NEUTRON RESONANCE CAPTURE ANALYSIS

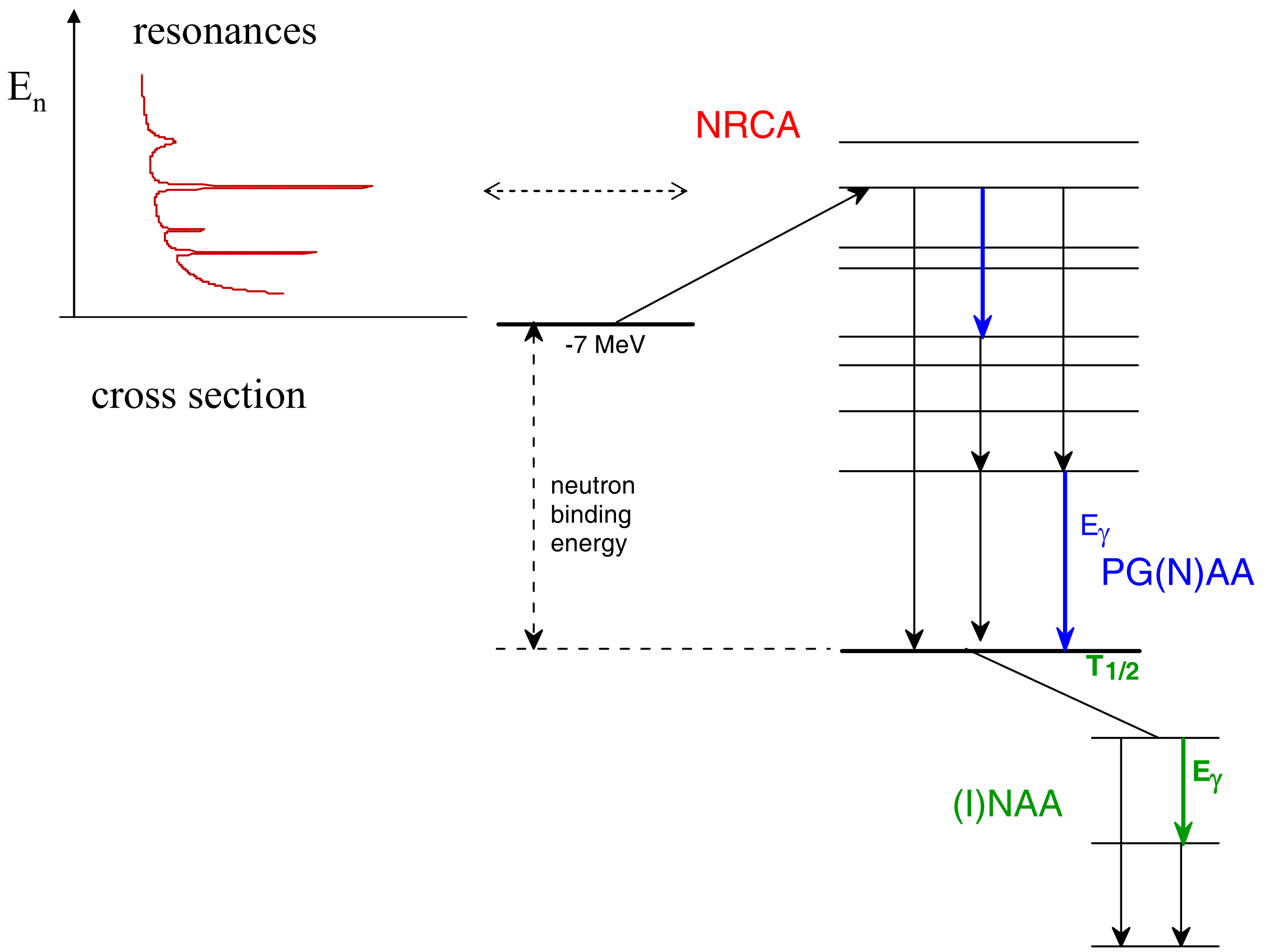
and

Artifacts studied at the GELINA facility

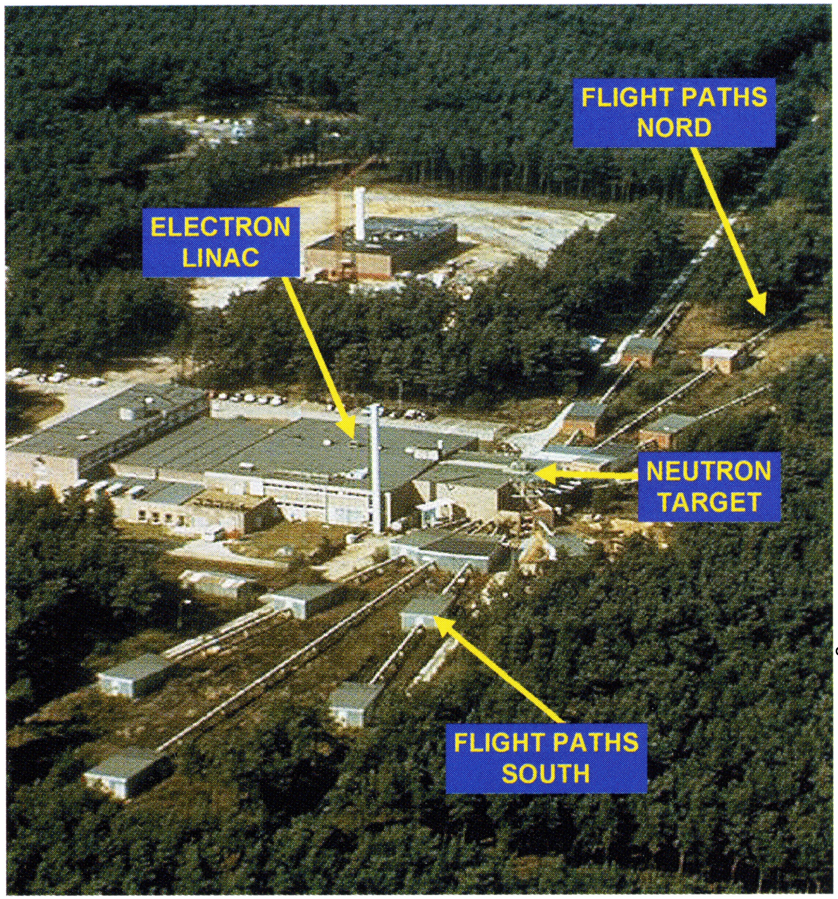
Hans Postma
University of Technology Delft

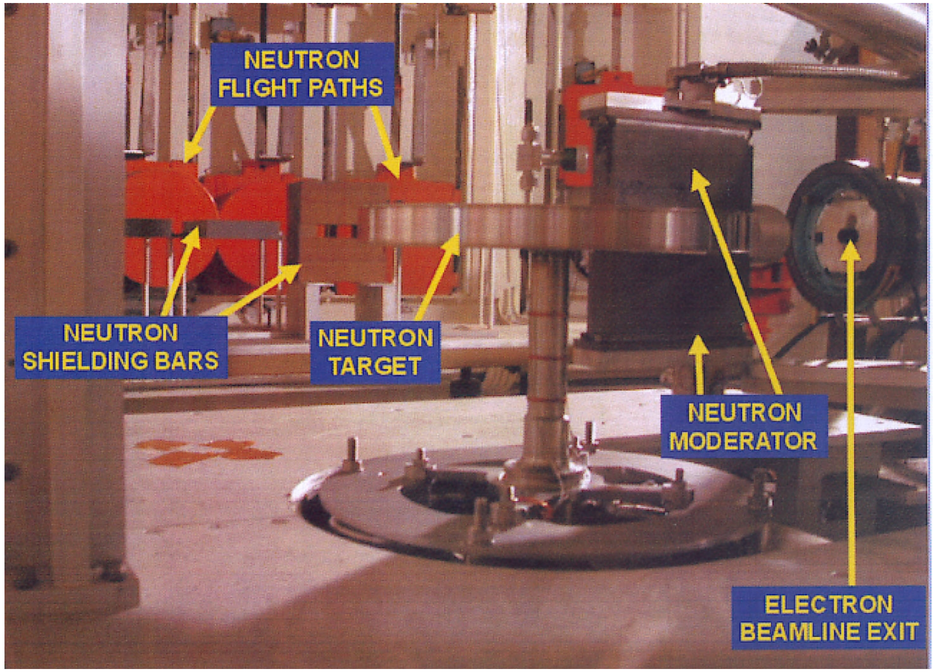
Budapest, EFNUDAT workshop
23-25 September 2009

TU Delft
Delft University of Technology

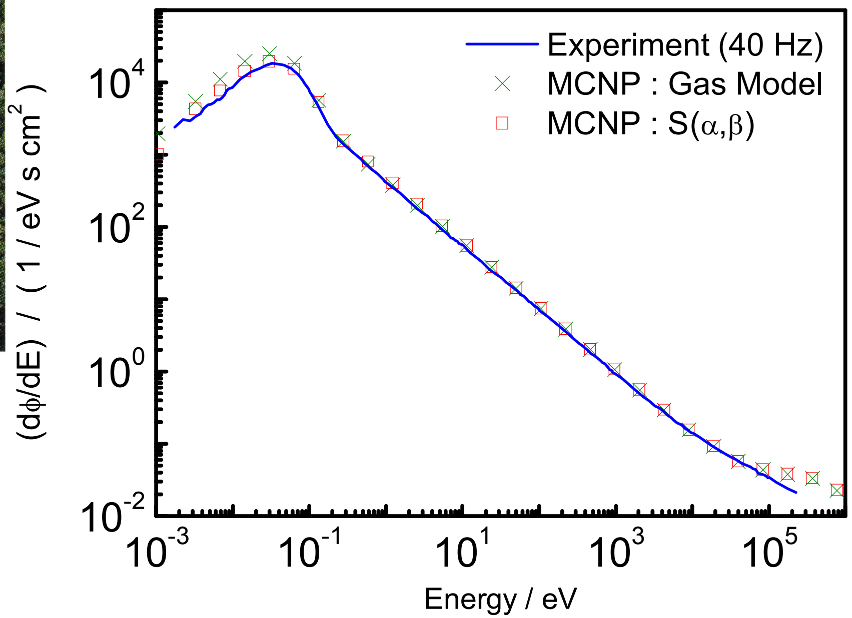

resonances
E_n
cross section
NRCA
-7 MeV
neutron
binding
energy
E_γ
PG(N)AA
$T_{1/2}$
E_γ
(I)NAA

GELINA facility





$$E = \frac{1}{2} m \left(\frac{L}{t} \right)^2$$



GELINA

Beam 5

Flight path

12.8 m

Two C_6D_6-

detectors

n-beam

diameter

7 cm

Capture followed by a cascade of gamma transitions

Detection efficiency ε_c for capture events

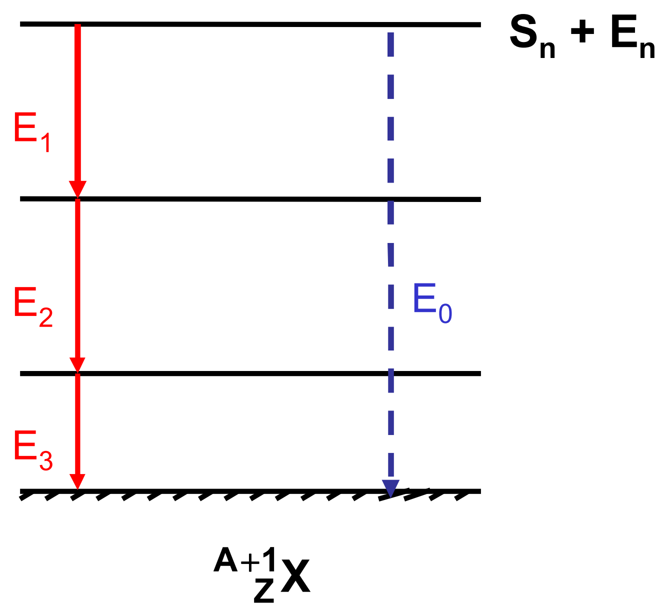
$$\varepsilon_c = 1 - \prod_i \left(1 - \varepsilon_{\gamma,i}\right)$$

for $\quad \varepsilon_{\gamma,i} << 1$

$$\varepsilon_c \cong \sum_i^{\nu} \varepsilon_{\gamma,i}$$

Suppose : $\varepsilon_{\gamma,i} = kE_\gamma$ proportional to E_γ

$$\Rightarrow \varepsilon_c = k(S_n + E_n) = kE^*$$



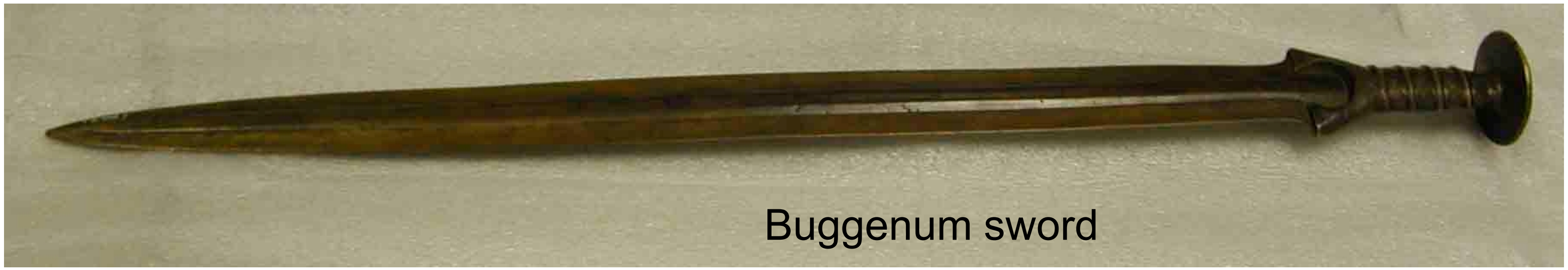
TU Delft
Delft University of Technology

Study of various kinds of bronze objects

Statuettes, e.g.
Etruscan

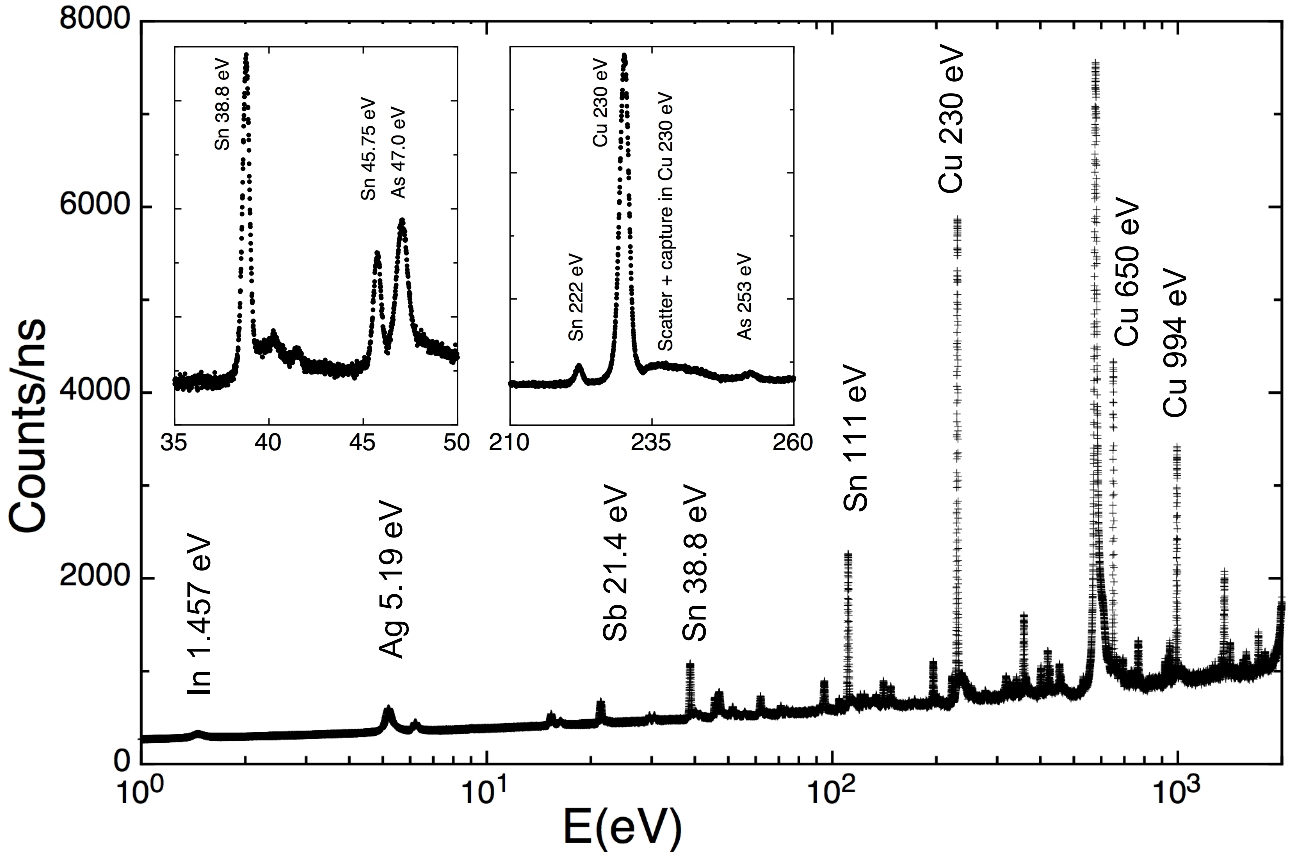
Roman, e.g.
water taps

Bronze Age:
Axes, swords



Buggenum sword

Resonance spectrum of the “Buggenum” sword



Number of capture events (yield) as function of neutron energy *E*:

$$N_{capt}(E)dE = O \cdot \Phi(E)\left(1 - \exp\left(-\sum_i n_i \sigma_i^d(E) D\right)\right) \frac{\sigma_\gamma^d(E)}{\sigma_{tot}^d(E)} \cdot \varepsilon_\gamma \cdot dE + N_{scat+capt}$$

sample area	integrated flux	absorption in sample	capture part	detection efficiency

n_i : isotope number density, *D*: sample thickness, *O*: sample area

Analysis of resonance spectrum with REFIT

A least square fitting program for resonance analysis of neutron transmission and capture data (M.C. Moxon)

-- Based on a complete description of the experiment

-- Use of R-matrix multi-level formalism

-- samples of simple shapes and compositions

Another approach: consider the total number of captures in a resonance

$$N_{capt} = O \cdot nD \cdot \Phi(E_o) \cdot R(nD) \cdot aA_\gamma \cdot \varepsilon_\gamma$$

n: number density element, a: abundance

The resonance capture area A_γ occurs in this expression:

$$A_\gamma = \frac{\pi}{2}\sigma_o \Gamma_\gamma = 4.097 x 10^6 \frac{g\Gamma_n \Gamma_\gamma}{E_o \Gamma} \text{ (barn.eV)}$$

Self-shielding factor:

$$R(nD) = \frac{\int_{res} dE \Phi(E) \left[1 - \exp\{-an\sigma_{tot}(E)D\}\right]}{nD \int_{res} dE \Phi(E) a\sigma t_{tot}(E)}$$

Compare ratios of counts, N_I and N_{II}, of two resonances of two elements in the object and calibration samples

Same experimental conditions, several terms cancel

Thin sample and/or weak resonances: $R(nD) \approx 1$

This leads to a relation between elemental weight ratios:

$$\left(\frac{W_I}{W_{II}}\right)_{obj.} \cong \frac{\left(\frac{N_I}{N_{II}}\right)_{obj.}}{\left(\frac{N_I}{N_{II}}\right)_{cal.}} \cdot \left(\frac{W_I}{W_{II}}\right)_{cal.}$$

Thick samples; strong resonances:

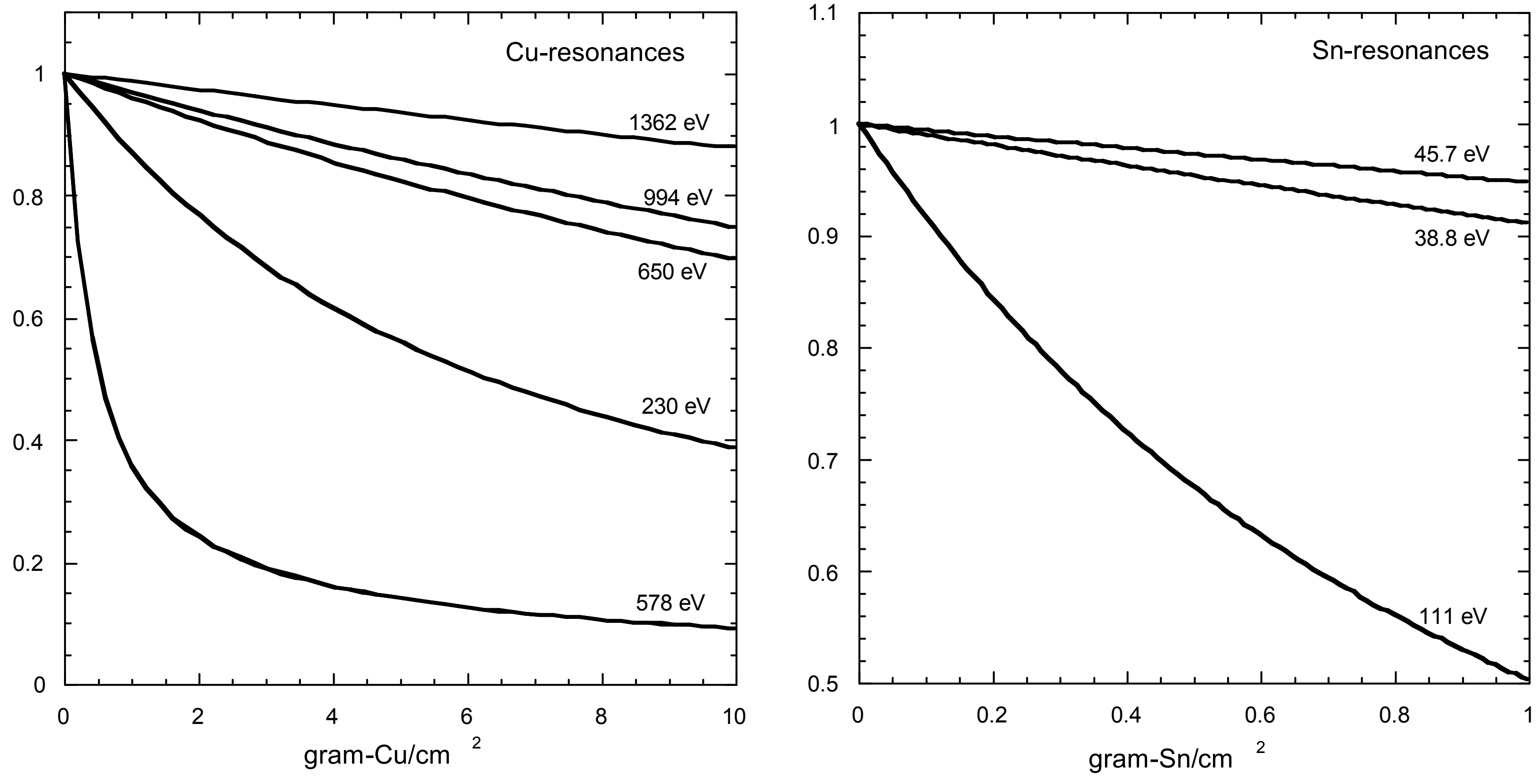
$$\frac{\left(\frac{R_{II}}{R_I}\right)_{obj.}}{\left(\frac{R_{II}}{R_I}\right)_{cal.}}$$

Plug in self-shielding factors

$$\left(\frac{W_I}{W_{II}}\right)_{obj.} = \frac{\left(\frac{N_I}{N_{II}}\right)_{obj.}}{\left(\frac{N_I}{N_{II}}\right)_{cal.}} \cdot \cdot \left(\frac{W_I}{W_{II}}\right)_{cal.}$$

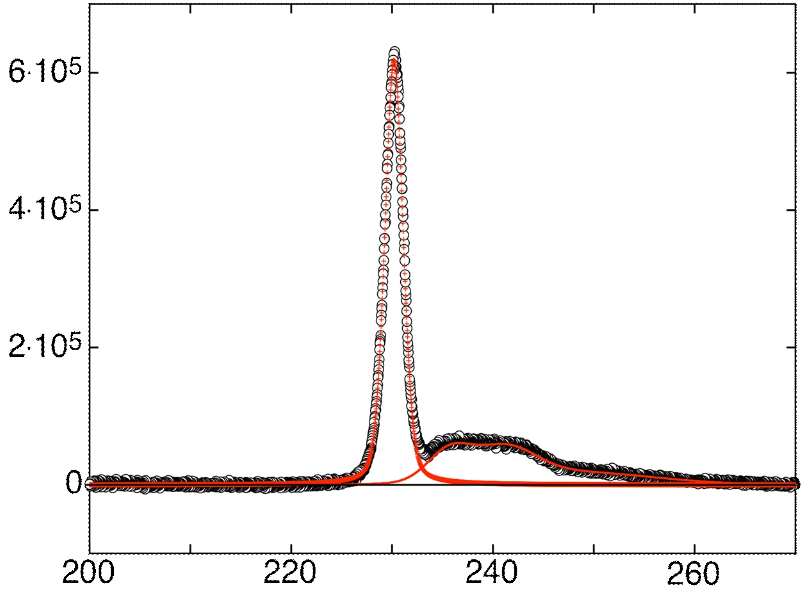
$$\left(\frac{W_I}{W_{II}}\right)_{obj.} = \frac{\left(\frac{N_I}{N_{II}}\right)_{obj.}}{\left(\frac{N_I}{N_{II}}\right)_{cal.}} \cdot \frac{\left(\frac{R_{II}}{R_I}\right)_{obj.}}{\left(\frac{R_{II}}{R_I}\right)_{cal.}} \cdot \left(\frac{W_I}{W_{II}}\right)_{cal.}$$

Self-shielding factors for some copper and tin resonances



230-eV Cu-resonance.

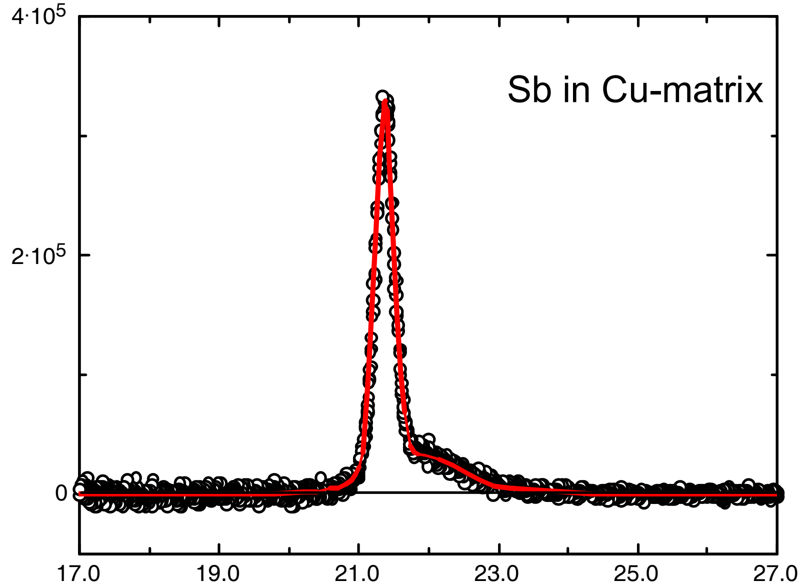
Background subtracted, capture peak, and scattering-capture structure



Native copper axe 14 mm thick
(Winfried Kockelmann)

21.4 eV Sb-resonance.

Background subtracted, capture peak and scattering-capture structure.

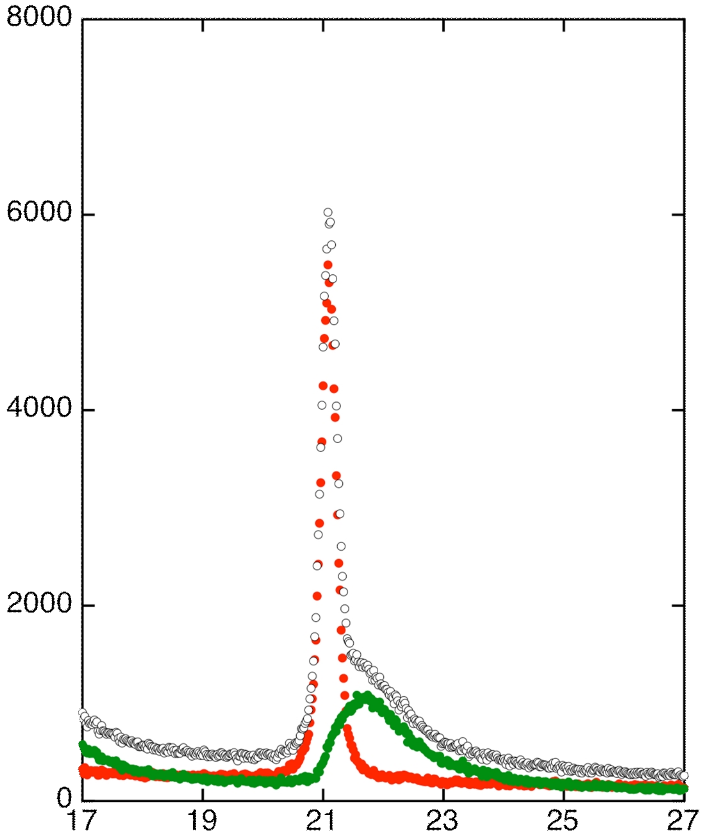


Bronze-age sword (2A)
(Vienna Museum)

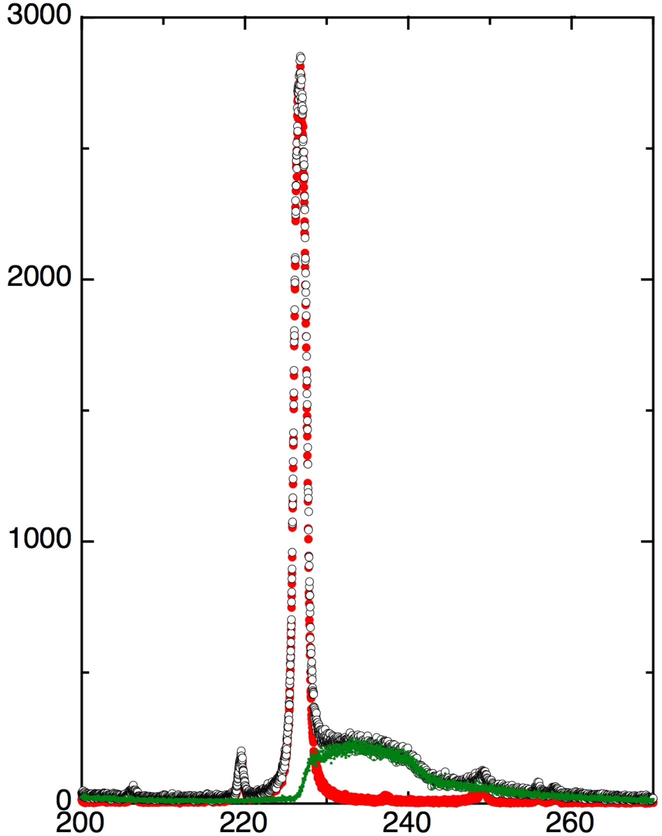
$$E_{recoil} = E\frac{2mM}{(M+m)^2}(1-\cos\theta)$$

Scattering at Cu nuclei, maximum recoil energy:
14 eV near 230-eV Cu-resonance,
1.3 eV near 21.4 eV Sb-resonance.

Simulated resonance spectrum; bronze 10 mm thick.



21.4 eV Sb-resonance



230 eV Cu-resonance

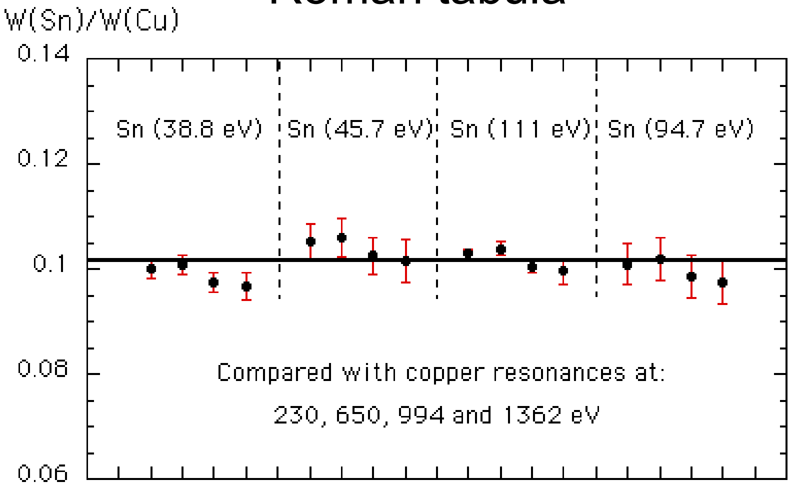
Delft University of Technology

thin and flat
versus
thick and irregular

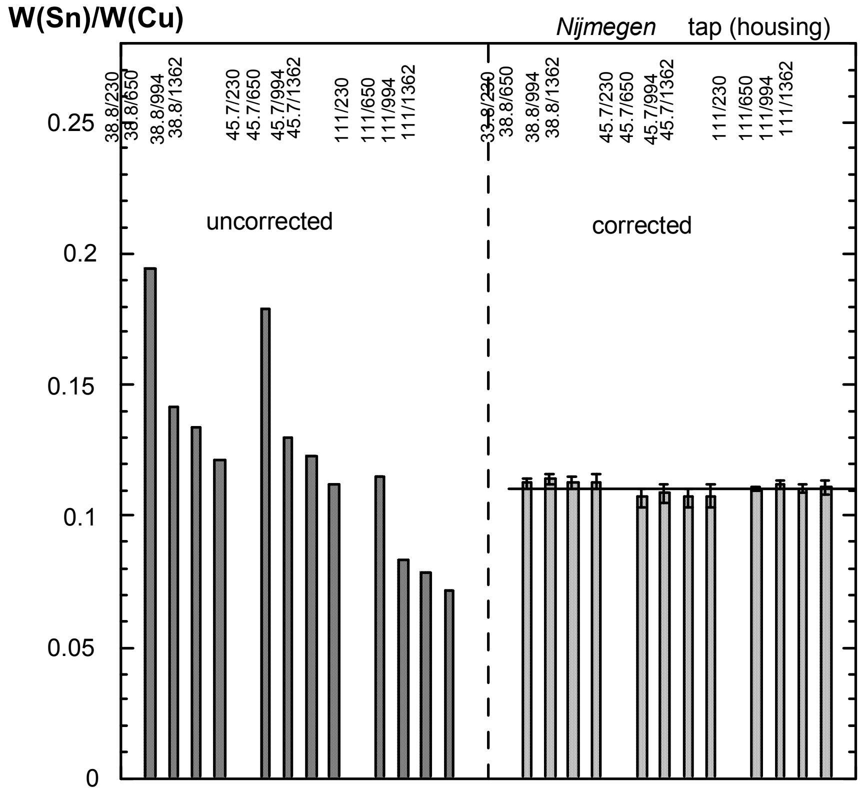
Roman bath tap
"Nijmegen"

Beam area

Roman tabula



16 pairs of resonances



12 pairs of resonances

Budapest, 23-25 Sept. 2009

TU Delft
Delft University of Technology

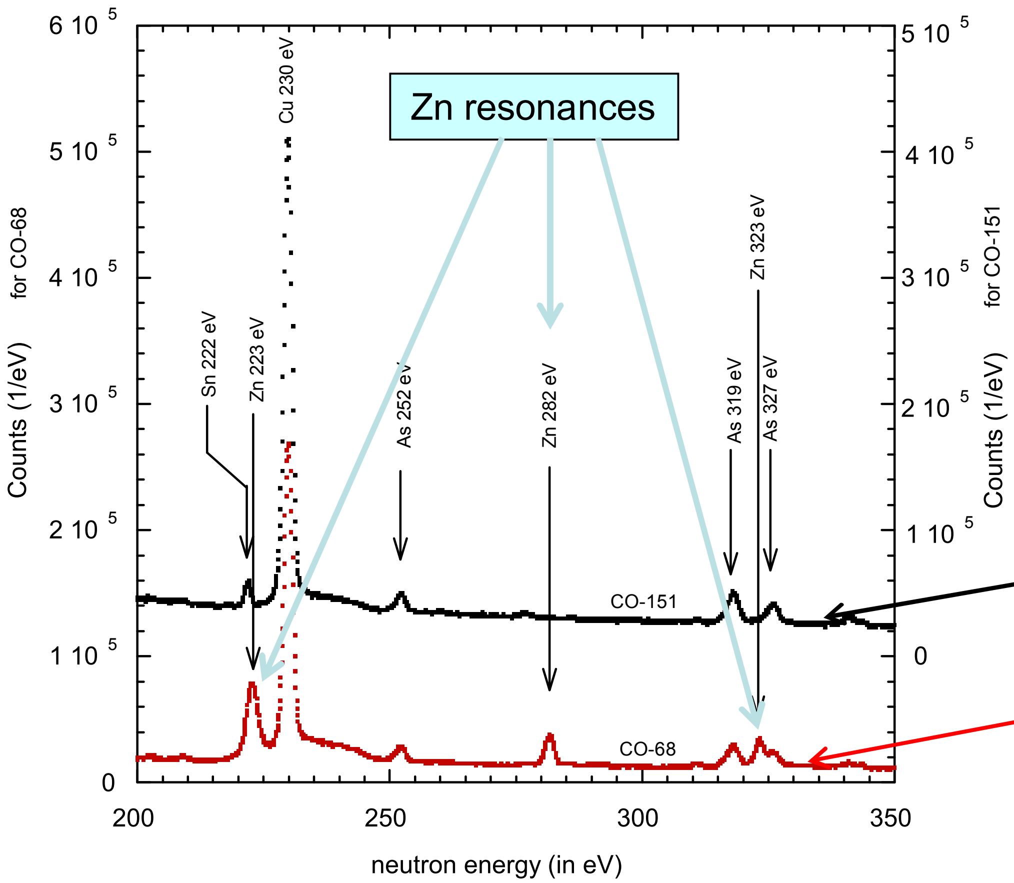
Study of various kinds of bronze objects

Statuettes, e.g.
Etruscan

Roman, e.g.
water taps

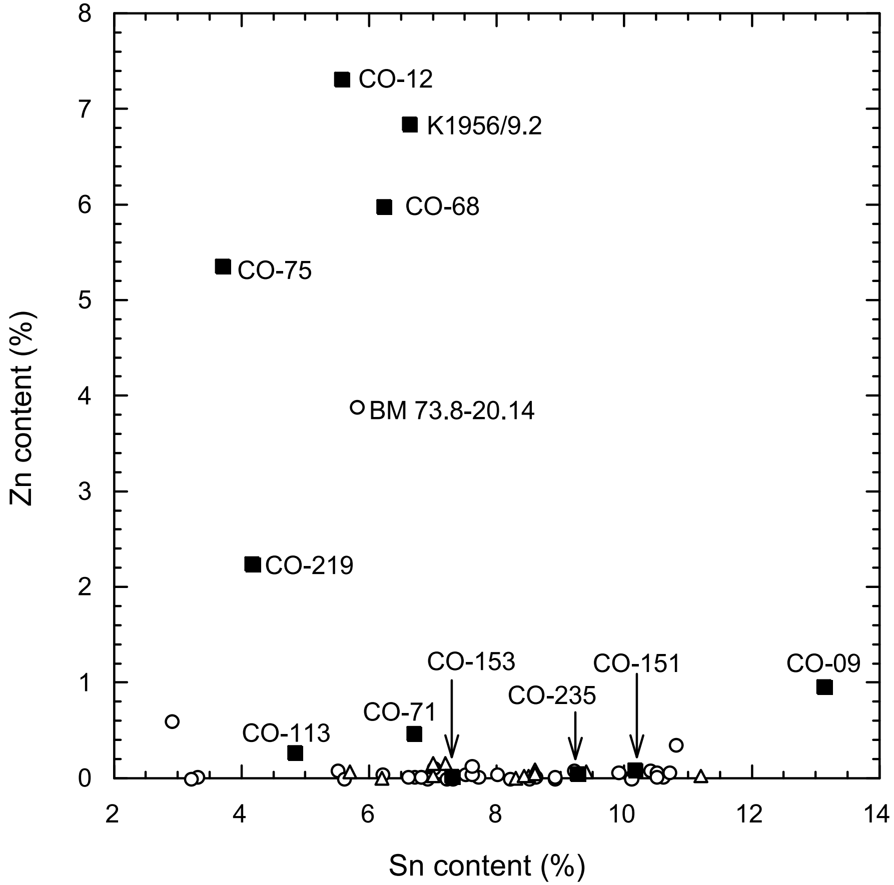
Bronze Age:
Axes, swords

"Corazzi" collection of Etruscan statuettes (from 2nd-7th c.BC)
Bought in 1826 by National Museum of Antiquities (Leiden, NL)
Are they all genuine Etruscan? Not those containing considerable amounts of zinc! (Zn does not dissolve easily in copper)



Zinc has a high vapor pressure at the melting point of copper, it evaporates during the smelting of copper if it occurs in the ore.

A comparison with studies by Craddock of bronze objects from the 2nd to 7th century BC in Italy and Sardinia

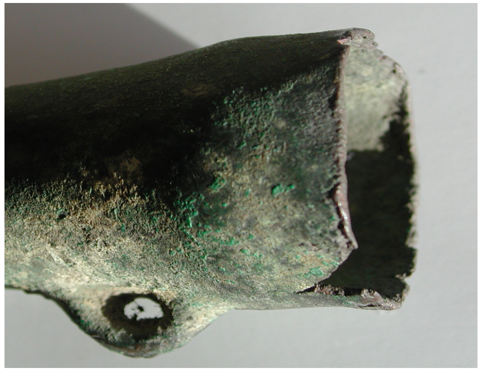


Black data points: from the Corazzi collection.

Open data points: from Craddock's study.

Geistingen type of socketed axes.

1,3 mm
dik



Purpose:

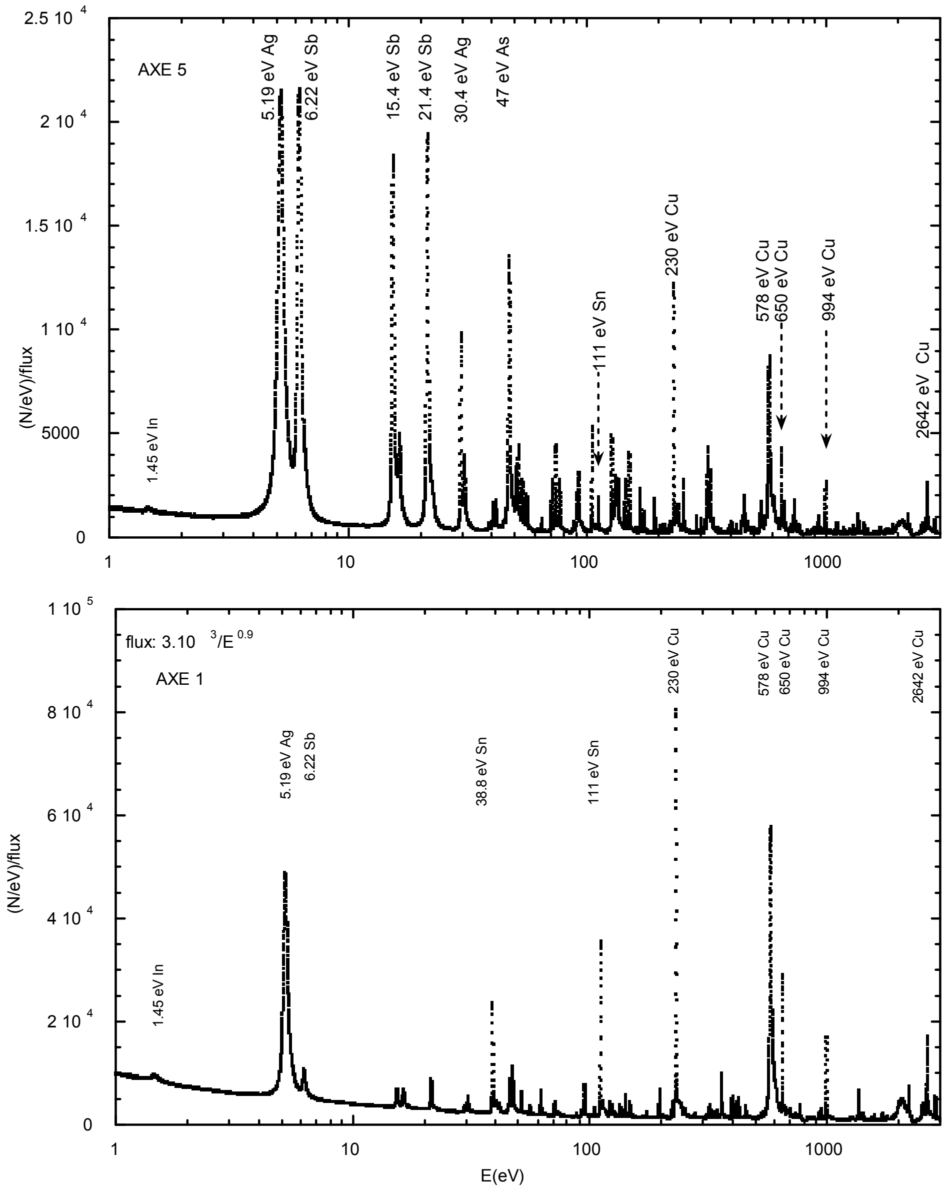
-- Ritual?

-- Ceremonial?

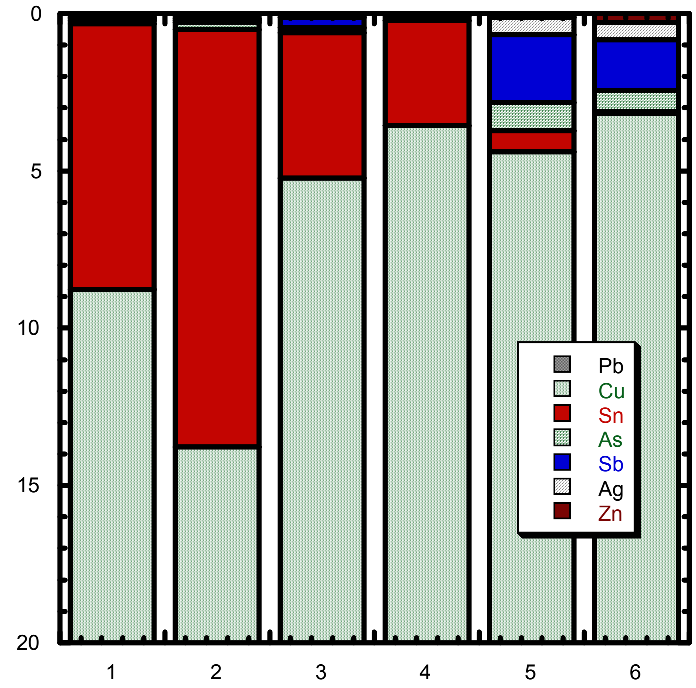
-- Status symbol?

-- Very thin walls

-- useless as axe

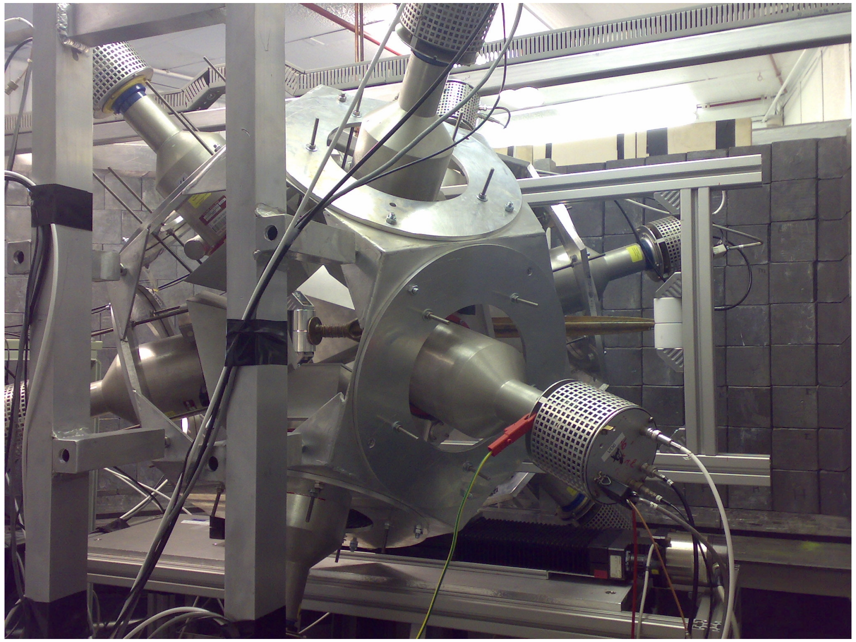


Very different compositions

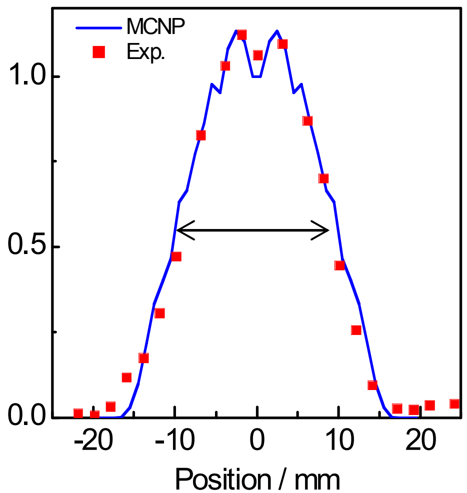


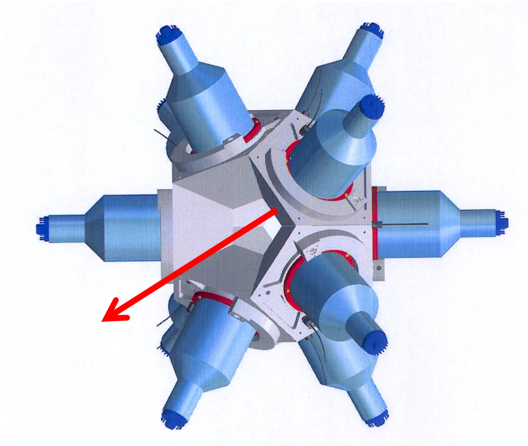
Delft University of Technology

Buggenum sword



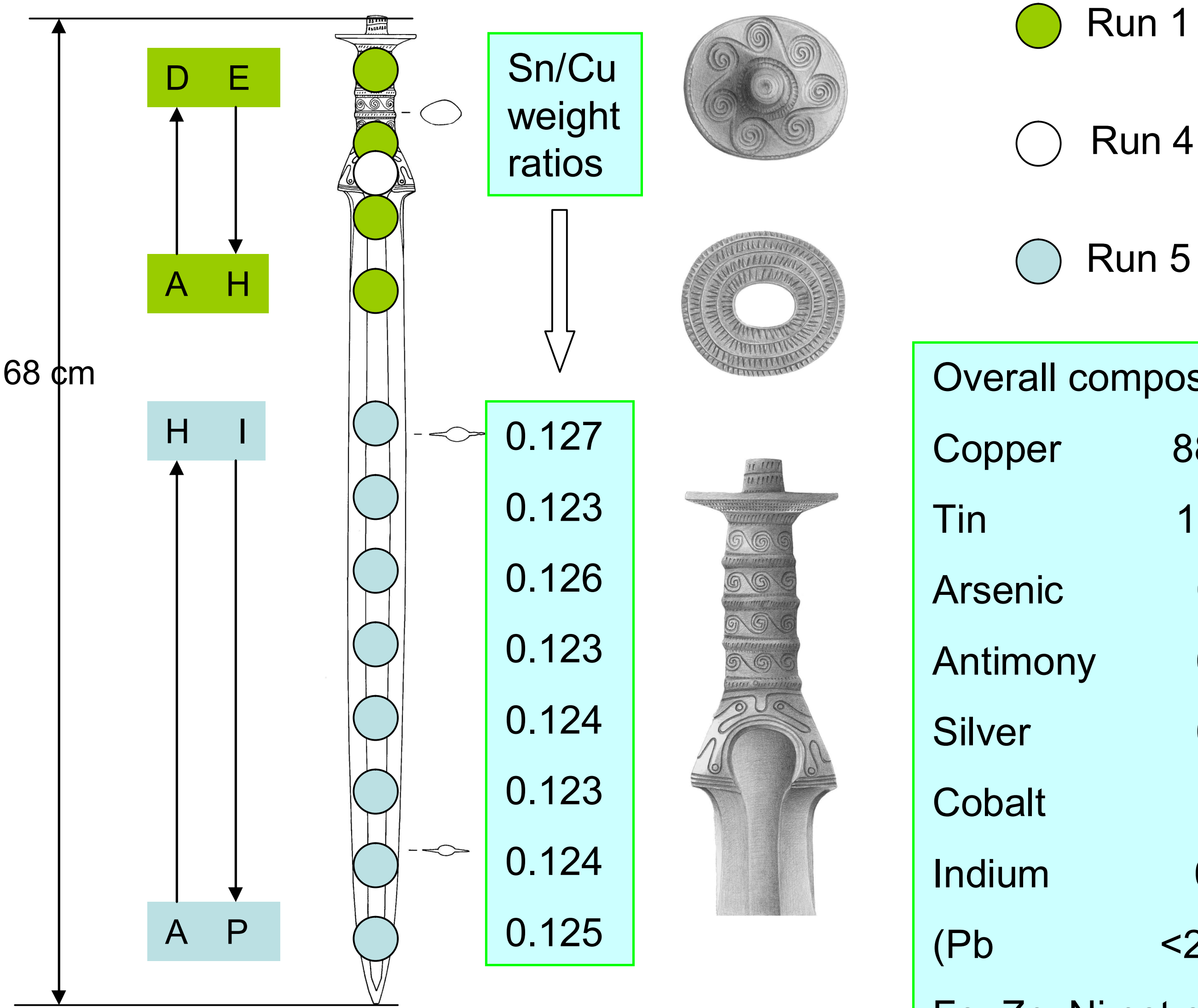
Beam width halve height 20 mm





Dodecaeder set-up

Budapest, 23-25 Sept. 2009



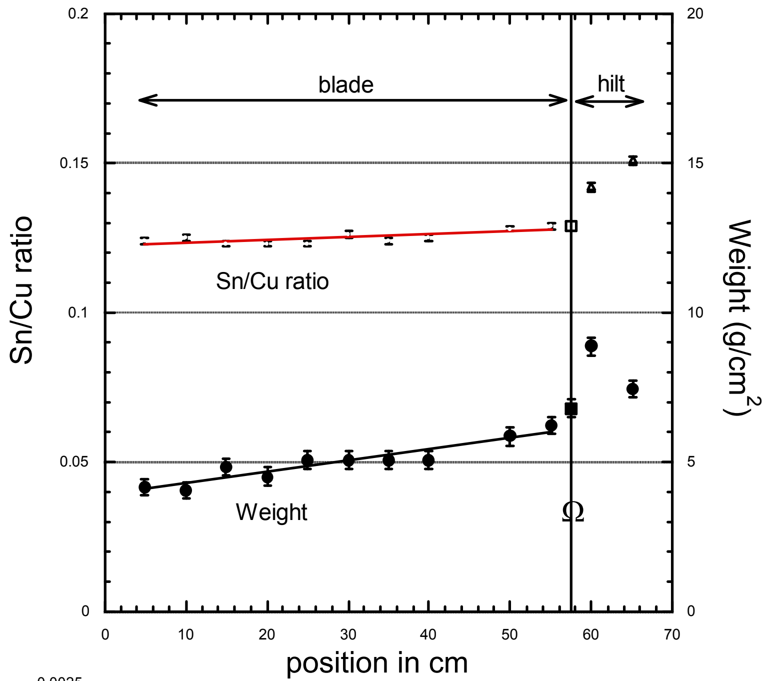
Overall composition:

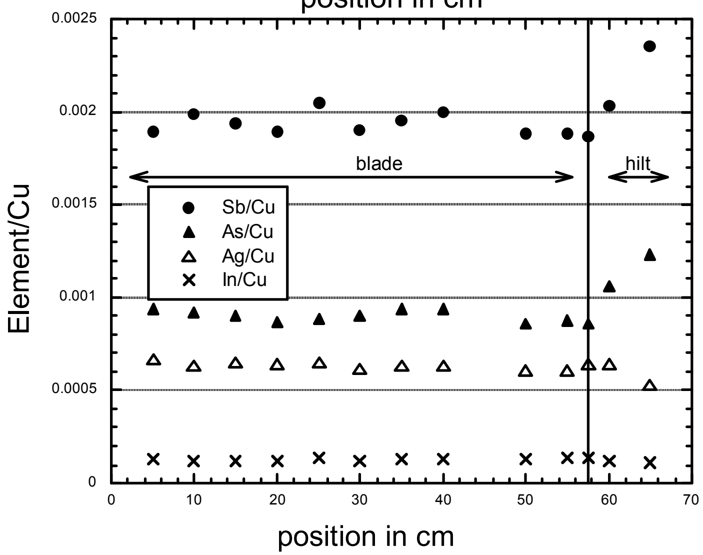
Copper	88.59 %
Tin	11.02 %
Arsenic	0.10 %
Antimony	0.20 %
Silver	0.05 %
Cobalt	0.03 %
Indium	0.01 %
(Pb	<2.5 %)

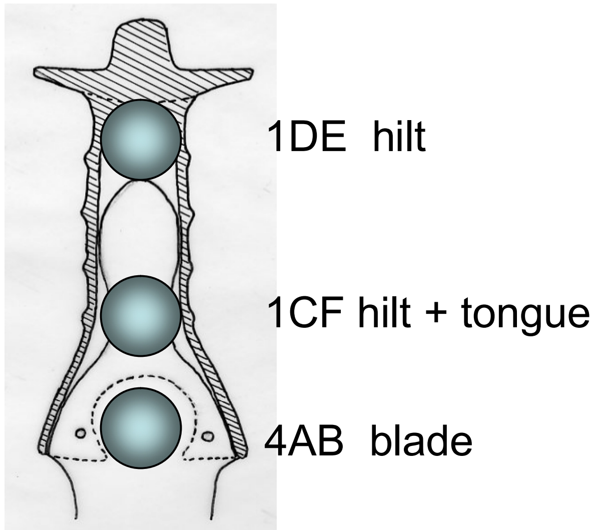
Fe, Zn, Ni not observed

Buggenum sword

variations of weight ratios X/Cu and weight with positions on the sword

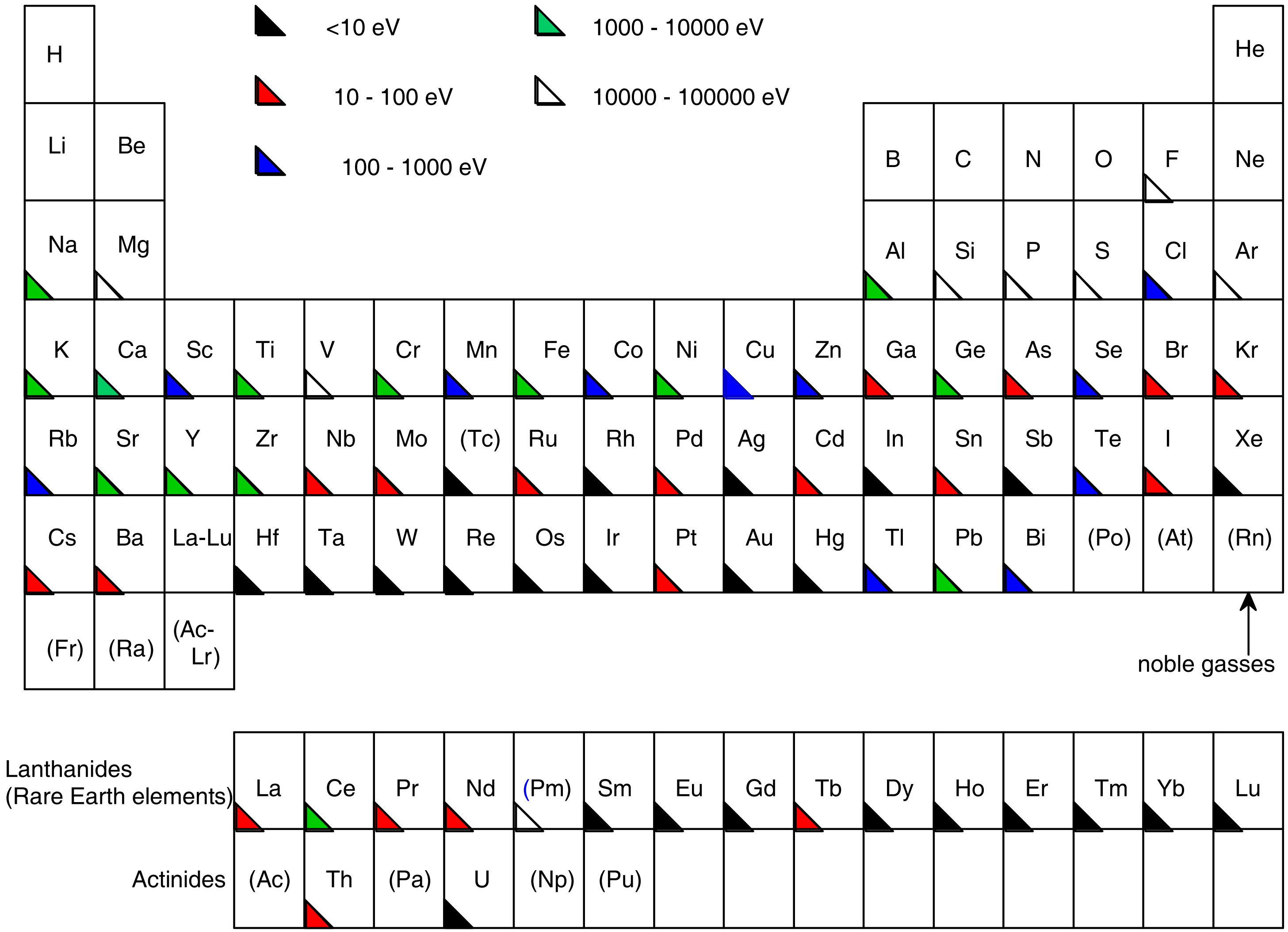






TU Delft
Delft University of Technology

Periodic system of elements. Usefulness of NRCA.



sensitivities for elements differ:

Generally high sensitivity for low energy resonances, e.g. In, Ag, Sb

low sensitivity for high energy resonances, e.g. Pb, Fe, Ni

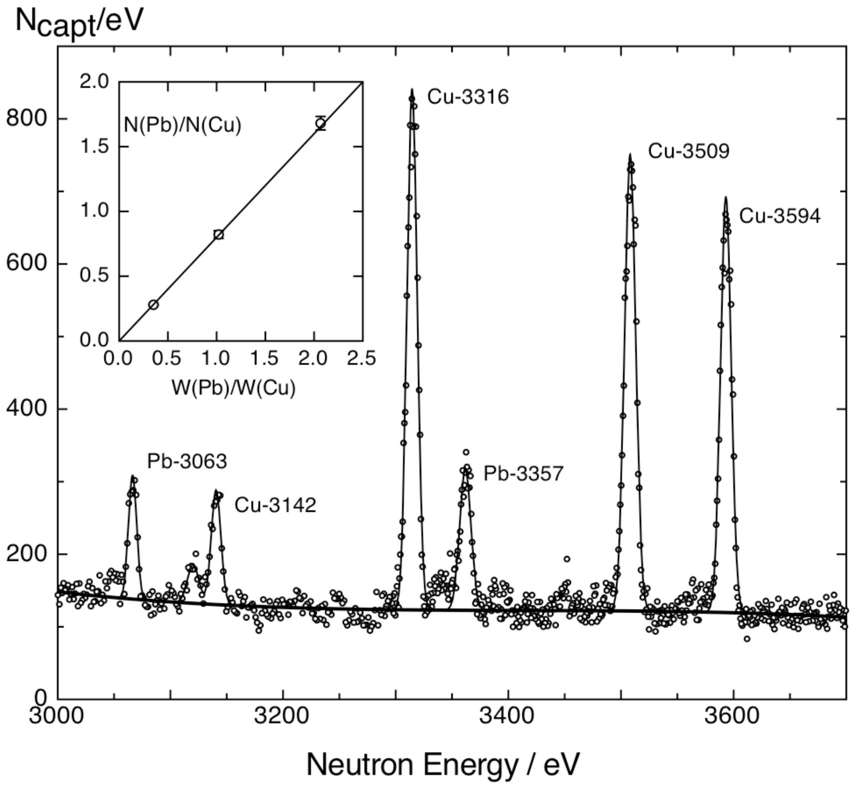
The case of Pb

Resonances at:

3063 and 3357 eV

Compared with

Cu-resonances



15-20 % Pb in

Roman water taps

The case of In.

Resonance at

1.457 eV

Indium seen in many bronze artifacts

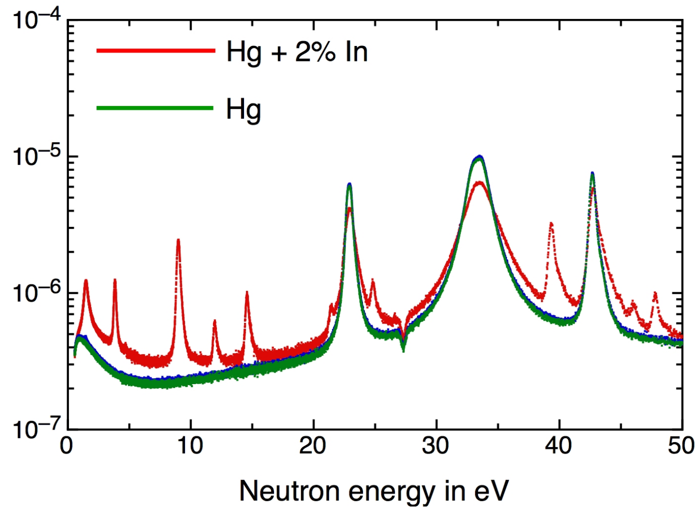
What can we learn from this?

Elemental compositions in weight % of some bronze artifacts

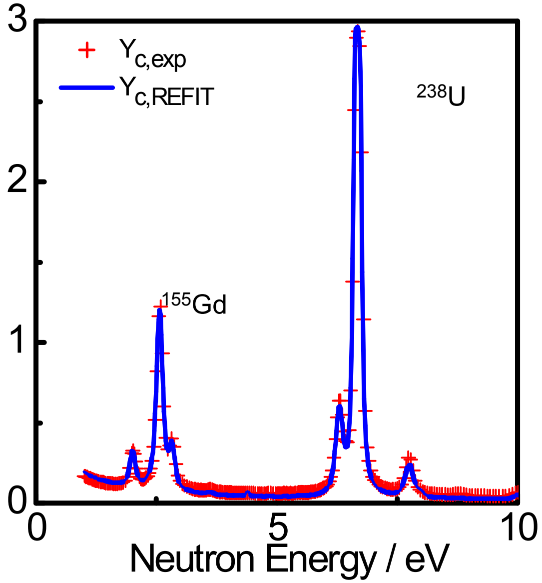
element	"Nijmegen" Roman water tap	Etruscan OC68 (fake)	Geistingen socketed axe (1938/X4)	Jutphaas sword (tip)
Cu	73.56	81.05	94.30	86.07
Sn	8.19	5.81	0.063	13.39
Sb	0.117	0.243	1.63	0.114
As	0.055	0.182	0.67	0.226
Ag	0.053	0.21	0.54	0.0124
In	--	--	0.0005	0.0028
Zn	0.883	5.57	--	0.153
Co	--	--	--	0.033
Pb	15.6	6.7	--	--

Studies for some applications of NRCA at IRMM

1) To follow the indium content during reprocessing of the Hg(+In) coolant used for the neutron production target of GELINA.



2) Test to determine gadolinium as a poison in nuclear fuel by NRCA.



$^{dep}U_3O_8$ + $^{nat}Gd_2O_3$

U (g)	Gd(g)	n(^{155}Gd)/n(^{238}U) nat. abundance	NRCA
20.988	0.0536	5.77×10^{-4}	$(5.76 \pm 0.04) \times 10^{-4}$
20.608	0.5205	5.71×10^{-3}	$(5.73 \pm 0.01) \times 10^{-3}$
18.656	2.6240	3.13×10^{-2}	$(3.14 \pm 0.01) \times 10^{-2}$

With ^{157}Gd the NRCA values are about 8% higher, incorrect resonance parameters of ^{157}Gd?

What do we like to see:

-- better resonance data:

(e.g. $^{63,65}Cu$, ^{75}As, ^{157}Gd)

-- Analyzing methods for the non-expert

-- more simulations, a.o. for scattering+capture

-- local distributions of inhomogeneous objects

Imaging methods, e.g. ANCIENT CHARM

Cu resonance parameters are poorly known

E_o	Cu	J	$2g\Gamma_n$	Γ_g	Γ_{tot}
230	65	2	0.0222	0.245	0.26276
579	63	2	0.7375	0.485	1.075
650	63	1	0.0127	0.35	0.36693
807	63	2?	0.0050	0.50	(0.504)
994	63	2?	0.01166	0.50	(0.51328)
1362	65	1?	0.022	0.395	(0.4243)
3316	63	2?	0.0224	0.50	(0.5179)
3594	65	1?	0.0470	0.395	(0.4577)

My thanks go to:

-- the GELINA staff,

-- collaborators in Geel,

-- to the Ancient Charm collaboration,

-- to museums for lending artifacts.

The next guy to look at