New neutron imaging techniques to close
the gap to scattering applications

Eberhard H. Lehmann, S. Peetermans, P. Trtik, B. Betz, C. Grünzweig
Neutron Imaging & Activation Group, Laboratory for Neutron Scattering & Imaging,
Paul Scherrer Institut, CH-5232 Villigen PSI
eberhard.lehmann@psi.ch

Abstract. Neutron scattering and neutron imaging are activities at the strong neutron sources which have been developed rather independently. However, there are similarities and overlaps in the research topics to which both methods can contribute and thus useful synergies can be found. In particular, the spatial resolution of neutron imaging has improved recently, which - together with the enhancement of the efficiency in data acquisition - can be exploited to narrow the energy band and to implement more sophisticated methods like neutron grating interferometry. This paper provides a report about the current options in neutron imaging and describes how the gap to neutron scattering data can be closed in the future, e.g. by diffractive imaging, the use of polarized neutrons and the dark-field imagining of relevant materials. This overview is focused onto the interaction between neutron imaging and neutron scattering with the aim of synergy. It reflects mainly the authors’ experiences at their PSI facilities without ignoring the activities at the different other labs world-wide.

1. Introduction: Neutron imaging vs. neutron scattering

Although the most prominent and powerful neutron sources for research are still dominated by neutron scattering devices world-wide, promising neutron imaging activities and installations have been started and completed in the recent years with success [1, 2].

Modern neutron imaging techniques are exclusively based on digital imaging detection systems which deliver a very high efficiency in data acquisition and enable performing advanced investigations (i.e. not only those based on the simple transmission through objects). This paper will describe some of the recent methodical improvements and developments where we identify a strong link to neutron scattering approaches, such as neutron diffraction, small-angle neutron scattering or neutron reflectometry.

Neutron Imaging is well established on the macroscopic length scale. A quasi-parallel beam is used to illuminate the sample. The highest spatial resolution is on the order of ~ 10 µm, some attempts goes for even higher resolution [3]. In the radiography mode, the direct beam produces a “shadow” image of the sample, thus integrating the entire thickness of the sample in beam direction. The parallel beam geometry is preferred compared to a divergent one with a micro-spot source, what is technically impossible with neutrons. The beam is observed by 2D planar pixilated detection systems of different dimensions and with different pixel size.

The scattering of interacting neutrons is considered in neutron imaging experiments as their removal. The scattered component towards outside the sample is often ignored – it is seen to be more a perturbation in the signal processing and in the quantification of the sample content.

Thermal and cold neutrons are preferred due to the high contrast variation among the different sample materials, their high detection efficiency and high achievable beam intensity. In most cases, the full energy spectrum (white beam) is applied.
Neutron imaging is isotope-sensitive (e.g. B-10/11; Li-6/7; He-3/4; H-1/2; U-235/238) which can be used in dedicated studies to tune the contrast according to the setup conditions (water in fuel cells, nuclear fuel enrichment, Li-battery studies, etc.)

Neutron imaging methods are very complementary to X-ray imaging ones (even if the underlying nuclear processes are completely different) and we can obtain similar performance in all respects (in particular image quality), while a synergy between both (data fusion) is of high interest in several research fields [4].

In the meantime, even advanced methods similarly to the X-ray ones have been developed and established (tomography on different length scales, neutron grating interferometry, phase-contrast imaging, real-time imaging, …).

Unfortunately, there are only a few neutron sources available where these new approaches have been installed and implemented into the user programs [13].

**Neutron Scattering** does not utilize the direct beam in forward direction, but takes care for the scattered components around the sample. Often the initial beam is tuned with respect to the applied energy band and the beam collimation onto the sample. The scattered neutrons are measured mainly by means of counting devices and high background sensitivity is often a problem.

This scattered component delivers information about the microscopic, atomic and molecular properties of the investigated materials (under different, even extreme conditions) like structure, lattice distances, phase transition, particles size and even quantum phenomena.

There is similarity to X-ray scattering, but the neutron methods are preferred for bulk materials, higher mass numbers and in particular magnetic sample behavior.

We can distinguish different classes of settings: diffraction, small angle neutron scattering (SANS), reflectrometry, spectroscopy (inelastic scattering) resulting in highly specialized facilities.

The related length-scale where information can be derived is on the order of nano-meters to micro-meters (Fig. 1). The real overlap is on the length scale for USANS vs. high resolution imaging and methodically for diffractive imaging [10].

The interpretation of the scattering data requires models and is therefore in all cases an indirect approach. The fitting between theoretical and experimental data is indicative how well a proposed system relates to reality.

![Application domains of neutron scattering and neutron imaging](Image)

**Fig. 1:** Application domains of neutron scattering and neutron imaging, respectively, with the intention of overlap and synergy (courtesy M. Strobl, ESS); results are from different studies and shown here only symbolically without references.
2. State of the art in Neutron Imaging
As stated above, imaging facilities are operated mainly with thermal and cold neutrons. Fast neutron imaging is a niche, but some applications (bulk materials studies) are very successful [5]. The time resolution can be on the order of 25 ms/frame, depending on the beam intensity and detector performance. In stroboscopic mode, 10,000 rpm have successfully been verified and clear quasi-static images were obtained.

The spatial resolution of user facilities is now on the order of about 10 µm at several facilities and the approach towards 1 µm is still a challenge, but about 5 µm resolution can be achieved using highly efficient scintillator screens (see e.g. [3, 12]).

Energy resolved imaging is a new approach, in particular in the cold range where a wavelength resolution between 10% and 2% can be achieved, depending on the selection device. The use of the time-of-flight options at pulsed sources will deliver a much higher flexibility with this respect [2].

Imaging with polarized neutrons has been implemented and tested at some places and interesting results were obtained for magnetic structures and phenomena – on the macroscopic scale [6]. This method will be further improved and made user capable.

Grating interferometry for getting phase contrast and dark field information about the setup has been developed in 2005 [8] and established at PSI since 2008 and is now on the level of a routine user device for the study of magnetic domain structures and of small-angle phenomena.

With the implementation of X-ray devices into neutron imaging beam lines it becomes possible to perform neutron/X-ray referencing on the pixel/voxel level. With such installations it becomes possible to enhance features or to distinguish material phases that cannot be discerned easily by other methods.

3. New approaches
Albeit this impressive methodical progress, there is further potential for improvement in order to extract even more information from the neutron experiments. Since neutrons have the ability to penetrate even large samples of some high-Z materials, while obtaining a profound contrast for hydrogenous materials. Materials like Pt, Pd, Pb or W are not accessible at all with X-rays for thicker layers.

The new approaches are mainly focussed on the study of the neutron scattering features next or in addition to the transmission data. For this purpose, the setup and conditions for the investigations have to be modified and sophisticated compared to the traditional radiography approach.

![Fig. 2: Neutron attenuation cross-sections of crystalline structural materials – idealized case for polycrystalline powder samples; the Bragg edge structure caused by the elastic scattering at the involved lattice planes is clearly visible – and can be used for imaging purposes](image-url)
3.1. Energy selective Neutron Imaging

As shown in Fig. 2, most structural crystalline materials behave with characteristic structures in their attenuation cross-sections due to the elastic scattering at crystalline lattice planes of their grains. Within a randomly oriented micro-grain distributed sample (quasi-powder) all possible lattice planes contribute determined to the transmission, ending up with the shown energy-dependent material attenuation. In order to study specifically some individual lattice plane orientations needs a reduction to the related energy band – to perform energy selective imaging.

Already with a relatively coarse energy resolution (about 10% in $\Delta \lambda / \lambda$), significant differences in the grain size distribution can be observed. As shown in the example of a rolled Al sample in Fig. 3[14], the different layers and individual grains become visible with the energy selective imaging approach.

The advantage of this method is the observation of the complete sample in one shot with the inherent spatial resolution of the setup (e.g. 0.05 mm). Compared to a real diffraction study where one can get the full crystalline information at the measurement position, deviations within the sample can be determined easily herewith.

Some additional features of energy selected imaging are summarized in Fig. 3: variation in contrast, phase separation, texture and even strain determination become possible when the right conditions are applied.

Fig. 2: Inspection of an 10 mm thick Al plate with weld, photo (left) “white beam” imaging (middle) and energy resolved methods (right); in this way, the grain size distribution and the different grain orientation can be visualized and measured [14]

Fig. 3: Information derived from studies in energy selective mode near Bragg edges of structural materials [15]; higher cross-sections: better detectability; lower cross-sections: higher transmission probability
3.2. Neutron Grating Interferometry: the dark field image

Similar to X-ray imaging research, neutron grating interferometers (nGI) [7] are available to derive information about phase effects and the so-called “dark field”, a composite data set of the investigated sample derived from the reduced amplitude in their sinusoidal inference pattern. This signal has been found sensitive for particle distributions on the length scale of a few micro-meters, the common domain of SANS and USANS.

However, since SANS describes the whole sample in a global/averaged way, the observation with the nGI delivers pixel-wise quantitative information with the same pixel size as the transmission image data.

This method has been used to derive detailed information about magnetic domain structures in bulk samples [8] and for polymer particles in solution [9]. Here, we show the example of three copper based alloys with some lead precipitations embedded. As visible in Fig. 4, there is no much attenuation contrast difference among the 3 materials, but much deviation in the dark-field data. This behaviour can clearly be explained by the scattering on the Pb precipitations, what was verified with destructive standard inspections.

![Transmission image (TI) and Dark-field image (DFI)](Fig. 4)

**Fig. 4:** Comparison of the transmission image data (left) of step-wedges of three different copper alloys with the corresponding dark-field signals (right); even only small lead precipitations are added to the brass materials, a strong effect to the dark-filed signal is induced.

3.3. Diffractive neutron imaging

Neutrons get diffracted in relevant materials at the lattice planes of the crystalline structures according to Bragg’s law. This behavior is used in neutron diffraction experiments to determine the crystal parameters such as lattice plane distances, orientation and scattering strength. Because the diffracted components are often much smaller than the transmitted one and the diffracted neutrons are sent in many directions, the data acquisition is time consuming.

If the samples are single crystals, the whole material has well-defined orientations and the diffracted neutrons from the whole sample can be observed in one determined direction at ones. In this way, the diffracted component can be used to investigate, next to the above mentioned main parameters, the homogeneity of the crystal with respect to a specific lattice plane.

Compared to a typical diffraction experiment (single-crystal or powder diffractometers), where only parts of a sample are under investigation, the whole object is studied (if the beam size and the field of view for the detector allow).

Fig. 5 shows the setup of diffractive imaging with a second imaging detector aside from the beam direction just where the diffracted component is expected. This configuration can be tuned either by the sample orientation or for a different relevant position of the second detector. Although the most investigations have been performed under white beam conditions with success (different spots occur
when the sample is rotated) or in energy-selected mode, where only one orientation is enhanced and the other contributions are suppressed.

The study of relevant samples has been performed by means of turbine blades from helicopters which were declared to be single-crystalline [10]. However, it has been demonstrated in the studies that this is only true in first order. Along the growing direction a preferred orientation has been found in the diffracted data as shown in Fig. 6. In this way, diffractive imaging is an elegant and relatively easy and straightforward method for material characterization even of bulk material samples.

Fig. 5: Setup for diffractive imaging with white beam and by using a second imaging detector aside; in forward direction the transmission image is obtained (see Fig. 6 middle); sideward image = diffracted component (see Fig. 6 right)

Fig. 6: Photo (left), transmission image (middle) and diffracted image (right) of a small single crystalline turbine blade; the preferred orientation in the crystal growing direction is clearly visible

3.4. Imaging with polarized neutrons

Neutrons are available in two different spin-states (up and down) since the nuclear spin is \( \frac{1}{2} \). As the spin is coupled inversely to the magnetic moment (\( \mu = -1.913*\mu_B \)), magnetic interactions with matter happens and are measurable.

Since the commonly used neutron beams have no preferred spin orientation it needs a separation process in a polarizer device to sort out preferentially one of them. In this manner, at least 50% of the beam intensity is sacrificed. The remaining polarized component can be used to investigate magnetic phenomena, materials and structures on the macroscopic scale with imaging methods. In particular, the depolarization of the neutrons in such processes is under investigation [6]. Since the spin rotation in magnetic fields is energy dependent, a certain energy selection is needed. With the PONTO instrument at BER-II (HZB) [11] a dedicated instrument was built in order to study effects of super-conductivity, complementing the research with neutron scattering methods which are focused onto the micro-scale.
3.5. Refractive and reflective imaging

Considering the neutron properties with respect to their wave behaviour, reflection and refraction can be interesting aspects for investigations. Such properties are used in neutron reflectometers in order to characterize the surface structure of solids and liquids.

In neutron imaging applications, the behaviour at edges of different materials and under certain conditions (sample-detector distance, neutron wavelength, geometry, and material combination) can be studied with high-resolution imaging detectors. Reflection and refraction can clearly be distinguished, as shown in Fig. 7.

![Fig. 7: Image and horizontal profile of a glass plate (2 mm thick) with mirroring coating tilted by 0.4° towards the beam direction: the reflected beam part and the refracted neutrons at the left outer edge becomes visible clearly and independently as two separate effects of the material.](image)

For the interpretation of the findings, the known relations for the refractive neutron index and the condition for total reflection can be taken into account [15]:

\[
\cos \theta_c = n \quad \text{with} \quad \theta_c \approx 0.1^\circ/\AA
\]

and

\[
n = 1 - \frac{\lambda^2 \cdot N \cdot b_c}{2\pi} = 1 - 10^{-6}
\]

(1)

and

(2)

where the scattering length \( b_c \), the nuclear density \( N \) and the wavelength \( \lambda \) have to be taken into account.

The imaging aspect for such kind of investigations is of high relevance since the whole setup is under control and imaged in high resolution (e.g. 0.02 mm pixel size) while the conditions can be changed and adapted. On request, the effects can be suppressed or enhanced and the visibility of edges or internal structures be tuned.

4. Applications and user demands

The new neutron imaging options enable improved material studies on the macroscale with feedback and information about the micro-structure. There is a clear demand for higher spatial resolution and time-dependent tomography, which will be the aim of further development. A new level of performance is already reached within the neutron microscope project [3].

7
Further combination of imaging and scattering devices are foreseen, e.g. at ESS (ODIN, HEIMDAL, BEER) and at ISIS (IMAT). The data interpretation needs the knowledge in both fields with strong overlap with simulations (MacStas tools).

5. Conclusions and outlook
The new imaging techniques bridging the range for neutron scattering have been implemented (at several places) already.
Most of them are already on the status of user instruments.
With the access to TOF options (at pulsed spallation sources) the energy resolution can further be tuned and the methods be improved.
The synergy between neutron imaging and scattering should further be enhanced in the practical work and within facilities interfacing both techniques. It will be important in the future to create a closer network between the different neutron imaging facilities and teams, involving also the aspects and options from the neutron scattering side.

References
[1] E.H. Lehmann, S.Peetermans, B. Betz, Instrumentation in neutron imaging – A world-wide overview, Neutron News, Vol. 26, Issue 2, p. 6-10
[2] T. Shinohara, T. Kai, Commissioning start of energy-resolved neutron imaging system, RADEN in J-PARC, Neutron News, Vol. 26, Issue 2, p. 11-14
[3] P. Trtik et al., Improving the spatial resolution of neutron imaging at Paul Scherrer Institut – The Neutron Microscope Project, Physics Procedia 2015, p.
[4] P. Vontobel et al., The X-ray option at the NEUTRA imaging beamline of the spallation neutron source SINQ, submitted to NIM A (2015)
[5] J.M. Bellosta von Colbe, G. Lozano, O. Metz, et al., Design, sorption behaviour and energy management in a sodium alanate-based lightweight hydrogen storage tank, International Journal of Hydrogen Energy Volume: 40 Issue: 7 Pages: 2984-8
[6] N. Kardjilov et al., Three-dimensional imaging of magnetic fields with polarized neutrons, Nature Physics 4, 399 - 403 (2008)
[7] B. Betz, C. Grünzweig, E. H. Lehmann, Advances in Neutron Imaging with Grating Interferometry, Material Evaluation, Vol. 72, Nr. 2, pp. 491-496
[8] C. Grünzweig, C. David, O. Bunk, M. Dierolf, G. Frei, G. Kühne, J. Kohlbrecher, R. Schäfer, P. Lejcek, H. Ronnow, and F. Pfeiffer, “Neutron decoherence imaging for visualizing bulk magnetic domain structures”, Phys. Rev. Lett. 101, 025504 (2008)
[9] B. Betz, R. P. Harti, M. Strobl, J. Hovind, A. Kaestner, E. Lehmann, H. Van Sloyen, and C. Grünzweig, Quantification of the sensitivity range in neutron dark-field imaging”, submitted to “Review of Scientific Instruments” (2015)
[10] S. Peertemans and E. H. Lehmann, Simultaneous neutron transmission and diffraction contrast tomography as a non-destructive 3D method for bulk single crystal quality investigations, J. Appl. Phys. 114, 124905 (2013); http://dx.doi.org/10.1063/1.4823741
[11] W. Treimer, Radiography and tomography with polarized neutrons, Journal of Magnetism and Magnetic Materials,Volume 350, January 2014, Pages 188–198
[12] P. Trtik, E.H.Lehmann, Isotopically-enriched gadolinium-157 oxysulfide scintillator screens for the high-resolution neutron imaging, NIM A: Volume 788, 11 July 2015, Pages 67–70, doi:10.1016/j.nima.2015.03.076
[13] http://www.isnr.de/index.php/facilities/user-facilities
[14] E.H. Lehmann, S. Peertemans, L. Josic, H. Leber, H. Van Sloyen, Energy-selective neutron imaging with high spatial resolution and its impact on the study of crystalline-structured materials, NIM A Volume 735, Pages 102-109
[15] S. Peertemans, Energy-selective neutron imaging for materials science, PhD Thesis 6515 (2014), EPFL, Lausanne