NEUTRON DIFFRACTION IMAGING OF CULTURAL HERITAGE OBJECTS

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Kivonat

A neutrondiffrakciós vizsgálatok alkalmazásának lehetőségeit régészeti tárgyakon (kerámián és fémtárgyakon) többször is bemutatták. A termális neutronok alkalmazásának előnye a mély behatolás az anyagba és az érintetlen tárgy roncsolásmentes vizsgálatának lehetősége. A neutron diffrakció információt szolgáltat a tárgy szerkezeti tulajdonságairól amelyek gyakran összefüggenek az anyag készítésének, használatának jellemzőivel és az egykori készítéstechnikával. A legtöbb neutrondiffrakciós vizsgálatot széles neutronsugárral végzik a tárgy egy vagy több pontján, így a térbeli felbontás elég gyenge. Ebben a cikkben áttekintjük a rendelkezésre álló lehetőségeket és a neutron sugárral végzendő finom szerkezeti vizsgálatok lehetőségeit.

Abstract

The capabilities of neutron diffraction for studying archaeological ceramics and metals have been demonstrated on many occasions. The main advantages of thermal neutrons are deep penetration and non-destructive analysis of intact objects. Neutron diffraction provides information on structural properties which are often related to the past material treatments and historical fabrication techniques. Most neutron diffraction analyses are normally performed on one or several points of an object with a large neutron beam, hence without much spatial resolution. In this paper we review the existing options and future perspectives of the systematic mapping of phases and microstructures with a neutron beam.

KULCSSZAVAK: REPÜLÉSI IDŐ NEUTRONDIFFRAKCIÓ, NEUTRONSZÓRÁS, DIFFRAKCIÓS KÉPALKOTÁS, ANCIENT CHARM

KEYWORDS: TIME-OF-FLIGHT NEUTRON DIFFRACTION, DIFFRACTION IMAGING, ANCIENT CHARM

Introduction

Neutron radiation is a versatile probe for obtaining information from the interior of undisturbed museum objects and archaeological findings. Neutrons penetrate easily through coatings and corrosion layers deep into centimetre-thick artefacts, a property that makes them suitable for non-destructive analyses. A particular attraction of neutron techniques for archaeologists and conservation scientists is the prospect of locating hidden materials and structures inside objects.

Neutron analysis techniques are based on a simple principle. A material is placed in a beam of neutrons which can interact with the atomic nuclei in two ways: the neutrons are either *scattered* or *absorbed*. These scattering and capture processes are material specific, that is, every material responds differently to neutron illumination. The response to the neutron irradiation is assessed using neutron and gamma detectors. Detectors can be used to measure the intensity of the transmitted or scattered radiation, the scattering angles, or the energies of both neutrons and gamma rays. From these measurements, details of the material properties are retrieved or reconstructed. For example, the gamma ray energies emitted during or after neutron irradiation are element specific, i.e. they allow to determining the elemental composition of a material. This is the basis of one of several neutron activation techniques. Scattered neutrons may be exploited to give information on the microscopic structure of a material in terms of the mineral or metal phase abundance, of the microstructure, of texture or porosity, to name some examples. Neutron radiography and neutron tomography make use of the selective attenuation (scattering and absorption) properties of neutrons.

A detailed, but by no means complete list of neutron diagnostic techniques in archaeological **Table 1.**

sciences   is   compiled   in   **Table   1**.

Table 1.

Overview of neutron techniques in archaeological sciences

Neutron Activation Analysis for isotope and element analysis is based on the capture of neutrons. Characteristic gamma radiation is emitted during (prompt γ's) or after (delayed γ's) the neutron capture.

INAA, Instrumental Neutron Activation Analysis **(**Glascock & Neff 2003): high sensitivity to many trace elements; usually requires sampling; delayed γ's;

NAR, Neutron Autoradiography (Schroeder-Smeibidl et al. 2006): cold neutrons; delayed γ's; non-destructive method for investigation of (mainly) paintings;

PGAA, Prompt Gamma Activation Analysis (Revay & Belgya 2004) is based on thermal and cold neutron capture; prompt γ's; applied non-destructively on intact objects; high sensitivity for some light elements (H, K , Cl);

NRCA, Neutron Resonant Capture Analysis (Postma et al. 2004): is based on epithermal neutron capture; prompt γ's; applied non-destructively on intact objects; good sensitivity for some heavy elements (Au, As, Ag, Sb, Sn).

Neutron Radiography/Tomography (Deschler-Erb et al. 2004; Materna et al. 2004): real space imaging based on the capture and scattering of thermal and cold neutrons to provide an inside view of objects with a spatial resolution down to 100 micrometers; exploits the attenuation of a neutron beam passing through an object; attenuation contrast for different elements, high sensitivities for some light elements (e.g. hydrogen); contrast variation by variation of neutrons energies (Kardjilov et al. 2003). Further imaging prospects are provided by phase contrast radiography which is based on neutron refraction (Treimer et al. 2003).

Neutron Diffraction (Kockelmann et al. 2001; Siano et al. 2006) is based on the elastic scattering of thermal neutrons by periodic, long-range ordered (crystalline) or non-periodic, short-range ordered (glass) arrangements of atoms. Many structural aspects can be studied: phase and structure analysis, texture analysis, microstructure analysis; residual stress analysis (Santisteban et al. 2002). Bragg-Edge transmission for mapping of strains and phases is based on Bragg scattering (Santisteban et al. 2001).

Small Angle Neutron Scattering (SANS) (Botti et al. 2006), based on the elastic neutron scattering of thermal neutrons. Porosity of a material, and size and surface characteristics of mineral aggregates can be studied.

Some of the neutron analysis methods are well established, some are still being developed. Other techniques have clearly shown their potential but have not found widespread use in archaeological sciences.

Neutron diffraction is capable of determining many structural aspects of a material such as phase composition and crystallographic texture. These properties are often related to the deformation and treatment history of the material. The use of neutron diffraction as a non-destructive

archaeometric tool to study ceramic and metal artefacts was proposed just a few years ago. The characterisation potentials were initially investigated on ceramics (Kockelmann et al. 2001; Kockelmann et. al. 2004) and archaeological bronze objects (Siano et al. 2003; Kockelmann et al. 2006a) on the powder diffractometer ROTAX at the pulsed neutron source ISIS at the Rutherford Appleton Laboratory, UK. During the first phase, the work was aimed at assessing and quantifying fundamental concerns, such as the typical

acquisition times to achieve satisfactory pattern statistics and pattern resolution, activation effects and the corresponding decay times. Depending on sample thickness and neutron spot sizes, the typical time needed to achieve satisfactory data statistics and resolution for a quantitative analysis ranged from a few minutes up to several hours for metal and pottery samples, respectively, using large neutron spot sizes of several square centimetres. This large beam illumination is considered an advantage of the neutron analysis because it provides a representative overview of the structural properties, averaged over a large sample volume. Neutron diffraction analyses are typically performed on one or several analysis points, either to check for homogeneity of the material or to survey a complex object composed of several parts of different materials. This non-destructive surveying of an object is limited by the available neutron flux, the available beamtime on the diffractometer, by the time it requires to collect a diffraction pattern and, last but not least, by the size of the diffracting volume, i.e. the size of the neutron sampling volume. It normally takes many hours to analyse several points on an object.

A complete mapping of objects by neutron diffraction is rather the exception than the rule, like the cross section mapping of a bronze sword (Bartoli et al. 2007), and it is done with comparatively low spatial resolution. This is to be compared with neutron tomography which produces images in the sub-millimetre resolution range in a matter of minutes. However, the attenuation images do not provide direct information on the type and structure of materials. That is to say radiography and tomographies often show high-contrast details of features inside objects but there is no information available on the elemental and structural consistency of the feature. The structural information can be provided by neutron diffraction which is why a combination of tomographic and scattering methods is generally desirable. The elemental information can be provided by activation methods (**Table 1**).

This paper focuses on the neutron diffraction techniques and surveys on the status and current developments of diffraction mapping techniques. Phase and structure mapping is investigated in the framework of the EU funded ANCIENT CHARM project that aims to develop and combine different imaging applications for Cultural Heritage materials, using neutron tomography, prompt gamma activation analysis, neutron resonance capture analysis, and neutron diffraction. The focus of this paper is on time-of-flight applications at the ISIS neutron source. Neutron imaging on bulk polycrystalline materials on a constant wavelength neutron source has been reported by Wroblewski et

al (1999) using a microchannel plate as secondary collimator between sample and detector. In chapter 2 the basic diffraction principles are recapitulated, followed by a short description of the time-of-flight technique. In chapter 3 the current experimental mapping principles are reviewed and compared. In chapter 4 and chapter 5 the advantages, limitations and future prospects of the diffraction mapping methods are discussed.

Neutron diffraction in archaeological sciences

Diffraction basics

Diffraction is a general phenomenon that occurs if waves are impinging onto an obstacle, be they sound waves, light waves, X-rays or particle waves. Diffraction is based on the superposition of waves that re-enforce each other (constructive interference) or cancel each other out (destructive interference). Diffraction is ideal for studying any periodic arrangements such as those of atoms in a crystal provided the wavelength of the radiation is just right, namely of the order of the interatomic distances, i.e. around 10^{-10} m. Intensity maxima occurring at characteristic positions (scattering angles or energies) in the diffraction pattern are called 'Bragg peaks' or 'Bragg reflections'.

The radiation may be electromagnetic (X-rays) or of particle nature (electrons, neutrons etc), the basic principles are the same. The incident electromagnetic or particle wave is elastically scattered by the objects of interaction (electrons in case of X-rays, atomic nuclei in case of neutrons), and the superposition of the scattered waves observed under a scattering angle 2θ and at a large distance from the sample results in an intensity distribution that is characteristic for the scattering object. This principally simple fact allows for modelling of the object's material and explains why diffraction is one of the most powerful tools in studying matter. X-rays see the electron shells of atoms, neutrons their nuclei. While thus for X-rays the proportionality between the atomic scattering power and the number of electrons (or the position in the periodic system) is a natural consequence, such a systematic relation does not exist for neutrons and nuclei whose capability to scatter neutrons can even have quite different values for isotopes of one and the same element. Also is the interaction between neutrons and nuclei generally much weaker than between X-rays and electrons, which follows basically from the facts that neutrons do not have an electric charge and that nuclei can be considered as practically point-like scattering centres that offer only very small 'cross sections' for interaction. As a consequence, neutrons can penetrate bulk matter much deeper so that in total

X-ray and neutron diffraction are not competing but complementary methods, particularly with respect to attenuation and to the response from different chemical elements. X-ray diffraction can hardly deliver information from the interior of a cm-thick sample and hardly distinguish elements adjacent in the periodic system, neutron diffraction can do both. Hence, for non-destructive studies of archaeological objects or historic artefacts, X-ray diffraction is a choice for studying surfaces whereas neutron diffraction sees the bulk. Since the neutron beam generally surveys a much larger sample volume than does an X-ray beam, the results of a neutron analysis have a good chance to truly represent the composition and material properties of the whole object.

Both types of radiation, X-rays and neutrons, probe not only the chemical elements in a sample but also the spatial distribution of the atoms represented by either their electron shells or their nuclei, respectively. If the distribution is not periodic as it is the case of liquids or amorphous material (e.g. glass) the intensity distribution of the diffracted radiation is a continuous function of the scattering angle (in case of monochromatic radiation) or of both angle and energy (in case of polychromatic radiation). Whichever applies, the observed intensity distribution reflects the geometrical arrangement of the atoms as well as their respective scattering powers and thus provides a 'fingerprint' of the scattering object which can serve to identify the material and to retrieve structural properties.

If the sample material is crystalline, i.e. characterised by an atomic arrangement repeated in three dimensions, the diffraction pattern becomes discontinuous. For a single crystal, all the scattered radiation is focussed into discrete spatially resolved directions with the consequence that a large number of very small and randomly oriented crystals of the same kind (a powder) produce reflection cones. Intersecting the cones with a detector one obtains a characteristic sequence of reflection lines with different intensities. The resulting diffraction pattern is again a 'fingerprint' of the scattering object, now of the kind and arrangement of the atoms in the repetition unit, its dimensions and symmetry properties, in short of the crystal structure. With data bases containing information for many thousands of structures, 'powder diffraction' is a very efficient method for the qualitative identification of crystalline phases and for the quantitative assessment of their structures.

This applies also to samples with more than one crystalline and/or amorphous phases which produce a diffraction pattern that is the superposition of the individual phase fingerprints, each weighted with its abundance. Then, provided the number of different phases is relatively small (up to 10), their abundances are not too different and their diffraction patterns do not overlap too much the diffraction pattern supplies also quantitative phase composition information. This holds under the caveat that the scattering response of a given amount of compound is continuously distributed in case of amorphicity, but focused into discrete reflections in case of crystallinity. Amorphous material is difficult to assess in presence of crystalline phases and even more so in case of amorphous mixtures alone.

Neutron diffraction information from archaeological objects

Neutron diffraction is a direct method for examining all structural aspects of archaeological materials. Many archaeological materials (e.g. pottery, marble, pigments, metals, alloys, corrosion products) are poly-crystalline and multi-phase opposed to single-crystalline and single-phase, respectively. The corresponding multi-phase diffraction pattern is a superposition of the single-phase patterns. The diffraction pattern of a pure metal with cubic or hexagonal symmetry may contain a rather small number of about 20 Bragg peaks. A multi-mineralic piece of pottery generates thousands of peaks. The task of diffraction analysis is to disentangle the different phase contributions, to determine their relative abundance, and to extract structural parameters for the individual phases.

The knowledge of the structural properties of a single- or multi-component material is of importance for understanding many of its physical and chemical properties and how it behaves under mechanical stresses or external environmental influences. The microscopic structure of a material often carries information about the mechanical deformation history. An important parameter is the crystallographic texture of a material. Polycrystalline samples are made of a large number of grains that are composed of tiny single crystals ('crystallites'). Each of the crystallites may have a size and orientation different from its neighbours. Often the grains are oriented at random, then the material is said to be free of texture. Otherwise, if the grains prefer certain orientations, e.g. from a particular mechanical treatment, then the material is said to exhibit texture.

More generally, the structure of a polycrystalline material may be characterised at different levels (Bunge 1999):

• The 'phase structure' describes the composition of a material from several mineral or metal components. The presence and the absence of certain minerals in a piece of pottery, for example, and a quantitative assessment of the mineral mixture may provide information about the initial clay

mixture and about firing temperatures and the firing atmosphere. Firing minerals like mullite or cristobalite are produced during high-temperature firing of the starting materials when the minerals undergo phase transitions and transformations into new compounds that are critically dependent on the firing processes. This means that the mineral phase compositions of different ceramics are generally dissimilar, which supports the concept of a fingerprint. However, the complexity of the phase transformations makes it almost impossible to reconstruct the firing processes in detail. Another important example is the phase analysis of metals. The detection of a particular phase can provide clues to production techniques. For example, archaeological bronzes may contain the beta bronze Cu-Sn phase which has to be hot-worked and quenched to ambient temperatures in order to be retained. For iron objects, the presence of iron carbides provides clues to intentional hardening.

• The diffraction peaks that belong to a particular phase are used to examine the 'crystal structure', i.e. the atomic arrangement of that phase. Unit cell dimensions can be determined with high precision by neutron diffraction. The lattice parameters of solid solutions, particularly of alloys (e.g. $Cu_{1-x}Sn_x$ bronze) often vary linearly in a wide composition range according to Vegard's rule when smaller atoms (Cu) are replaced by bigger atoms (Sn). Then, by measuring the lattice parameters, the concentration of the substituting atoms (Sn in Cu) can be determined.

• The 'grain structure' refers to sizes, shapes and mutual orientations (texture) of grains. Neutron texture analysis is an elegant method to determine the orientations of grains (more precisely: of the crystallites) in an object. A polycrystal with a preferred orientation of crystallites is said to have texture. If the grains in an object are oriented at random and if all grain orientations are equally realised then the material is said to be free of texture. Texture is an important material characteristic of metals, alloys and ceramics. Since texture is usually a result of the solidification and manufacturing processes it can contain information about the production history of an object. Well defined textures are produced under specific conditions, during primary crystallisation from a melt as well as by thermal and mechanical treatment of the workpiece such as annealing, drawing, rolling, and hammering. Neutron diffraction can therefore provide information on the creation and deformation history of an object, be it a modern engineering component, a mechanically deformed workpiece of archaeological interest or even a geological sample deformed by tectonic processes.

• The 'microstructure' describes deviations from the ideal crystal structure within a grain. The diffraction data contain, to some extent, information on microstrains. Structural defects like missing atoms or extra impurities, but also lattice defects in otherwise 'ideal crystals' may be responsible for local distortions and lattice deformations. Defects and microstrains are induced by mechanical and thermal working processes. Slow heating and 'annealing' at high temperatures may reduce crystal defects and relax lattice strains whereas a rough treatment like hammering and quenching generates microstrains. Diffraction and ensuing reflection profile analysis can provide valuable clues to the working processes, particularly useful if microstructural trends are revealed by comparison with suitable reference samples that are produced in a controlled way.

In addition to these four structural levels also residual strains (or macrostrains) should be mentioned. Residual strains are induced by macroscopic compressive or tensile stresses exerted during working on a material. While microstrains are inside microscopic grains and are often isotropic, macrostrains are different and directional in different parts of a piece, for instance on either side of a bent metal bar. Residual strain-stress analysis is a strong domain of neutron diffraction and hence an important tool in engineering sciences regarding non-destructive materials testing and quality control. The same technique can be applied to archaeological objects (Siano et al. 2006).

It is important to underline that these structural features are obtained non-destructively. Neutron diffraction allows for separating corrosion and alteration phases from the alloy phases, and hence to obtain an unobstructed view onto the original alloy components, even in the presence of substantial surface corrosion and mineralisation. The use of neutrons is made even more attractive by the recent development of neutron diffraction techniques such as 'single-shot' texture analysis (Kockelmann et al. 2006b) and Bragg edge transmission (Santisteban et al. 2001) that can be applied to stationary, intact objects. On an instrument like GEM at ISIS identification and quantification of crystalline phases inside archaeological objects can be achieved, along with the characterisation of crystallographic textures and microstructural features of each phase in one single experiment and acquisition. Structure, texture and residual strain analyses can be utilised for identifying the raw materials used, for distinguishing various levels of sophistication of the material treatments and, thus, for unveiling fabrication methods. Once the making techniques are known, texture and microstructure information may help distinguishing between genuine and fake objects.

Time-of-flight (TOF) neutron diffraction

The experimental concepts presented in this paper are based on the time-of-flight (TOF) method, i.e. the energies of the neutrons are determined by a measurement of the flight times. The TOF techniques use a 'white neutron beam', containing neutrons with a wide range of velocities. TOF diffraction has some special advantages for analysis of archaeological objects (Kockelmann et al. 2006a), one of them being that phases and textures of objects can be studied in a stationary set-up. TOF measurements are well performed at a pulsed spallation source where the natural time structure of the neutron production can be exploited. The diffraction imaging concepts presented in this paper have been realised at ISIS. For the neutron production at ISIS, a bunch of protons is accelerated in a synchrotron to 800 MeV and directed onto a composite tungsten/tantalum target. 50 pulses of neutrons per second are generated inside the target material by a 'spallation', i.e. a splintering, process. The generated neutrons are slowed down in a moderator (water or methane) in order to make them useful for material studies. The pulsed nature of the neutron beam is the essential feature for the study of objects by the time-of-flight technique. Energies of neutrons are measured via the flight time (TOF)

$$E = \frac{1}{2} m_n (v)^2 = \frac{1}{2} m_n \left(\frac{L}{TOF} \right)^2$$

(1)

given that the velocity is given by v=L/TOF, where L is the flight path of the neutrons and m_n is the mass of a neutron. With the deBrogie relation v=h/($m_n \cdot \lambda$) the equation reads:

$$E = \frac{1}{2} m_n \left(\frac{h}{m_n \lambda} \right)^2$$

(2)

h is Planck's constant and λ is the neutron wavelength. Diffraction involves reflection of neutrons by crystal planes which is described by Bragg's equation $\lambda = 2d \sin(\theta)$ thus yielding

$$TOF = \frac{2m_n}{h} L \cdot d \cdot \sin(\theta)$$

(3)

relating the time-of-flight (TOF) to the interplanar spacings (d) for a neutron detector at a scattering angle 2Θ. Neutrons scattered by the sample produce diffraction patterns (intensity versus crystallographic d-spacings). The lattice plane distances (d) are characteristic material parameters for a crystalline material. They correspond to the positions of the Bragg peaks in the diffraction patterns, used to identify phases and structures, or to derive strains. **Fig. 1** shows the suite of diffraction instruments at the ISIS neutron source. Several of the stations, like ROTAX, INES, GEM and ENGIN-X, are frequently used for studies on archaeological materials.

There are basically two ways to set-up a time-of-flight experiment. The time of flight technique can be applied with a neutron detector in the incident beam behind the sample (radiography type set-up with 2Θ=180 degrees) to measure the transmitted neutrons or with a neutron detector out of the primary beam (diffraction set-up with 2Θ=0 - 180 degrees) to measure the neutrons scattered by the sample. The neutron diffractometer ROTAX at ISIS (Kockelmann et al. 2001) for instance, is installed on a cold liquid 110K-methane moderator. The flight path is about 15 meters, extending from the moderator to the sample and further on to the detectors. The TOF method makes use of the polychromatic beam of neutrons possessing wavelengths ranging from about 0.5 to 5 Å which correspond to neutron velocities from approximately 8000 to 800 m/s and flight times of the order of 1 to 20 milliseconds. On ROTAX, the scattered neutrons are registered by three position-sensitive detector banks. The banks at backscattering angles have a special relevance in TOF diffraction because diffraction patterns of bulky samples and of objects with irregular shapes can be straightforwardly collected at 'back-reflection' angles. Actually, peak widths of diffraction peaks, measured at backscattering angles, are to a large extent independent of the sample thickness. This special feature of TOF diffraction is of advantage for multiphase analyses and for studying fabrication related peak broadening effects (Siano et al. 2003). It is worth noting that the neutron energies for diffraction are in the (thermal) milli-electron volt (meV) regime, i.e. small in comparison to the epithermal energy regime (eV-keV) of neutron resonance capture analysis, NRCA.

The short-time induced radioactivity of samples on TOF diffraction instruments at ISIS is rather moderate. The time-averaged integrated neutron fluxes on the instruments are rather low compared to reactor neutron sources, whilst the data acquisition times are short in the order of a few hours. In the worst cases the objects can leave the facility after a few days after which any residual activity has disappeared.

***Diffraction imaging: mapping of
structural properties with neutrons***

Diffraction imaging, being a rather underdeveloped field in archaeological sciences, is more commonly applied for the study of engineering materials on neutron strain and stress scanners.
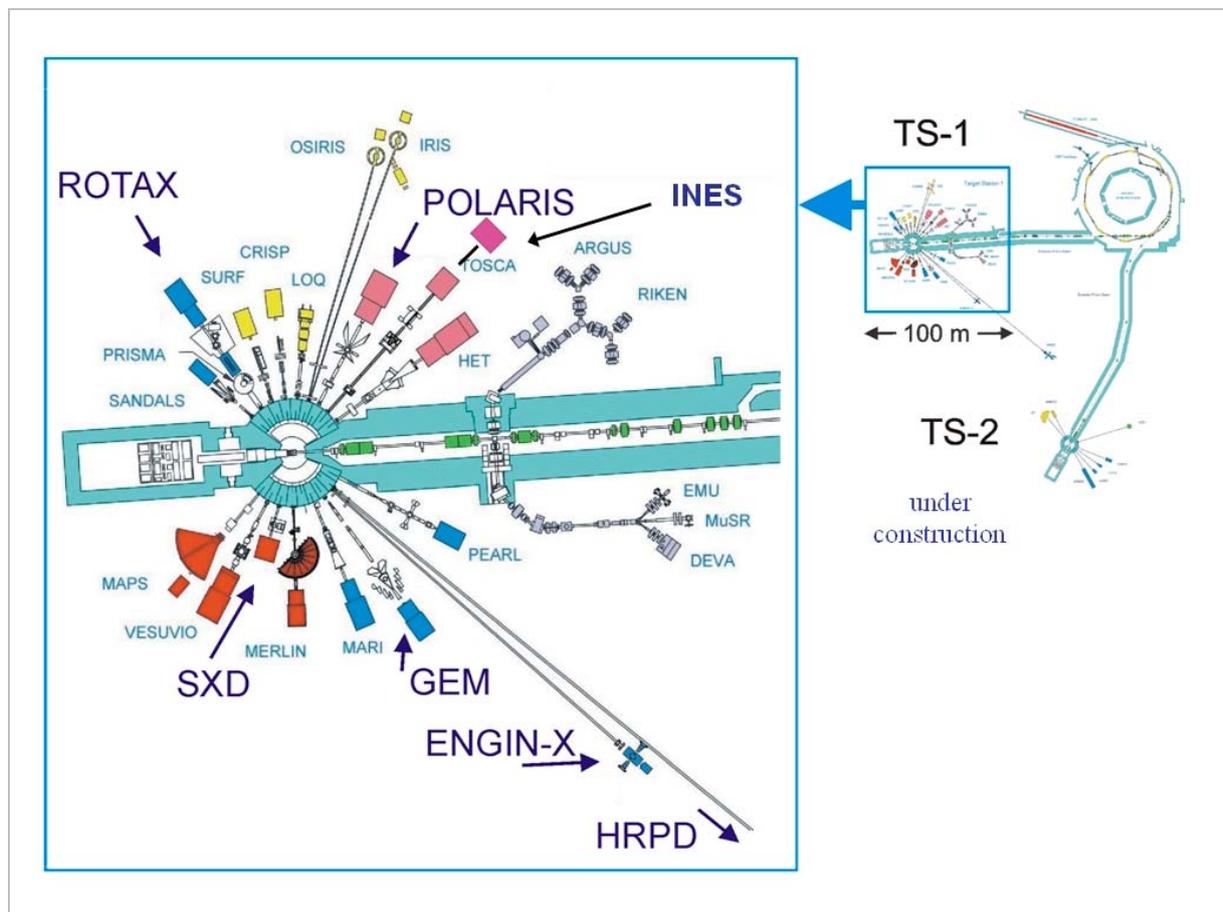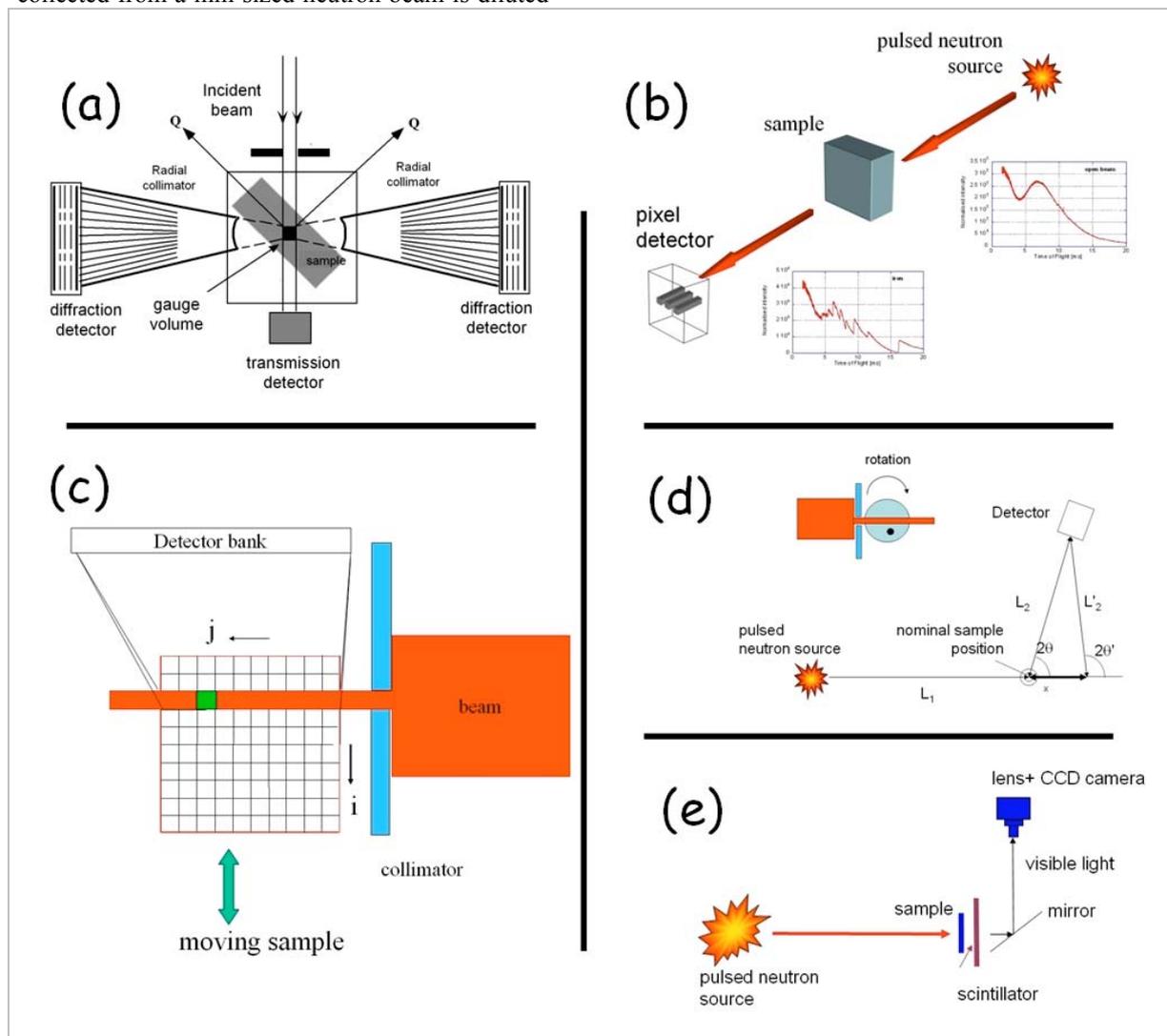

Figure 1.

The instrument layout of the ISIS neutron spallation source. The diffractometers GEM, POLARIS, INES, HRPD and ROTAX are frequently used for studies on polycrystalline materials. SXD is a single crystal diffractometer. ENGIN-X is used for strain and stress analysis of engineering components as well as for archaeological objects. The ISIS target station 1 (TS-1) is operated since 1984, target station 2 (TS-2) is under construction and will be available from 2008.

Many engineering applications involve producing linear scans or two dimensional spatial maps of strains and phases (Santisteban et al. 2003), for instance of mechanically deformed, worked or welded materials. The instrumental requirements for archaeological objects are quite similar to those of engineering applications. In archaeological sciences diffraction analysis provides structural information that is not available from other neutron methods like tomography (contrasts) and activation analysis (elemental contents). Neutron diffraction measurements, performed on existing powder and materials diffractometers, are typically performed on one or several points on an object, to check for homogeneity and for compositional and structural variations of ceramics and alloys. Different points are surveyed by repositioning the object on the instrument. Neutron beams are typically large, in comparison to X-ray and synchrotron applications. For most applications the cross section of the incident neutron beam can be adjusted using diaphragms, typically ranging from 5x5 to 40x40 mm^2. The neutron beam penetrates the whole thickness of the sample, that is to say averaged structural information is collected from the whole illuminated diffracting volume. This averaging of information can be a desired effect if representative information from a homogeneous sample is required or if, for instance, good grain statistics for a texture analysis is needed. The averaging of information can be rather unfavourable if a sample is inhomogeneous, made up of different materials or exhibiting microstructures varying across the sample in terms of phases, inclusions, grain

structure, and microstructures. For instance, archaeological iron generally has a pronounced heterogeneous microstructure, so that information collected from a mm-sized neutron beam is diluted

to such an extent that the results are rather misleading than informative.

Figure 2.

Experimental schematics. (a) ENGIN-X with 2 diffraction detectors at 90 degrees. The gauge volume is selected by the incoming beam collimation and the radial collimators; (b) Bragg edge transmission on ENGIN-X; 3 pixels are representative for a total of 100 pixels of the transmission detector. With no sample in the beam, a smooth distribution of neutron energies is observed (right insert). With the sample inserted into the beam one observes Bragg edges that are characteristic of the crystal structure of the material (left insert); (c) diffraction imaging on ROTAX using a collimated beam; (d) diffraction imaging on SXD by exploiting time-of-flight aberration effects; the insert on the upper right symbolises that the method requires a rotation or translation of the object in front of a collimated beam; (e) set-up for energy selective imaging with a CCD camera.

For many archaeological objects, a systematic mapping of phases and structures, rather than a single analysis point, is therefore highly desirable, and often required in order to obtain useful information. In many cases, high spatial resolution of sub-millimetre range is required. It is obvious that the requirement for high-resolution diffraction imaging is difficult to fulfil because neutrons are highly penetrating, which is why they are used for

non-destructive testing in the first place. There are essentially two approaches to achieve diffraction imaging, depending on the signal used to reconstruct the structural maps. This signal can be extracted from the direct beam (transmission), or from the scattered beam (diffraction). Transmission techniques, essentially modifications of conventional radiography/tomography, are fast, since the full object can be simultaneously

illuminated. Scattering techniques are typically slow because of the low neutron fluxes compared to X-ray sources and because of limited and expensive coverage of the space around a sample with detectors.
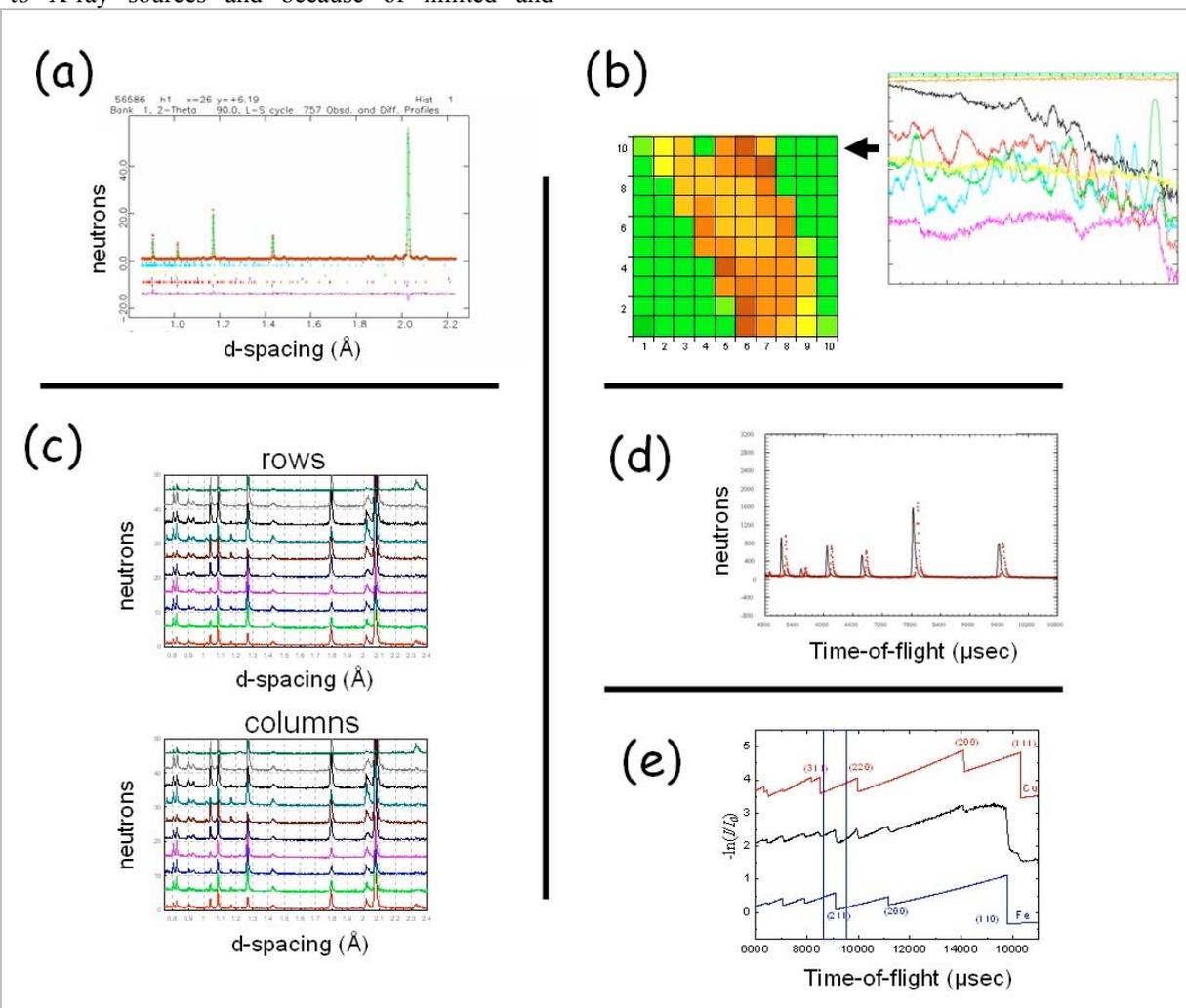
Figure 3.

Illustrations of the type of data collected for the set-ups in Figure 2. (a) typical diffraction pattern collected in 30 minutes from a gauge volume 2x2x10 mm on ENGIN-X on an iron sample displaying ferrite and cementite peaks; (b) example of a Bragg edge transmission map on ENGIN-X; each pixel provides also a fully time-resolved Bragg edge spectrum; (c) sequence of diffraction patterns from a row and column scan on a copper-iron test object on ROTAX; the intensities of the Cu and Fe Bragg peaks are evaluated in order to determine the positions of the phases in the object; (d) diffraction peak shifts due to geometrical aberration as measured on SXD; the positions of Bragg peaks are evaluated in order to determine the positions of the phases in the object; (e) contrast variations by energy-selective imaging are achieved by collecting radiographies below and above Bragg edges of materials. The figure compares a measured Bragg edge spectrum (middle) with calculated spectra of Cu (top) and Fe (bottom).
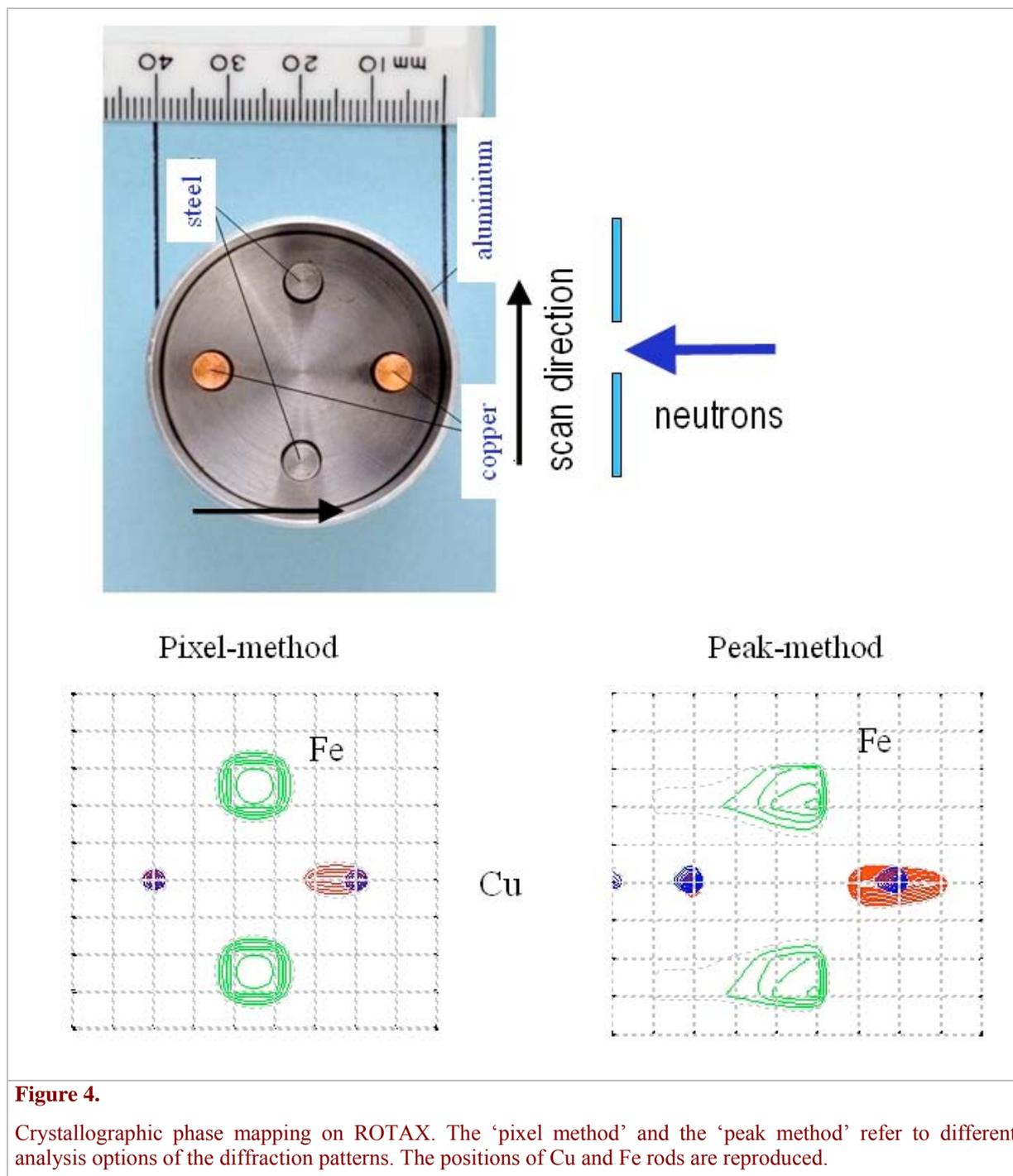
For mapping of structure parameters by neutron diffraction the following components and variables within the instrumental set-up have to be considered in order to achieve spatial resolution:

Conditioning of the incoming neutron beam: The shape, size and divergence of the incoming beam affect and limit the achievable spatial resolution of the maps. An 'open beam' geometry (large beam in the order of square centimetres) for transmission experiments requires a position-sensitive area detector, set-up behind the sample. The real-space resolution of the maps cannot, in principle, be better than the pixel size of the transmission detector. A 'pencil beam' (small beam in the order of square millimetres), defined by a circular or rectangular primary beam diaphragm, is required for scattering applications. The real-space resolution of the resultant maps cannot be better than the size of the pencil beam. The divergence of

the incident neutron beam (i.e. if the beam is parallel or not) is crucially important for both the transmission and pencil beam geometry. A largely diverging incident neutron beam has a blurring effect on the maps.

Figure 4.

Crystallographic phase mapping on ROTAX. The 'pixel method' and the 'peak method' refer to different analysis options of the diffraction patterns. The positions of Cu and Fe rods are reproduced.

Conditioning of the scattered neutrons: A secondary collimator may be used to restrict the diffraction volume along the beam direction: it defines the 'gauge volume' in the sample from which information is collected (**Fig. 2a**). The secondary collimator is usually designed as 'radial collimator' in order to be able to use large diffraction detector areas.

The choice of detector type (transmission or scattering, position-resolving or not, continuous or discrete) is important. The detector resolution is given as pixel size, and is typically in the order of several square-millimetres for neutron detectors. For the transmission mapping, the pixel sizes of the transmission detector limit the spatial resolution.

The choice of dimensionality of the maps to be recorded (linear scan: 1D, plane map: 2D, volume map: 3D), depends on the object to be analysed and on the specific questions to be addressed.

Choice of scan grid for a sample. For producing 1D, 2D or 3D maps with a pencil beam, the sample has to be translated or rotated in small linear or angular steps. The stepwidths limit the achievable resolution. For the transmission set-up the distance of the object from the detector affects the achievable resolution, dependent on the primary beam divergence.

The neutron flux should be as high as possible in order to achieve sufficiently high signals in reasonable data collection times. On present day neutron sources, neutron guides are often used to transport the neutrons and to achieve a high neutron flux at the sample position. This is, however, at the expense of higher beam divergence which may blur the maps.

The scattering of neutrons inside the object itself can have a drastic effect on the achievable resolution (Kasztovszky & Belgya 2006). This effect is particularly pronounced for samples containing hydrogen, e.g. water, plastic, organic material.

In the following different ideas of diffraction mapping are presented. All applications have been realised and tested at ISIS on different neutron diffractometers (**Fig. 1**). **Fig. 2** shows the experiment schematics, **Fig. 3** illustrates the mapping ideas and instrument schematics with data.

Point-by-point scanning on a neutron strain scanner (ENGIN-X)

Structure and phase mapping is a standard procedure on an engineering instrument like ENGIN-X at ISIS (Santisteban et al. 2003). This point-by-point diffraction technique is conceptually the simplest method of imaging. The structural information is obtained from the diffraction signals. The spatial resolution is achieved by tight collimation of both incident and scattered radiation, which define the gauge volume (**Fig. 2a**). On ENGIN-X diffraction patterns are collected with two detectors at scattering angles of 90 degrees. The sample is moved on a positioning table in front of the beam, point by point, in three directions (X, Y, Z), in order to shift the gauge volume through the object. Only neutrons scattering in the gauge volume enter the detectors. Diffraction scans can be performed along arbitrary trajectories, for example into the thickness of a sample, or along the surface of an object to map the strain distribution, the phase content, or the texture of an engineering component or an archaeological artefact. The method is ideal for the analysis of heavily restored objects or heavily corroded objects covered with millimetre thick corrosion crusts, because the signal from the interior of the sample, for instance the original alloy, is not mixed up with the signals from corrosion layers. ENGIN-X has been used for a number of archaeological studies on bronzes (Siano et al. 2003; Siano et al. 2006; Bartoli et al. 2007) and iron objects (Godfrey et al. 2006).

On ENGIN-X the gauge volume is in the order of cubic millimetres, as low as 0.5x0.5x0.5 mm^3. For most applications on ENGIN-X one of the dimensions is kept larger (e.g. 10 mm) for mapping strains and phases in 1D or 2D. It is very (beam-) time consuming to produce 2D maps with high resolution. Due to flux limitations, the recording of three dimensional maps is very slow, in the order of many hours and days, and therefore not considered economical. More appropriate are linear depths scans and profile scans on parts of the sample that have been beforehand surveyed by neutron tomography. ENGIN-X is optimised for determining lattice parameters with high precision. The d-spacing range is rather limited and applications are therefore mostly restricted to metals.

Bragg Edge Transmission (ENGIN-X)

Bragg edge transmission analysis (Santisteban et al. 2001) is performed in a radiography type set-up. The technique uses a broad wavelength distribution of the incident beam, and an open beam geometry with a transmission detector behind the object. The method makes use of the fact that the transmission varies because neutrons are removed from the primary beam due to Bragg scattering. **Fig. 2b** illustrates the technique. Once the sample is inserted into the beam, neutrons are absorbed and scattered. The transmission detector measures a modified spectrum which contains the Bragg edges, which are drastic steps in the transmitted intensity distribution. The Bragg edges occur if the wavelength increases and for a given family of crystal planes the Bragg angle 2Θ equals 180 degrees. Beyond that wavelength these crystal planes don't diffract neutrons anymore and, as a result, the transmission increases. Bragg edges are equivalent to Bragg peaks. Bragg edge spectra can be indexed and analysed like diffraction patterns. The position of Bragg edges are related to lattice parameters of the crystalline phases in the object, and therefore can be used to identify the phases and to map strains. The magnitudes and shapes of the edges are related to the phase contents and the textures of the phases in the sample, respectively. By mapping the Bragg edge spectra, a 2D image of the spatial variations of the structural features are obtained. The Bragg edge transmission technique has been developed in collaboration between ISIS, Manchester University and Open University

(Santisteban et al. 2003). Bragg edge transmission is preferably performed with slow ('cold') neutrons in the wavelength range between 1-8 Angstrom wherein most metals have their first Bragg edge.

ENGIN-X has a neutron transmission detector with 100 pixels. It consists of a 10x10 array of 2x2 mm^2 scintillation detector pixels, arranged on a 2.5 mm pitch (distance between two pixels). The detector is directly placed behind the stationary sample in transmission, covering an area of 25x25 mm^2. This pitch defines the minimal spatial resolution of the set-up. The full spectral time-of-flight information (i.e. a Bragg edge spectrum) is available for each pixel of the detector, averaged over the thickness of the sample in beam direction. One measurement thus provides a map of 100 Bragg edge spectra (**Fig. 3b**), each of them can be analysed in terms of phases, strains and texture. Moreover, for each pixel the transmitted intensities can be averaged over all neutron energies in order to obtain integral attenuation values, as illustrated in the pixel matrix in **Fig. 3b**. Applications are restricted to objects with a small number of metal phases. The Bragg edge transmission technique was used to determine the strain distributions in archaeological bronze samples (Siano et al. 2006; Santisteban et al. 2006). Rotation of the sample in front of the Bragg edge transmission detector allows in principle to produce 3D phase maps, however this technique has not been applied so far.

Pencil beam scanning for crystallographic phase mapping (ROTAX)

Crystallographic phases in a three-dimensional object can be mapped on a conventional neutron time-of-flight powder diffractometer by scanning a sample in front of a collimated beam along three directions (x, y, z). The method, as it is illustrated in **Fig. 2c** for a 2D scan, uses a laterally collimated beam and diffraction signals to reconstruct the positions of phases. By contrast to the point-by-point method, there is no secondary collimator to confine the diffracting volume along the beam direction. Diffraction intensity from the whole illuminated path is recorded by one or more detector banks. For a 2D object, a collimated beam illuminates N equidistant positions (row scan). After rotation of the object by 90°, the scan is repeated (column-scan). In the TOF mode one obtains a full diffraction pattern for each sample position (**Fig. 3c**). Normalising each spectrum with respect to the sum over all spectra, diffraction signals above background are correlated in order to locate phases in terms of row- and column numbers. This is to say, that the methods evaluates the peaks intensities and ignores the peak shifts due to displacement out of the diffractometer centre (in contrast to the SXD method below).

Fig. 4 shows the imaging results for a 2D test object and a laterally collimated beam of 5 mm dimension: a 1 mm thick hollow aluminium cylinder of 40 mm diameter contains 2 iron and 2 copper rods of 5 mm diameter each. Row and column scans each consisting of 10 single acquisitions were evaluated using one or more distinctive Bragg peaks of the phases Cu and Fe, for instance, (211) at 1.17 Å for Fe and (200) at 1.8 Å for Cu. A 'pixel method' employs a flagging (0 or 1) of pixels if the phase under consideration is observed. The 'peak method' uses the Bragg peak heights to determine the probability for the presence of a phase in a particular pixel. Fig. 4 shows the reconstructed positions for both Cu and Fe rods for the two analysis methods. The Al peaks of the housing were not evaluated. The total acquisition time was about 2 hours. The apparently weaker presence of Cu in the left part of the crystallographic phase map is probably due to attenuation by the Cu rod on the right.

The scanning method can straightforwardly be extended for a three-dimensional object, using 2 90-degree rotations and 3 linear scans. This imaging approach does not require any conditioning of the diffracted radiation, hence it preserves the virtues of the experiment, namely the full detector coverage and the considerable d-spacing range and resolution. The resolution is limited by the size and divergence of the primary beam and is currently limited to 3 mm for practical applications. A disadvantage of the method is the requirement for two orthogonal rotations which can be difficult to achieve for museum objects.

Exploiting geometrical aberration for crystallographic phase imaging (SXD)

A different method to reconstruct the phase distribution was used on the single crystal diffractometer SXD at ISIS (Gutmann et al. 2006). The positions of the phases were derived from the shifts of diffraction peaks due to geometrical aberration, using a formula that is usually employed to correct for sample offset. A laterally collimated neutron beam is used for the 2D imaging. If the secondary flight path L_2 from sample to detector is short compared to the primary path L_1 of the incident neutrons (**Fig. 2d**) then the TOF shift Δt due to a longitudinal sample displacement x is:

$$\Delta t / t \sim x \cdot \cos^2(\theta) / L_2$$

(4)

In comparison to the previous, one scan-direction is elegantly replaced by the time-of-flight variable,

i.e. for a 2D object just one translational or rotational scan of the sample is required. Test measurements were carried out on the same 2D demonstration object of two crossed Cu and Fe bars inside an aluminium cylinder (**Fig. 4**). The incoming beam was collimated to 2x2 mm. The cylinder object was rotated about the vertical axis in 6°-steps to cover 180° (**Fig. 2d**). The data collection time per step was 30 minutes. **Fig. 3d** illustrates that a shift of a phase along the beam direction corresponds to a shift of diffraction peaks. Vice versa, by measuring the time-of-flight offset of Bragg peaks one derives the sample displacement along the beam direction according to equation 4. For the data analysis only a very narrow band of detector pixels around a scattering angle of 90 degrees was used. The reconstruction of the phases is directly obtained by plotting the rotation angle as a function of the sample displacement variable. The result of the scan, details of the data analysis, and the reconstructed map have been reported recently (Gutmann et al. 2006). The spatial resolution of the phase map for this pilot experiment was just below 5 mm. With smaller detector pixel sizes, a less divergent beam and a somewhat longer primary flight path, it is estimated that a resolution of 1-2 mm can be obtained with this imaging method.

Energy-selective radiography at a pulsed source (ENGIN-X + OSIRIS)

A rather new imaging technique combines the hardware used for conventional neutron radiography with the Bragg edge transmission characteristics of time-of-flight methods. The main component of an energy-selective radiography set-up is a digital neutron area detector system (Fig. 2e) (Vontobel et al. 2006). The time structure of the source allows collecting radiographies for many different neutron energies. It is well known that energy selection in neutron radiography provides a means for contrast enhancement and contrast variation by exploiting the Bragg edge effect (Kardjilov et al. 2003). By measuring radiographies at two wavelengths, slightly shorter and slightly longer than the Bragg edge (**Fig. 3e**), one can produce ratios of radiographies in order to achieve a contrast enhancement for a particular crystallographic phase. In the same way one can choose a particular pair of wavelengths that correspond to equal attenuation coefficients, and produce ratios in order to render a particular phase transparent.

The important aspect here is that, due to the relationship to Bragg edges, structural features produce contrast effects in the wavelength-dependent radiographies. With this technique microstructural variations can be mapped with an unprecedented sub-millimetre resolution, a spatial resolution that is otherwise only obtained by attenuation neutron tomography. The potential of mapping crystallographic texture of a material is a particular promising prospect. Preliminary results of direct imaging of structural features have been obtained with a light-intensified CCD camera system (Kockelmann et al. 2006c).

Discussion

In this paper, several diffraction imaging techniques have been presented, all of them based on the time-of-flight method. All five diffraction mapping schemes are capable of providing internal structural features of archaeological objects, properties that are difficult if not impossible to obtain by other methods if non-destructiveness is required. For all methods, the spatial resolution is low and counting times are very long, compared to conventional attenuation neutron tomographies. The required beamtime is therefore in the order of many hours for phase mapping, compared to minutes for conventional radiographies. The scattering techniques use signals emitted from the sample and are naturally weaker, also due to limited detector coverage. The transmission methods have much higher counting statistics, but since the information is averaged over the whole thickness of the objects, structural effects are much more difficult to disentangle. Scattering methods require scanning of the objects whereas transmission techniques can be applied to stationary objects for 2D maps which is a considerable advantage if precious and delicate museum pieces are to be analysed.

The main limitation of the diffraction imaging techniques is the resolution. The presently achieved spatial resolutions are adequate for a course surveying of the phases of objects but are far from providing microstructural information that are obtained by conventional microscopic and metallographic methods. Technically, the size of the incident neutron beam can be reduced but at the prize of much reduced neutron flux which makes the collection times even longer. Also, neutron beams cannot be focused to sub-millimetre spot sizes. Likewise, for the open beam geometries, the pixel size of a transmission detector can be reduced. But this is at the expense of counting statistics in the individual pixel spectra, which again renders the method little efficient in terms of acquisition times.

For all diffraction techniques, tomographic reconstruction as known for conventional neutron tomography is not required. The mapping is achieved directly by plotting a transmission or diffraction signal as a function of the scanning variable or pixel. On the other hand, software tools are not well developed, and data analysis is often a

lengthy and intricate hands-on process, what can be considered as a disadvantage.

Conclusions and future perspectives

Neutron tomography is ideal to produce contrast images of the inner parts of complex engineering and archaeological artefacts. The attenuation maps, however, do not provide information of chemical and structural compositions of the contrast features. Supplementary diffraction analyses are therefore desirable if not necessary in order to identify the materials and to deduce information about working techniques. Hence, neutron diffraction is the ideal complementary technique to X-ray and neutron tomography. Neutron diffraction techniques will always have a disadvantage compared to attenuation tomography if high spatial resolution is required. Imaging of a complete object by neutron diffraction is in many cases not the first choice of analysis. A complete and big object is much faster surveyed by tomography, which then can be followed by neutron diffraction analysis of some parts where contrast variation was observed. In many cases a single linear scan or a 2D mapping may be sufficient. Neutron diffraction mapping can be of much benefit to extrapolate the results from a single-point conventional (destructive) analysis to other points of the object. Destructive or micro-destructive analysis may in many cases be required for obtaining the detailed microstructure with high resolution. The diffraction analysis, once calibrated with the conventionally acquired information, can be used to survey the object for inhomogeneities and structural variations.

Neutron diffraction imaging methodologies and experimental capabilities are underdeveloped but they are now rapidly improving. Since the discussed diffraction mapping techniques lack high spatial resolution, an imaging in the tomography sense is therefore not achievable at present. But with the advent of new intense neutron spallation sources elsewhere, there are opportunities to realize improvements of both resolution and scanning times of the diffraction imaging techniques.

Acknowledgements

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