IL NUOVO CIMENTO Vol. 30 C, N. 1 Gennaio-Febbraio 2007
DOI 10.1393/ncc/i2006-10050-x

The non-destructive study of museums objects by means of neutrons imaging methods and results of investigations(*)

E. H. Lehmann, P. Vontobel and G. Frei

Paul Scherrer Institut - CH-5232 Villigen PSI, Switzerland

(ricevuto il 31 Ottobre 2006; pubblicato online il 22 Febbraio 2007)

Summary. — Based on experience from many kinds of investigations with neutron imaging methods (radiography, tomography, time-dependent studies) and in comparison to conventional X-ray methods, the authors discuss the potential of future improved studies for cultural-heritage purposes. Whereas the focus of the paper is on the imaging aspect, other established techniques as neutron activation analysis (NAA), prompt gamma activation analysis (PGAA), neutron-induced autoradiography and neutron scattering are mentioned too. Although a great potential for studies similar to those described in the paper exists, a considerable effort is needed to define the best-suited methods for the dedicated cultural historical request. A real barrier between the experts at large research facilities (as, *e.g.*, neutron sources are) and the partners from the museums side has to be overcome in order to solve the problems. A joint European approach will help in this respect.

PACS 87.59.Fm – Computed tomography (CT).
PACS 29.25.Dz – Neutron sources.
PACS 07.05.Pj – Image processing.

1. – Introduction

The investigation and preservation of objects from our cultural heritage is an important task for archaeologists, historians and restoration experts. For this purpose, most of the larger museums and collections operate units with natural scientific background. The kind of the objects at different sites is diverse due to the individual history of the country or region. Therefore, different materials have to be studied in order to solve the specific historical, scientific or restorative question.

The investigation of objects from cultural heritage should be done non-destructively per definition to enable their transfer to later human generation without loss. Neu-

(*) Paper presented at the Workshop "RICH—Research Infrastructures for Cultural Heritage", Trieste, December 12-13, 2005.

tron investigations are another option among the non-destructive methods with specific characteristics as will be explained in the following paper.

The investigations with neutrons should never be considered isolated and completing, but as a supplement to other information about the objects under study. Here, the *a priori* knowledge of the museums experts has the same value as the natural scientific studies can provide. Only the joined information about the museums objects will deliver a new and enhanced level in the knowledge.

The use of the large existing installations (as strong neutron sources are) for the study and preservation of cultural heritage should be an important aim within Europe. A tighter and improved contact between the experts from this research infrastructure with the museums people should be an important task for the future. A dialog is needed to bring the options and the needs closer together.

2. – Neutron interaction with matter in comparison to X-ray

X-ray techniques have been available as standard tools since many years. They are in use in hospitals and facilities for non-destructive industrial investigations. The individual installation differs mainly in the energy spectrum of the source and therefore in the different options for the transmission of objects and the contrast obtained in the transmission image from the involved materials.

The principle of a radiography set-up is similar for both X-ray and neutron inspection: from the source of the radiation a beam is guided through the collimator to the object and the shadow image is registered in the two-dimensional detector placed perpendicular to the beam direction directly behind the object. The interaction of the radiation with the object delivers the needed contrast to enable the visualisation of material distributions as its shadow image on the detector screen.

It is well known that X-ray collision with matter happens via an interaction with the electrons in the atomic shell as Compton scattering, photo effect and pair production. Therefore, the number of electrons per atoms is a clear measure for the interaction probability—that means heavier elements deliver much higher contrast than light ones. On the other hand, the X-ray interaction with matter is depending on the initial photon energy. This is important for the samples' contrast on the one side, but for the detector response on the other side. The energy dependence of attenuation coefficients is shown in fig. 1 for some relevant materials. The typical photon energy range is from few keV (especially synchrotron radiation) up to 450 keV (for X-ray tubes) and some MeV for gamma sources.

Similar conditions in transmission are valid for neutrons too. However, the interaction mechanism with the atoms is completely different: neutrons interact with the nucleus and do not recognise the electrons. Two interaction mechanisms are mainly relevant for imaging purposes—neutron absorption and neutron scattering. In first order, all neutrons missing in the direct line behind the sample are considered as lost. This is in principle true for all neutron capture reactions. Scattered neutrons are often more difficult to handle when they contribute to the resulting image behind the sample in the form of a diffuse sky shine.

In the practical neutron radiography and tomography thermal neutrons (E_n around 25 meV) play the most important role. There are two major reasons in respect to this neutron spectrum: reactor-based sources (and this is still the majority) need for the chain reaction in the core the moderation down to the thermal energy; the largest community

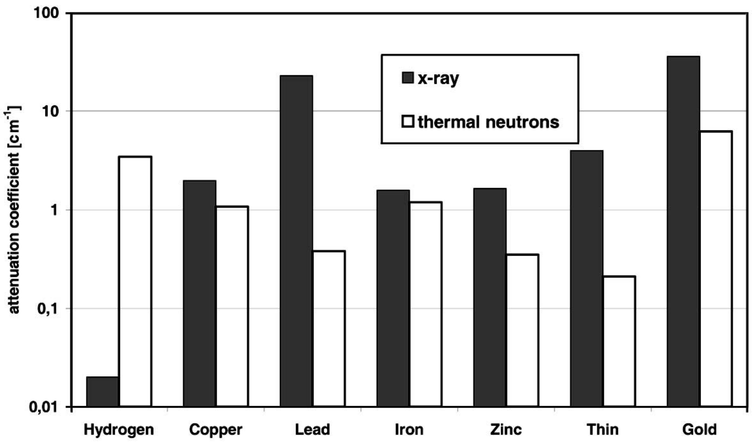

Fig. 1. – Attenuation coefficients for relevant sample materials (comparison of 150 kV X-ray with thermal neutrons). Please, take into account the logarithmic scaling. The values for X-ray are generally higher than for neutrons, beside the case of hydrogen. That means thicker material layers can be transmitted by neutrons, whereas hydrogenous materials will contribute with large contrast to the images.

dealing with the neutron research—the neutron scattering people—require this energy range because of the most efficient interaction with matter.

In the recent years, the neutron energy could be shifted by cold moderators (temperature about 25 K) towards lower values (E_n around 3 meV). Due to the higher interaction probability with matter compared to thermal neutrons both the material contrast and the detection efficiency is increasing with cold neutrons. A variation of the radiation energy as it is possible for X-ray by changing the high-voltage can happen therefore for neutrons in a limited amount only.

With respect to individual material interaction there is no clear systematic as in the case of X-rays. Four regions in the periodic table of elements are important, where large differences exist between interaction probabilities of thermal neutrons and X-rays:

– most of the light elements (H, Li, B, C) have stronger neutron interaction than X-ray;

– heavy elements (Pb, Bi, U, W) have only low interaction with thermal neutrons compared to X-ray;

– there are some especially strong neutron absorbers (Gd, In, Cd) with extremely high cross-sections;

– most of the technically relevant metals are more transparent for neutrons than for X-ray.

Due to these differences in the attenuation behaviour (see fig. 1) of the individual materials it becomes very clear that both kinds of radiation are complementary to each other even if the radiography and tomography methods and procedures are similar in principle. Therefore, the fields of applications must differ—X-rays are ideal, *e.g.*, for the investigation of large hydrogenous assemblies as the human body, where neutrons must fail completely due to the strong scattering on hydrogen.

3. – Different options for neutron investigations

The interaction mechanism for neutrons as described before is mostly relevant for the imaging methods (radiography, tomography) and can be directly attributed to the obtained contrasts in the images. Therefore, a quantification of the sample content can be derived in some cases directly or after the application of correction algorithm (*e.g.*, for the secondary scattered neutrons [1]). The majority of examples described below are based on the imaging techniques established at the dedicated beam lines at the spallation neutron source SINQ [2] at the Paul Scherrer Institut. Similar studies might be possible at other neutron radiography stations within Europe too [3]. The image quality depends very much on the beam properties and the detection system, respectively.

There are some further methods in use for studies of samples from cultural heritage by using neutrons. They are using either dedicated neutron beam lines or neutron fields very close to the source at points of high intensity (up to $10^{14}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$).

Neutron Activation Analysis (NAA) and *Prompt Gamma Activation Analysis* (PGAA) are multi-elemental analysis methods, where the induced radioactivity is measured by means of gamma-spectrometry. Due to the individual decay scheme, each activated material emits separated characteristic energies (spectral lines), which can be used to identify and to quantify the sample content. Depending on the irradiation-decay-history, many different materials (even isotopes) can be measured more or less simultaneously.

Whereas a certain amount of sample material is needed in the case of NAA to be irradiated in a highly intense neutron field, the PGAA acts as an *in situ* method, where the gamma-spectrometer observes the sample directly during the neutron exposure. Both methods complete each other by the sensitivity for mutual different materials. Because the neutron exposure is much higher in the case of NAA, its sensitivity is even higher for less absorbing materials.

Neutron-induced activation radiography is another imaging technique, mainly in use for the study of paintings [4]. The whole area of a painting is equally exposed for this purpose and the induced activity is measured with highly sensitive film methods. In this way, a 1:1 image of the activity distribution is obtained. Due to the different decay properties of, *e.g.*, cobalt, phosphorus or manganese, measured in different sequences, several deviating distributions can be derived from one exposure. Because of the different pigment compositions in the colours, the art historians can derive very valuable information about the painting history and the layers below that visible at the surface.

Neutron diffraction and scattering methods are the dominant ones at the most prominent places for neutron research, located at the strongest neutron sources. However, such techniques have been used only rarely for the investigation of objects from cultural heritage until now. The scattering methods can provide structural information on the atomic level, derived from the patterns of the scattered neutrons in the individual experimental set-up. Because of the large number of different scattering techniques and the special performances, it is impossible to describe the various options in very detail. The dialog between the experts of the scattering technique with the museum people is much more needed compared to the application of imaging methods, where the relation to the object is much more obvious.

Some first results of investigation of the metallic micro-structure in a Celtic helmet were reported recently [5].

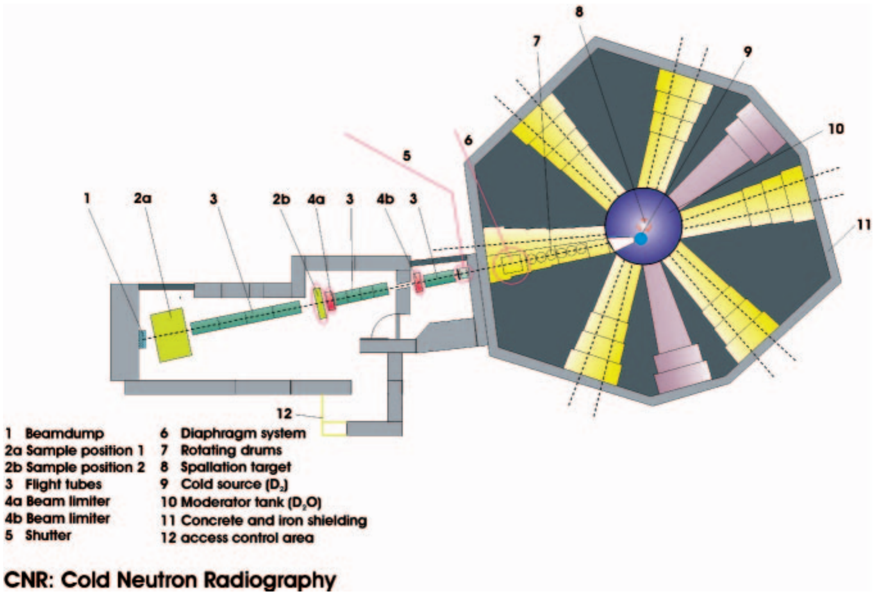

Fig. 2. – Set-up of the ICON station at the spallation neutron source SINQ, completed in 2005.

4. – Set-up of a neutron imaging device

The beam lines used for imaging purpose must be designed in a specific way. With the aim of high image sharpness, an extended field of view and equal illumination a set-up is required as shown in fig. 2 for the ICON beam line, completed in 2005. The neutrons from the source are selected at the inlet aperture and spread with low divergence into the collimator downstream to the different image positions. Certainly, the whole set-up has to be surrounded by a shielding of about 1 meter of concrete in all directions, acting also against the scattered neutron and gamma component from the sample interaction.

The key element of the imaging system is of course the detector. A couple of new options has been developed in the recent years, mostly digital ones, enabling much higher sensitivity and new options like tomography and image post-processing. The results in this report were obtained either with the CCD-system [6] or with neutron sensitive imaging plates.

There are two beam lines available at SINQ—NEUTRA for thermal neutrons and ICON (fig. 2) for cold neutrons. It has to be decided case by case, which set-up is most preferable for the individual investigation task. Thermal neutrons can provide higher penetration, whereas cold neutrons deliver higher contrast for most of the materials.

Recently, NEUTRA was completed by an additional option—the insert of an X-ray tube in front of the beam tube. Given by the small spot size of the electron focus, the collimations of both the neutron and the X-ray fields are similar. Using the same detection system for both kinds of radiation (CCD-camera in conjunction with a Gadox scintillator), a pixel-wise referencing is possible. Imaging plates are also suitable for investigations with both kinds of radiation.

Fig. 3. – Neutron images of the same object, a Jewish finger ring with menorah sign from the Augusta Raurica settlement (Basel, CH): left—transmission image, middle—outer structure obtained by neutron tomography, right—virtual slice in the tomography data set near the boundary between the two parts of the ring.

5. – Examples from national and international studies

The starting point for the use of the neutron imaging methods in cultural-heritage investigations in Switzerland was the participation in the COST action G8 [7] (entitled "Non-destructive testing of museums objects"). Since 2002, individual inspections were done for six museums sites in Switzerland with success. Examples are shown in pictures and descriptions in particular below. The results can never be only a simple service, but substantial support to new knowledge or the confirmation/rejection of existing hypotheses.

Of much higher importance has been a project about the completion of a catalogue of Roman bronze sculptures found and stored in East, South and Central Switzerland [8]. First of its kind, it will contain natural scientific results as neutron radiography, tomography and elemental analysis based on X-ray fluorescence and atom absorption spectroscopy. This project has been supported by the national COST secretariat and will be completed in 2006. About 200 different objects have to be described and attributed.

In the case of the bronze sculptures, neutron imaging methods are preferably to X-ray techniques due to the high involved lead content and the much better transmission for neutrons therefore.

5˙1. *Finger ring from Augusta Raurica with a Menorah sign.* – This small object with a diameter of only 15 mm about was spectacular because the menorah sign was the first indication of Jewish influence (and maybe settlement) in the region around the Roman Camp Augusta Raurica [9]. The museum experts in charge wanted to know how it was manufactured—from a single piece or made from two ones.

Whereas the transmission radiography (fig. 3, left) gave almost no information because of the low material amount of the object, some more information could be derived from the tomography data (fig. 3, middle). Here, the outer view of the ring can reproduce the menorah sign again. Artificial, means virtual slices with special software tools were able to look into the finger ring. As shown in the right part of fig. 3, there are clear indications that the sample was original made from two parts.

5˙2. *Block excavation in the region* Zug *(*CH*).* – During preparation work for the construction of new houses in the Zug region of Switzerland, ancient found pieces were excavated. Due to the needed hurry a very careful segmentation and preparation as in a laboratory was impossible and a so-called block-excavation was applied (the important parts were covered and sealed in plaster layers together with surrounding soil and stones).

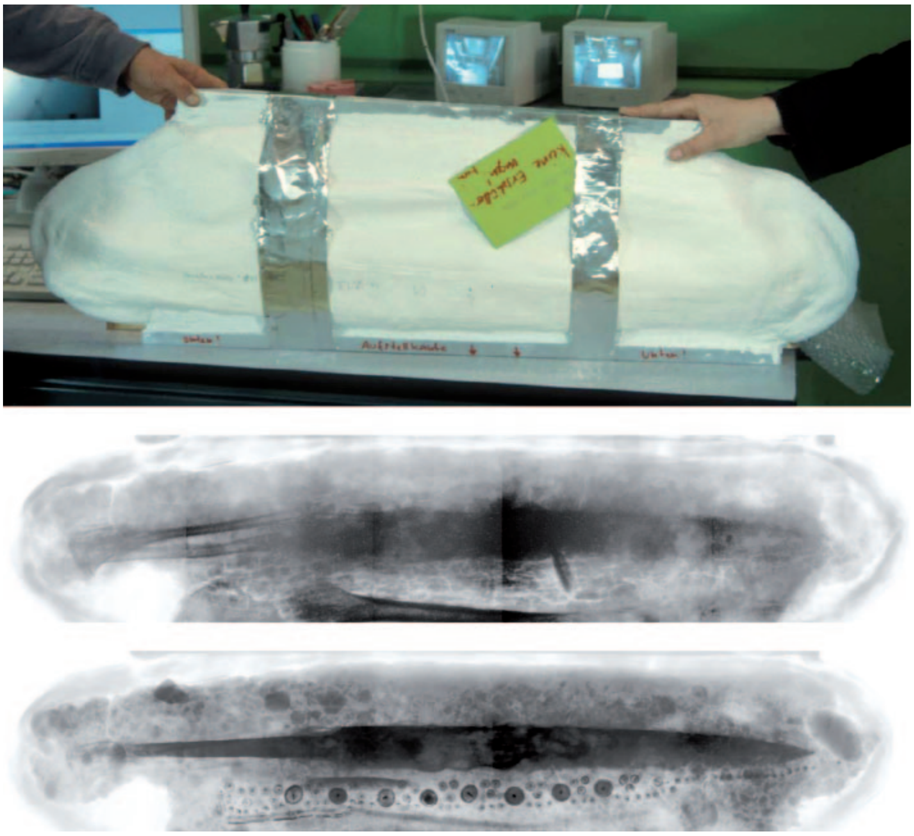

Fig. 4. – Transmission images with neutrons (middle) and 320 kV X-ray (bottom) of the bulky plaster covered antique assembly containing a sword, a knife, bones and knops at a leather band. The two imaging methods provide different information from the same sample. The image data were produced in 5 segments and placed together with post-processing tools.

After the transportation to the museum site in Zug, the problem was to decide how to continue the segmentation between the valuable materials and the soil particle in a minimal-invasive way. The knowledge about the content of the covered sample and the position of the most relevant parts has been of high importance for a professional separation and conservation procedure.

The results of the investigation with neutrons and X-rays are shown in fig. 4. In complementarity it was possible to separate metals, stones, bones and leather in advance to the preparative work.

5˙3. *Dagger from the Vindonissa Camp*. – In the case of this find in an excavation near Brugg (CH) [10], both neutron and X-ray transmission images were made. In the comparison, the two images in fig. 5 look similar. But a deeper analysis shows structural differences and some effects of the conservation work. So, the cracks in the metallic structure are obviously be filled with resin or oil and show high contrast in the neutron images. Furthermore, the corrosion effects are better visible in the neutron images. On the other hand, the remaining metal in the object gives higher contrast in the X-ray images.

Fig. 5. – A dagger from excavations near the Roman Vindonissa camp (photo left) were inspected with neutrons (middle) and X-ray (right). Similar in first order, specific differences can be found out in respect to conservation measures and corrosion status.

The safe transportation and the insurance costs can be a problem in case of objects from abroad, when they have to be transferred to the inspection site over borders. This was really the case for the famous "Sky disk from Nebra" [11], a bronze plate with 32 cm diameter, considered as one of the very first descriptions of astronomical observations. The basic idea to investigate the whole object in the neutron beam, which fits very well to its size, in order to make a non-destructive analysis of hidden structures and the general material distribution could not yet realised for this reason. Additional data and information to the already done studies [12] could be provided in this way.

Instead, a test sample was send by the owner (Landesmuseum für Vorgeschichte Sachsen-Anhalt, Halle/Saale) in order to demonstrate the promised feature of neutrons—to transmit bronze easily and to visualise organic material with even higher contrast in small concentrations. As shown in fig. 6, both the outer contours and the adhesive layer behind the gold plate were identified easily by means of neutron radiography and tomography. Therefore, we wait for another chance to perform the proposed neutron inspection.

Less valuable but also very interesting samples were analysed with neutrons and with X-ray for comparison. The pieces submitted by the Bavarian Museums Foundation were taken from a private collection and represent Roman bet buckles, produced in the special

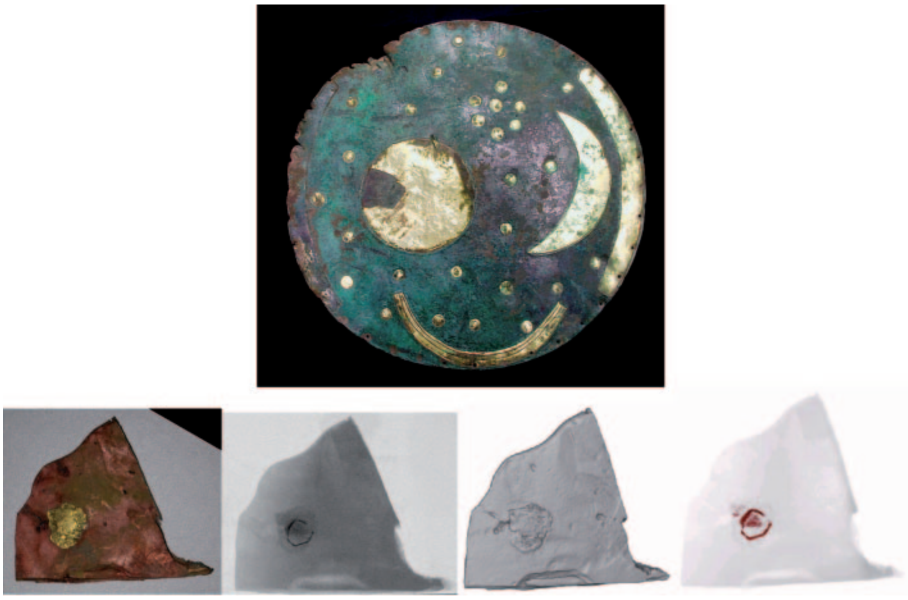

Fig. 6. – The famous "Sky disk from Nebra" (above) contains gold symbols on the bronze surface. In order to check whether organic resin or adhesives might be visualized with neutrons a copper dummy (photo right) was investigated with transmission radiography and neutron tomography. The glue behind the gold plate was easily identified in both cases.

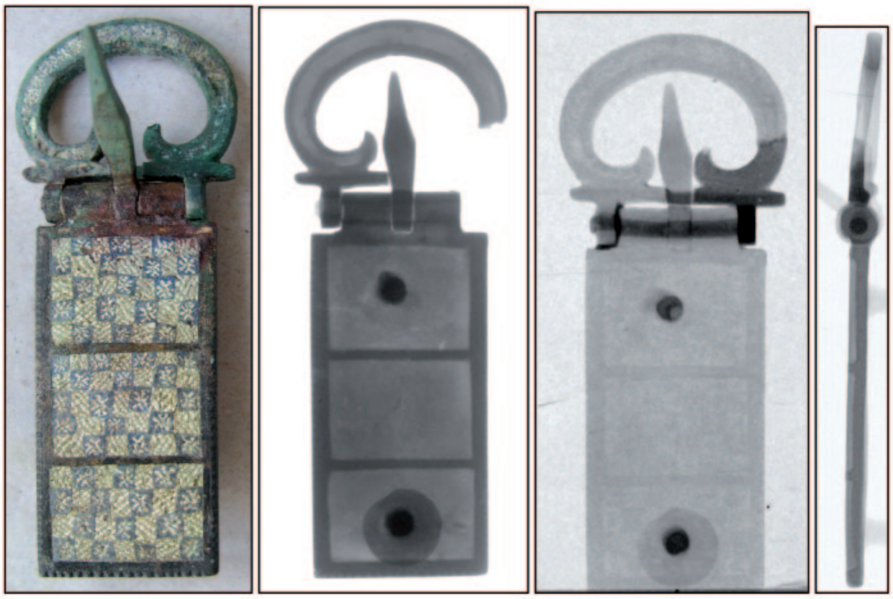

Fig. 7. – A Roman belt buckle was investigated with X-ray and neutrons: due to the high sensitivity of neutrons in respect to hydrogenous materials, the artificial completion with an epoxy substitute was identified. This part is completely missing in the X-ray image (second from left).

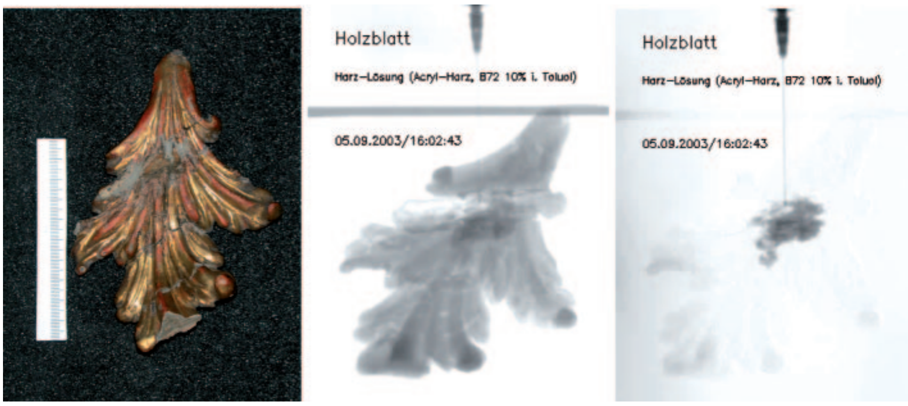

Fig. 8. – Inspection of the resin treatment of a wooden leaf where an injection takes place: left: photo of the object, middle: direct image of the arrangement, right: structure of the leaf withdrawn—the resin solution becomes directly visible.

Millefiori technique. As demonstrated in fig. 7, the comparison between the neutron and the X-ray images identified a clear restorative work—the completion of the outer ring with an organic structure (epoxy resin). On the other hand, the layer of the ceramic ornaments can directly measured in the neutron image, when a projection is chosen perpendicular to the flat surface. This would be impossible with X-ray due to the limited transmission in this direction.

6. – Contributions to conservation work

One of the most common methods to stabilize and to protect historical objects (wooden ones, stones, ceramics and even metals) is the treatment and injection with resins. It is often hard to verify how much resin is applied and where it remains at the specific place in the object. This is caused by the complicated structure of the matrix and by the evaporation of the solvents of the resins in different way.

Neutron imaging methods were found to be a very suitable tool for the inspection and verification of the resin treatment, also in time-dependent sequences [13]. As demonstrated in the example shown in fig. 8, resin can be visualised, followed and quantified with neutron imaging methods. Because the exposure time per image is 10 s only, time sequences can be obtained for minutes, hours or even day. Of high importance is a image processing step, where the matrix of the material under restoration is withdrawn from the resulting data set and the net resin amount results in the end.

7. – Future improvements in neutron imaging

The demands for non-destructive inspection of museums objects are very different. Neutron methods in high quality are never in-house ones for museums due to the need for strong source, located in big institutions. Samples and objects have to be transported to the neutron source in every case.

The imaging methods as the focus of this paper are in continuous improvement, where the museum partners can take profit. Some years ago, the step from film to digital data sets was performed. This enabled the start in neutron tomography [14] and time-depending investigations.

Further development in the improvement of spatial resolution and higher inspection speed will take place. More advanced methods as phase contrast enhancement and energy selective neutron imaging will be available on routine base at some sites in the next few years. The shift of the neutron spectrum towards epithermal and fast energies will enable higher transmission and the inspection of thicker material layers. The direct comparison and the pixel-wise referencing with X-ray data will be another and promising approach [15].

8. – Problems and experiences

People at the museums are not ever interested in collaboration with natural scientists *ab initio*. The first contact has to be initialized on more or less private basis. In first trials the applicability of the methods has to be demonstrated.

The experimental facilities for neutrons are big stationary installations—impossible to bring to the museums site. Therefore, the objects for investigations have to be transported to these neutron facilities in the institutions. Insurance and custom can play important role then, especially when the samples have to move over borders.

The residual radioactivity from the neutron exposure can delay the return of samples by hours or days. However it has been no real problem indeed in all cases described above.

The examples of investigations shown in the paper can demonstrate that powerful tools for interesting studies with neutrons exist.

The potential is by far not been exploited, new options have to be discussed between the experts from both sides.

If a scientific background for studies exists, the investigations might be performed with only low costs for the museum in form of a "scientific proposal". The proof of authenticity and the certification with a commercial interest have to be handled differently.

9. – Outlook for further options in using research infrastructure

It might be of high importance to create networks to serve for investigations of cultural heritage on higher level, when other European research centers than PSI operating strong neutron facilities becomes involved. In this way, the specific advantages of the facilities can be exploited and the dialog with museums experts can be intensified.

Because such kinds of networks already exist (*e.g.*, NMI3 for neutrons and myons), it is a challenge to find the best way for communication with people from the museum side.

* * *

The authors want to express their thanks to M. Jaggi and M. Estermann for their contributions to analytical work in many tomography tasks and to K. Hunger for their work on the Roman bronzes, mentioned in the paper. Furthermore, the good collaboration with M. Wöhrle and P. Wyer (Swiss National Museum) and E. Deschler-Erb (Uni Zurich) is appreciated herewith. We thank L. Berger (Basel), C. Flügel (Munich, D) and K. Müller (Zug) for the submission of valuable objects and interesting discussion.

REFERENCES

[1] Hassanein R., Lehmann E. and Vontobel P., *Nucl. Instrum. Methods Phys. Res. A*, **542** (2005) 353.
[2] Bauer G., *Nucl. Instrum. Methods Phys. Res. B*, **139** (1998) 65.
[3] Lehmann E. H., *Facilities for neutron radiography in Europe: Performance, applications and future use*, in *Proceedings of the 15th World Conference on Non-destructive Testing, Rome, October 2000.*
[4] Fischer C.-O. *et al.*, *Autoradiography of large scale paintings—Titian, Rembrandt, La Tour*, in *Proceedings of the 6th Word Conference on Neutron Radiography, Osaka, 1999*, pp. 563-570.
[5] Kockelmann W., Chapon L. C. and Radelli P. G., *Physica B*, **385-386** (2006).
[6] Pleinert H., Lehmann E. and Körner S., *Nucl. Instrum. Methods Phys. Res. A*, **399** (1997) 382.
[7] `http://srs.dl.ac.uk/arch/cost-g8/`.
[8] Deschler-Erb E., Lehmann E. H. and Pernet L. *et al.*, *Archaeometry*, **46** (2004) 647.
[9] Berger L., *The Kaiseraugst Menorah Ring, Forschung in Augst*, Vol. **36**, August 2005.
[10] Hagendorn A., *Zur Frühzeit von Vindonnissa, Pro Vindonissa*, Vol. **XVIII**.
[11] `http://www.archlsa.de/sterne/`.
[12] Mommsen H., *Physikalische Methoden in der archäologischen Forschung, Physik-Journal*, Dezember 2005, p. 41.
[13] Lehmann E., Vontobel P., Hartmann S. and Wyer P., *Neutronenradiographie-untersuchungen zum Test des Eindringverhaltens von Harzen bei Massnahmen zur Holzfestigkeitserhöhung*, *Restauro*, **3** (2005) 194; 111. Jg.
[14] Vontobel P., Lehmann E. and Frei G., *Performance characteristics of the tomography setup at the PSI NEUTRA thermal neutron radiography facility*, in *Proceedings of Computed Tomography and Image Processing for Industrial Radiology, June 23-25, 2003, Berlin, Germany.*
[15] Lehmann E. H. *et al.*, *The approach of X-ray enhanced neutron imaging and its realisation at the NEUTRA facility*, PSI-Annual Report (2004), Vol. **III**, p. 218.