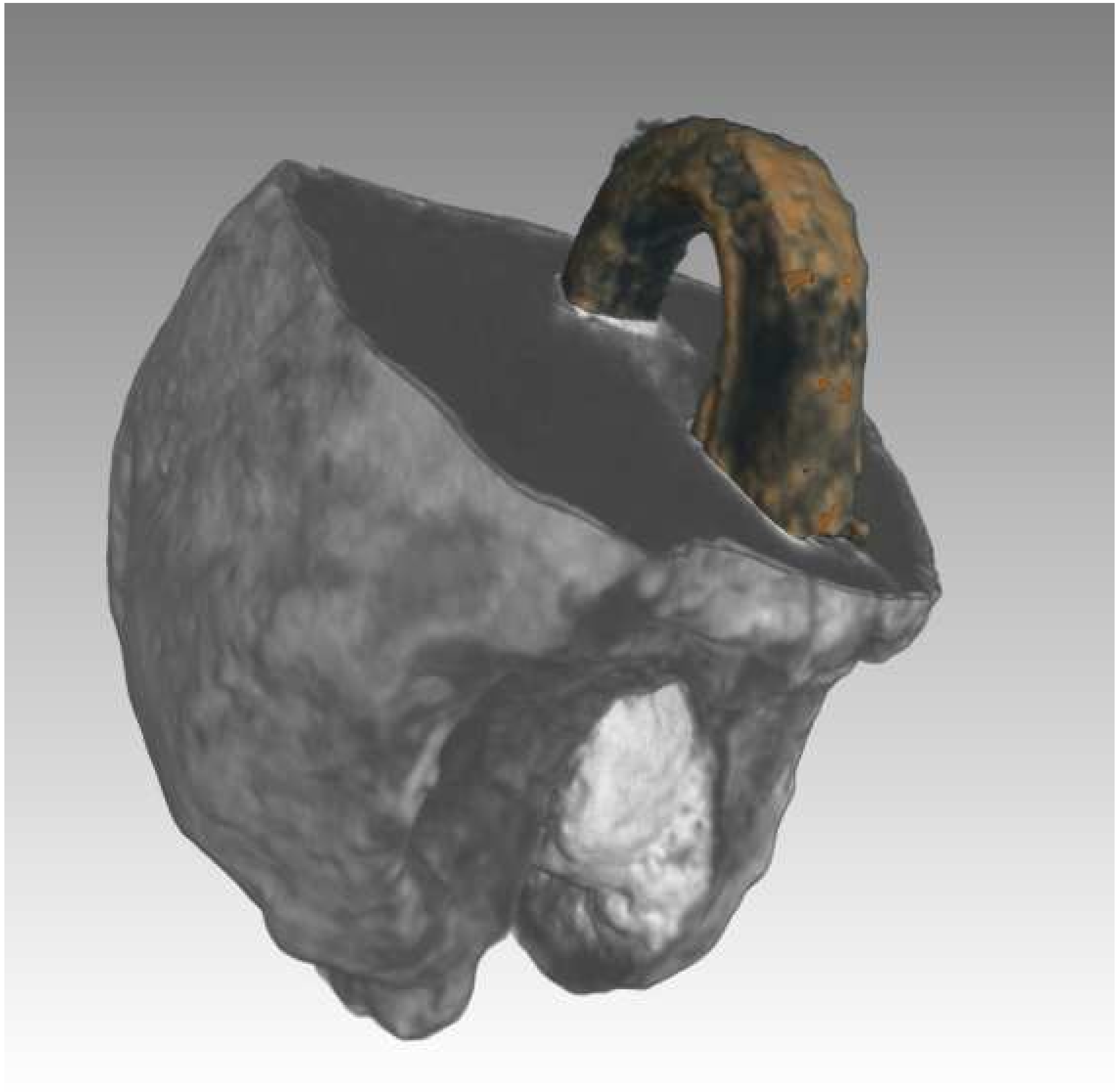

impossible to perform with X-rays

Origin: England in the Middle Age → Cultural Heritage

canon ball – iron core embedded in lead

impossible to perform with X-rays

Origin: England in the Middle Age → Cultural Heritage

Asian Buddhist bronze sculptures

size: ~ 20 cm

age: ~ 500 years

material: bronze

filling: unknown

X-ray inspection

Neutron transmission imaging

Buddha Sakyamuni, Bhumisparsa Mudra, Westtibet, 14.- 15. century

Buddha Sakyamuni, Bhumisparsa Mudra, Westtibet, 14.- 15. century

radiography

tomography slice

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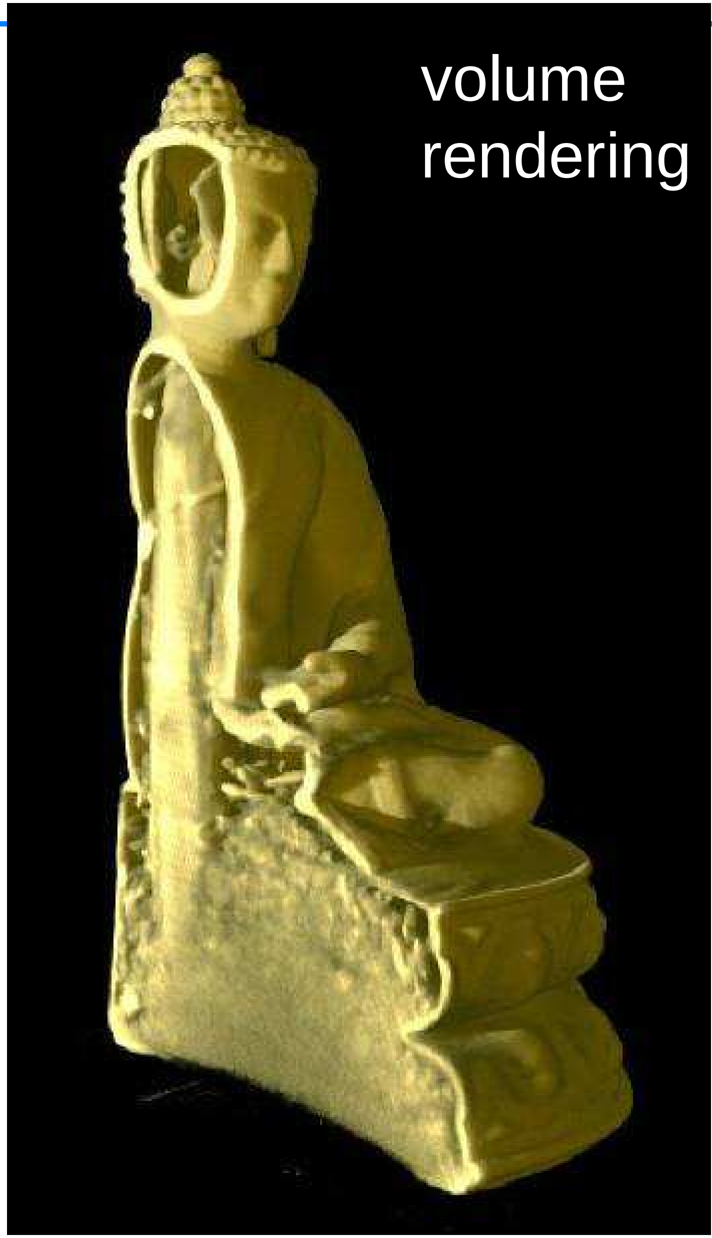
Buddha Sakyamuni, Bhumisparsa Mudra, Westtibet, 14.- 15. century



E.H. Lehmann, S. Hartmann, M.O. Speidel,
Investigation of the content of ancient Tibetan metallic Buddha statues by means of neutron imaging methods, Archaeometry 52, 3 (2010) 416-428

Neutron imaging of *The violinist,* lead sculpture by Pablo Gargallo.




Registration no: MNAC 010972-000
title: The violinist
author: Pablo GARGALLO
date: 1920
Material/technique: 2mm thick lead sheets, hammered and assembled with soldering and nails on a wood structure
dimensions: 55,3 x 31,8 x 21,6 cm
weight: 11,9 kg

Neutron imaging of *The violinist,* lead sculpture by Pablo Gargallo.

State of conservation: **broken or splitting solderings**

Neutron imaging of *The violinist,* lead sculpture by Pablo Gargallo.

State of conservation: **lead corrosion going on**






Questions

1. Degree of damage

2. Origin of the corrosion

3. Strategy for restoration

→ neutron imaging as only and final hope for answers

Neutron imaging of *The violinist,* lead sculpture by Pablo Gargallo.

✓ **Location, types and number of fixing nails**

34 iron nails and tacks hammered into the wood sculpture (there seems to be also a little screw). These nails can be seen in the radiographs, the tomographies and in the 3D reconstruction.

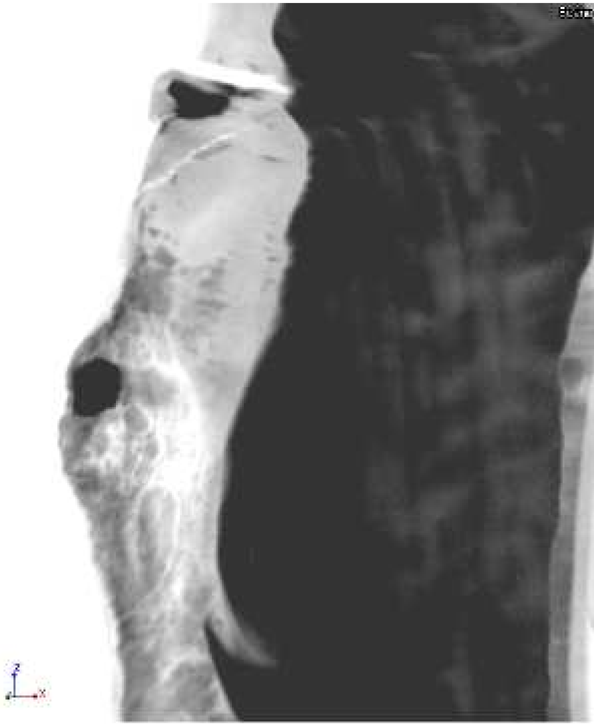
These nails can fix 1 to 3 layers of lead

There are 22 nails and tacks in the head only

Neutron imaging of *The violinist,* lead sculpture by Pablo Gargallo.

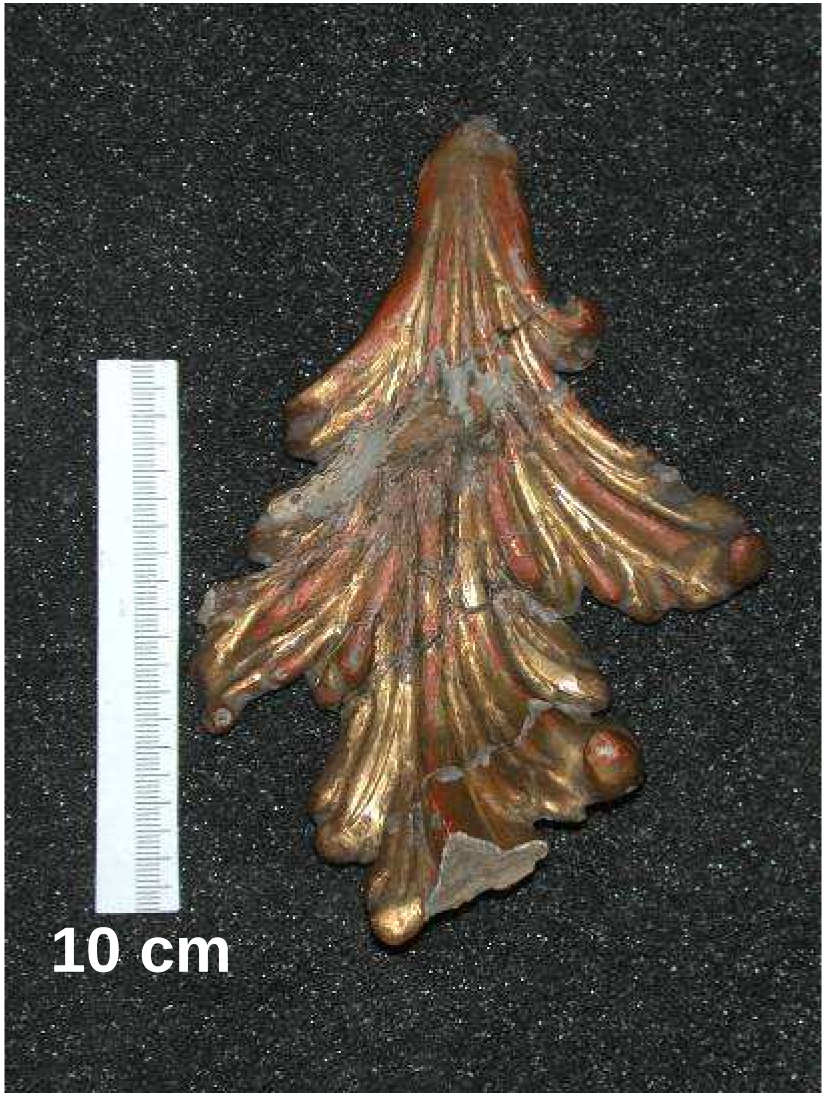
✓ Location of carbonate corrosion on the external side of right leg

•**Spot of higher attenuation (dark colour) inside the big blister on the right leg, possibly due to the sinthetic stucco used to fill in the hole left by sampling in that zone.**



Restoration work:

infiltration of resin into a leaf like wood sample



Resin injection into
a wooden sample:

leaf 15 cm long

direct run

Resin injection into
a wooden sample:

leaf 15 cm long

referenced to original state

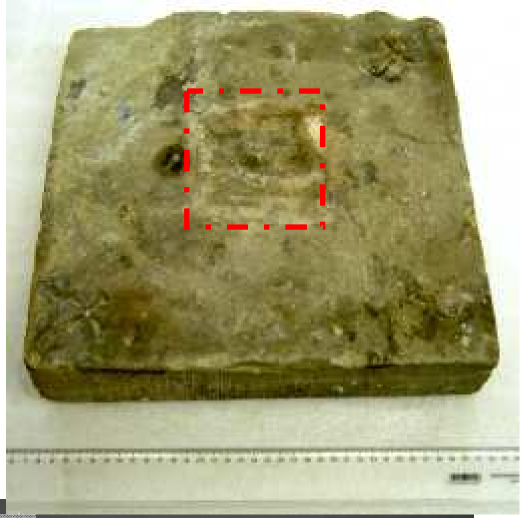
Combined neutron and X-ray imaging of an altar stone from the former Augustinian church in Fribourg/Switzerland

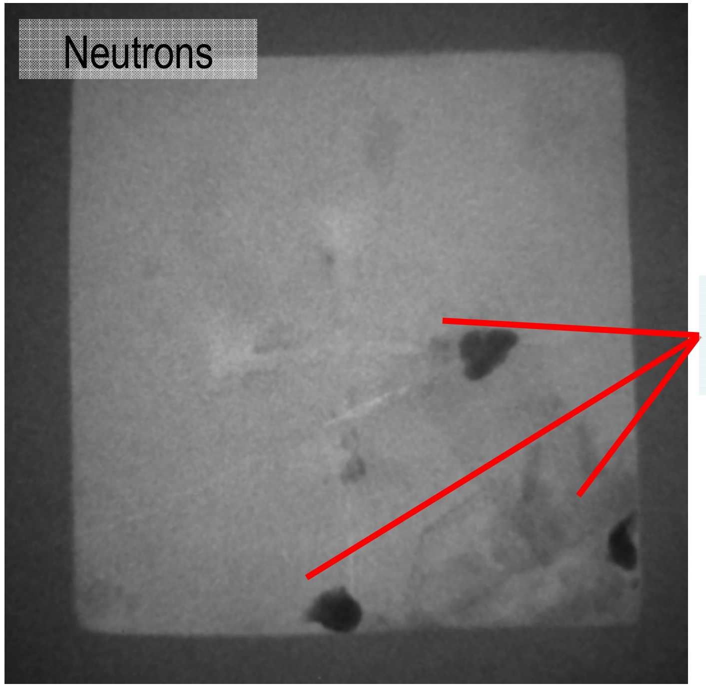
The object

Altar table (33,5cm x 33,5cm x 7cm) from a church (église des Augustins) in Fribourg (CH) with an inlay bearing a cross symbol.

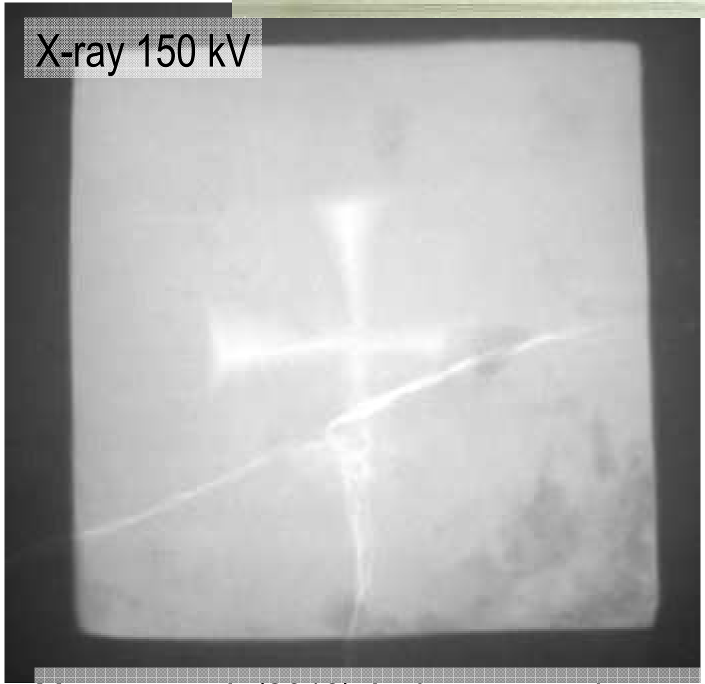
Questions

- Is beneath the inlay a hollow space / void?
- What is in the presumed void?
  Possibly mortal remains / some sort of relic





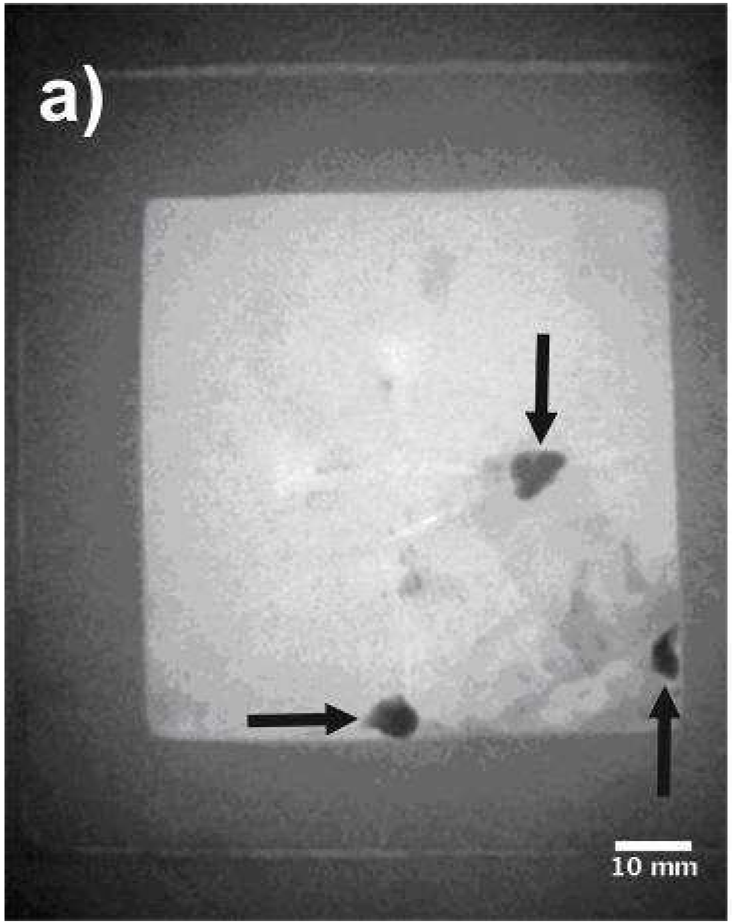
Organic particles/fragments ?



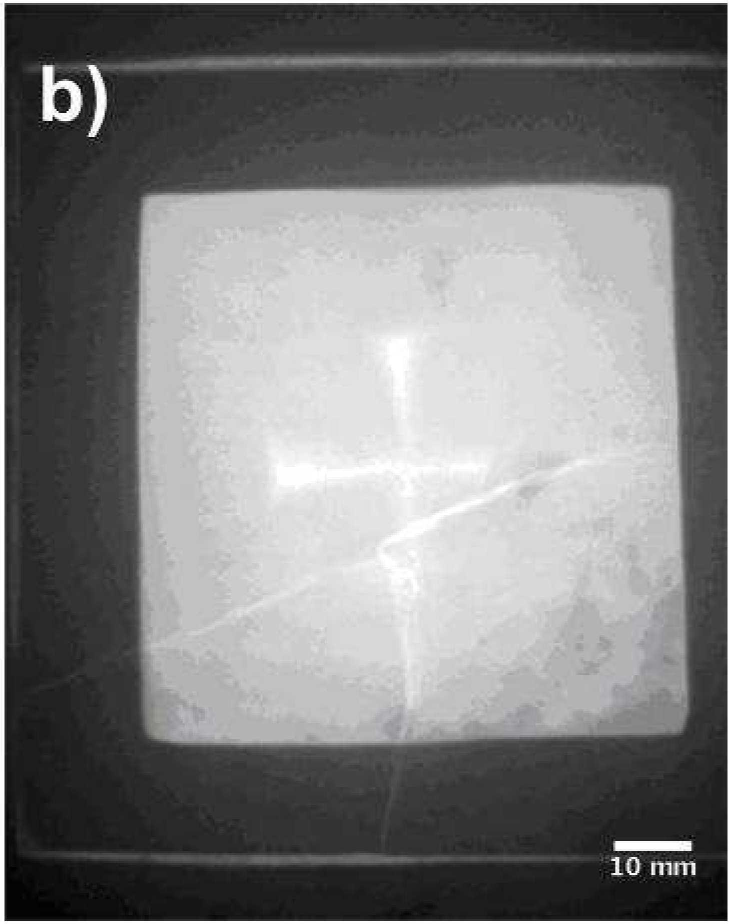
Mannes et al. (2013) Archaeometry *in press*

Transmission Image data

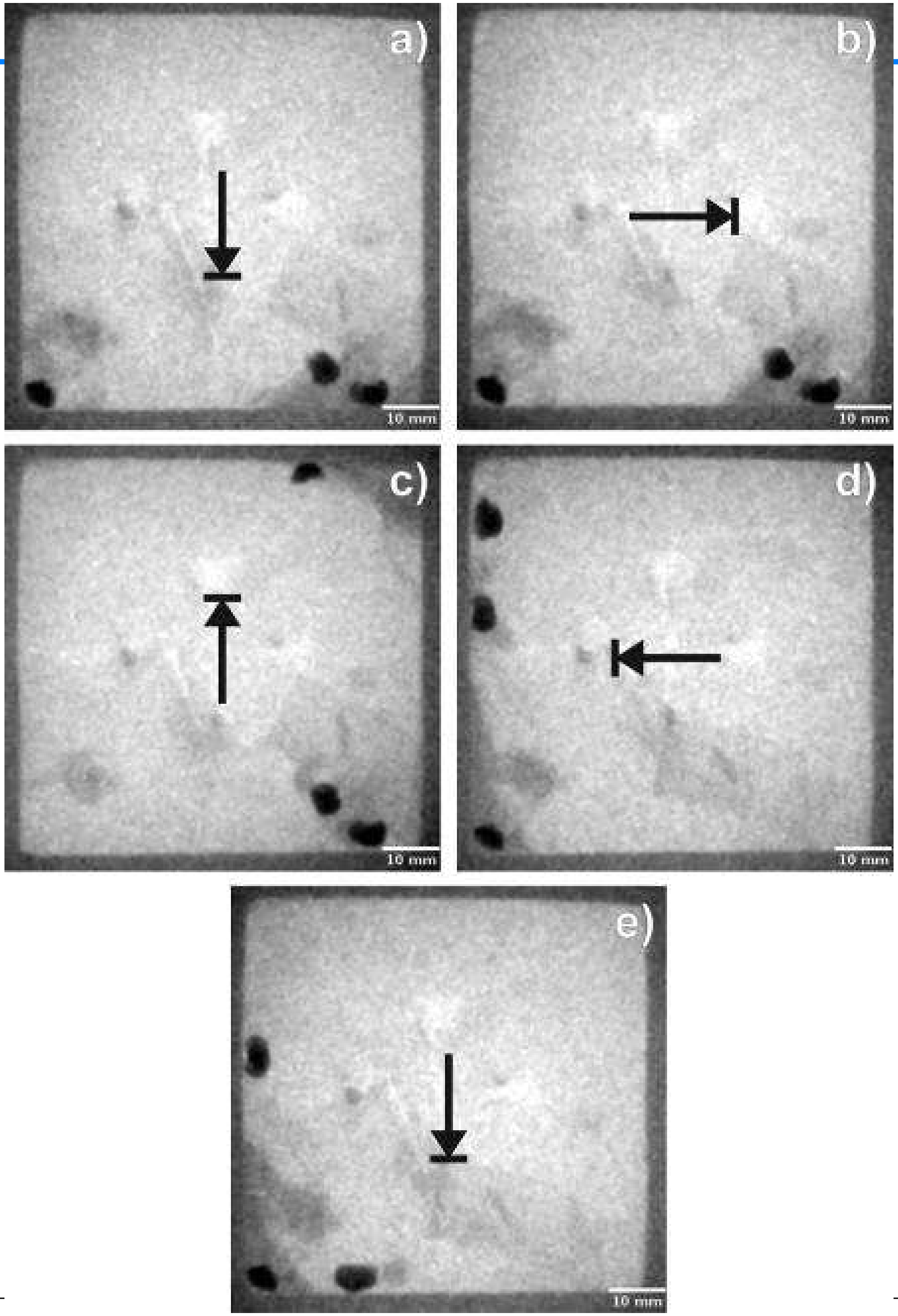
NEUTRONS



X-RAYS

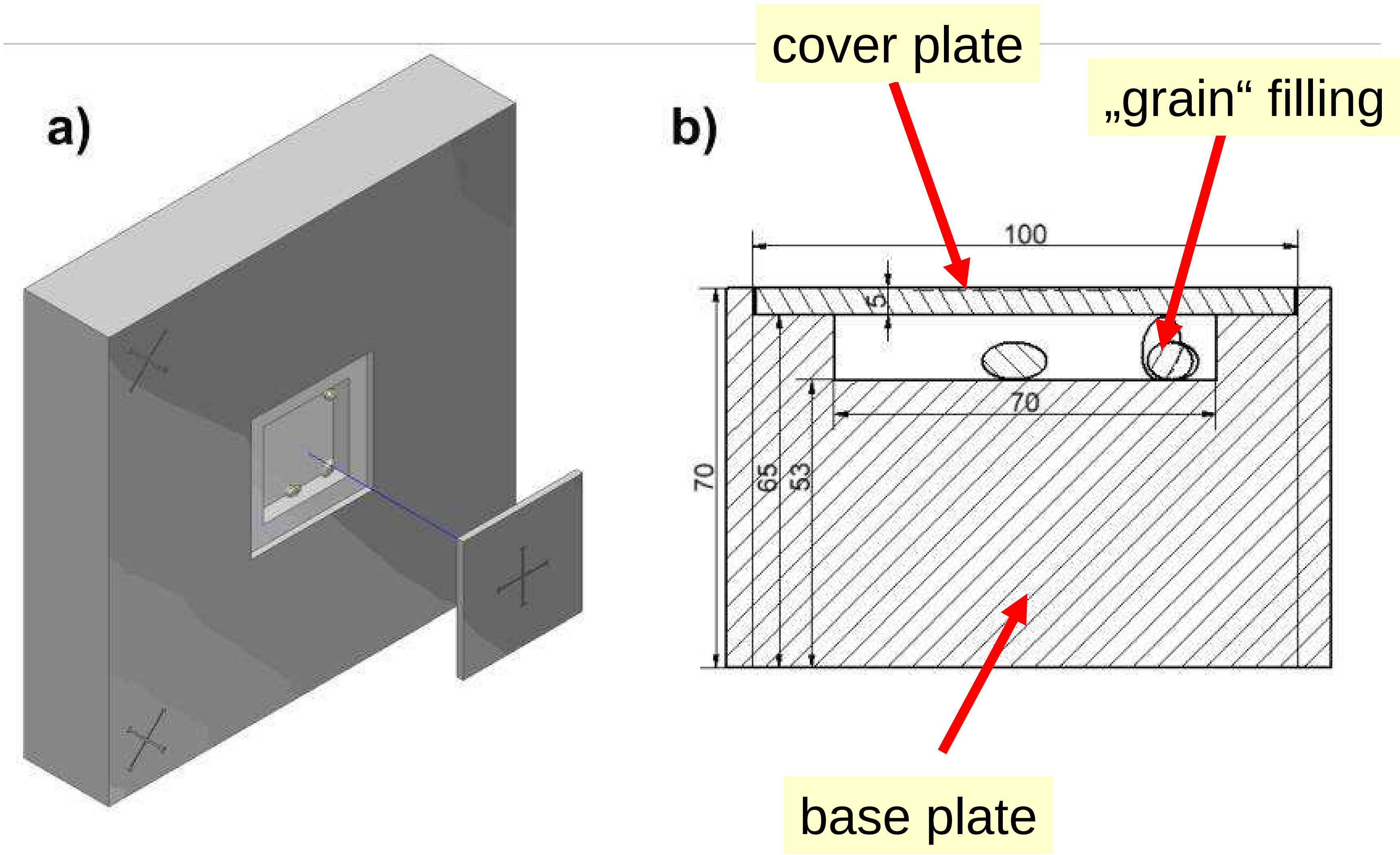


Conclusion: some **organic** material is placed inside the void space

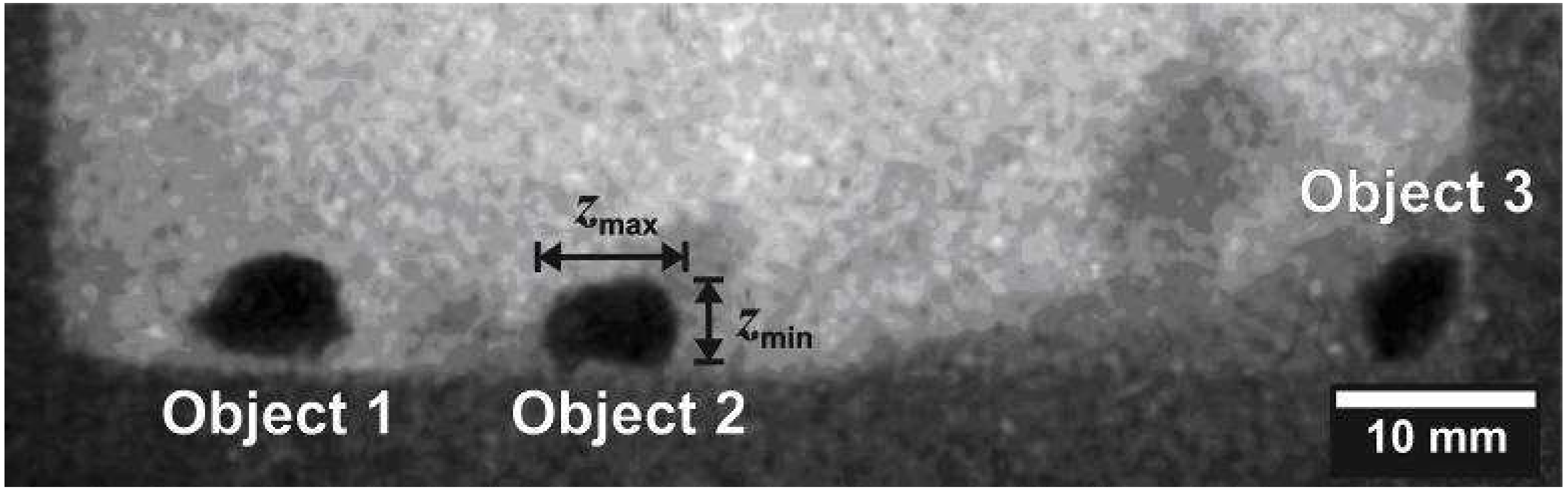


a rotation shows: the „grains“ are mobile

Reconstruction of the plate structure from the transmission data



What is the „grain“ filling material ?



	Transmission (=I/I$_0$) [-]	z_{max} [cm]	z_{min} [cm]	z_{mean} [cm]	Σ [cm^{-1}]
faceplate	0.46				
object 1	0.38	0.67	0.40	0.54	0.35
object 2	0.39	0.60	0.33	0.46	0.37
object 3	0.38	0.48	0.30	0.39	0.51

	z [cm]	T (=I/I$_0$) [-]	Σ [cm^{-1}]
Leather	0.34	0.45	2.36
Parchment	0.02	0.96	2.44
Tooth	0.90	0.29	1.38
Bone	0.87	0.65	0.57
Incense	1.08	0.09	2.23

Filling: parts of bones

Investigation of Renaissance bronze sculptures

- more than 15 sculptures from Rijksmuseum Amsterdam (NL)
- were studied with neutron tomography
- bulky structure of bronze much better to transmitt with neutron than with X-rays
- metal, ceramic remains, soldering, corrosion, repair work were identified
- information about provenance, manufacturing, damage, conservation state derived
- in a completely non-invasive way

Johan Gregor van der Schardt (1530-after 1581),
cast by Georg Labenwolf (?-1585)

Sol (The Sun)

Nuremberg, c. 1570-1581

Height 45.7 cm

Rijksmuseum, Amsterdam

Inv. no. BK-1977-24

PROJECT COORDINATION

Robert van Langh
Dick Visser

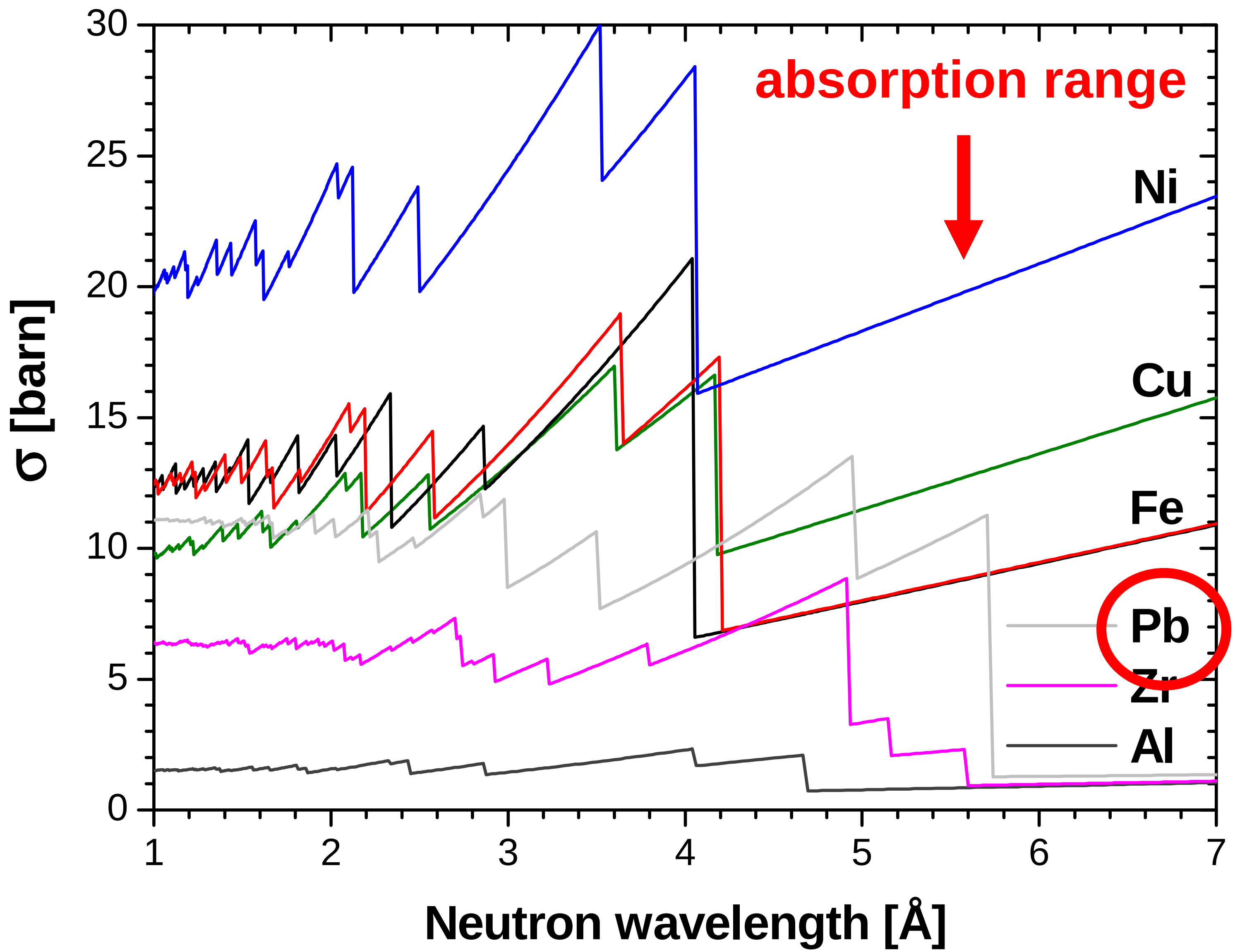
SPECIAL THANKS TO

Eberhard Lehmann
Peter Vontobel
Frits Scholten

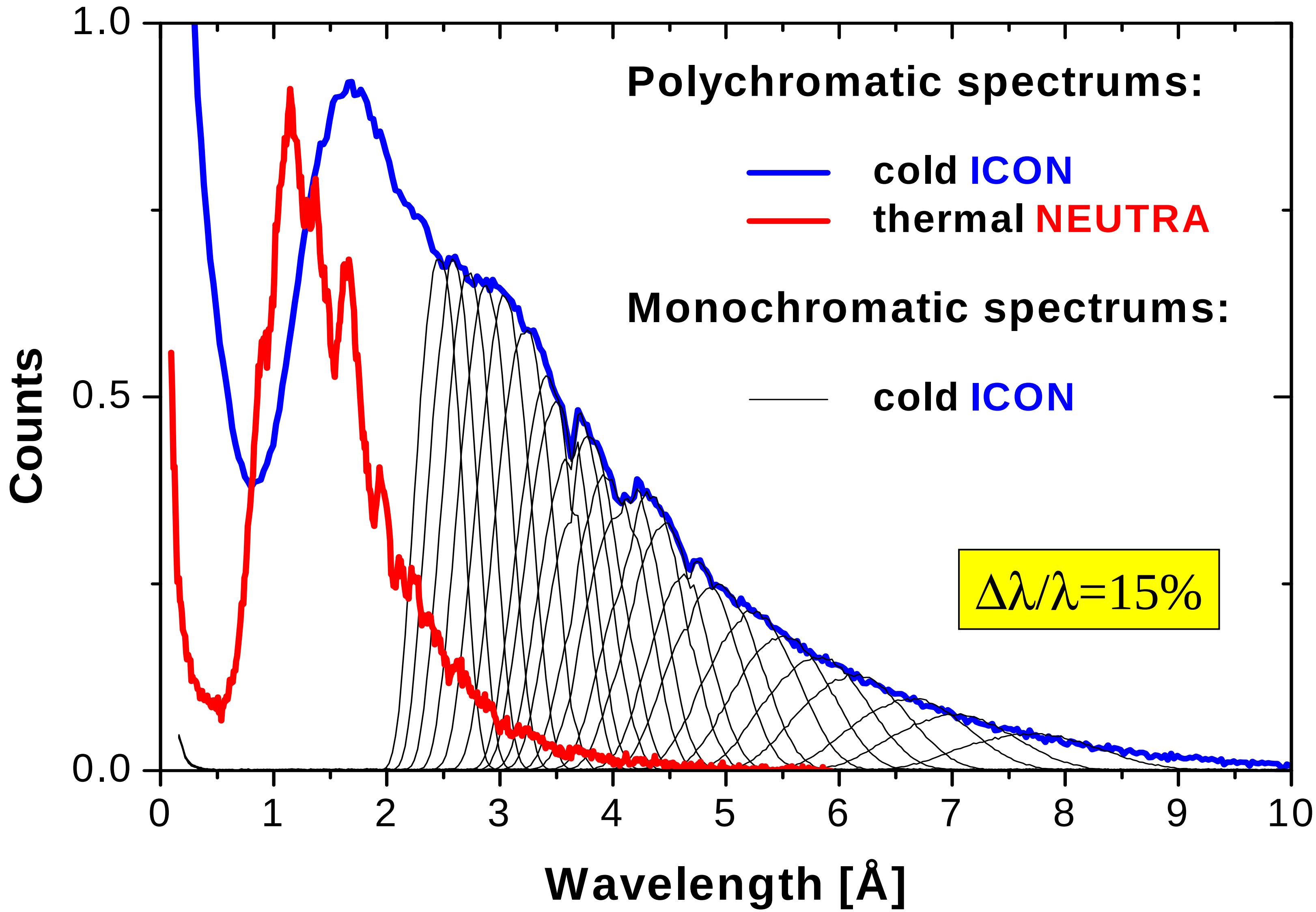
animation by Mirko Estermann

Energy-selective tomography

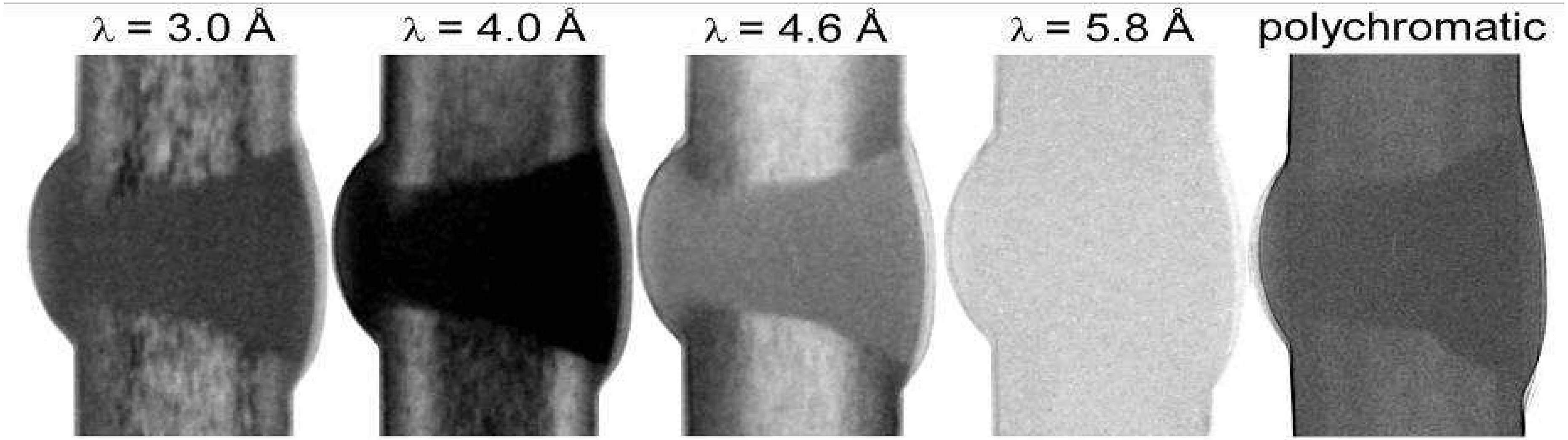
Cross-sections of crystaline structural materials



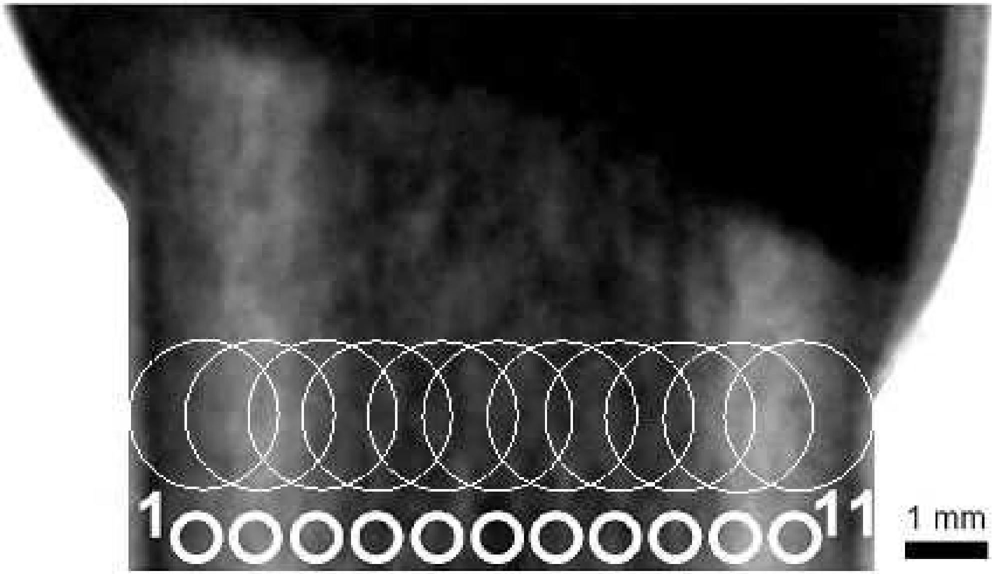
Conditions with the turbine type E-selector



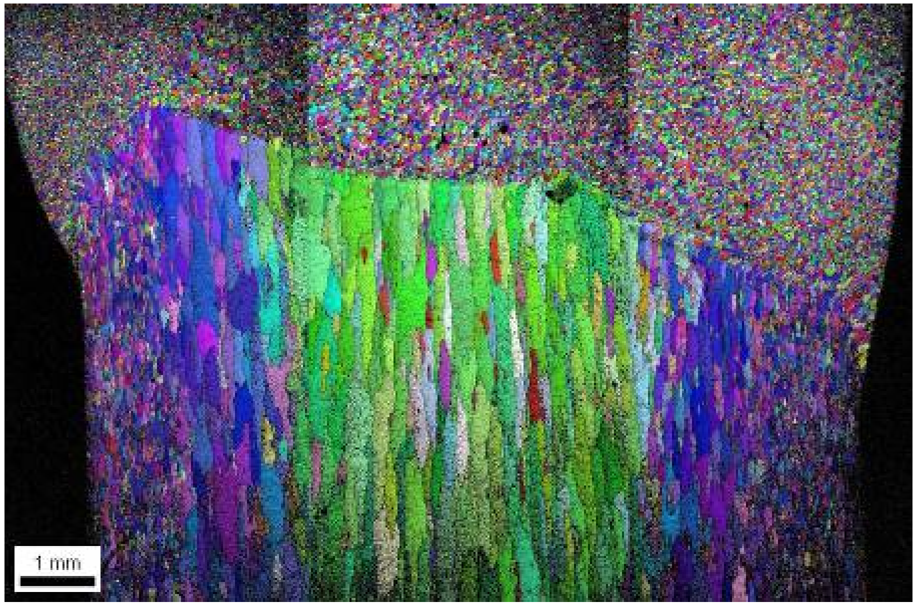
energy selective radiography of rolled aluminum



Al structure – a possible interpretation



transmission image at 4 Å



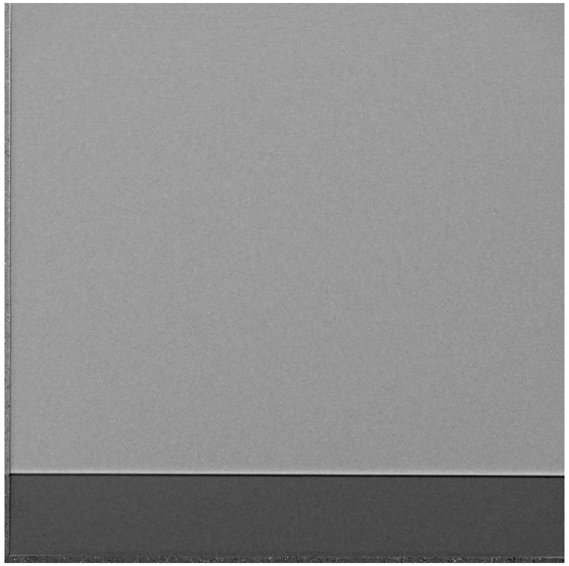
Electron Backscattering Diffraction (EBSD) measurement at the surface

→Different grain size and orientation in bulk and weld

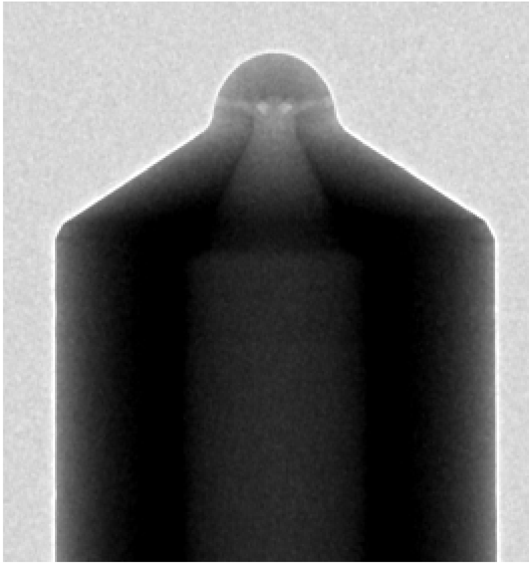
Data: L. Josic, H.Leber, PSI

Phase based neutron imaging

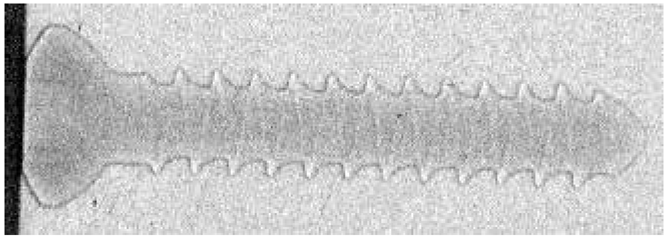
Features



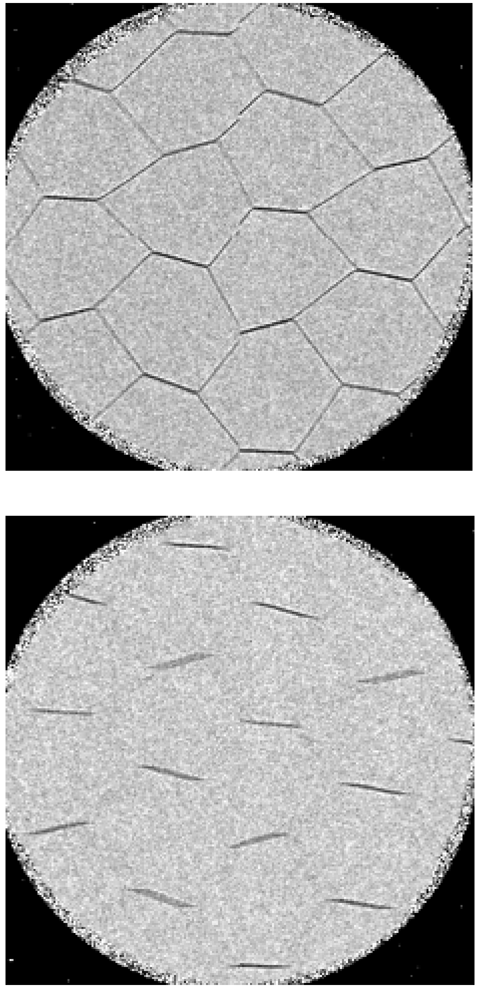
Al plate 20 mm thick



steel nozzle

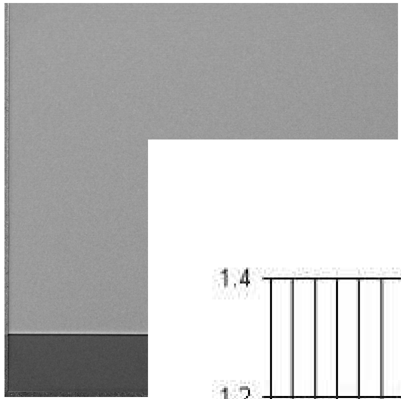


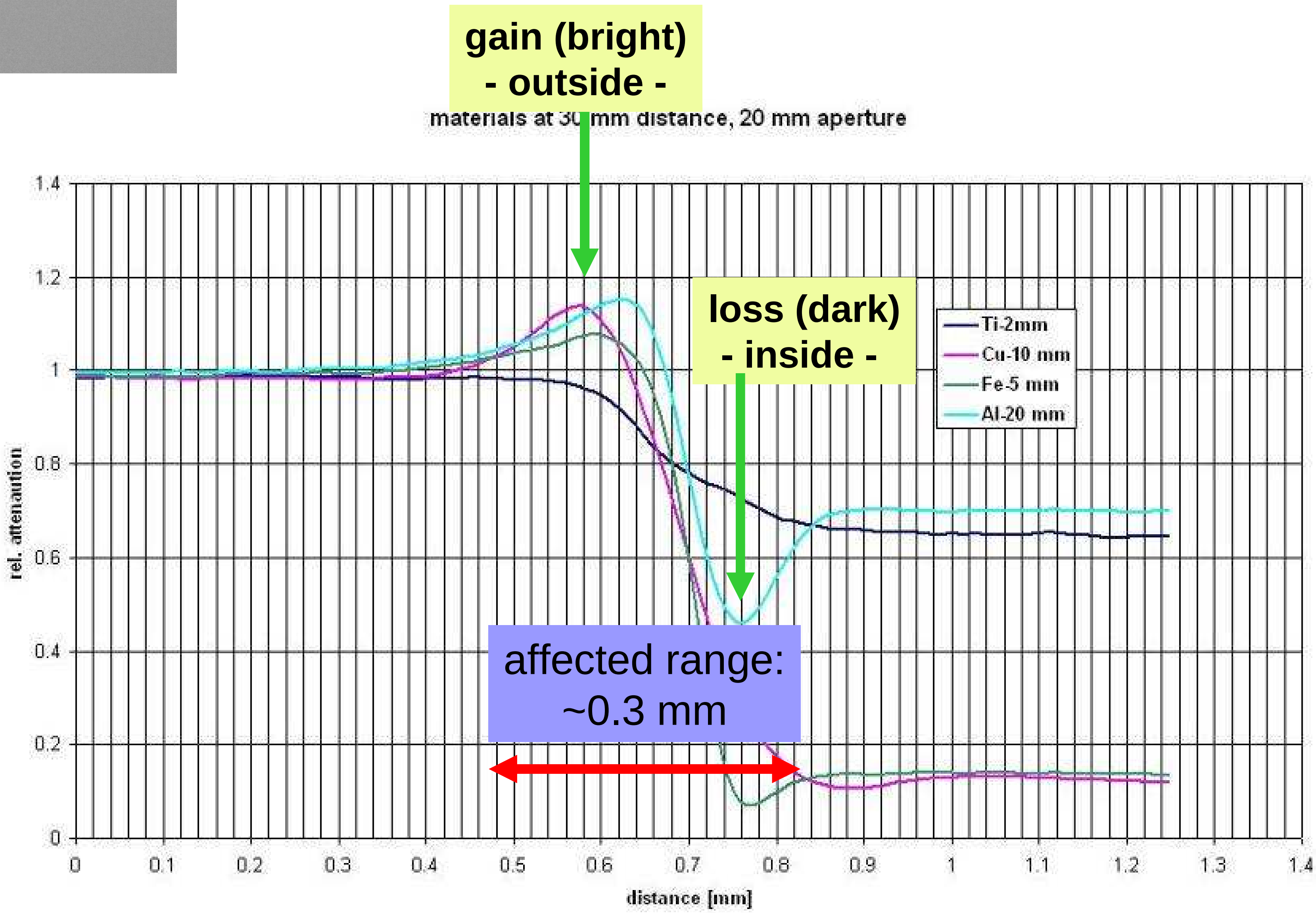
Ti screw



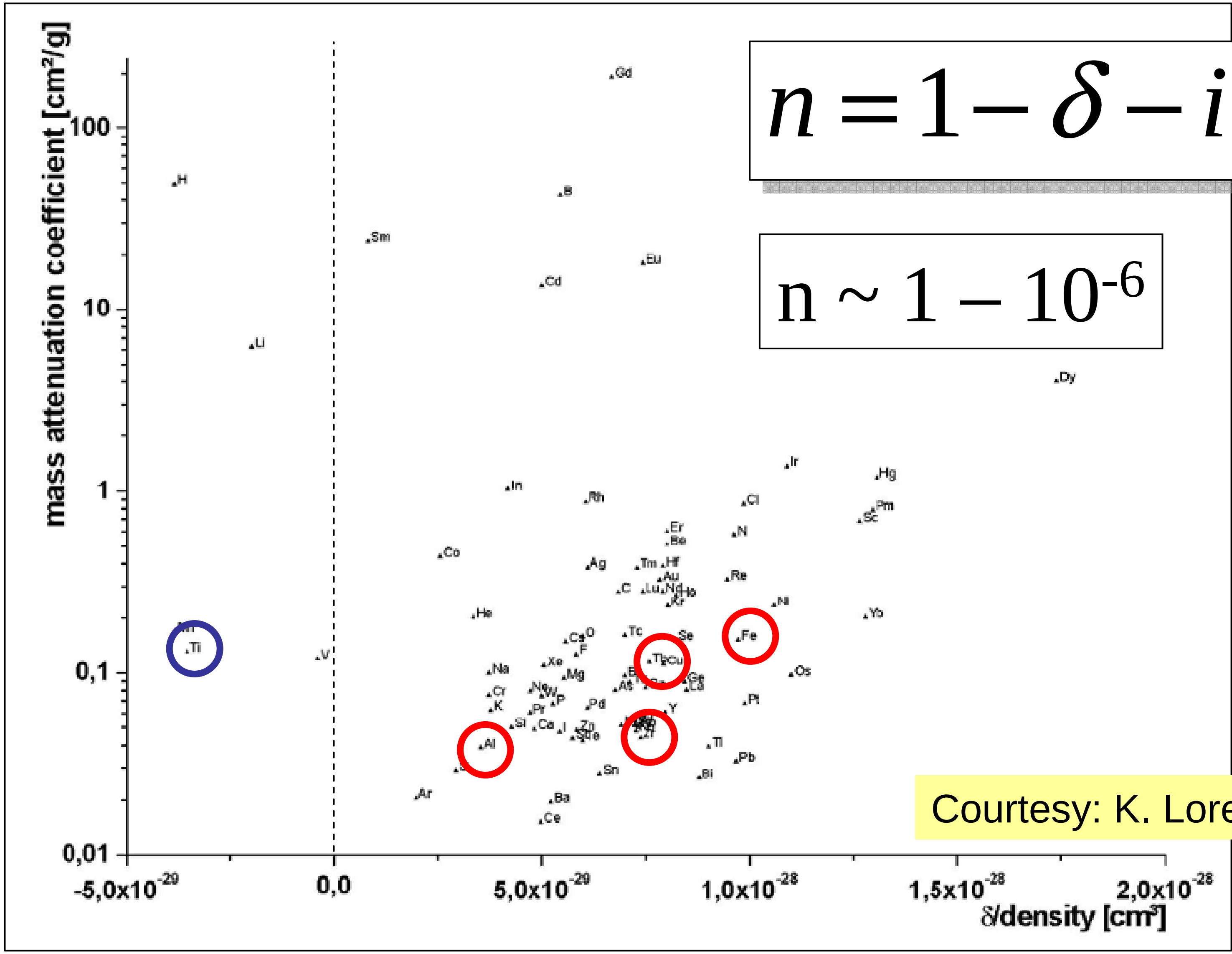
Al honeycomb

Edge effects (plates)





Material properties Abs. / Phase

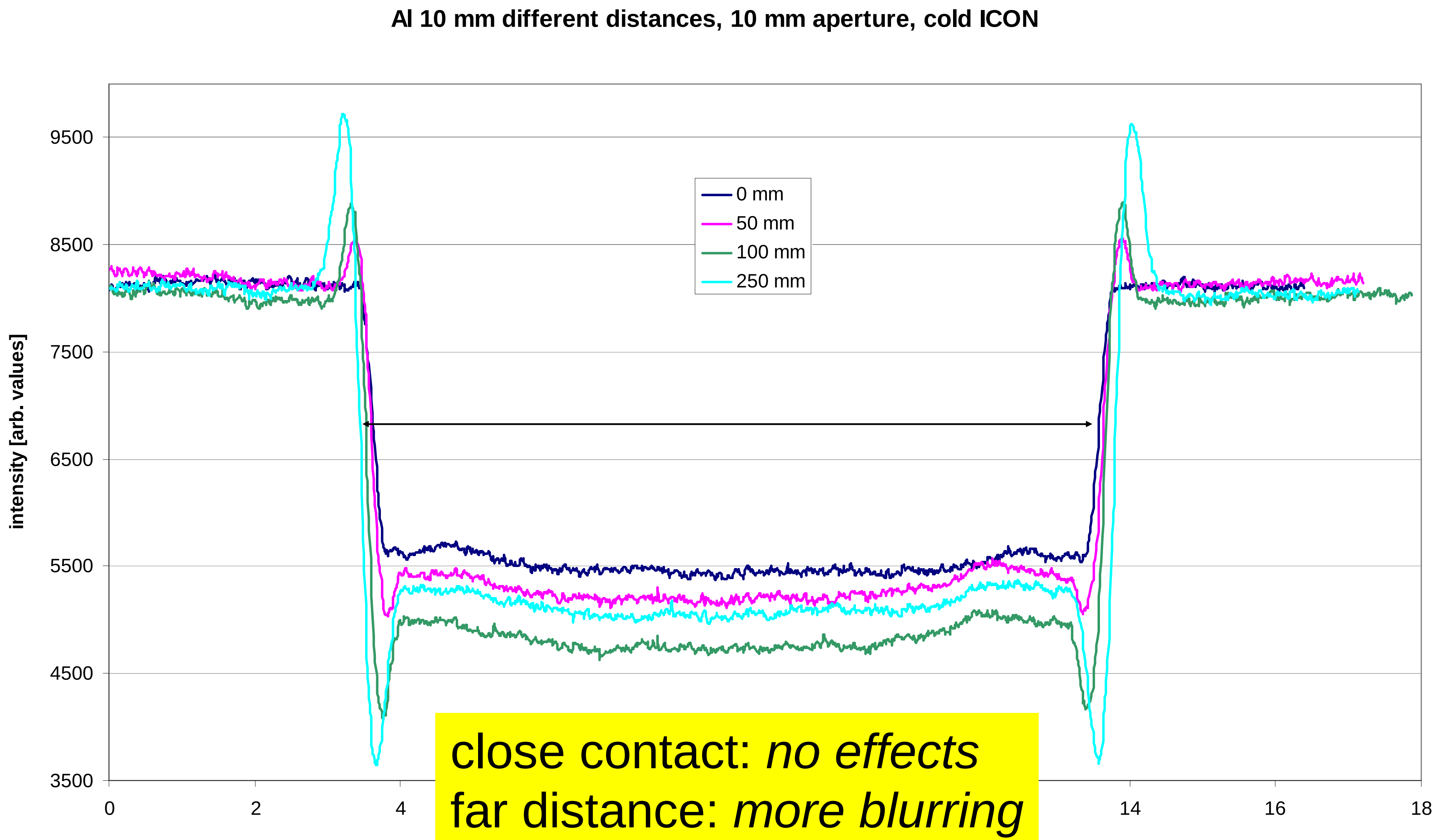


$$n = 1 - \delta - i \cdot \beta$$

$$n \sim 1 - 10^{-6}$$

Courtesy: K. Lorenz, TUM

Variation of sample – detector distance



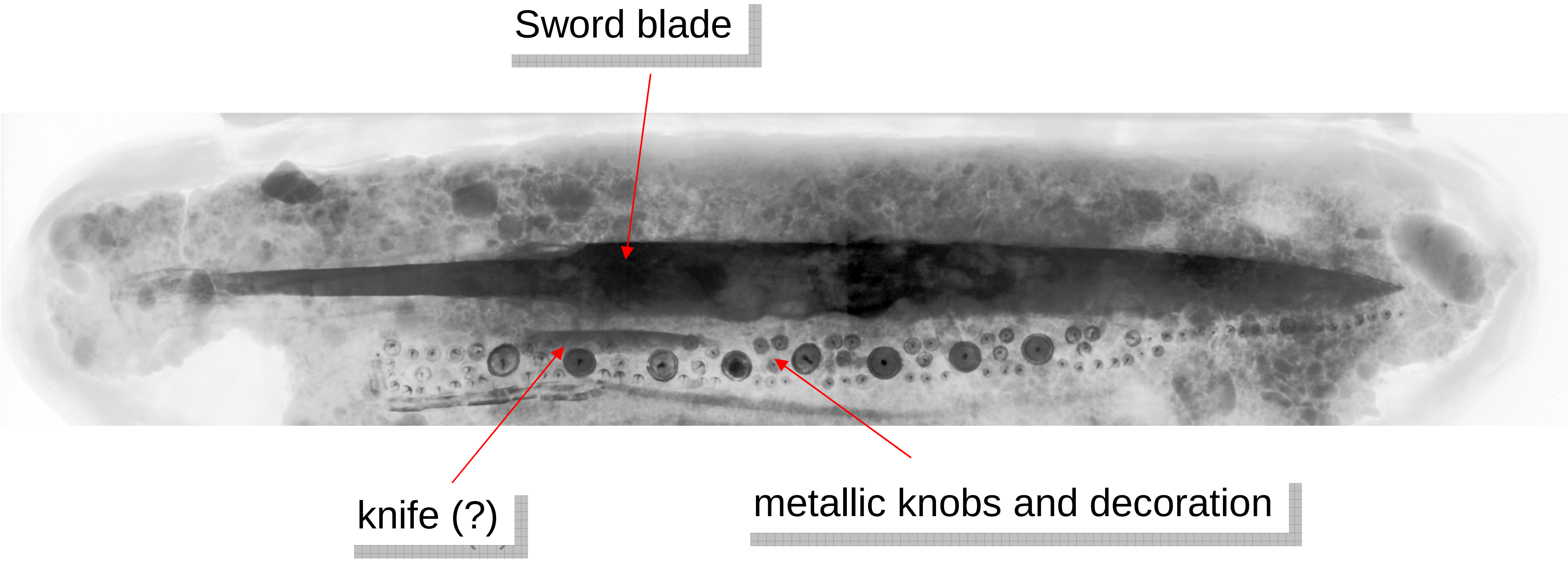
close contact: *no effects*
far distance: *more blurring*
→ optimization needed

Neutron – X-ray data fusion

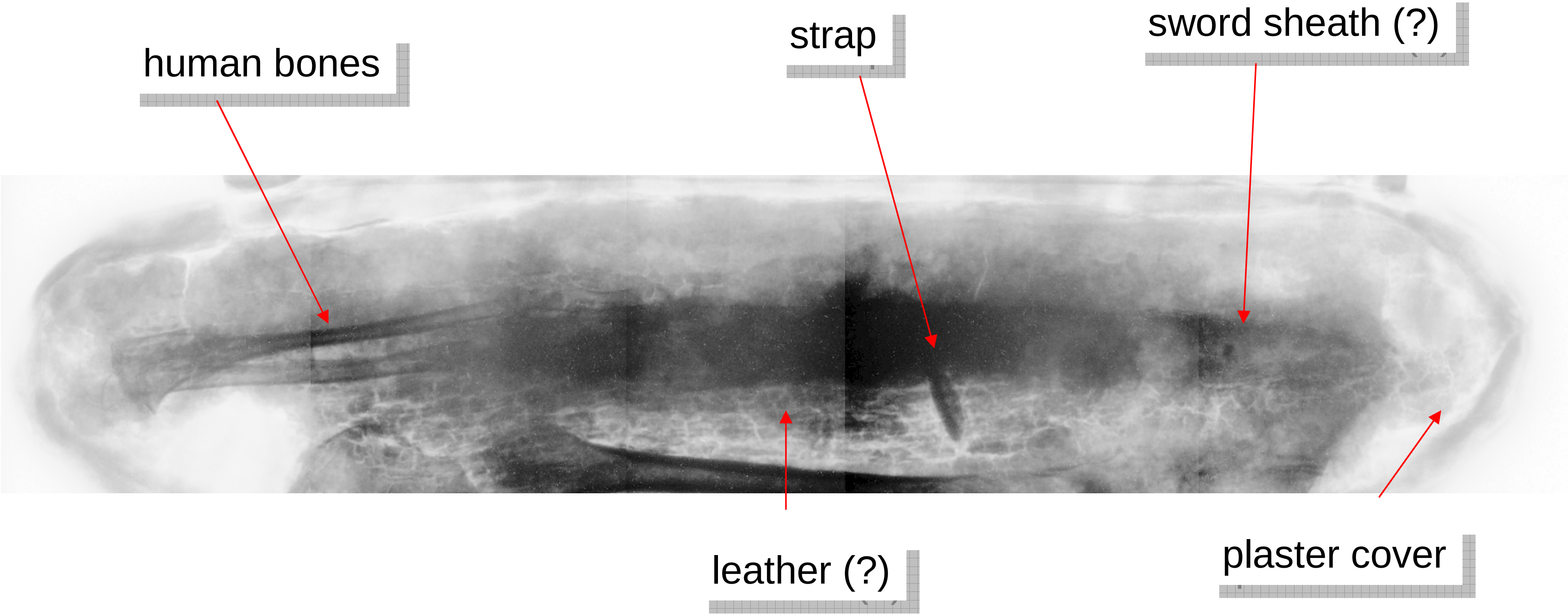
Sealed excavation part of archaeological finds observed on demand from archaeological service Canton Zug (CH)

top view of the object

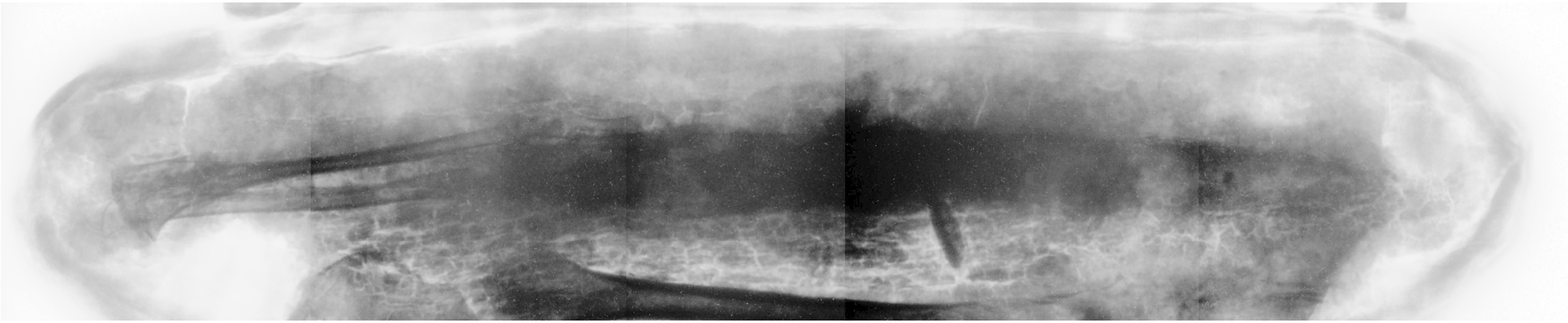
Features of the X-ray image

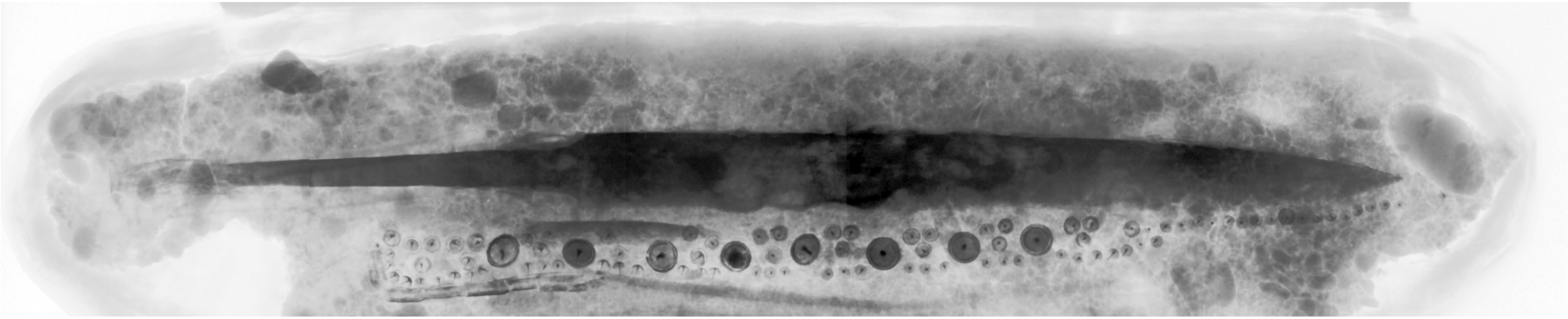


Featutes of the neutron image

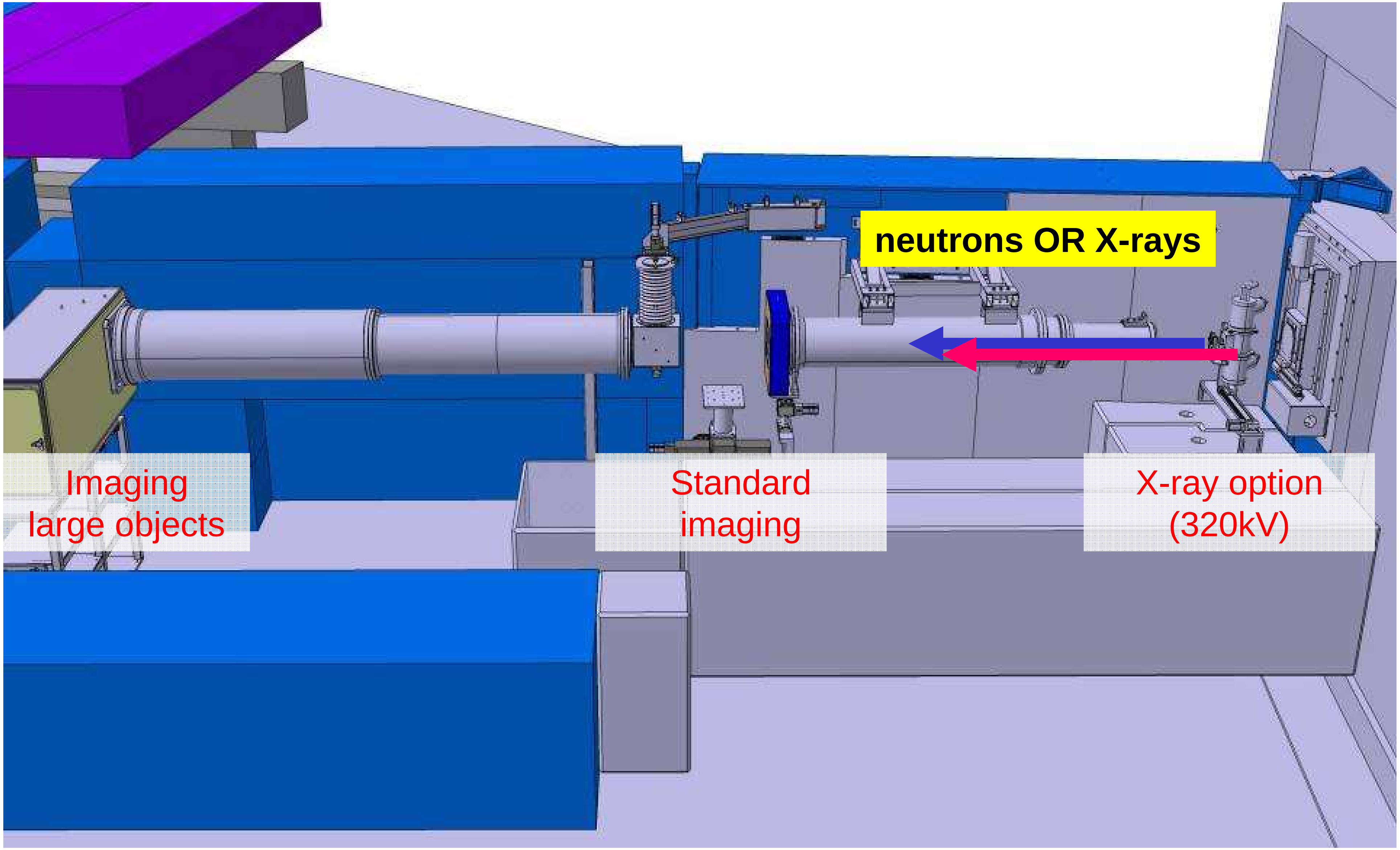


Comparison





Infrastructure: NEUTRA → XTRA



Sword from Oberwil (Lake Zug, Switzerland) 15th Century

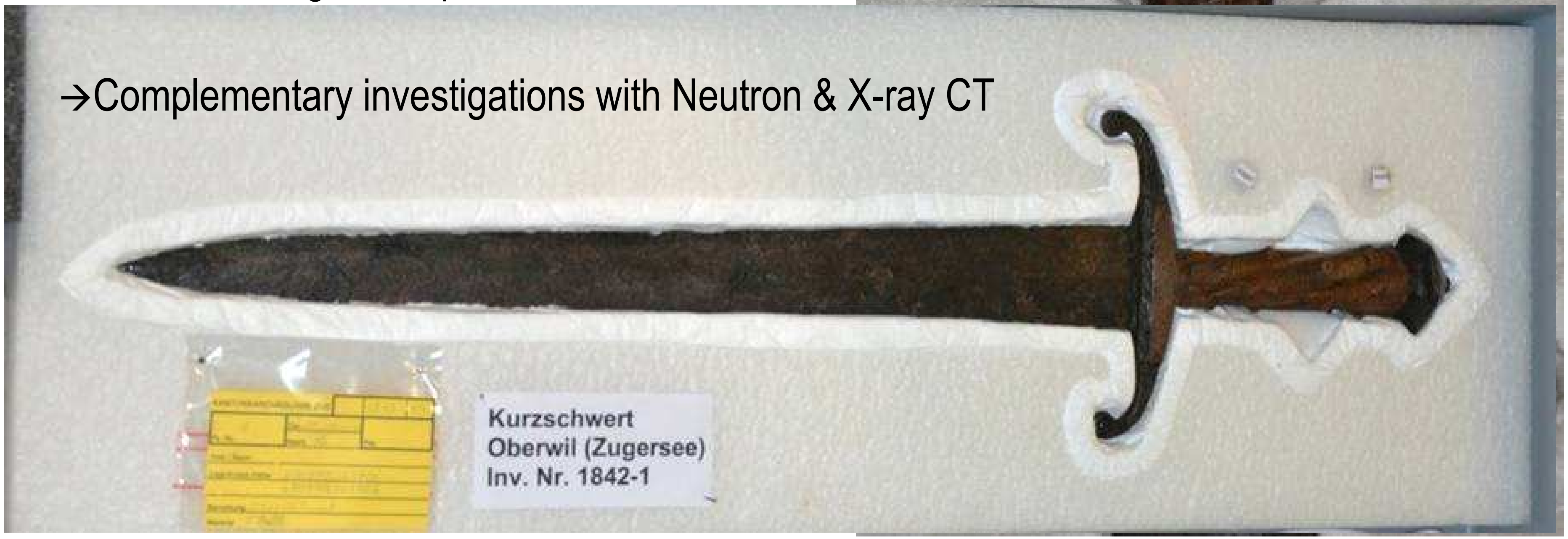
Department for Ancient Monuments and Archaeology of the Canton of Zug & Swiss National Museum

Documentation as complete as possible:

- State / condition
- Build-up (organic part & metal parts)

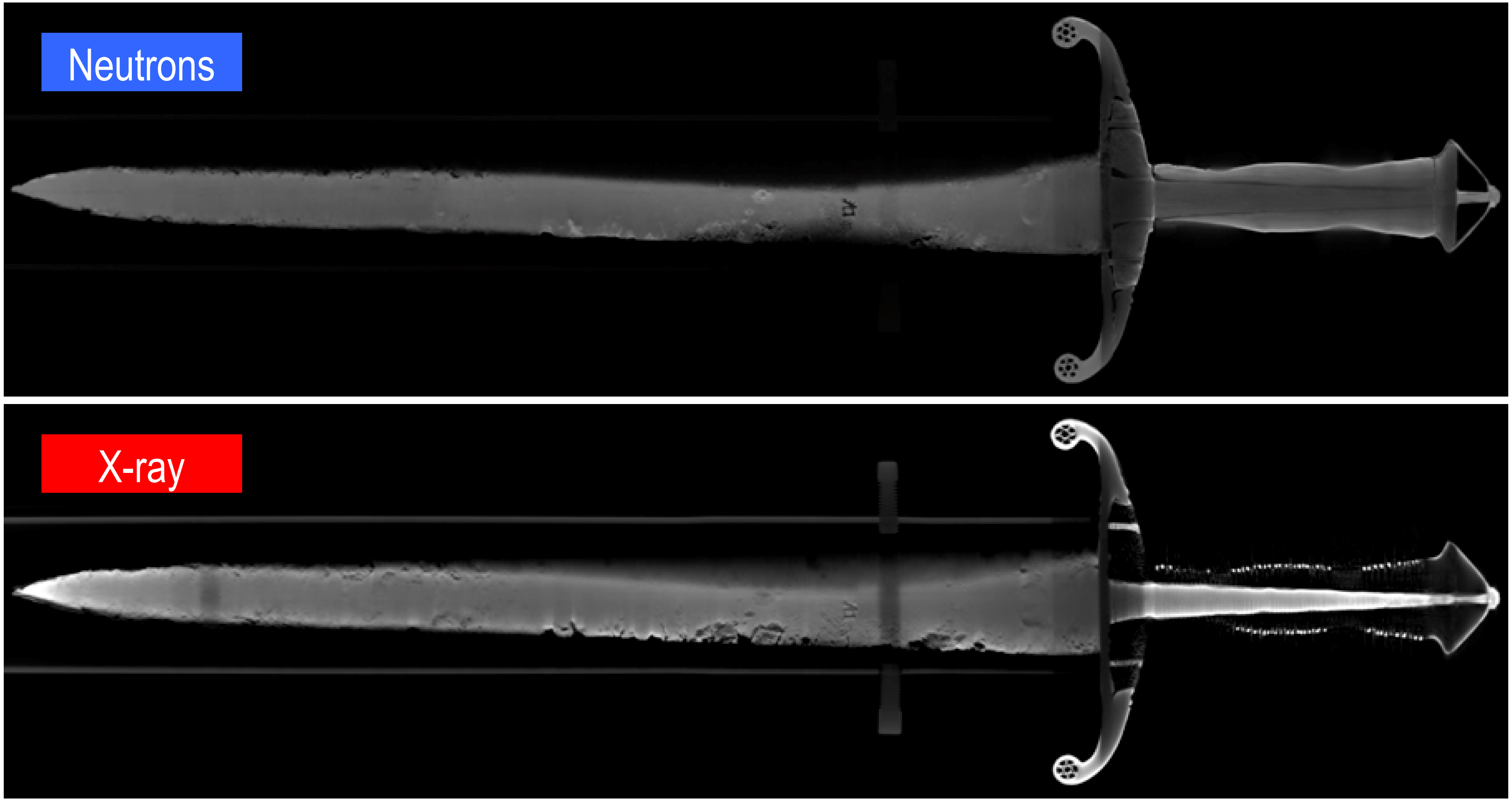
→Manufacturing of a replica

→Complementary investigations with Neutron & X-ray CT



Tomography slices

Sword from Oberwil (Lake Zug, Switzerland) 15th Century



Examples – Combination of Neutrons & X-ray

Sword from Oberwil (Lake Zug, Switzerland) 15th Century
Fusion of X-ray & Neutron data sets

The relation to archaeology and museums experts

1. The investigation results from studies with neutrons and X-rays can **only added** to the a-priori knowledge of the experts in cultural heritage

2. Stable and **interactive links** have to be established between the two knowledge areas to be most efficient and successfully

3. There are already **networks** for mutual interaction (COST-G8, COST-IE0601, conferences ART'11, TECHNART); however, they are dominated by natural scientists

4. Therefore, these approaches often do not follow the real demand

5. On the other hand, there is often minor knowledge in the museums about the developed sophisticated new methods, more **exchange of information** is required

Neutron imaging for cultural heritage studies in Italy?

Neutron Imaging in Italy (1)

Neutron Sources:

Research reactors: from initially 13 reactors only 3 remain:

- Pavia: TRIGA LENA, 250 kW, University of Pavia
- ENEA Casaccia, TRIGA RC-1, reactivated 2010
- ENEA Casaccia, TAPIRO, 1 kW fast, reactivated 2010

Accelerator driven:

- DAØNE Beam Test Facility
- e-Linac Uni Messina?
- medical photo-converted source PhoNeS (for BNCT)
- RFQ based source (INFN-LNL) (for BNCT)

→ *all not yet considered for Neutron Imaging*

Neutron Imaging in Italy (2)

Neutron Imaging Facilities:

- Facility at TRIGA Casaccia became never operational in the user mode
- Contributions to detector development (Uni Messina for IMAT beam line in UK, ISIS Oxford)

Activities in Neutron Imaging

- Students from various Italian universities as visitors

Activities for Cultural Heritage Studies using Neutrons:

- F. Grazzi (Florence) – studies with imaging and diffraction at several facilities

→ MORE PARTICIPATION IS WELCOME! ←

CONCLUSIONS

- Neutron tomography has it application range mainly on the **macroscopic** scale (given by the penetration power of neutrons and the detector/beam performance)

- There are attempts to come close to 1 micro-meter resolution

- The high **sensitivity to light elements** can be exploited favorably

- New unique techniques are under development (combination with diffraction, polarized neutron imaging, energy selective imaging, …)

- Fusion between X-ray and neutron data looks very promising

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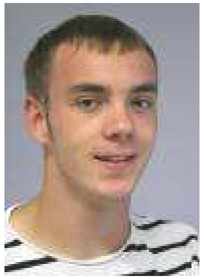

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