BURKHARD SCHILLINGER

WHY USE NEUTRONS?

Sufficient intensity of free thermal neutrons can be generated either at a research reactor or a spallation source – mobile generators usually do not deliver sufficient flux for high-resolution measurements. While X-rays interact with the electron shell of the atoms, and thus are more attenuated the more electrons an atom possesses, the uncharged neutrons interact with the nuclei themselves, either by absorption or by scattering. For absorption, the probability depends on the inner structure of the nuclei and the number of nucleons, both protons and neutrons in the nucleus.

The periodic system of the elements depends on the number of electrons of each element and provides classes of chemical reactivity, where noble gases are mostly inert, and alkali metals most reactive. Similar, but more complicated models exist for nucleons, and the probability for specific isotopes to absorb a neutron. Since these depend on the numbers of both the protons and neutrons, it often means that even isotopes of the same element show very different absorption cross sections and thus different contrast. Huge differences exist for neighboring elements in the periodic system. Absorption of neutrons is usually accompanied by the emission of prompt or delayed gamma radiation, which is the effect used in auto radiography and prompt gamma activation analysis (see below).

Figure 1 shows the attenuation coefficient (consisting of both scattering and absorption) for X-rays, high energy gammas, fast and thermal neutrons for the elements of the periodic system as a function of the atomic number (Bücherl/Lierse von Gostomski 2004). While the attenuation of X-rays rises monotonously with the number of protons of the elements, attenuation decreases for fast neutrons. Since do mainly single-body scattering, the attenuation is most prominent for hydrogen that has the same mass as a neutron. For thermal neutrons, the behavior is at first sight totally unpredictable even for neighboring elements. There is also strong attenuation by scattering on hydrogen, and some other light elements, while most metals, especially aluminum, iron and lead, are easily penetrated. Coherent scattering occurs on crystalline structures and causes attenuation as well. The main strength of thermal neutron radiography lies in the ability to detect hydrocarbons, i. e. organic materials, in combination

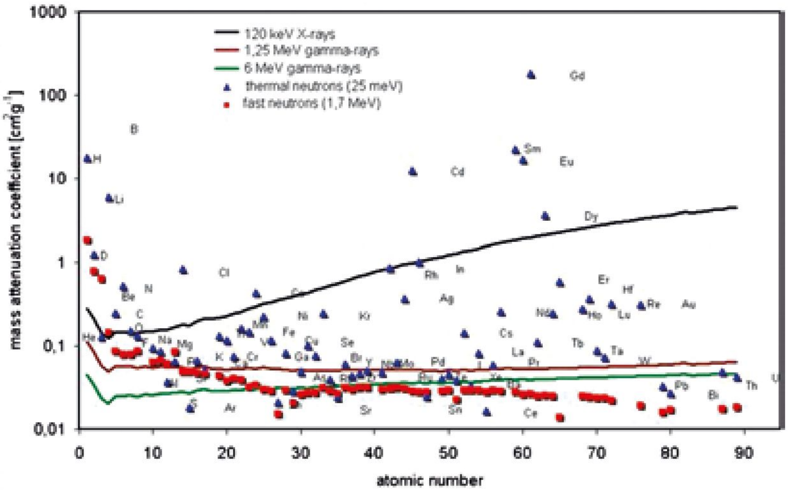

Fig. 1 Attenuation coefficient for X-rays, fast and thermal neutrons for different elements. – (TU München – FRM II).

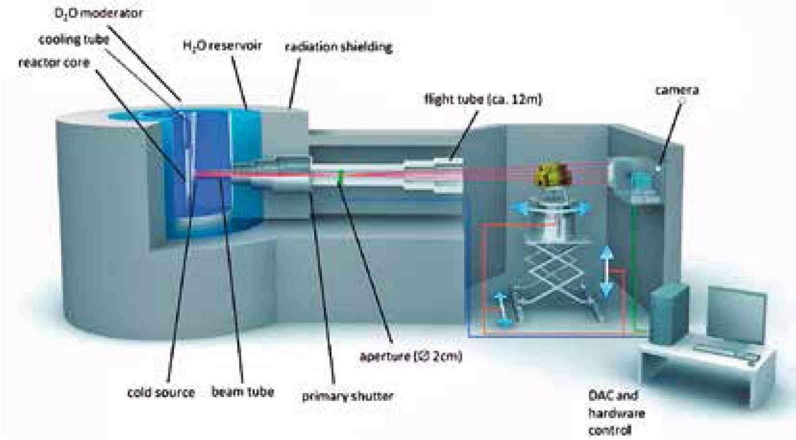

Fig. 2 Schematic setup for neutron radiography. – (TU München – FRM II).

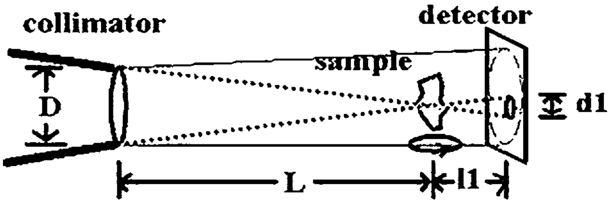

Fig. 3 Beam geometry in neutron radiography. – (TU München – FRM II).

with metal casings, like textiles on sword handles and scabbards, or bone fragments as relics enclosed in metal containers.

Neutron imaging: radiography and tomography with thermal neutrons

Neutron Radiography produces a shadow image of the sample, just like X-ray radiography. But while X-ray radiography mostly uses an X-ray tube with a tiny focus and a resulting cone beam that magnifies the projection of the sample, neutron radiography uses a pinhole-camera setup that results in an approximated parallel beam without magnification. **Figure 2** shows the principal setup for neutron radiography.

The neutron beam from the reactor or spallation source enters a convergent collimator up to its smallest aperture with diameter D, then diverges again on its way of length L to the sample. **Figure 3** shows an exaggerated view of the beam geometry (Schillinger 2006).

Since the effective source is not a point source, but has the diameter D of the aperture, every point of the sample is blurred to a little disk of diameter d1 on the detector in distance l1. The image quality thus depends on the collimation ratio L/D and on the sample to detector distance. (The same is, in principle, true for imaging with X-ray tubes, but the inherent magnification of the cone beam mostly wipes out the effect of the finite size of the focal spot on the X-ray tube.)

Early reactor installations for use with film had a typical collimation ratio of L/D = 50, which allowed only for blurred images of large samples. Modern installations have collimation ratios from 300 to several thousand, as new sensitive electronic detectors allow to sacrifice intensity for higher quality. Typical fields of view can be adapted from about (5 cm)^2 to (35 cm)^2.

Figure 4 shows a typical neutron radiography of a bronze statue provided by Archäologische Staatssammlung München (FRM II 2009a-b).

Computed tomography can be performed by rotating the sample and recording several hundred different angular views, which can be reconstructed into a three-dimensional distribution of attenuation coefficients. **Figure 5** shows a neutron computed tomography of a small textile fragment of about 4 mm size where the sensitivity of neutrons for

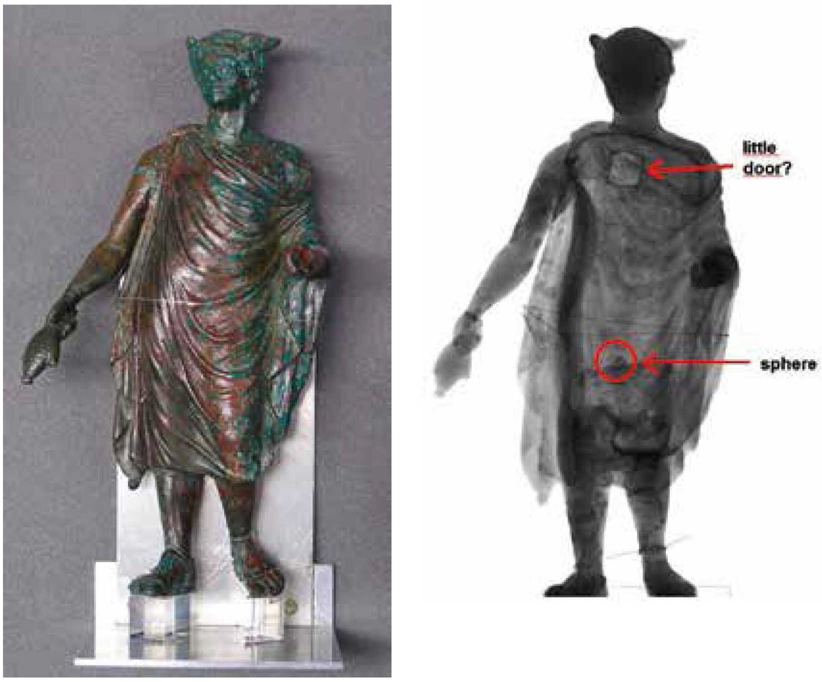

Fig. 4 Neutron radiography of a little bronze statue. The radiography shows a little door that was apparently used to remove the casting core, and a little sphere, possibly clay, left within. – (TU München – FRM II).

organic material was used to study details of the weaving technique. For X-rays, the fragment is transparent.

Limits in resolution

For a parallel beam without magnification, the limit for the resolution is the detector resolution in both scintillation screen and camera. Since neutrons are uncharged particles, they can only be detected by a nuclear reaction, like the absorption in lithium-6, which produces an alpha particle and a tritium particle with a lot of kinetic energy. These will produce visible light within a zinc sulfide scintillation screen, which in turn is recorded by a scientific CCD camera. Stopping the reaction products within the screen takes several thousand collisions, which produces an extended light spot in the order of 80 µm size for a screen with 100 µm thickness. The same is true for gadolinium-based scintillation screens. With thinned screens (5-20 µm), resolutions in the order of 10-20 µm are possible.

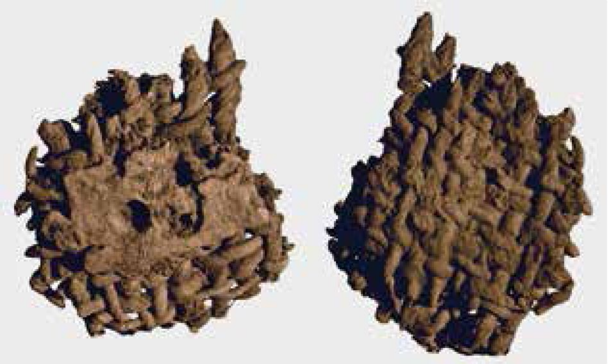

Fig. 5 3D Images of textile fragments from the Lauchheim excavations in Southern Germany (Stelzner et al. 2010) allow for the identification of production technique and the resulting textile characteristics. The images show a double layered well-used ribbed body of average quality. Thickness and torsion of threads could be measured as well as textile densities. – (TU München – FRM II).

Fast neutron radiography and tomography

Thermal neutrons (5-30 m-eV, this corresponds to the energy of the thermal motion of water molecules) can penetrate about 1 cm of water, 4-5 cm of

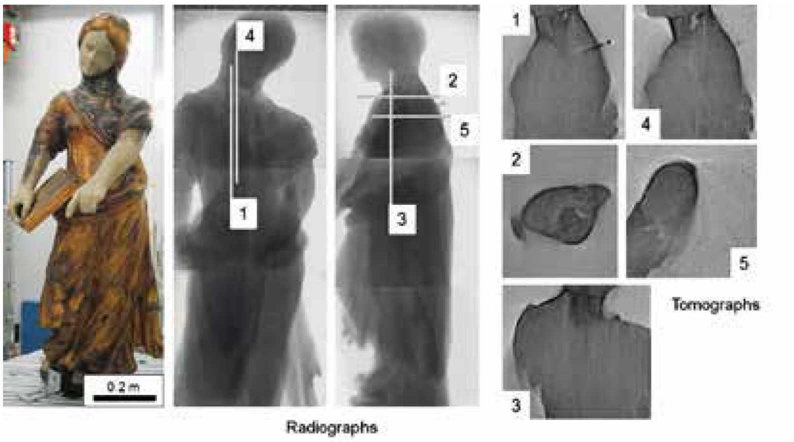

Fig. 6 Cracks and layers of glue in a wooden sculpture from an epitaph in the St. Laurentius Church in Tönning (Holstein/ Northern Germany). – (BAM and TU München – FRM II).

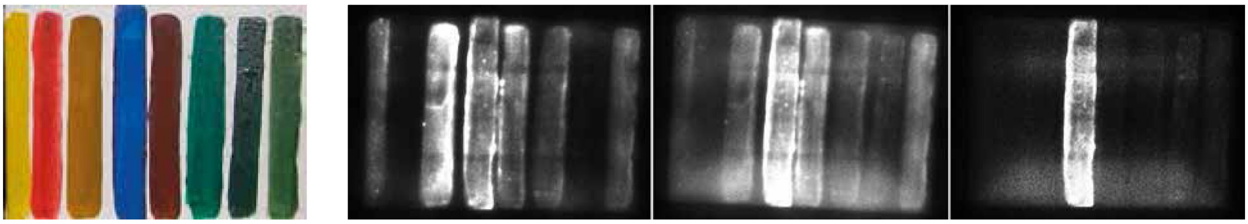

Fig. 7 Photo of paint stripes and auto radiographs after 20 min (2.5 h exposure), after 3 h (22 h exposure) and after 25 h (20 h exposure). – (TU München – FRM II).

iron, and 20 cm of aluminum. For larger samples, the FRM II reactor of Technische Universität München possesses a unique converter facility for fast fission neutrons, where 1-2 Mega-eV neutrons can be extracted for radiography without moderation. Fast fission neutrons penetrate up to 10 cm of water and 10 cm of iron, or wooden samples up to 50 cm diameter while being sensitive to hydrocarbons, i. e. glue or oils. Due to the high penetration for most materials, the achievable detector resolution lies between 0.5 and 1 mm. **Figure 6** shows a fast neutron CT of a wooden statue, where layers of glue are revealed within the wood (Osterloh et al., this volume).

Autoradiography

Most materials that can absorb neutrons reach an excited state in their nuclei which decays by emission of prompt or delayed gamma radiation. Hence, nearly all irradiated materials become radioactive for a certain time, which can be anything from milliseconds to days or in rare cases, even months. The emitted gamma radiation consists of several gamma quanta of different energy, which are characteristic for a specific nucleus. This can either be used to sample material (see next paragraph), or to e. g. extract in-depth information about the pigments used in paintings, even in hidden deep layers.

For this so-called Autoradiography, a painting is irradiated with thermal neutrons for several hours, so part of the pigments become radioactive. The painting is then removed from the beam and covered by X-ray film or storage plates. The first set of film is removed after two hours, catching the short-lived activations and most intense gamma radiation, then the process is repeated for e. g. twenty hours and another twenty hours to catch longer-living activation. The resulting images are caused by different pigments contained in the different paints from all layers of the painting, often revealing hidden over-

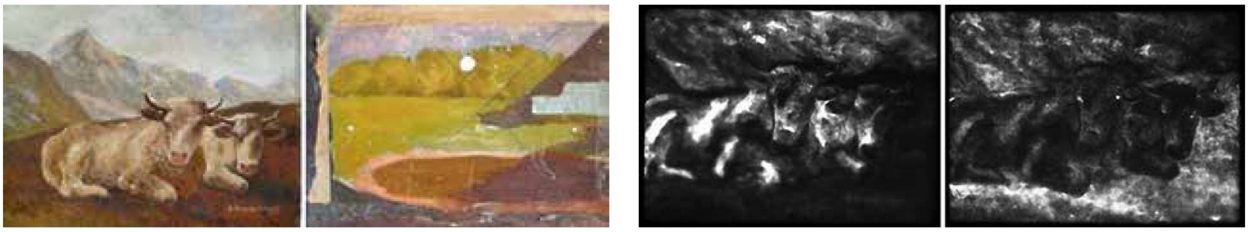

Fig. 8 A cheap double-sided painting front and mirrored back and its auto radiographies after 20 min (2.5 h exposure) and after 40 h (50 h exposure). – (TU München – FRM II).

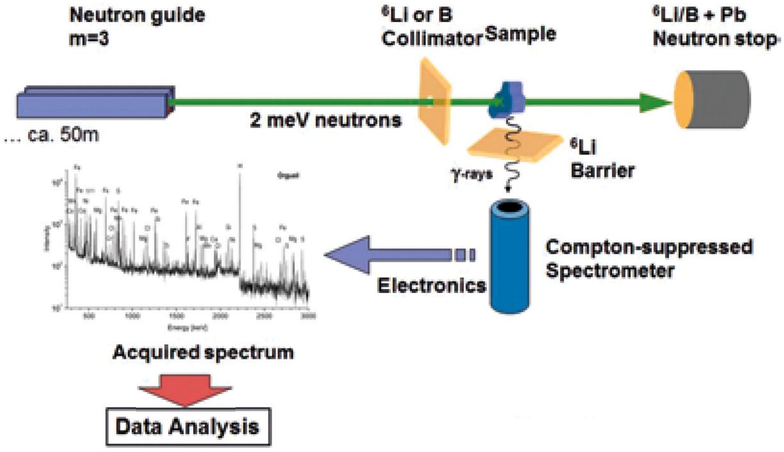

Fig. 9 Principle setup of a PGAA experiment. A collimated fine neutron beam hits the sample. The generated gamma radiation is detected in 90 degrees to the neutron beam. Stray neutrons are filtered out by a light-element neutron filter. The acquired spectrum gives information about the bulk composition of the sample. Spatial information can be obtained by moving the larger samples across the pencil neutron beam. – (MLZ/TU München – FRM II).

painted details beneath the surface of a painting. Contrary to neutron autoradiography, X-ray fluorescence gives information only about the surface of paintings and cannot reveal hidden deep layers. **Figure 7** shows a photo and three auto radiographs of different paints (Scheungraber 2008). **Figure 8** shows a cheap double-sided painting front and mirrored back and its auto radiographies after 20 min (2.5 h exposure) and after 40 h (50 h exposure) (Scheungraber 2008).

Prompt gamma activation analysis (PGAA)

PGAA is a nuclear analytical technique for determination of elemental/chemical composition of samples. Samples are irradiated by thermal or cold neutrons, and the generated gamma radiation is deecected and analyzed to identify the chemical, elemental, even isotopic composition of the sample. PGAA can detect amounts expressed in parts per million or even less; it can be employed on samples of less than 1 mg, but can also be used on large bulky samples. PGAA is sensitive to many light elements like H, B, C, N, F, S, and delivers information from the depth of the bulk, not only surface information. **Figure 9** shows the principle setup of a PGAA experiment (Kudejova 2015).

Neutron scattering

As sub-atomic particles, neutrons also have a wavelength and can produce interference and scattering effects on crystal lattices, just like X-rays. The possible methods and instruments (powder diffractometer, single crystal diffractometer and engineering diffractometer) are too many to be treated in detail here. They all use coherent scattering on crystal lat-

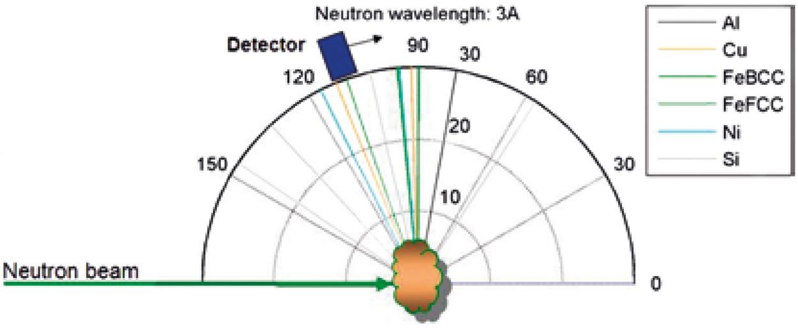

Fig. 10 A collimated monochromatic neutron beam that hits a crystalline sample produces several reflexes under different angles that are characteristic for the crystal structure. – (After Peetermans/Van Swygenhoven/Lehmann 2015).

tices and the reflexes emitted under different angles (**fig. 10**) to determine the crystalline structure of the samples. Compared to X-rays, neutrons can penetrate deeper into materials, especially metals, and employ a longer wavelength range (1-20 Ångström, 10-200 nm) than X-rays. Neutron scattering is treated in much detail at most neutron conferences, but it goes beyond the scope of this special publication.

Activation of samples

Most samples become activated by neutron irradiation and must stay for some days, rarely a few weeks at the neutron facility before their activation has decayed and they can be released to public use again. Undue activation can be avoided by preliminary short-time irradiation and a consecutive gamma scan to predict activation. Neutron methods are not detrimental to a later ^{14}C scan of organic material, because the isotope ^{14}C is not generated by neutron irradiation.

Access to neutron sources

Most major neutron sources (e. g. Helmholtz Zentrum Berlin, Heinz Maier-Leibnitz Zentrum at Technische Universität München, Paul Scherrer Institut Switzerland, ISIS in UK, ILL Grenoble, KFKI Budapest) run user programs where scientific proposals for experiments and beam time can be submitted about twice a year. Proposals are evaluated only by scientific quality, granted beam time is free, provided that the results are published. The European Union even provides financial support for researchers for travel and accommodation. The required radiation protection surveillance is usually done by the home institution of researchers. Local staff at the neutron sources assists in conducting experiments. Overall, access to neutron experiments is rather straightforward and simple for public research.

Conclusion

X-ray methods are always first choice because of their easy availability. Neutrons come in where X-rays reach their limitation in penetration depth, penetration of metals and sensitivity for light elements. Although powerful neutron methods are available at only a few centers in Europe, access is rather straightforward, and can be easily established.

Even though the use of neutron methods has seen a steep rise in the past years, their potential has hardly been tapped, and in many fields, they are still waiting to be employed for the first time and to provide unique new information. Whenever X-rays fail, it is worth asking: »Can’t we do it with neutrons?«

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Summary / Zusammenfassung

Why Use Neutrons?

Thermal and fast neutrons can penetrate most metals while delivering high contrast for many light elements, which is often the opposite behavior of X-rays. Hydrogen, i. e. organic materials, can be detected even in lead enclosures. Neighboring elements, even isotopes of the same element, may deliver very different contrast. Neutron activation analysis can deliver in-depth information about material composition, auto radiography can even reveal hidden layers in paintings. X-ray methods are always first choice for easy application and availability, but neutron methods are great tools where X-rays fail, not only in cultural heritage. This paper provides an overview about neutron methods, which are treated in further detail in the other publications in this issue.

Warum Neutronen verwenden?

Thermische und schnelle Neutronen können die meisten Metalle durchdringen, während sie hohen Kontrast für viele leichte Elemente erzeugen, was oft das Gegenteil des Verhaltens von Röntgenstrahlen ist. Wasserstoff und damit organische Materialien lassen sich selbst in Bleibehältern sichtbar machen. Benachbarte Elemente im Periodensystem und selbst Isotope desselben Elements können sehr verschiedene Kontraste hervorrufen. Die Neutronenaktivierungsanalyse kann Tiefeninformation über Materialzusammensetzungen liefern und Autoradiographie gelingt es, selbst verborgene, übermalte Schichten in Gemälden sichtbar zu machen. Röntgenmethoden sind aufgrund ihrer einfachen Anwendung und Verfügbarkeit stets erste Wahl, doch Neutronenmethoden liefern großartige Werkzeuge, wenn die Röntgenmethoden versagen, und das nicht nur bei Anwendungen aus »Cultural Heritage«, also Kunst und Archäologie. Dieser Beitrag gibt eine Übersicht über Neutronenmethoden, die in den weiteren Beiträgen dieses Hefts im Detail erklärt werden.

Keywords

neutron imaging / neutron radiography / neutron computed tomography / X-ray computed tomography / thermal neutrons / cold neutrons / fast neutrons / auto radiography / prompt gamma activation analysis / neutron scattering / neutron activation / neutron sources