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# Study of Ancient Metallic Artifacts by Using Neutron Imaging Techniques

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with a contribution of Alex R. Furger

## Summary

This article describes results of successful investigations of metallic and ceramic find pieces with archeological and culture historical importance. Since objects of our cultural heritage are often unique and of high value the studies have to be performed completely non-destructively. Although methods with X-rays are very common and available at many places, they often fail when larger amounts of metals are involved. In such cases, neutron imaging methods can be very useful since the penetration of neutrons through (heavy) metals is much higher and clear information about the inner content and structure of inspected objects can be obtained. In the same way, corrosion of metal can be studied very carefully due to the high sensitivity of neutrons for hydrogen, a component in the corrosion products. Also organic remains and agents from conservation treatment can be inspected easily in this manner.

Neutron imaging with state-of-the-art performance and image quality is established at only a few facilities world-wide, given by the limited number of suitable strong neutron sources and the needed experimental infra-structure. The Paul Scherrer Institut operates the Swiss spallation neutron source SINQ since 20 years with success and has available dedicated neutron imaging beam lines which are satisfying highest standards and flexibility.

We present the physical background of neutron imaging, the principle layout of the imaging facilities and describe the experimental techniques in radiography and tomography modes.

Based on this knowledge it becomes possible to describe and to analyze the data obtained by the different examples found in Switzerland in this paper: the golden Marc Aurel bust from Avenches, a sword from the lake of Zug, a *gladius* from *Vindonissa*, further find pieces from Avenches (bronze bracelet, sanitary equipment) and ceramic casting remains from *Augusta Raurica*.

## Introduction

Many objects from our cultural heritage need deeper analysis in order to get knowledge about their provenance, origin, manufacturing process and conservation status. Since the objects should be preserved for future generations, the utilized research methods have to be as non-destructive and non-invasive as possible. There are many analytical tools available and established in museums and dedicated analysis labs like X-ray fluorescence, Raman spectroscopy or even X-ray tomography.

Since the discovery of X-rays at the end of the 19<sup>th</sup> century cultural heritage objects have been investigated with this penetrating kind of radiation where the inner content becomes visible in many cases. This technique has been continuously improved by means of higher performance of the X-ray sources with respect to photon energy, source spot size, intensity and the implementation of new detection systems (replacement of film by digital imaging systems). There is also an access to very intense synchrotron sources enabled in a few cases<sup>1</sup>. However, their beam size is often small and the photon energy too low for the investigation of larger objects (on the order of cm to dm).

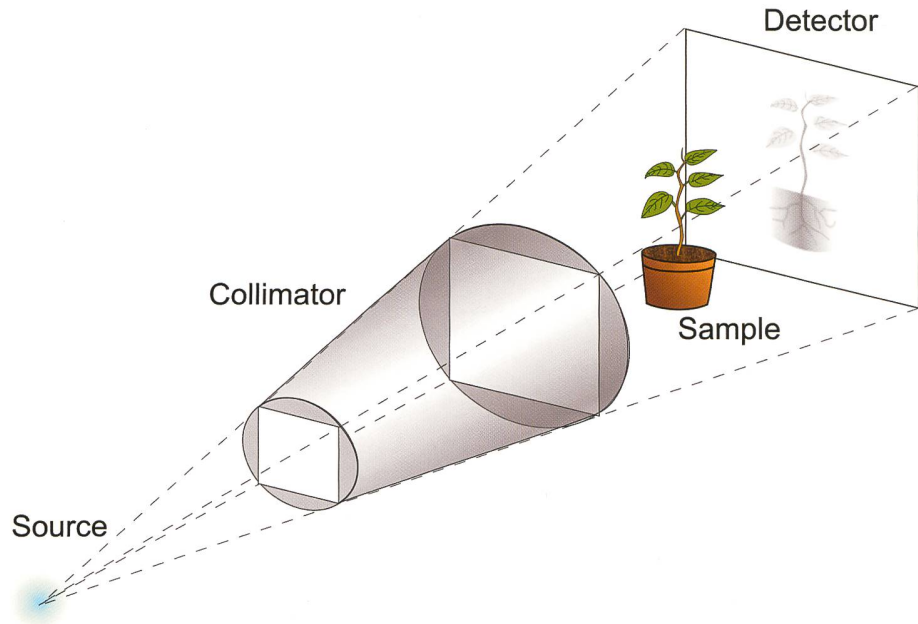
An alternative and until now not yet very much established technique for non-invasive analysis in the area of cultural heritage research is given with the use of neutrons. Neutron imaging works similarly to the more common X-ray imaging (fig. 1), but the source, the beam line layout and the detector for the detection of the radiation are different. Below we will describe the principle differences in the two kinds of imaging techniques caused by the interaction mechanism of the radiation with the sample materials. With

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<sup>1</sup> Stampanoni *et al.* 2007.

**Fig. 1**

Simplified setup of a transmission imaging facility where the source and the detector can be attributed to either neutrons or X-rays, respectively. The collimator length  $L$  and the aperture  $D$  define the characteristic collimation ratio  $L/D$  which influences the image quality with respect to inherent image blurring. Other parameters like the field of view and the content of accompanying gamma radiation can determine the facilities' characteristics too.



knowledge of the interaction properties it is quite easy to decide which option (neutrons or X-rays) is preferential for the particular non-invasive investigation. In several cases, it is useful to have the information of both methods with the same image quality to enable the «data fusion», that is pixel/voxel wise referencing and correlation.

Neutron imaging techniques are available with best performance at large scale facilities because high beam intensity is a requirement for suitable image quality in reasonable low acquisition time. Such sources are either based on a nuclear fission reactor or driven by a proton accelerator, causing spallation<sup>2</sup>.

We take the opportunity of this article to explain the method of neutron imaging and its useful application to cultural heritage objects with examples of Swiss finds. Based on the imaging data, but also under augmentation of other information and with confirmation of comparable knowledge the objects get a deeper analysis. Many questions can only be answered in this way.

## Neutron Imaging principles

As presented in fig. 1, the beam from the initial source is guided towards the object of interest via the collimator system while the detector (as two-dimensional area across the beam) behind the sample registers the transmitted part of the beam intensity. The sample material undergoes interaction with the applied radiation in the beam by either absorption or scattering.

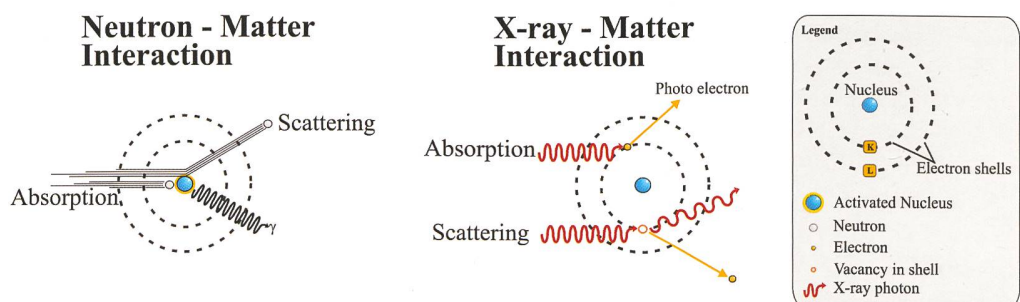
As indicated in fig. 2, neutrons interact exclusively with the atomic nuclei and «ignore» the electrons at all. In contrast, X-rays have no strong impact to the nuclei, but a strong interaction with the electrons in the atomic shell. Therefore, the heavier atoms systematically attenuate the beam more than light elements can do.

In the case of neutron interactions, there is no such systematical behavior in the interaction scheme: even different isotopes of one specific

<sup>2</sup> Lehmann/Trtik/Ridikas 2017.

**Fig. 2**

Interaction scheme of neutrons (left) and X-rays (right) with the atom: while neutrons have their impact on the nucleus only, the X-ray mainly interact with the electrons in the shell. This complete different mechanism results in images with alternative contrasts even if the imaging detector performs very similar with respect to spatial resolution (see e.g. fig. 3).

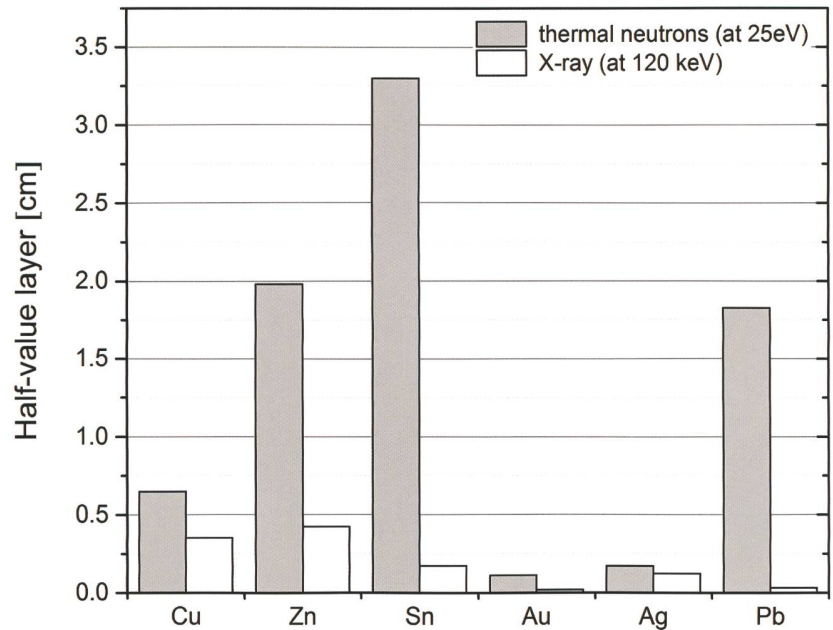


element can have strongly deviating properties. A deep knowledge of the materials behavior is needed to decide about the best experimental strategy. The cross-section data, which are values for the interaction probability with matter, are tabulated in specific data bases<sup>3</sup>.

To compare the degree to which a certain element attenuates a specific radiation the so-called «half value thickness» can be used. The half value thickness represents the material thickness at which the intensity of the radiation is reduced by 50% behind the sample layer. Fig. 3 shows these values for elements where the differences between the attenuation for X-ray photons (in this example 100 keV) and thermal neutrons (at 25 meV) is especially pronounced.

The example of the bust sculpture from Avenches, where we compare the transmission radiography images in fig. 4, shows the practical relevance of penetration power of the neutrons. While a transmission is only possible with the X-ray tube voltages higher than 200 kV high voltage, the dynamic range of the neutron imaging data allows much better and detailed studies, including neutron tomography (see below).

Today, neutron imaging is based on digital detection systems where a pixelated matrix delivers the map of the transmitted neutrons in data files. These systems are similar to the very common digital photography but the involved sensors have to be more sensitive by orders of magnitude since the light from the neutron converting scintillator screen is very weak in intensity (see fig. 1).



Based on the digital data it is possible to apply more advanced image processing like tomography, real-time imaging or direct quantification of the sample content. Most of these processes are taking into account the Beer-Lambert's law of the beam attenuation, which is valid on first order<sup>4</sup>.

**Fig. 3**

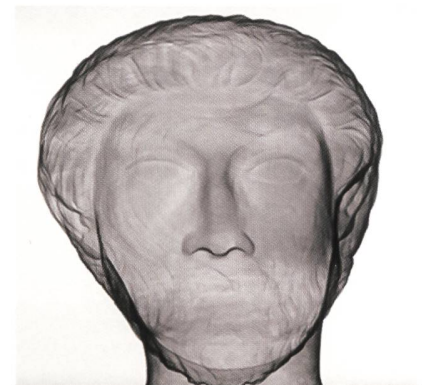
Half value thicknesses for relevant heavy metals for thermal neutrons in comparison to X-rays with 100 keV photon energy. These data demonstrate the high potential of neutrons to penetrate thicker layers of the involved materials.

## Facilities for Neutron Imaging at the Paul Scherrer Institut

The Swiss national neutron source SINQ is based on the spallation principle where high energy protons (590 MeV) are sent to a heavy metal target (e.g. lead) and about 10-15 neutrons are emitted per spallation act. This large scale facility with a strong proton accelerator (MW range) and the spallation target station is the host of neutron research in Switzerland based on about 15 beam lines for neutron scattering and imaging (see fig. 5). It is open for scientific

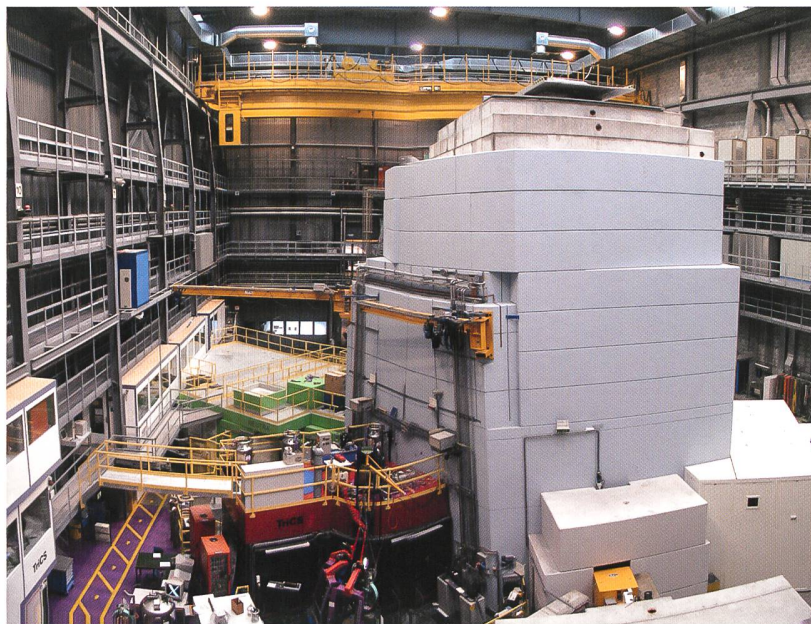
**Fig. 4**

The golden bust of Marc Aurel found in Avenches (photo left), studied by X-rays (200 kV - middle) and thermal neutrons (right).



<sup>3</sup> <https://www.nist.gov/pml/x-ray-mass-attenuation-coefficients>; <https://www.nds.iaea.org/exfor/endl.htm>.

<sup>4</sup> Hassanein/Lehmann/Vontobel 2005.



**Fig. 5**  
The spallation neutron source SINQ, accelerator driven and host of about 15 beam lines for many applications for neutron research in Switzerland, including the NEUTRA and ICON facility for neutron imaging.

**Fig. 6**  
Thickness map of the golden bust of Marc Aurel determined by neutron tomography investigations at the NEUTRA neutron imaging beam line at SINQ (PSI). Cf. De Pury-Gysel/Lehmann/Giumlia-Mair 2016.

and industrial users and enables unique studies, including those for cultural heritage observations if feasible and justified.

The neutron imaging techniques are established at two different beam lines: NEUTRA for thermal neutrons (and X-rays up to 320 kV) and ICON for cold neutrons (and X-rays up to 150 kV). The field-of-view can be tuned according to the object size from 40 cm in diameter down to about 1 cm since the initial beam from the source covers these dimensions. Since the detection systems have 2000 and more pixels in the two dimensions each, the nominal spatial resolution is between 200 and 5 micro-meters accordingly. This high flexibility is needed and essential to satisfy the very different requests from the wide range of users and applications.

Next to the detector properties, the achievable spatial resolution is also determined by the beam divergence, the scattering in the sample and the camera optics. The current technical limit with all parameters involved optimized is on the order of 1 to 2 micro-meters<sup>5</sup>.

Samples can be manipulated by either rotation around the vertical axis (as needed for tomography), tilted or shifted with high precision and repetitively in all three directions for scanning purposes. With the help of an in-situ climate chamber, specific conditions can be provided and tuned during an ongoing experiment in the temperature range from -10°C to 100°C and the air humidity 10 to 95% r.h. In this way, also processes in conservation can be followed, where the uptake and loss of moisture is of special importance due to the high sensitivity of the method to hydrogen contents.

There are some arguments against studies of metals with neutrons due to a possible activation risks. This holds for a very few materials (silver, Ag ; cobalt, Co) only since the activation probability is normally low and the half-life of the induced activation is short. Even very bulky metallic structures, which are exposed over several hours, e.g. during a tomography, are in most cases activated only for a short period and can be returned after the investigation.

## Results of studies

This article contains objects and their investigations from Swiss origin only. Because our facilities have been requested from teams around the globe many other studies have been performed already in addition. However, it is our deep intention and motivation to serve Swiss customers preferentially and most professionally since PSI is governmentally funded and a little return can be given in this way.

### The Marc Aurel from Avenches

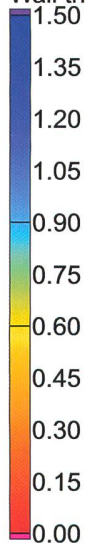
A photo of this object is shown already in fig. 4. The very pure golden object was found in 1939 and has survived in the soil for many centuries without heavy corrosion<sup>6</sup>.

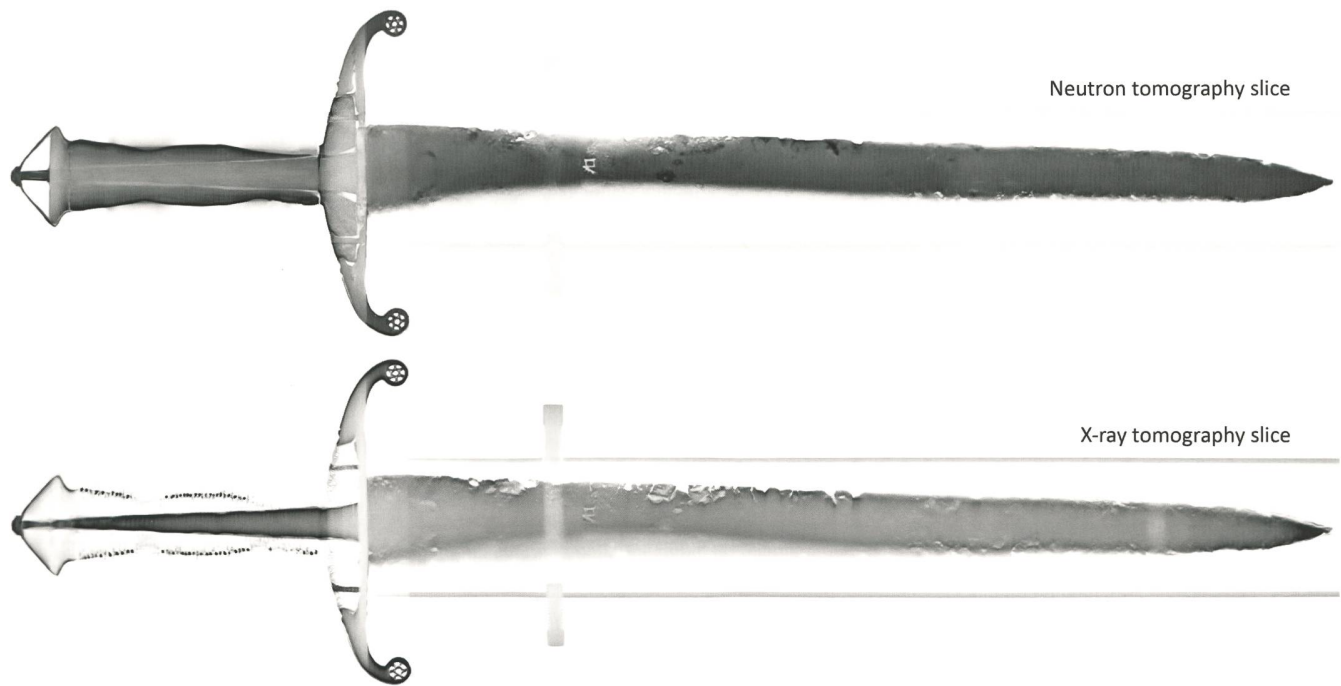
The aim of the neutron investigation was to obtain a global overview on the objects material thickness with a precision on the order of 0.2 mm over the whole object volume. Based on this knowledge some principle questions about

5 Trtik/Lehmann 2016.

6 Details of the sculpture are summarized in: Hochuli-Gysel/Brodard 2006.

Wall thickness [mm]





the manufacturing process should be possible to answer: has the whole bust been made in only one piece... or combined by several individual pieces.

The study was done in the tomography mode where the object has to be inspected from about 300 individual perspectives while rotating around the vertical axis. After the volume reconstruction with the «filtered back-projection» method and the assumption of parallel beam approximation the data were analyzed by special software tools. There is one option to measure the wall thickness of the bust and to present it in a color coded map. These data are given in fig. 6. Neutron imaging shows the different thickness of the gold sheet in the various parts of the bust, some small traces of soldering and tiny repairs, but provides strong evidence for manufacture in one piece<sup>7</sup>.

### The sword from Oberwil

In 2010, a medieval sword was found in the lake Zug which was attributed to a period in the 15<sup>th</sup> century. Stored in the water over the long period it is in a surprisingly well preserved condition. Nevertheless, some conservation treatment was proposed and a replica was intended to be build. For these purposes, a detailed three-dimensional investigation (tomography) with non-destructive measures was initiated using the neutron and X-ray capabilities at the PSI facilities.

Because the length of the sword (about 70 cm) exceeds the field-of-view of the imaging systems, the study was performed in three steps by moving the object in vertical direction. The obtained three volume data sets were combined into one without any gaps at the interfaces. Slices from the middle plane of the sword, obtained by thermal neutrons and 150 kV X-rays are presented in fig. 7.

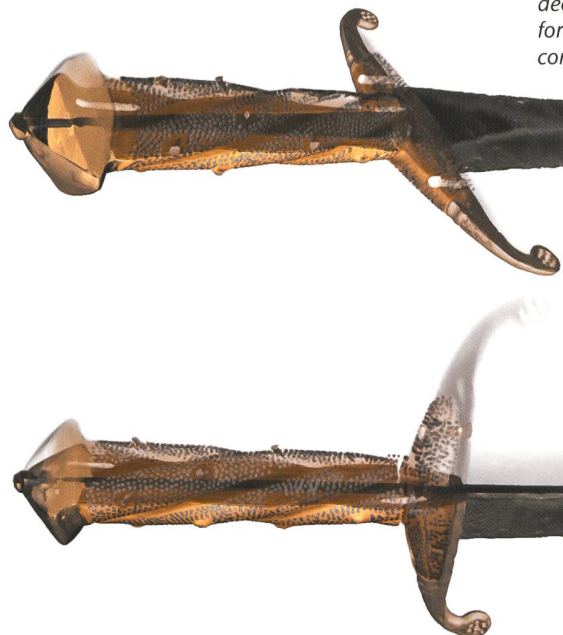
This comparison shows clearly the differences in the observation, in particular in the area of the sword hilt which consists of a boxwood grip with decorative tin-amalgam inlays: neutrons can visualize the wooden structure while X-rays show

**Fig. 7**

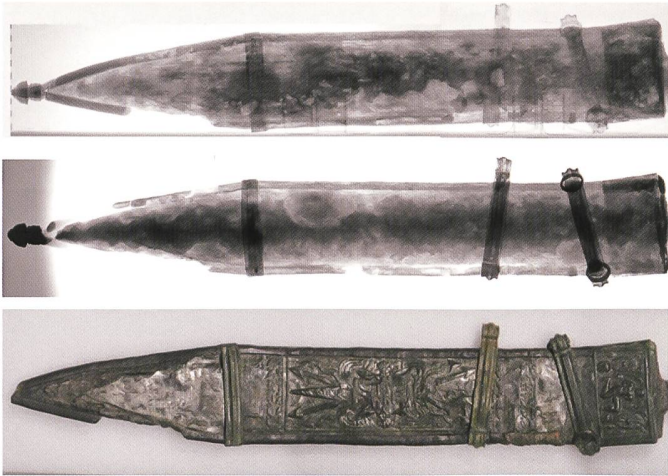
*Results of the tomography reconstruction of the sword from lake Zug, taken with thermal neutrons and 150 kV X-rays, delivering complementary information on the object.*

**Fig. 8**

*3D-visualisation of the combined X-ray and neutron CT data sets. Assembly of the different features of the sword from lake Zug into one data set as template for the replica, including the decoration parts and bolts for the internal fixation of the components.*



<sup>7</sup> The detailed conclusions of the study are summarized in: De Pury-Gysel/Lehmann/Giumlia-Mair 2016.



**Fig. 9** (above)  
Neutron (top) and X-ray (middle) radiography images of the gladius from the Vindonissa settlement. Vindonissa Museum in Brugg (AG).

exclusively the metallic pieces at the surface of the handle. Only by combining the two data sets a complete «construction plan» was derived. On the other hand, the degree of corrosion and aging was determined with the aim to have the best strategy for conservation measures. Next to the cleaning of the surfaces, the protection of the metallic structures has been an important issue. The exhibition in Zug contains in the showcase now next to the original sword the newly produced copy, based on the tomography data obtained at the imaging beam lines at PSI<sup>8</sup>.

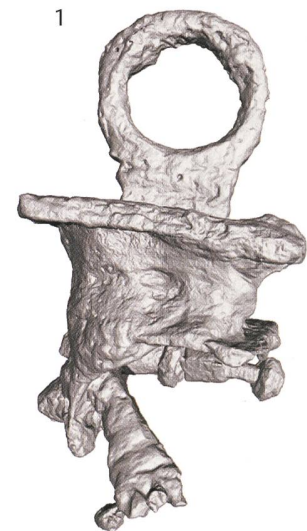
### The gladius from Vindonissa

This object was one of the first cultural heritage objects studied in collaboration with the experts from the University Zurich, Institute for Archaeology. This sword type was very common in the Roman empire while the age is on the order of 2000 years about. While the status of preservation is reasonable the knowledge of inner structures and their composition was quite limited. Therefore, radiography images were taken with thermal neutrons and X-ray, using imaging plates. The results are given in fig. 9. On the first glance, both image data sets look similar and show the structure quasi-transparent. While the X-ray data in the middle in the figure gives a clear picture how heavily damaged the sword itself (metal parts) is presently, the neutron data (top) demonstrated that next to the metal two other components are inside the sample: the remaining parts of the wooden sheath and the resin amount which was used in the previous conservation treatments. At the outer layer, some pieces were preplaced by plastic components, visible by neutrons, hidden for the X-rays. Further treatment and conservation measures can be planned on the basis of the image data.

**Fig. 10** (right)  
Two objects from a necropolis (À la Montagne) in Avenches (VD): a sanitary tool (1) and a bracelet (2) made of copper alloy and presenting different questions.

### Finds from Avenches

Two samples from recent excavations next to the Avenches settlement (fig. 10) were studied by means of high resolution tomography with the aim to understand their composition, production process, usage and purpose next to the conservation status. With modern tools of image processing and data handling, virtual slices and views were enabled in order to look into encapsulated structures and the segment individual pieces<sup>9</sup>.



<sup>8</sup> Detailed descriptions of the sword can be found in: Bernasconi/Binggeli/Sager 2014; Frey 2014; Schmidt-Ott/Hunger/Mannes 2014; Mannes et al. 2015.

<sup>9</sup> The results and findings are summarized in more detail *supra*: A. Duvauchelle, M. Krieg, Un nécessaire de toilette de la nécropole d'Avenches/À la Montagne. L'apport des analyses, p. 139-148; A. Duvauchelle, M. Krieg, Un bracelet-étui de la nécropole d'Avenches/À la Montagne. L'apport des analyses, p. 149-156.

## Casting tools from *Augusta Raurica*

### Roman crucible fragments analyzed by neutron imaging

A few samples of a series of 893 crucibles from *Augusta Raurica* (Augst, BL) were examined by neutron imaging at the Paul Scherrer Institut (fig. 11)<sup>10</sup>. The aims were to visualize the assembling of a very rare, small crucible with handle (T851) and the traces of hot metalworking executed in these cups of refractory clay. The handle turned out to be added secondarily to a ready-to-melt crucible with another, very sticky mixture of clay (*lutum*). The few analyzed sherds of used crucibles all showed many prills of «bronze», not only in the inner surfaces, but outside as well. The more the crucible fabric is porous, the more prills are detected even in the inner ceramics (e.g. T867).

### An intact Roman «lost wax»-casting mould analyzed by tomography

A completely preserved casting mold, 128 mm in height, was excavated in 1966 in the Roman town of *Augusta Raurica* (Augst, BL). The pear-shaped hollow body is made of sandy, low-fired clay, but was never filled with molten metal. A three-dimensional X-ray microtomographic imaging performed with the XTRA setup at the NEUTRA facility of SINQ provided the negative of a crouched down divinity to be cast in this mould, like they are known from some Roman bronze lamps. A separate article in this volume<sup>11</sup> lists a number of these rare hollow moulds of the «cire perdue»-technique which have – for reasons we cannot reconstruct – never been used by pouring metal in them and not broken up.

## Outlook to further methodical options and applications

This overview paper can only show few examples of studies with neutron (and X-rays) in the radiography and tomography mode<sup>12</sup>.

Although many questions can be answered by means of these routine methods, there were

10 Furger 2017, fig. 22 and 49-52.

11 A. R. Furger, Eine römische «Gussbirne» aus *Augusta Raurica* (Augst, BL): durchstrahlt und enttarnt, *supra*, p. 157-170.

12 More details are given in the references and can be found in related other publications: Deschler-Erb *et al.* 2004; Lehmann *et al.* 2005; Lehmann/Hartmann/Speidel 2010.

13 Kockelmann *et al.* 2007; Lehmann *et al.* 2009; Mannes *et al.* 2014.

14 Trtik/Lehmann 2016.

developed more sophisticated ones, where profit can be seen also for cultural heritage studies. Some of such approaches were already applied successfully in related studies<sup>13</sup>.

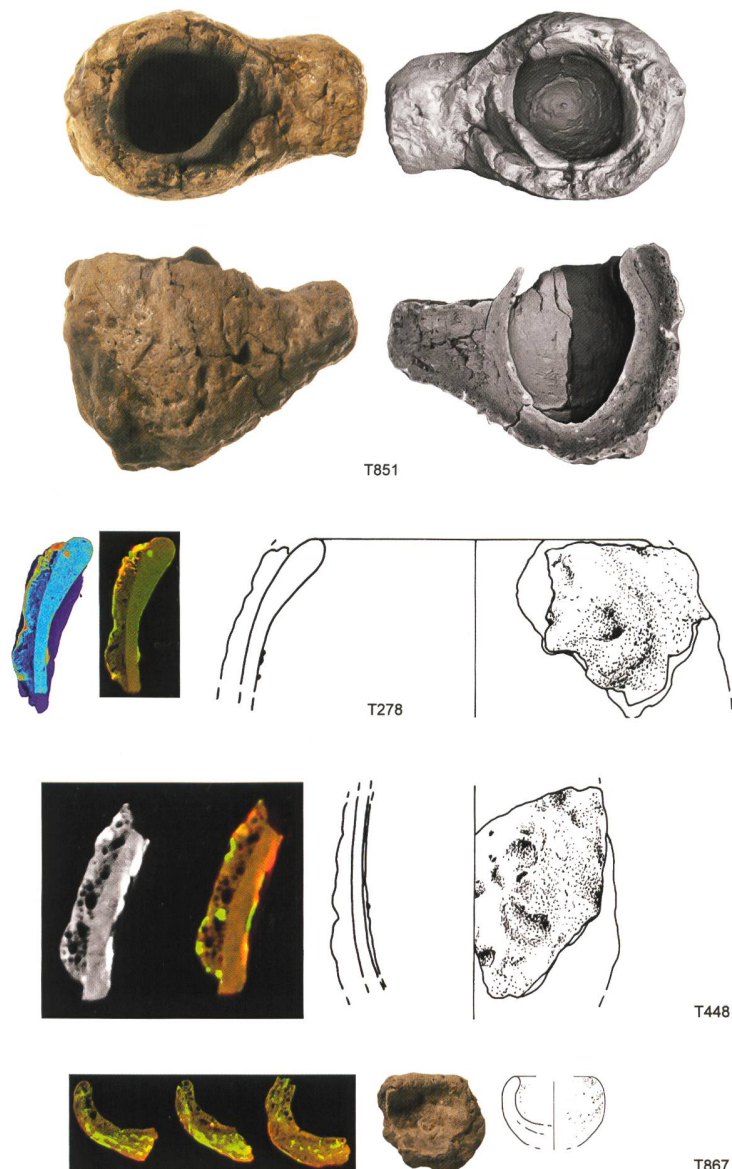
In particular, the reduction of the energy band enabled the access to diffraction features and the crystalline properties of metal can be studied on the macroscopic scale.

With the help of high resolution detection systems (project «neutron microscope»<sup>14</sup>) the access to the micro-meter range, however with the unique contrast of neutrons to hydrogenous samples can be enabled.

Neutron imaging can also be performed in different settings on the time scale with exposure time from seconds down to milli-seconds per image. In this way, time dependent studies like the ingress of preserving agents into the matrix of a porous material can be followed very precisely.

**Fig. 11**

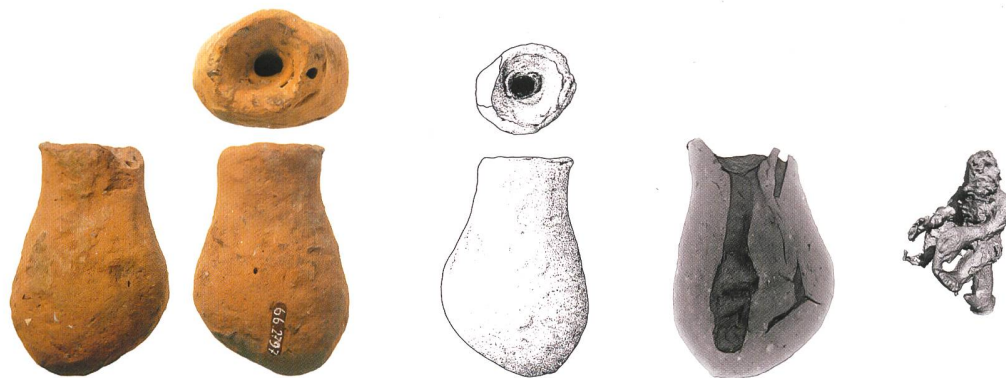
Four crucibles for non-ferrous alloys from *Augusta Raurica*, examined by neutron imaging. The numbers refer to Furger 2017. The inner structure of the clay (top right) and many metal droplets on the vitrified surfaces and in the porous ceramics are clearly visualized by this method.





**Fig. 12**

The completely preserved casting mould from Augusta Raurica; from left: photographs, drawing, longitudinal tomography slice and the virtual reconstruction of the hollow cavity (= statuette of a male divinity to be cast) by tomographic 3D-X ray imaging; height: 128 mm.



Very interesting and promising is the combination and dedicated merge of neutron and X-ray data in the optimal manner when the pixel/voxel-wise information is experimentally obtained. This approach enables the determination and specification of most of the contained materials in complex samples<sup>15</sup>.

### Conditions for the access to the facilities

The imaging facilities are open for research projects with partners from museums and archeological institutes if the purpose and the relevance are justifying it. All results have to be published in adequate manner. However, the proposals for related beam time are in competition with other applications and the success cannot be guaranteed a priori.

<sup>15</sup> Kaestner et al. 2017.

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Doc. MRA

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SMRA

Site et Musée romains d'Avenches.

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## Illustration credits

Fig. 1, 2 (Mannes 2009, modified), 3, 4 (middle), 5-8, 9 (top, middle), 10  
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Fig. 4 (left)  
Site et Musée romain d'Avenches.

Fig. 9 (bottom)  
Vindonissa Museum, Brugg.

Fig. 11-12  
Augusta Raurica, Augst / Paul Scherrer Institut, Villigen (AG).