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THE ANCIENT CHARM PROJECT: NEW NEUTRON BASED IMAGING METHODS FOR CULTURAL HERITAGE STUDIES

1. The project

The Ancient Charm (Analysis by Neutron resonant Capture Imaging and other Emerging Neutron Techniques: new Cultural Heritage and Archaeological Research Methods) Project (Gorini 2007) was a European research programme, financed by the European Community in the frame of the "New and Emerging Science and Technology" initiative. Ten institutes from the fields of physics, archaeology and restoration, located in Italy, Hungary, Germany, the Netherlands, Belgium and the United Kingdom were involved in the project. It was started in 2006 with the aim to develop new 3D, non-destructive, neutron-based imaging techniques for the analysis of cultural heritage objects.

Most of the methods employed to characterize the large variety of chemical, physical and microstructural properties of objects are invasive or have limited penetration depth in matter (such as X-rays and charged particles). However, as chargeless particles, neutrons can deeply penetrate objects and non-destructively provide information about the inner structure and composition of materials (Chadwick 1932). In matter they undergo nuclear reactions, which can be categorized into two main groups: absorption and scattering. The probability of the various reactions and the depth of the penetration are highly dependent on the energy of the neutrons and on the material along the path of the beam. As neutrons interact with the atomic nucleus, where the interaction probability varies even for neighbouring nuclei, they provide bigger contrast for neighbouring elements in the periodic table of elements than X-rays. The use of neutrons permits e.g. inspection of hydrogenous materials, which can be transparent for X-rays. On the other hand some heavy elements, such as lead, are almost transparent to neutrons.

While the potential of neutron-based techniques is large, their development is recent in most cases. Especially, few attempts have been made to use neutrons for quantitative 3D imaging. The motivation of the project is to benefit from the great potential of neutron-based analytical methods, because they:

– are non-destructive,
– can be element-sensitive and structure sensitive,
– can have a good spatial resolution, and
– are able to investigate bulky objects, that are too thick for e.g. X-ray analysis.

The obtained results are intended to answer questions of historical interest, like e.g. the state of craftsmanship during a selected epoch or the origin of the investigated objects as well as aspects of restoration and conservation procedures.

2. THE COLLECTION OF 3D DATA

Two fundamental ways to measure and reconstruct the 3D spatial distribution of sample properties will be discussed here. The first approach is called the direct method that uses a collimated irradiation and detection to localize the source of information, while the other applies mathematical algorithms to determine the spatial distribution from the measured integral quantities.

2.1 *The direct method*

This is the most natural transition from a bulk analytical technique to a 3D-capable method. It is preferred if the detector(s) used do not provide intrinsic position information. An existing instrument can provide spatially-resolved data if the size of the irradiating neutron beam and also the solid angle of the gamma-detection is being reduced. Such collimated neutron beam is often called "pencil beam", and in case of thermal neutrons, its dimension can be reduced down to 2-3 mm. The gamma-ray collimator is usually made to similar size.

Although the beam irradiates the sample along a chord, the origin of the useful analytical signal is only the intersection of the beam path and the detection angle through the gamma-collimator. This is called double-collimated geometry (Fig. 1, a). The achievable size of the active volume is usually limited by the intensity of the beams and the availability of beam time. The data required to fill up the 3D grid is measured one by one, applying a systematic scanning of the whole sample, which is placed on a moving table, or at least of a region of interest.

The data reduction in this case is straightforward: the measured raw data are plotted against the coordinates of the moving table, i.e. the positions in the 3D space.

However, this raw image is affected by biases from self-absorption and self-shielding, and the distortion effects of the gamma-ray collimator transmission function, which have to be point-wise corrected for. A correction method is under development using a large amount of random numbers (Monte Carlo method) to yield quantitative results.

If 2D results are enough for meaningful answers, the gamma collimation may be omitted in favour of a higher event rate (Fig. 1, b). This configuration is called the chord geometry. The main advantage of the direct method over the

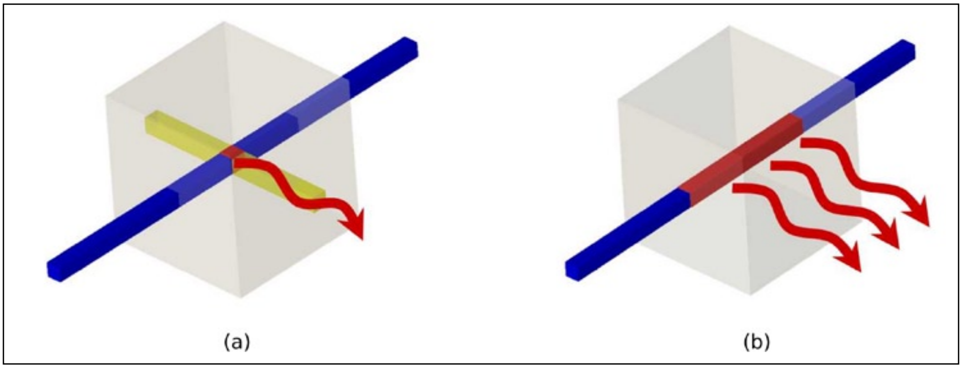

Fig. 1 – Visualisation of an active measurement volume for the scanning based approach. (a) 3D setup with gamma collimator (double collimated geometry). (b) 2D setup without gamma collimator (chord geometry). The blue line represents the path of the neutron beam. The yellow line shows the collimated view of the gamma detector. The intersection of the two lines gives the currently measured sample volume for (a) while for (b) the whole volume irradiated by the neutron beam is measured.

second type of detection technique, i.e. the tomographic reconstruction, is that one does not need to inspect the whole sample. For real samples it is usually safe to assume that they are made up from a few uniform parts, and useful results can be made even if the full scanning with 2 mm resolution is infeasible.

2.2 *The tomographic reconstruction*

The roots of this theory go back to the 1920's, when the mathematical background was worked out. Several decades passed until the electronic detectors and the increasing computing power made the routine application of this method practical in medicine and also in material science.

This is best applicable if there are pixelated detectors, which provide inherently a dataset with some resolution, determined by the size of the pixels and other physical processes. In this reconstruction method the sample must be fully illuminated and integral information along the beam path is recorded with a detector. Such a measurement is repeated at several angles, and mathematical calculations lead to the 3D results. It is theoretically equivalent if the sample or the detector is rotated.

In tomography, the goal is to determine a measure of the interaction probability between the material and the neutron as a function of spatial coordinates (x, y, z) (Kak, Slaney 1999). This quantity can deliver structural or even elemental information about the interior of the sample, depending on the chosen method.

An arbitrary object $O(x,y,z)$ is considered to comprise n "slices", of equal thickness dz, that all lie in planes parallel to the (x, y) plane (i.e. horizontal) and thus perpendicular to the vertical *z*-axis. Our goal is to obtain *f(x,y)*, the quantity of interest in a horizontal plane. These planes are handled separately from each other. By stacking such slices along the *z*-axis we finally get to the data in 3D space. In 1917, Radon gave a formula to express a detected signal related to integral quantities. The probability of interaction along a straight line in a plane *p(t)*, is given by the following formula:

$$p(t) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \delta(x\cos\theta + y\sin\theta - t)\mu(x,y)dxdy$$

where δ is the Dirac delta function, θ is the angle between an axis fixed to the sample (e.g. *x*-axis) and the *t*-axis (which is perpendicular to the direction of the beam), and *t* is the perpendicular offset of a line from the origin, along which one of the line integrals through *f(x,y)* at different offsets is taken. The Dirac delta function is zero everywhere except its argument is zero, which is true along the line $t = x\ cos\theta + y\ sin\theta$. Corresponding to the different value of *t*, many line integrals through *f(x,y)* can be done at a fixed θ, which constitutes a projection of *f(x,y)* onto a line at the angle θ. Each *t* detector element is hit by a number of neutrons, depending on what fraction of neutrons was absorbed during the passage through the sample. Such integral quantities detected in each detector element are called raysums.

The next step of the process involves the so-called Fourier-transformation. This is a mathematical construction to relate two complex functions defined on inverse quantities, such as time-frequency, or wavelength-lattice parameters. Data can be equally well represented in the real-word coordinates and in an abstract Fourier space. Although the conventional presentation of the information is easier to comprehend, the information content of the two representations is exactly the same and can be converted back and forth at any time.

The core idea in the reconstruction of $f(x,y)$ is the so-called Fourier slice theorem that makes a connection between the quantity of interest and its representation in the Fourier space. It can be worded as follows: the one dimensional Fourier transform $P_\theta(\omega)$ of a parallel projection of a distribution $f(x,y)$, taken under an angle θ, is equivalent to a single line within the 2D Fourier transform of the distribution $f(x,y)$ that encloses the angle θ with the *u*-axis (Fig. 2).

This means that if we obtain raysums (projections) at multiple angles, it is possible to fill up adequately the Fourier space with data. If these are then transformed back to the real-word, the spatial distribution of the quantity of interest is obtained. It can be proven that the minimum number of projections needed is $\frac{\pi}{2}N$, where *N* is the number of pixels along the t axis.

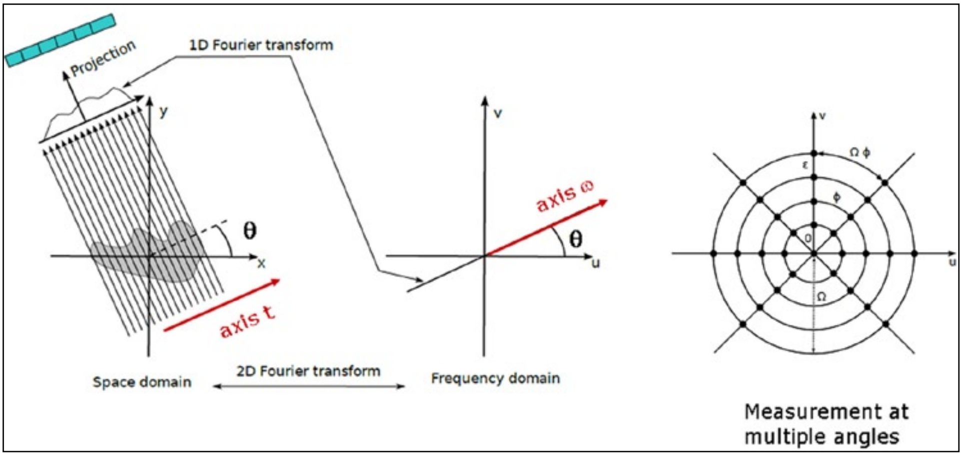

Fig. 2 – Reconstruction process with the Fourier-slice theorem. The parallel projection taken at an angle delivers via 1D Fourier transform a slice in the frequency domain. The combination of multiple Fourier transformed projections at different angles fills up the frequency domain. The 2D Fourier transform back in space domain yields the wanted 2D distribution.

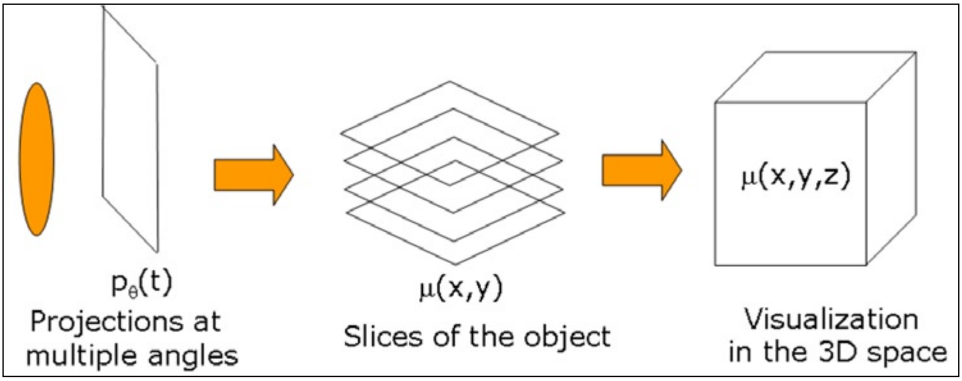

Fig. 3 – The filtered backprojection algorithm yields slices of the measured object. These slices are finally stacked with the help of a visualisation software, to deliver the 3D image of the object.

The practical way to the back-transformation of the information is the "Filtered Backprojection algorithm" (Kak, Slaney 1999). This uses a so-called ramp filter in the frequency domain to eliminate high-frequency noise, and thus outperforms the competing methods.

The advantage of the tomographic reconstruction is that the raysums can be detected with a pixel detector independently of each other, thus there is no need to collimate the beam. This substantially speeds up the experiment.

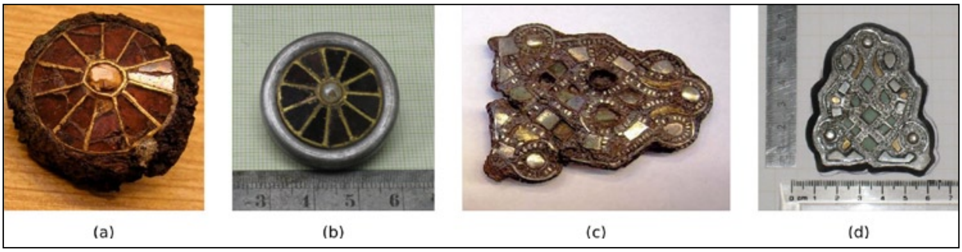

Fig. 4 – Some of the samples investigated during the Ancient Charm Project. (a) Fibula found at the Kölked-Feketekapu site, Hungary. Dated to the end of 6th century. (b) Replica of the fibula. (c) Belt mount found in Környe, Hungary. Dated to the middle of 7th century. (d) Belt replica (original objects provided by the Hungarian National Museum; replicas provided by the Mineralogisch-Petrologisches Institut, Universität Bonn).

One important issue to note is that completely absorbing parts of a sample will break the reconstruction of the slices covered by these parts, because the backprojection algorithm fails for these occasions (Fig. 3).

3. OBJECTS

Archaeological objects have to be handled very carefully by trained personnel. This is normally achieved by having a curator at the experimental site who takes care of the proper treatment of the objects. During the development, test, and fine-tuning stages of a new method, which take a long time, this is not a practical approach. For this reason replicas of interesting archaeological objects were created, which were designed to match the original objects as close as possible in shape and material and the first tests were performed on these replicas.

The size of the selected objects is in the order of a few centimetres (Fig. 4), which should be a typical size for many interesting archaeological objects and which makes the handling during transportation and experiment feasible. The selected objects have adornments of different sizes in the millimetre range to investigate what sizes can be resolved by the different methods.

4. METHODS

The methods involved utilize neutrons of different energy ranges to provide various kinds of information for the study of cultural heritage objects. While some of them are based on corresponding established methods for bulk analysis, others, as e.g. Neutron Resonance Transmission (NRT), are newly developed. The combination of existing, enhanced, and new methods should give the most fruitful results for archaeological studies. The aim of the Ancient Charm project was to combine and enhance Neutron Tomography

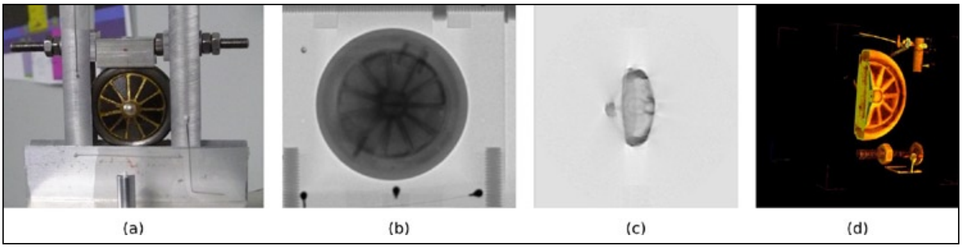

Fig. 5 – Steps in the tomography process. (a) Fibula replica in sample holder used for the measurements. (b) Radiography of the fibula replica at 0°. (c) Reconstructed slice obtained with the filtered backprojection algorithm. (d) Visualisation of the complete tomography.

(NT), Prompt Gamma Activation Analysis (PGAA), Time-of-flight Neutron Diffraction (TOF-ND), Neutron Resonance Capture Analysis (NRCA) and Neutron Resonance Transmission (NRT) in order to generate 3D images of the elemental and phase compositions of complex museum objects.

4.1 *Neutron Tomography (NT)*

Neutron Tomography (NT) is a well established method for the investigation of inner morphological structures of objects (SCHILLINGER *et al.* 1999). The technique is based on the measurement of the attenuation of a neutron beam passing through an object.

The object is placed in a neutron beam in front of a neutron sensitive screen on a turntable. Radiographies (a radiography could be considered as a set of parallel line projections) of the object are taken at different rotational angles. These radiographies, showing the 2D integral neutron absorption for one direction of illumination, are the basis for the reconstruction of the 3D image. The result shows the neutron absorption properties of the objects, i.e. the morphology, but no elemental information is obtained (Fig. 5).

For the Ancient Charm project NT with cold neutrons was utilized to get a general overview of the inner and outer structure of the objects. This overview can give us hints to locate positions of interest, where further measurements are reasonable with elemental or phase sensitive neutron techniques. If the analysed object is manufactured from a few distinct parts, which each for itself can be treated as nearly homogeneous, it may be sufficient to investigate only a few spots on these distinct parts. For the methods based on scanning, this can give a considerable benefit in measurement time.

The resolution that can be obtained with NT is in the order of ~100μm, which is very sharp compared to other methods used in the Ancient Charm project. It provides information within a relatively short time (from few minutes to few hours).

4.2 *Prompt Gamma-ray Activation Imaging (PGAI)*

Neutron-induced Prompt Gamma-ray Activation Analysis (PGAA) (Paul, Lindstrom 2000; Molnár 2004) is based on the detection of characteristic, immediate gamma-rays from radiative neutron capture. It enables a non-destructive analysis for the determination of elemental and even isotopic composition of the irradiated samples on major as well as minor level.

PGAA utilizes thermal or cold, i.e. lower energetic, neutrons, because the probability for a neutron capture process is decreasing with higher neutron energies. A typical PGAA setup consists of a sample holder at the end of a neutron beam guide and a shielded Compton-suppressed gamma spectrometer, which is placed perpendicular relative to the beam direction. This way, the number of neutrons hitting the gamma detector is minimized, reducing neutrons damages of the detector crystal. The energies of the measured gamma-rays determine the elemental composition of the complete sample while their intensities are a measure for the elemental concentrations inside the sample. Typical PGAA measurements can be relatively short, i.e. in the order of an hour, depending on the elements involved and their concentrations. The analysis can be started on-line during the measurement to give quick first results.

Though conventional PGAA provides no spatial resolution, a strong collimation of the neutron beam and the gamma-detection can reduce the sampling volume, i.e. the origin of the detected gammas is localized. This provides the basis to construct the element distribution of the sample by scanning, i.e. measuring a set of gamma spectra at different positions in the sample. This technique was named Prompt Gamma-ray Activation Imaging (PGAI) (Kasztovszky, Belgya 2006). Two measurement configurations can be applied (Belgya *et al.* 2008). In the "chord geometry" only the neutron beam is collimated so the resolution along the beam direction is lost, giving 2D elemental distribution information. However, the data from a set of rotations and translations of the sample illuminated by a chord of neutron could serve as an input to tomographic reconstructions. If both the neutron beam and the gamma detector are collimated, the intersection of the neutron beam and the solid angle of the collimated gamma-detector are defining the probing volume, which gives the elemental distribution at a position in 3D space. Ideally, it is a small fixed volume in the space, which is the source of the analytical information. Due to counting rate considerations the smallest feasible volume was a few tens of mm^3 so far. The sample is placed on a xyzω-moving table, to move the desired measurement positions into the spatially fixed probing volume. In principle, a complete 3D scan of the sample can be performed in this way, giving a full 3D elemental map. As a unique approach, this method gives a direct image and does not involve a mathematical reconstruction procedure.

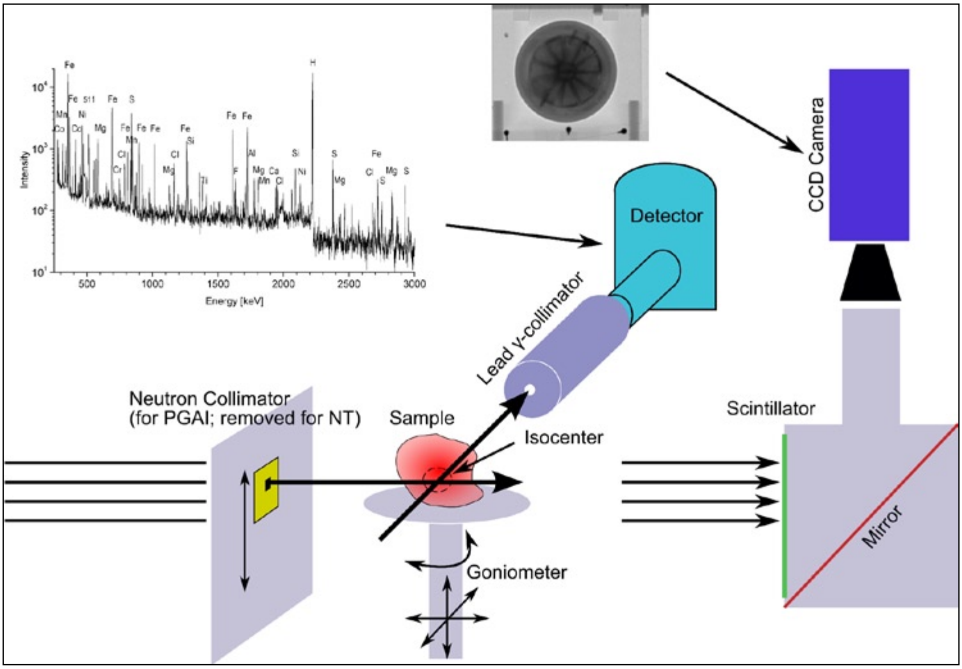

Fig. 6 – Illustration of a combined PGAI/NT setup. For PGAI measurements the neutron collimator is put into the beam, while for NT an open neutron beam is needed.

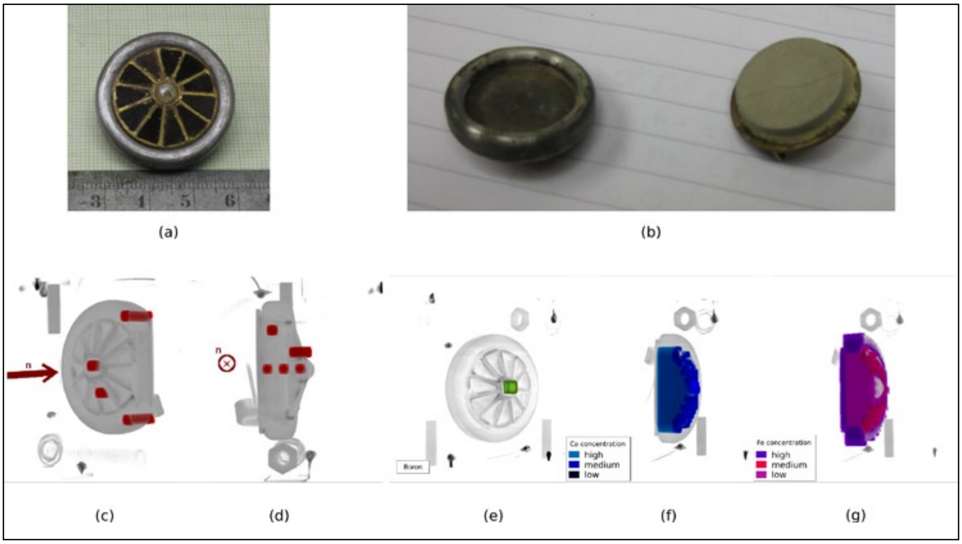

Fig. 7 – PGAI measurement on: (a) Fibula replica with chord geometry. (b) Replica taken apart after analysis; left: top of replica, right: back-plate with filling material. Measurement positions can be seen in (c) for frontal orientation and in (d) for lateral orientation. In combination with the earlier performed NT a 3D elemental distribution can be deduced: (e) Boron in the pearl of the fibula; (f) Calcium in the filling material; (g) Iron mainly in the outer ring of the fibula.

Because PGAI and NT both utilize a steady cold neutron beam, it is sensible to combine these two methods in one measurement setup (Fig. 6), which can easily be switched from PGAI configuration to NT configuration by removing the neutron collimator. The possibility to take neutron radiographies at the PGAI setup also eases the process of sample positioning.

The main disadvantage of PGAI compared to bulk PGAA is the limited statistics, which results in much longer measurement times. Thus it is desirable to restrict the measurements to some previously selected positions. Due to the missing gamma collimation the chord geometry is performing better in terms of statistics. A well-thought selection of measurement positions and angular orientations of the sample might as well give limited 3D information for special sample geometries.

In Fig. 7, c-g an example of such a measurement, performed at the PGAA station at the research reactor FRMII in Garching near Munich (Kudějová *et al.* 2008), is shown. The replica of a fibula was measured at 11 different positions from the front and the side with a pencil beam of 2 mm diameter in chord geometry. The combination with the previously performed NT gives a good, although not completely correct, understanding of the 3D elemental distribution in the fibula. While the boron and calcium distributions are accurately reproduced, iron seems to be distributed all over the replica. This is explained by the fact that it is impossible for the given geometry to irradiate the inner part of the fibula without hitting a part of the iron ring or back-plate. Although the 3D distribution cannot be determined with this chord configuration as precisely as with the double collimated setup, the much shorter measurement times might outweigh the loss in accuracy in many circumstances.

4.3 *Neutron Resonance Transmission (NRT) Tomography*

Neutron Resonance Transmission (NRT) is one of the main analysis modes to be developed by the Ancient Charm project. NRT reveals the elemental composition of samples by measuring neutron absorption resonances in the transmitted neutron beam. Resonances appear as a sharp peak or dip in the measured data representing a physical quantity. They are studied with the so-called neutron time-of-flight (TOF) technique and thus are ideally operated at pulsed spallation neutron sources with high fluxes of epithermal and high-energy neutrons. Due to their different velocities the neutrons spread during their flight, which allows discrimination based on their energies.

The start of a beam pulse is recorded electronically. Because the neutron's energy is proportional to the square of the neutron velocity, it can be derived from the time interval from the start of the beam pulse to the moment the neutrons hit the detector. This turns the energy determination into a time measurement, which can be performed more accurately for neutrons.

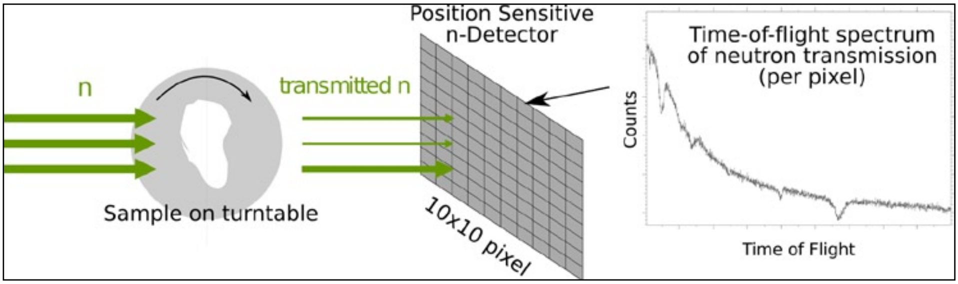

Fig. 8 – Principle of NRT. The sample is rotated in front of a position sensitive neutron detector and turned. For each angle time-of-flight absorption spectra are recorded.

The aforementioned resonances in the transmission process appear now as "dips" in the count-rate vs. time-of-flight spectra taken behind the sample, i.e. from the normal neutron energy distribution some neutrons at distinct energies are removed resulting in lower count-rates at the corresponding measurement times (Fig. 8).

At the ISIS neutron spallation source in Oxfordshire, United Kingdom, a position sensitive neutron detector (PSND) for time-of-flight measurements has been developed (Schooneveld *et al.* 2009). It consists of a 10×10 array of 2×2 mm^2 lithium glass pixels, which are separated by a 0.5 mm optically insulating layer, resulting in an overall size of 2.5×2.5 cm^2. When hit by a neutron the lithium inside the glass emits a light flash, which is electronically recorded. A motor-driven moving system is used to position the samples in front of the PSND. Samples that are larger than the 2.5×2.5 cm^2 can be measured by moving the sample in front of the detector and combining the measured sections afterwards.

Each spectrum of the 100 lithium glass pixels gives the elemental distribution of the spot located in front of the pixel. The 2D plot of the concentrations of a single element then results in an "elemental picture" of the investigated object. In Fig. 9, b the silver distribution of a belt replica measured with NRT is shown. Due to the size of the object a 3×3 array had to be scanned for a full elemental picture of the object, resulting in 900 spectra and a picture of 30×30 pixels. The use of a standard interpolation algorithm can improve the subjective picture quality (Fig. 9, c), which may help in the interpretation of the object; however it is important to note that the real resolution is not enhanced by interpolation.

The final goal is again to obtain a 3D elemental distribution by rotating the sample in front of the NRT detector and acquiring "element radiographies" for multiple angles, which can then be reconstructed with the back-projection algorithm to a 3D image.

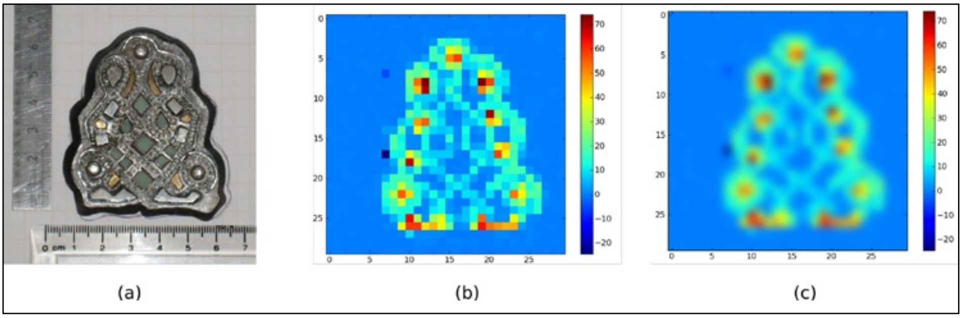

Fig. 9 – NRT on the replica of a belt point shown in (a) measured at the ISIS spallation source. (b) Silver intensities measured with the pixelated neutron detector. The picture is a combination of 3×3 different distinct NRT exposures to cover the whole sample. (c) Bilinear interpolation provides subjective better resolution and a "clearer" image but cannot improve the real resolution (Intensities in arbitrary units).

4.4 *Neutron Resonance Capture Imaging (NRCI)*

Neutron Resonance Capture Imaging (NRCI) is the imaging extension of Neutron Resonance Capture Analysis (NRCA) (Postma *et al.* 2001; Postma, Schillebeeckx 2005), a technique that uses the time-resolved detection of the capture gamma-rays as an indication of the absorption process of epithermal neutrons. While for NRCA an open neutron beam is used and a single gamma-detector is sufficient, the spatial information for NRCI is obtained by using a collimated pencil beam of neutrons of about 5mm diameter and a so-called detector bank with several detectors, which surrounds the investigated object to extend the solid angle for gamma-ray detection. The sample is then scanned with the collimated neutron beam in chord geometry to produce a projection map. Rotating and moving the sample with pencil beam geometry can be used to reconstruct the 3D element distribution with tomographic methods. The NRCI measurement setup at the ISIS facility is shown in Fig. 10.

The principle difference between NRT and NRCI is that the former uses an open beam, while the second requires a narrow, collimated beam of neutrons. The two detection techniques complement each other.

One big problem of NRCI is, like it is for PGAI, the reduced statistical precision compared to the bulk method NRCA. The strong collimation of the neutron beam reduces the measured signal significantly but unfortunately the detected background is not reduced in the same magnitude.

4.5 *Neutron Diffraction Tomography (NDT)*

In addition to elemental information the phase composition and the microstructural properties of the analysed samples may give valuable informa-

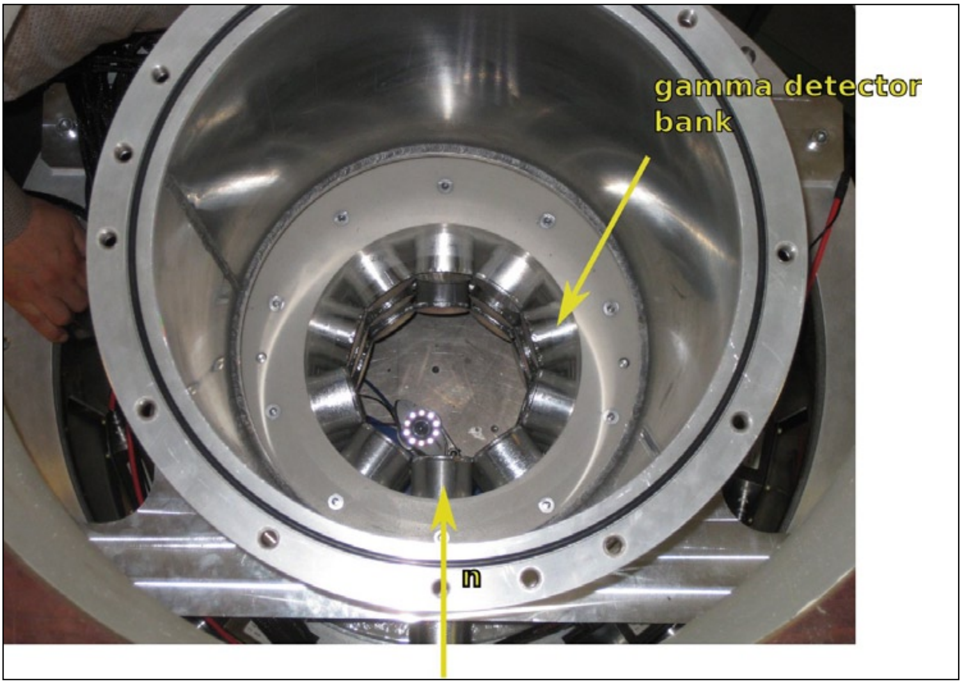

Fig. 10 – Picture of the NRCI measurement chamber at the ISIS facility. The arrow labeled with "n" indicates the direction of the incoming neutrons.

tion about the manufacturing process or other mechanical stresses the objects were exposed to (Gutmann *et al.* 2006; Kockelmann, Kirfel 2006).

These properties can be investigated by analysing the diffraction properties of the materials. The scattering of neutrons on crystal planes produces characteristic diffraction patterns, which result from the constructive and destructive interferences of neutrons reflected from the crystal lattice planes. One may use the reflected or the transmitted neutrons to derive the microstructural information. For the reflected neutrons, the positions and intensities are used to deduce the lattice parameters and atom positions, respectively. For transmitted neutrons one gets sharp edges in the transmission spectra occurring for longer neutron wavelengths that do not fulfil the scattering conditions anymore and thus are fully transmitted by a given family of crystal planes. This results in a sharp increase in neutron transmission, which occurs as so-called Bragg edges in the transmission spectra.

The position sensitive detector from NRT may be used to measure angular dependant Bragg edge intensities, which can in principle be used to

	PGAI	NRCI/NRT	NDT	NT	X-ray Tomography
Neutron source	Research reactor (steady-state)	Spallation source (pulsed)	Spallation source (pulsed)	Research reactor (steady-state)	X-ray tube, synchrotron (steady-state)
Information	Elemental composition	Elemental composition	Microstructure	Neutron attenuation	X-ray attenuation
Sensitivity					
High	B, Cd, Sm, Gd	Cu, As, Zn, Ag, Sb, Sn, Sm, Gd, Au, Co	Single crystals	H, Li, B, Cd, Sm, Gd	Increases for heavy elements
Medium	H, Cu, Ag, Au, Na, K, Mn, Fe, Al, Ti	Al, Fe, Ni, Ti, Ca, Na, K, Cl, Si	Polycrystals	K, Mn, Fe, Ti, Cu, Ag, Au	
Low	C, N, O, Mg, Si, Sn, Pb	H, B, C, N, O, Pb	Amorphous	C, N, O, Na, Al, Sn, Pb	
Typical objects	pottery, stones, metals, glass	metals, copper alloys, pottery	copper alloys, iron, pottery	metallic objects, wood, organics	pottery, metals, alloys, organics
Penetrating depth	1-2 cm	~ 10 cm	~ 10 cm	few cm	mm – cm
Spatial resolution	1-3 mm	~2-5 mm	~2-5 mm	~100 um	~1-5 um

Tab. 1 – Overview of the described methods.

obtain the 3D distribution of crystallographic properties via tomographic reconstruction. This method is called Neutron Diffraction Tomography (NDT) (Gorini 2007). It should complete the element sensitive methods with complementary microstructural data.

4.6 *Summary of the methods*

The key features of the above discussed methods are summarized in Table 1. One can see that the methods can provide complementary information. The joint use of various methods is the key to characterize the objects in great details. The curator and the analyst should cooperate to identify the most suitable methods in order to answer adequately the archaeological questions. Please note that these methods may not be available at a single laboratory, thus the transportation of the object is an issue to be addressed (Tab. 1).

5. Sample positioning and alignment

5.1 *Requirements*

A crucial task is the correct placement of the samples in the desired measurement positions. It is essential that the accuracy of the sample positioning has to be better than the resolution of the measurement method itself. This means that the alignment has to be accurate to about 1 mm or better.

To define measurement positions lying on the surface of sample, a laser beam can be used that is coincident with the path of the neutrons. The sample can be easily positioned by eye in such a way that the laser hits the desired part of the sample. This method fails when it is desirable to investigate inner parts of a sample, e.g. the glue or resin used during the manufacturing process of an object. For this demand another method has to be used.

5.2 *Sample holders and reference markers*

It had been decided to construct universal sample holders for the objects, which protect the valuable objects from any possible damage during transportation from one facility to another and which ensure the unique, firm positioning on the different instruments (Kudějová 2008). Reference markers were attached on these sample holders at precisely determined positions relative to the sample (Fig. 11, a). These reference markers have to be detectable by all measurement methods and should be small enough to allow accurate determination of their position. For the unique alignment of these markers in 3D space at least four markers have to be used. For the tomography based methods it is essential that the markers, which have high neutron absorption, never appear in front or behind the samples during the rotation, because then the filtered back-projection algorithm would fail.

For the cold neutron based methods, i.e. PGAI and NT, needle hats, which were painted with gadolinium-loaded white nail polish, were used to mark the reference positions. Due to the high cross-section of gadolinium for cold neutrons they were easily detectable with these methods. Because of the nearly spherical shape the determination of their position could be performed very accurately by using the centre of the spheres as reference.

Unfortunately, for the methods utilizing neutrons of higher energies, these gadolinium markers turned out to be not as visible as desirable. A reason for this could be that the gadolinium concentration in the nail polish used for painting the needle hats was too low. This flaw could be fixed by using small silver pieces as markers, which could be detected easily with higher energy neutrons. An optimal solution for the future may be the combination of these two markers by placing small pieces of silver in the middle of gadolinium painted spheres.

5.3 *Positioning with the help of reference markers*

By performing a Neutron Tomography of the sample in the frame with attached references markers one gets a 3D “map” of the inner and outer parts of the object. The reference markers provide landmarks, which can be used

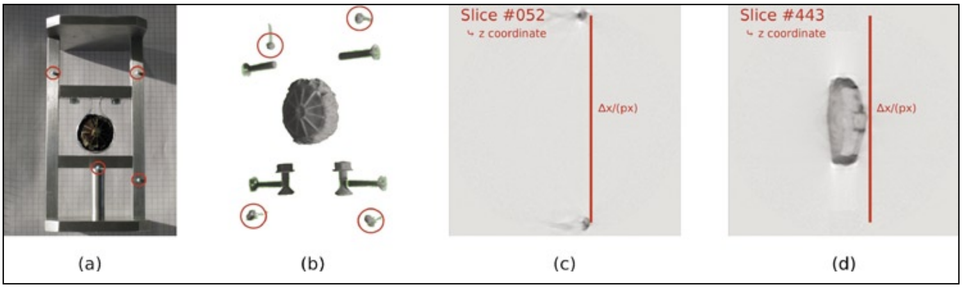

Fig. 11 – Use of reference markers for sample positioning. (a) Fibula sample in holder with reference markers. (b) NT of fibula. The references markers are clearly visible in the reconstruction. (c) and (d) Slices of the NT reconstruction. The slice number gives the z-coordinate. (c) A slice in the middle of the two upper reference markers. The known distance in mm of the reference markers and the distance in pixels in the reconstruction slice give the conversion factor for the transfer of positional information from reconstruction to laboratory system. For the y-axis the lower front reference marker can be used. (d) Slice in the middle of the fibula.

to convert from 3D voxels inside this map to millimetre coordinates in the laboratory coordinate frame.

The determination of the reference marker positions in the laboratory coordinate system, which is usually given by motor steps of the xyzω-tables used for sample positioning, can be achieved by different means, e.g. by measuring the distance from fixed known positions in the laboratory system with a ruler or calliper, utilizing a laser to place the markers to a known position or by doing an in-beam scan. Taking NRT as an example, the scanning was performed by moving a small silver piece (~0.5 mm) horizontally in the direction of one axis in front of the position sensitive detector in small steps of 0.5mm until the depths of the silver resonances were minimal. For minimal silver signal the marker is located in front of the inactive frame between two pixels. For the other axes this procedure is repeated with the moving table turned by 90°, and the vertical axis is aligned by doing a vertical scan. The motor positions obtained from this scans give a fixed relation between moving system and detector and therefore the detected data.

With the known positions of reference markers in the laboratory coordinate system sample positioning is now a task of selecting a desired measurement position in the NT, determining the offset of these positions in NT coordinates, i.e. pixel numbers and slices in the reconstruction, from the position of the reference markers in the reconstruction (Fig. 11, c, d), and transferring these offsets to the laboratory coordinate system by applying translations vectors, scaling factors and a rotation matrix.

The transferred coordinate offsets are finally added to the known position of one of the reference markers in measurement position, which gives the final coordinates for the moving system.

6. Combination and registration

An important aspect of the project was the combination of the results gained from the different measurement methods. In the end a full 3D representation of the object, containing the morphological structure, elemental distribution and crystallographic properties is desirable. For this task a 3D visualisation software was used, which allows the import and combination of different 3D data-sets and the navigation inside the combined data-sets for easy interpretation of the results.

For the alignment of the different data-sets the reference markers used for the positioning of the samples can be used. The collected data is inserted in a 3D voxel space, where the voxels size is chosen according to the resolution of the measurement method. The position of the single data-points in this "voxel space" is determined relative to the reference markers, which are also included into these 3D data-sets. The final registration is then done by aligning the position of the reference markers.

7. Conclusion and outlook

It has been shown in the frame of the Ancient Charm project that neutron based methods have a great potential for the investigation of cultural heritage objects. Several measurements on replicas and real objects have been performed, some still being under analysis. The main disadvantage of the 3D methods compared to their bulk counterparts is their reduced statistics, which limits sensitivity and increases the measurement time that is needed to get meaningful results. If one limits oneself to 2D space this drawback is reduced. As was shown for the 2D-PGAI the combination with a beforehand performed neutron tomography, together with some reasonable assumptions about the sample, can be used to deduce the 3D elemental distribution for not too complex objects. For relatively flat objects, like the belt mount that was shown here, the additional information obtained from a real 3D measurement may be negligible compared to the information gained from increased statistical significance.

For the future the results of a full 3D scan of the real fibula and the 3D reconstruction from a NRT tomography can be expected.

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ABSTRACT

The development of novel non-destructive, neutron based, 3D elemental imaging methods for the analysis of cultural heritage objects was the aim of the European Ancient Charm Project. From 2006 on, many European institutes have worked on the extension of existing and the development of new neutron based analysis methods, and on procedures for the combination of the results from these measurement methods. Several objects were measured and analysed, and have already given important results for archaeologists and conservators. The spatial resolutions that could be obtained so far for elemental imaging are in the order of about 3mm. Some exemplary measurements are shown here to point out the possibilities and resolutions of the different techniques.

An important method in imaging is tomography, which can be applied to angular related projections providing integrated sample properties, like e.g. the transmission of cold and epithermal neutrons or the detection of gamma-ray distributions and intensities.

In addition to the development of various imaging methods, the tasks of data registration and object transportation had to be addressed. The joint use of different dataset has advantages over their separate evaluations. A marker-based approach was chosen in combination with the development of universal sample holders, which aims to solve the problems of safe transportation, reproducible positioning and unique data registration.