Recent progress of pulsed neutron imaging in Japan

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Characteristics of spectroscopic imaging using a pulsed neutron source

Traditional neutron radiography: Contrast

- Engine
- Lily
- Fuel cell

Spectroscopic imaging (diffraction like)

We can get image showing the crystallographic characteristics of a sample.

Lattice spacing image of a single crystal like Nb sample
Principle of pulsed neutron imaging and analysis of the pulsed neutron transmission data
Experimental setup for spectroscopic imaging at a pulsed source and obtained information

\[ \Sigma_{tot}(\lambda) = -\frac{\ln\left(\frac{I(\lambda)}{I_0(\lambda)}\right)}{b} \]

- **b**: Sample thickness
- **\lambda**: Neutron wavelength

(Bragg edge analysis) + (2D-detector)

Position dependent crystal and texture information
Rietveld-type Transmission-Spectrum analysis code: RITS    (by H. Sato)

Transmission spectra

\[ \text{Tra}(\lambda) = \exp \left[ - \sum_p \sigma_{\text{tot},p}(\lambda) \rho_p t_p \right] \]

\[ \sigma_{\text{tot}}(\lambda) = \sigma_{\text{coh}}^{\text{el}}(\lambda) + \sigma_{\text{inh}}^{\text{el}}(\lambda) + \sigma_{\text{coh}}^{\text{inel}}(\lambda) + \sigma_{\text{inh}}^{\text{inel}}(\lambda) + \sigma_{\text{abs}}(\lambda) \]

\[ \sigma_{\text{coh}}^{\text{el}}(\lambda) = \frac{\lambda^2}{2V_0} \sum_{d_{hkl}=0}^{2d_{hkl}<\lambda} \left| F_{hkl} \right|^2 \times d_{hkl} \times P_{hkl}(\lambda, d_{hkl}) \times O_{hkl}(\lambda, d_{hkl}) \times E_{hkl}(\lambda, F_{hkl}) \]

\( V_0 \): Volume of a unit cell

\( d_{hkl} \): Lattice plane distance of \((hkl)\) plane

\( F_{hkl} \): Crystal structure factor

Profile function near the edge (Lattice space)

March-Dollase function (Preferred orientation)

Extinction function (Crystallite size)
Examples of evaluation of crystallographic characteristics of materials by pulsed neutron transmission
Experimental setup at Hokkaido linac and BL10 at J-PARC

**HOKKAIDO LINAC**

~$10^{12}$ n/s

Coupled moderator

Evacuated beam tube

$10\text{cm} \times 10\text{cm}$

~7 m

$B_4C$ collimator

Sample

B shield

256 pixel $^6\text{Li}$-glass or GEM detectors

**J-PARC**

~$10^{16}$ n/s @100kW

Decoupled moderator

Evacuated beam tube

$10\text{cm} \times 10\text{cm}$

$7.05\text{m}$

$B_4C$ collimator

$7.27\text{cm} \times 7.27\text{cm}$

$8.49\text{cm} \times 8.49\text{cm}$

$12.755\text{m}$

$14\text{m}$

Sample

B shield

256 pixel $^6\text{Li}$-glass or MCP detector
Aim: to observe the change of preferred orientation and crystallite size between welded and original areas.
Hokkaido electron linac neutron source

Cold neutron source
GEM detector

- Pixel size: 0.8mm x 0.8mm, 14400 (120 x 120)
- Effective area: 10 cm x 10 cm
- Counting rate: >1MHz/Detector
- Time stamp: ~20ns

GEM detector developed by S. Uno (KEK)
{110} orientation analysis (ND) at outside of the welded region

Rietveld refinement parameters obtained by fitting

- Degree of anisotropy (March-Dollase coefficient) $r = 0.54$
- Preferred orientation $A = <111>$ along ND ($r < 1$)
- Surface density $\rho \times t = 4.79 \times 10^{22} \text{ cm}^{-2}$
- Crystallite size $S = 4.65 \mu m$
{110} orientation analysis (RD) at outside of the welded region

Rietveld refinement parameters obtained by fitting

Degree of anisotropy (March-Dollase coefficient) \( r \) : 0.65
Preferred orientation \( A \) : \(<110>\) along RD (\( r < 1 \))
Surface density \( \rho \times t \) : \(4.90 \times 10^{22}\) cm\(^{-2}\)
Crystallite size \( S \) : 6.09 \(\mu\)m
**Anisotropy**

Degree of preferred orientation
(March-Dollase coefficient)

Pole figures extracted from a Bragg edge transmission

![Diagram](image_url)
Preferred orientation

Preferred crystal orientation parallel to the beam direction, \(<HKL>\)

Position \(y / \text{cm}\)

Position \(x / \text{cm}\)

\(<111>\) \(\gamma\)-fiber
\(<110>\) \(\alpha\)-fiber
\(<100>\)
\(<221>\)
\(<211>\)
\(<210>\)
Crystallite size

We observed the changes clearly.
b. Quenched iron rod


Aim: to confirm the quenched region, 3 mm.

Sample: Quenched iron
Diameter: 26mm
Thickness: 20mm
Quenched thickness: 3mm
MCP detector with Medipix/Timepix readout

MCP detector developed by Dr. A. Tremsin at UC Barclay.

- 256 x 256 array of 55 μm pixels
- 100 kHz/pxl
- Frame rate: 1-3 kHz
- Low noise (<100e⁻) = low gain operation (10 ke⁻)
- ~1 W watt/chip, abuttable
- Developed at CERN

Stack of MCPs is placed above Medipix2/TimePix readout

Muros readout electronics (NIKHEF) and PixelMan software (IEAP, CTU Prague) with Labview and C++ plugins (UCB)
NOBORU beam line at J-PARC

- DNA (protein dynamics, JAEA)
- HI-SANS (Nano structure, JAEA)
- Reflectometer (KEK)
- Proton Beam
- High Reso. Chopper (KEK)
- AMATERAS (low E chopper, JAEA)
- Muon port for μSR (KEK)
- TAKUMI (stress analysis, JAEA)
- Bio-Molecule Diff. (Ibaraki Pref.)
- 4-SEASONS (JAEA, KEK, Tohoku U)
- Versa. Total Scatt. Diff. (KEK)
- Versa. Powder (Ibaraki Pref.)
Transmission image obtained by whole neutrons
More detailed image of transmission around 4.5 Å

We observed an enhanced image of the boundary between quenched and un-quenched regions.
Transmission spectra

Distance from sample center

Sample center

edge

Transmission spectra for different distances from the sample center.
Lattice spacing at the quenched area is larger than the un-quenched one.
c. In-situ tensile test
(strain, texture, crystallite size of a iron plate during tensile test)

Aim: to see change of the crystallite size, anisotropy, strain depending on load, and check reliability of strain obtained by the transmission.

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‘TAKUMI’ beam line at J-PARC

DNA (protein dynamics, JAEA)

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Varsa. Total Scatt. Diff. (KEK)

Versa. Powder (Ibaraki Pref.)

TAKUMI (stress analysis, JAEA)
Setup for measurements under the tensile test

(1) Bragg edge transmission
- Pulsed neutron beam
- Slit: 4mm width, 4cm height
- Spatial res.: 3mm × 3mm
- Sample-detector: 2m
- Meas. time: 5min/meas.

(2) Diffraction and mapping
- Pulsed neutron beam
- Slit: 4mm width, 4cm height
- Slit (mapping): 4mm × 4mm
- Sample-detector: 2m
- Meas. time: 3hrs/meas. (2min/pos.)
Photos of experimental setup

Sample (5mm thick α-Fe with notches)

Tensile test machine

Sample setup

Slit (B$_4$C)
Macroscopic strain measured by a strain gauge

![Graph showing macroscopic strain measurements](https://via.placeholder.com/150)

- **No load**: 
  - Strain: -14 με

- **10kN**: 
  - Strain: 167 με

- **20kN**: 
  - Strain: 235 με

- **25kN**: 
  - Strain: 264 με

- **30kN**: 
  - Strain: 293 με

- **32.5kN**: 
  - Strain: 314 με

- **37.5kN**: 
  - Strain: 276 με

- **40kN**: 
  - Strain: 388 με

- **49kN**: 
  - Strain: 511 με

- **Load released**: 
  - Strain: -116 με
Change of Bragg edge shapes at three loads

(1) Near notch

(2) Center area

(Expanded)
The strain data showed in the following figures were calculated by an assumption of the lattice spacing data obtained here to be $d_0$. 

March-Dollase coefficient

Crystallite size

Lattice spacing \{110\} 

\[ d \text{-spacing of } \{110\} \text{ crystal-lattice-planes (nm)} \]

\begin{align*}
\text{Position } y / \text{ cm} & \quad 1.8 \\
\text{Position } x / \text{ cm} & \quad 0.0 \quad 1.05 \quad 2.1
\end{align*}

\[
\begin{array}{c}
0.0 \\
0.20241 \\
0.20241 \\
0.20244 \\
0.20244 \\
0.20247 \\
0.20247 \\
0.20250 \\
0.20250 \\
0.20253 \\
0.20253 \\
0.20256 \\
0.20256
\end{array}
\]

(10^{-5} \text{ nm/div} \sim 50 \text{ \mu e/div})
March-Dollase coefficient

Crystallite size

Strain \{110\}
Macrostrain of \{110\} crystal-lattice-planes
(\(\mu e = 10^{-4} \% = 10^{-6}\))
+ 325
- 725
(50 \(\mu e/\text{div}\))

Position y / cm
0.0 1.8

Position x / cm
0.0 1.05 2.1

Macroscopic strain obtained by a strain gauge
March-Dollase coefficient

Crystallite size

Strain \{110\}

Macrostrain of \{110\} crystal-lattice-planes 
($\mu e = 10^{-4} \% = 10^{-6}$)

25 kN

Macroscopic strain obtained by a strain gauge
March-Dollase coefficient

Crystallite size

Strain \{110\}

Macrostrain of \{110\} crystal-lattice-planes

(\mu e = 10^{-4} \% = 10^{-6})

Position y / cm

0.0 1.8

Position x / cm

0.0 1.05 2.1

Macroscopic strain obtained by a strain gauge
March-Dollase coefficient

Crystallite size

Strain \{110\}

Macrostrain of \{110\} crystal-lattice-planes \((\varepsilon = 10^{-4} \% = 10^{-6})\)

Macroscopic strain obtained by a strain gauge
The crystallite size become smaller over the sample at this load.
After releasing load

March-Dollase coefficient

Crystallite size

Strain \{110\}

Macrostrain of \{110\} crystal-lattice-planes
\( (\mu \varepsilon = 10^{-4} \% = 10^{-6}) \)

Position \( y / \text{cm} \)

1.8
0.0

Position \( x / \text{cm} \)

0.0 1.05 2.1

3.6

Macroscopic strain obtained by a strain gauge

The strain was released and crystallite size was kept small over the sample.
The compression strain data obtained by the diffraction sit in the range of the transmission data. The trend is almost the same.

The diffraction data are average value over elastic and plastic deformation areas.
Magnetic field imaging using neutrons

We detect spin ration in a magnetic field due to precession.

Rotation angle depends on the neutron speed (the wavelength) and path integration over the magnetic field.

Spin rotation corresponds to polarization.

\[ \frac{d}{dt} S_\alpha(t) = \frac{g \mu_N}{\hbar} [S(t) \times B(t)]_\alpha, \quad \alpha = x, y, z \]

Rotation angle due to precession

\[ \varphi = \omega_L t = \frac{\gamma_L}{\nu} \int_{\text{path}} B ds \]

\[ \omega_L = \frac{g \mu_N}{\hbar} B = \gamma_L B \]
Test experiment for magnetic field imaging using a solenoid coil

Beam line: BL10 at J-PARC
Polarizer/ analyzer: Bended magnetic mirror
    + Solid collimator (HMI)
Spin flipper: AFP type
    (AC field: 173kHz, ~20G)
Sample: solenoid (Outer diam. 5mm)
Detector: 5 inch-RPMT
(Schintilator: ZnS(Ag)/6LiF 0.25mm)
    (\(\Delta x, \Delta y = 0.5\text{mm}, \Delta t = 0.5\text{msec}\))
Beam size: 20mm(H) x 10mm(W)
Measuring time: \(~ 30\text{ min. (50000 pulses)}\)

Spin flipper OFF (N-) and
Spin flipper ON (N+)

Monitor slit Polarizer spin flipper solenoid coil analyzer collimator detector
By fitting wavelength dependence of polarization we can get information on $BL$.

$$\frac{P_i}{P_0} \propto \cos \phi$$

$$\phi = \gamma Bt = \gamma B \frac{L\lambda}{3956}$$

$B = 2.2 \text{mT/A}$
Summary

1. The pulsed neutron transmission method can give crystallographic information such as crystallite size, preferred orientation, strain, phase transition, magnetic field and so on for a thick (bulk) sample over the wide area.

2. The data analysis code ‘RITS’ was developed, and it has been proved to be very useful for deducing quantitative data.

3. The improvement in the experimental method and in the analysis code is required to give more precise data and expand the application field.