

中性子ビーム利用基盤技術開発

加倉井和久

日本原子力研究開発機構 量子ビーム応用研究部門

1932年、Chadwickによる中性子の発見は、一方で、原子核物理の急速な発展と共に原子力技術や近代素粒子物理の創成を促した。他方で、短期間の間に中性子の質量、スピン、中性子磁気モーメント、中でも電荷が存在しないなどの中性子の本性が明らかにされると、中性子こそが物性物理（Condensed Matter Physics）のナノ構造を研究する為の理想的な道具であることが認識された。実際1950年以後、原子炉や加速器技術の急速な発展によって、研究用原子炉をはじめとする大強度の中性子ビーム源の施設が建設されて、中性子散乱による物性物理・化学の研究が一挙に花開いた。その後高束定常中性子源施設の建設、加速器を用いたパルス中性子源の開発、中性子ビームデバイスや検出技術の開発により、中性子利用は従来の固体物理のみではなく、ソフトマターを含む広い意味での物性研究、産業利用を含む材料研究、タンパク質の構造やダイナミクス等を対象とするバイオ研究において重要な観測子としての利用が拡大して来た。しかしこのように利用研究の領域が広がれば広がるほどに、定常中性子源或はパルス中性子源のみを所有する中性子利用研究施設では幅広い基礎研究や産業利用を含む利用研究からの需要に十分答えられる質的及び量的中性子データ収集及び解析力の実現が困難である事が明らかになって来た。

幸いに我が国では日本原子力研究開発機構 東海研究開発センター 原子力科学研究所の敷地内に世界でも稀な定常中性子源施設（JRR-3）とパルス中性子源施設（J-PARC/MLF）を備え持つ中性子量子ビーム利用環境の基盤が整った。この両中性子源の複合的ビーム利用の高度化を目標に、文部科学省の量子ビーム基盤技術開発プログラムの枠組みの中で高度化ビーム技術開発課題として「中性子ビーム利用高度化技術の開発」が採択され、偏極中性子、集光、検出技術を柱とするビーム利用技術の基礎基盤技術開発が行われている。

本講演ではこのプログラムの枠組みの中で得られた技術開発の成果を中心に、中性子ビーム技術の最新動向及びこれらの基盤技術を活用することにより可能となる新機能材料や創薬開発に向けた中性子利用研究の例を紹介したい。

文部科学省 量子ビーム基盤技術開発プログラム「中性子ビーム利用高度化技術の開発」は平成20年度より幹事機関の日本原子力研究開発機構とネットワーク型研究開発拠点の北海道大学、東北大学、高エネルギー加速器研究機構、東京大学、京都大学との共同研究として行われております。各機関の研究代表者及び研究協力者の方々に謝意を表します。

東北大学多元物質科学研究所-九州シンクロトロン光研究センター 合同シンポジウム
2012.7.30 仙台

中性子ビーム利用基盤技術開発 Neutron Beam Fundamentals Development

Kazuhsa Kakurai
Quantum Beam Science Directorate (QuBS)
Japan Atomic Energy Agency (JAEA)

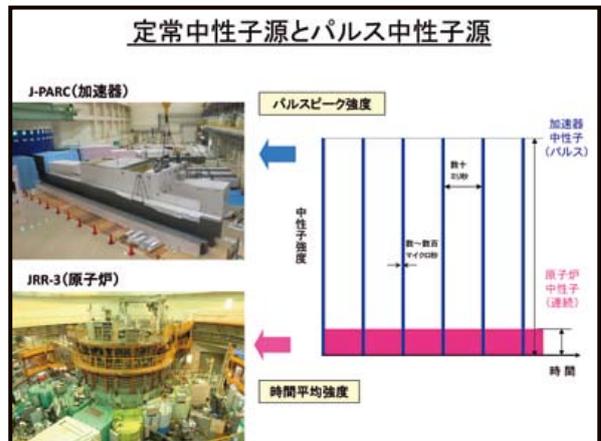


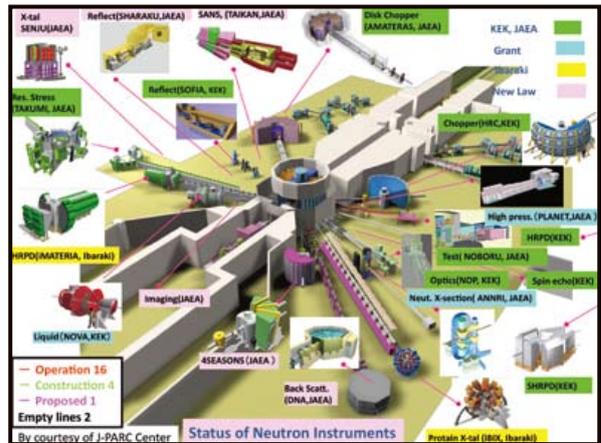
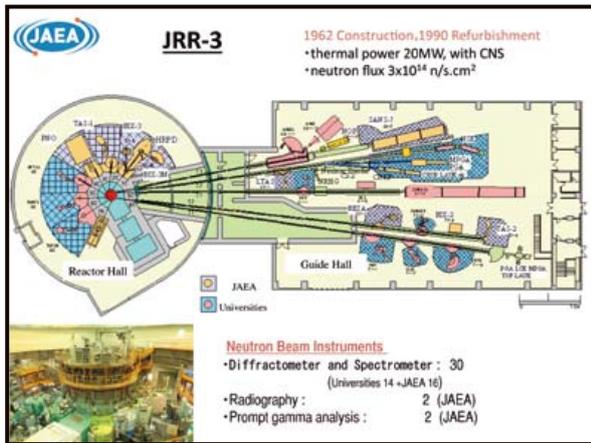
Outline

- Neutron Beam Fundamentals Development
- Development concerning the utilization of polarized neutrons
- Development of focusing devices
- Development of detecting devices

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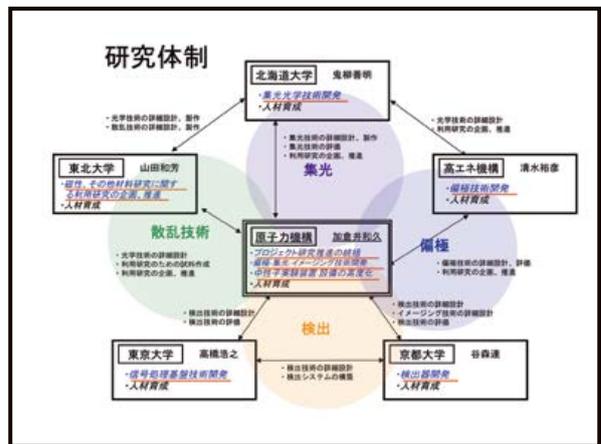


Development of Neutron Beam Techniques at JRR-3

- Development of neutron imaging plate
- Extreme sample environments (low T, high H, high P)
- SANS upgrade
- Development of neutron optics (magnetic lens, mirror, detector)
- Polarized neutron upgrade (higher flux, 3-D polarization analysis)
- Installation of reflectometer
- LTAS upgrade
- High energy transfer (Cu monochromator)

MEXT Quantum Beam Technique Development Program
 "Development of Neutron Beam Fundamentals Techniques"

Utilization of polarized neutrons e.g. ³He polarization filter
 High precision focusing device
 High resolution detection device



MEXT Quantum Beam Technique Development Program
 "Development of Neutron Beam Fundamentals Techniques"

Subject: Development and Application of Advanced Neutron Optics and Detectors Including Polarizing, Focusing, and Imaging Systems

(1) Polarizing device (2) Focusing device (3) Imaging technique

5 years (2008-2012)
 Members : JAEA, KEK, Hokkaido Univ., Tohoku Univ., Tokyo Univ.
 Kyoto Univ., L.J. Chang (NTHU)

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Polarized neutron

Neutron spin 1/2

2 eigenfunctions for $S_z = +1/2$ and $-1/2$

$$|\frac{1}{2}, +\frac{1}{2}\rangle \equiv u \quad |\frac{1}{2}, -\frac{1}{2}\rangle \equiv v$$

'spin up' state 'spin down' state

If a beam of neutrons has a fraction f of neutron in the state u

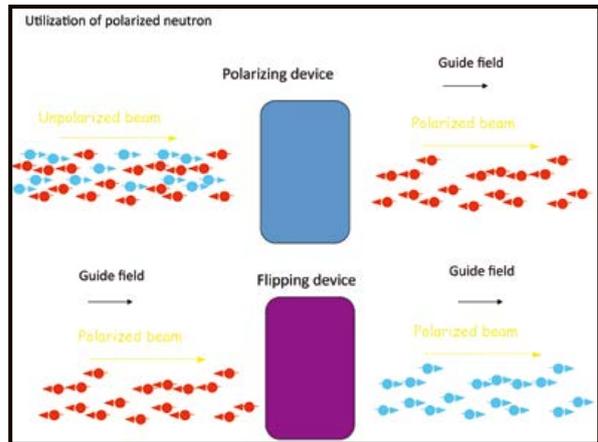
Polarization P (in the quantization direction) with magnitude $P = 2f - 1$

Unpolarized beam $P=0$;
 completely polarized beam with all the spins up $P=1$, with all the spins down $P=-1$

Polarization dependent cross section

$$\frac{d\sigma}{d\Omega} = NN^* + N^* (\vec{P} \cdot \vec{M}_\perp) + N (\vec{P} \cdot \vec{M}_\perp^*) + \vec{M}_\perp \cdot \vec{M}_\perp^* + i\vec{P} \cdot (\vec{M}_\perp^* \times \vec{M}_\perp)$$

Assuming collinear structure and inversion symmetry

$$\frac{d\sigma}{d\Omega} = N^2 + P(2N M_\perp) + (M_\perp)^2$$


Polarization dependent cross section

$$\frac{d\sigma}{d\Omega} = NN^* + N^* (\vec{P} \cdot \vec{M}_\perp) + N (\vec{P} \cdot \vec{M}_\perp^*) + \vec{M}_\perp \cdot \vec{M}_\perp^* + i\vec{P} \cdot (\vec{M}_\perp^* \times \vec{M}_\perp)$$

Assuming collinear structure and inversion symmetry

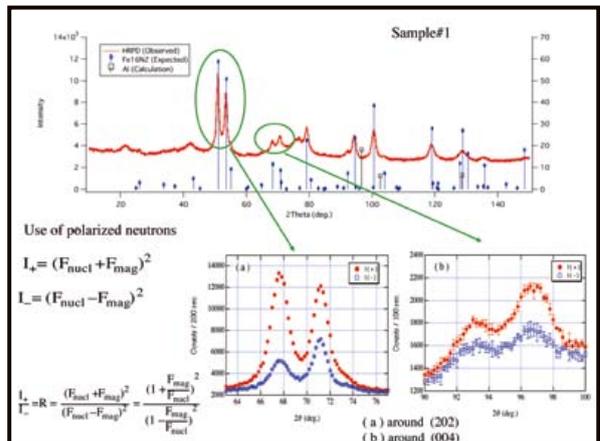
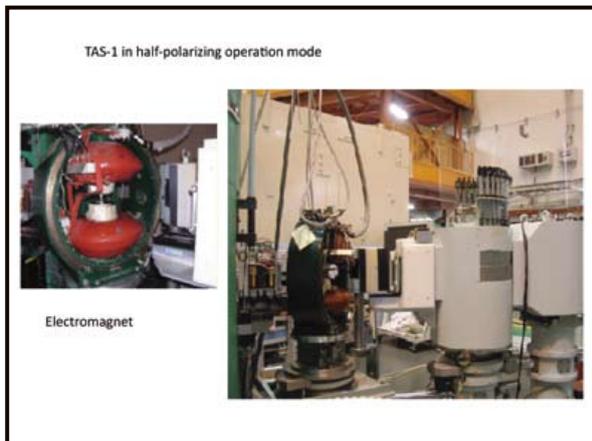
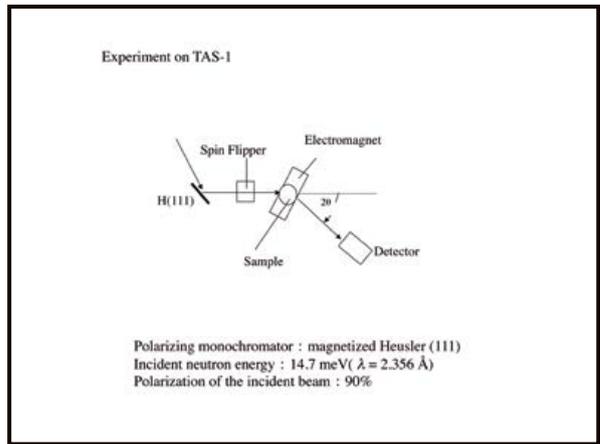
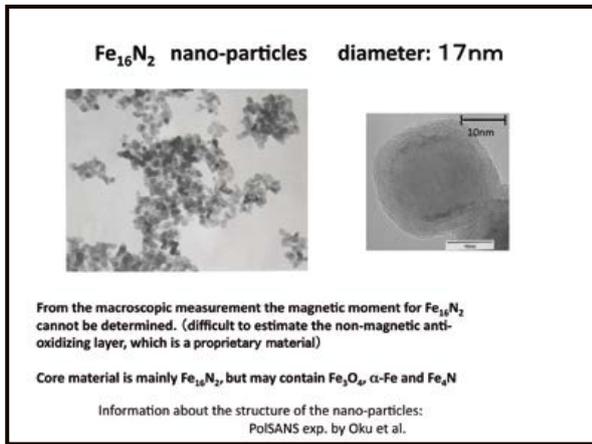
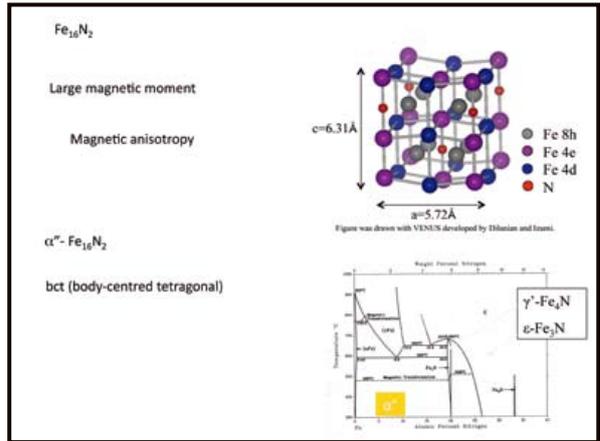
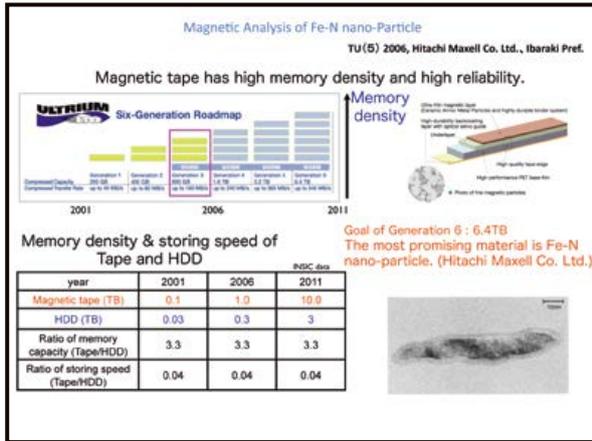
$$\frac{d\sigma}{d\Omega} = N^2 + P(2N M_\perp) + (M_\perp)^2$$

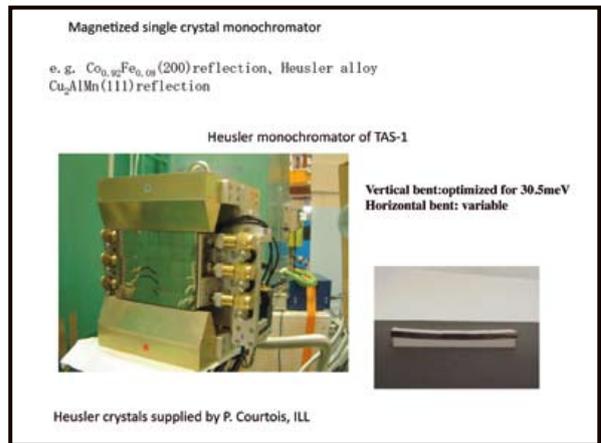
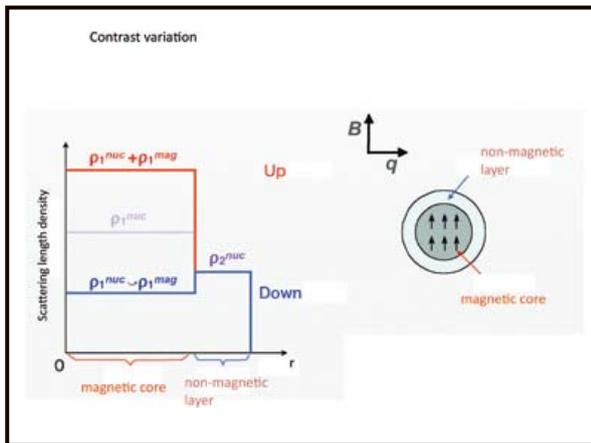
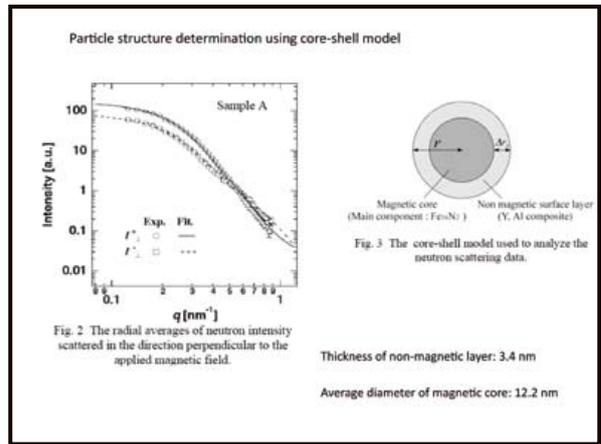
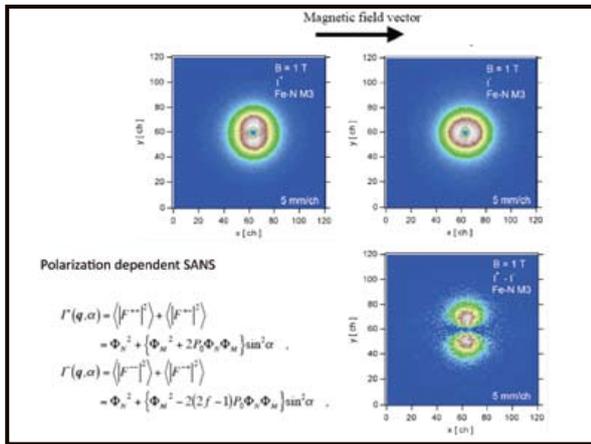
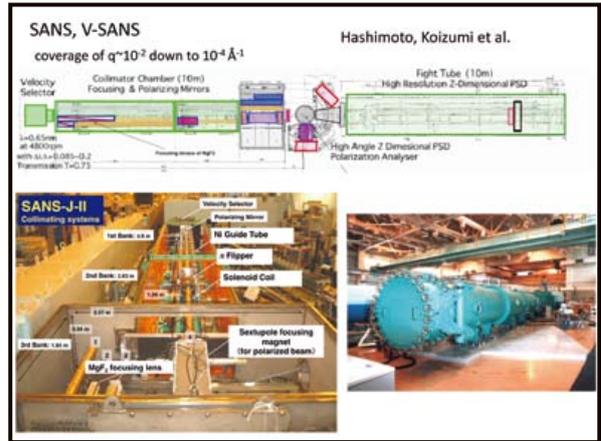
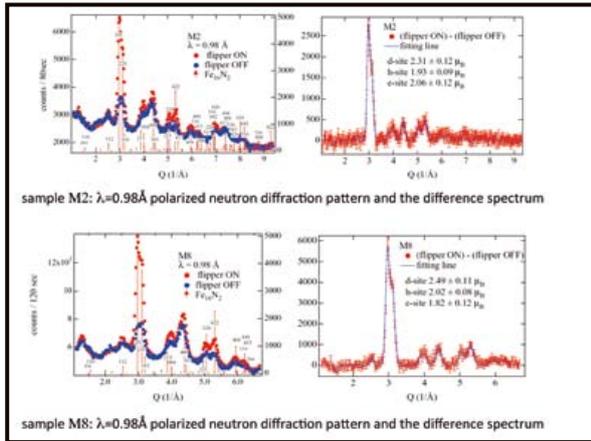
$R = \frac{I_\perp}{I_\parallel} = \left(\frac{N + M_\perp}{N - M_\perp} \right)^2$ when N is known → M_\perp

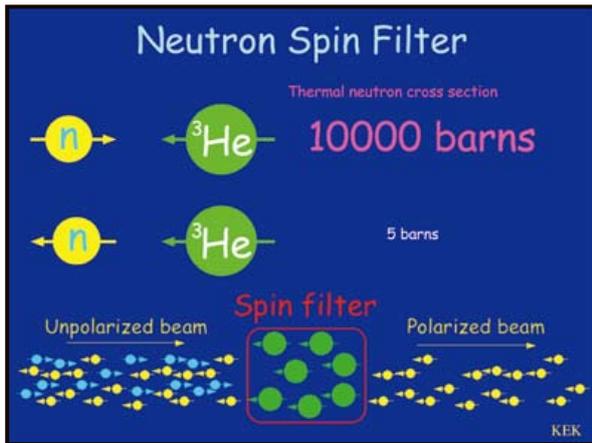
Polarized Neutron Powder Diffraction Experiments on $Fe_{16}N_2$ Nano-Particles

Y. Ishii¹⁾, M. Takeda¹⁾, T. Kikuchi¹⁾, T. Shinohara²⁾, T. Oku²⁾, J. Suzuki²⁾,
 Y. Sasaki³⁾, M. Kishimoto³⁾, M. Yokoyama⁴⁾, Y. Nishihara⁴⁾ and K. Kakurai¹⁾

¹⁾Quantum Beam Science Directorate, Japan Atomic Energy Agency (JAEA)
²⁾Materials and Life Science Division, J-PARC Center
³⁾Hitachi Maxell, Ltd.,
⁴⁾Physics Department, Ibaraki University







Japan Science and Technology Agency under auspices of MEXT

Funding for the development of neutron beam fundamentals techniques from the MEXT

Project Leader: K. Kakurai (JAEA) in collaboration with Hokkaido University, Tohoku University, KEK, Univ. of Tokyo and Kyoto University

Development of polarizing devices, e.g. ${}^3\text{He}$ filter

T. Ino, H. Kira, Y. Sakaguchi, L.J. Chang, T. Oku

Development of focusing devices, e.g. magnetic lens

T. Oku, J. Suzuki, M. Shimizu

Development of detecting devices, e.g. high time- and spatial-resolution detector

Taniori, Kiyonagi, Matsubayashi, Yasui, Sakai

${}^3\text{He}$ filter (SEOP) Development

(By courtesy of Dr. Ino, KEK)

Polarization

Intensity (Counts/200 sec)

Time (h)

72.3°C

180°C

175°C

Preliminary

Drs. H. Kira, Y. Sakaguchi, L.J. Chang, T. Oku, T. Ino et al.

New Laser Optics with VHG¹⁾ optics

¹⁾VHG: Volume Holographic Grating

laser sink

laser exit

135 mm fan

AFP-NMR : ${}^3\text{He}$ polarity controlled

T. Ino et al., J. Phys.Conf. (2012)

On Line Pumping SEOP system

Solenoid Coil

${}^3\text{He}$ Cell

GE180 Cell with a 10 cm-diameter fabricated by Tohoku Univ.

Pick up coil

RF coils

High angle detectors

90 degree bank

Small angle det.

Neutrons

L=26.5m

${}^3\text{He}$ polarizing filter

Ibaraki prefecture high intensity powder diffractometer IMATERIA

Drawing by courtesy of Prof. Inagaki et al., Ibaraki Prefecture

中性子小角散乱装置大観 (TAIKAN) への応用

J-PARC

中性子レンズ

真空散乱槽

超小角検出器

光学デバイス

小角検出器

${}^3\text{He}$ 偏極フィルター

By courtesy of Dr. J. Suzuki (J-PARC/CROSS)

中性子小角散乱装置大観 (TAIKAN) の観測対象

金属材料、磁性材料、ソフトマター、タンパク質等のサブナノからミクロンスケールの構造解析

Focusing Option

Small-angle bank

Simulation

Medium-angle bank

60A

金属材料ノ粒子

タンパク質溶液

lysozyme

myoglobin

β -lactoglobulin

By courtesy of Dr. J. Suzuki (J-PARC/CROSS)

Pulsed Neutron Imaging (BL22): Construction have started 2012.
(energy selective imaging, Magnetic Field Imaging with polarized neutrons.)

Polarization Analysis Neutron Chopper Spectrometer (POLANO, BL23)

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Development of the compact hybrid cell for low T experiments
T. Osakabe et al. JMMM 310 (2007) 2725; JJAP 47(2008) 6544

Fig. 1 (a) Cross sectional view of the hybrid cell. (b) MC cell with a cone-shaped coil. (c) MC cell (left) and MP35N steel support (right).

Fig. 2 Generated pressure and diameter of a sample chamber as a function of applied load.

Development of a thermal neutron focusing device
T. Osakabe and K. Soyama, Rev. Sci. Instr. 76(2005)073102

FIG. 2. Schematic illustration of the focusing device. The device has many neutron multilayer supermirrors with the shape of a circle. The shape of each mirror is a circle arc and the extension of each mirror meet at a focal point, namely at the sample position.

hybrid steel cell Focusing device

(a) 30.5 meV (1.64 A) (b) 13.7 meV (2.44 A) (c) 4.9 meV (4.1 A)

By courtesy of Dr. T. Osakabe (QuBS, JAEA)

Ground state of the rare earth filled skutterudite $\text{PrFe}_4\text{P}_{12}$

Temperature (K) vs. Pressure (GPa) phase diagram. Magnetic moment (μ_B) vs. mixing parameter d diagram. Energy level diagram for PrFe₄P₁₂.

Synchrotron radiation x-ray and neutron diffraction studies of phase separation on rare-earth metal dihydrides under high pressure

Dr. A. Machida et al.

Hydrogen in Materials

Various H-M bonding state

- metallic bond
- ionic bond
- covalent bond

Various valence state

H^{\cdot}, H^0, H^+

High-speed diffusion

Hydrogen related properties

- Metal-Insulator Transition
- Structural transition
- Hydrogen Storage
- etc.

J. N. Hubers et al. Nature (1996)

Experimental –SR XRD –

To investigate the variations of metal lattice

Diffractometer for Diamond Anvil Cell

BL22XU Exp. Hutch1

Detector
Imaging Plate (R-AXIS V, Rigaku Co.)
Size : 400×400mm²
100×100μm²/pixel
Sample-IP distance: 200mm-730mm

Sample

LaH₂ (40μm×20μm×10μm)
LaD₂ (35μm×25μm×10μm)

DAC
X-rays
Pressure marker
sample
Pressure medium
Hydrogen
Hydrostaticity
Hydrogenation reaction
Helium
Hydrostaticity

By courtesy of Drs. Aoki, Katayama et al. (QUBS, JAEA)

Phase separation from LaH₂ into LaH_{2+δ} and LaH_x

A. Machida et al., Phys. Rev. B 83,054103 (2011).

Additional Bragg spots appeared just outside of the original ones.

AP HP
LaH₂ LaH_{2+δ}

Formation of small-fcc lattice
Volume reduction ~ -17%

The small fcc phase correspond to the H-poor phase.

Observed change is reversible.

$LaH_2 \rightleftharpoons (1-\eta)LaH_{2+\delta} + \eta LaH_x$

Interstitial H atoms transfer from the T-sites to O-sites.

Experimental –NPD–

To investigate the hydrogen positions

High Intensity Total Diffractometer (NOVA)

BL-21, MLF, J-PARC

Paris-Edinburgh Press

NPD experiments above 10GPa became available.
Max. pressure reached 17 GPa

NOVA@J-PARC
Incident neutron
sample
anvil
90 degrees scattering geometry

By courtesy of Drs. Katayama, Hattori, Sano, Utsumi et al.

The H position determined using neutron diffraction

In NPD patterns, odd number reflection peaks are not observed.

Formation of the NaCl-type monodeuteride, not solid solution!

LaD₂ XRD 13.1GPa
NPD 13 GPa
Simulation: LaD₂⁰

Neutron scattering length
 $b_{La} = 8.24$ fm
 $b_D = 6.671$ fm

Form factor of NaCl-type
Odd : $4(b_{La} - b_D)$
Even : $4(b_{La} + b_D)$

The simulated patterns strongly indicate the concentration is close to 1.0.

First observation of the formation of LaD using neutron diffraction.

Different concentration with common metal lattice

	LaH	LaH ₂	LaH ₃
Structure			
fcc metal lattice			
Nearest neighbor	H ⁰ -La	H ^T -La	H ^T -La H ⁰ -H ^T

Different bonds should be made in the different state.

Important information to clarify the H-M interactions.

PLANET

Pressure Loading Apparatus for NEutron diffraction

Moderator	Decoupled H ₂ (para)
Source to sample (L _s)	25 m
Sample to detector (L _d)	1.5 m
Angular coverage	horizontal: $\pm 11^\circ$, vertical: $\pm 35^\circ$
Wavelength range	0.1 - 5.8 Å
Q _{max}	30 Å ⁻¹
Resolution at 90° bank	$\Delta d/d \leq 0.005$

6-axis Multi-Anvil Press

- Diffraction ³He PSD
- Imaging Neutron Image Intensifier

By courtesy of Drs. Hattori, Sano, Utsumi et al.

6-axis multi-anvil press ATSUHIME 庄姫 (Pressure Queen)

(Advanced TOF SUIted High-pressure Multi-anvil Equipment)

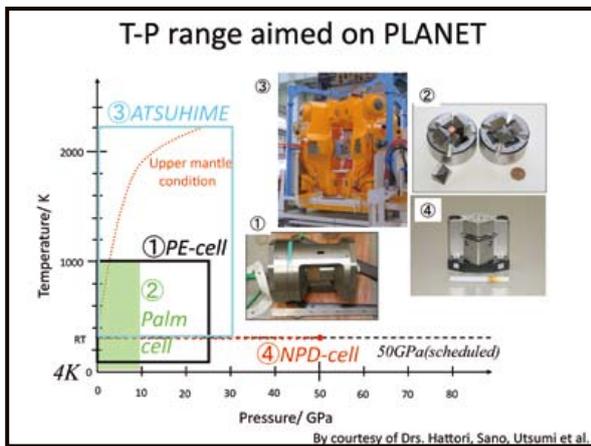
max. load 500ton/axis

- relatively large access windows
- variety of pressurizing hydroscopic uniaxial deforming

庄姫 (weight: 29ton)

Central part

By courtesy of Drs. Hattori, Sano, Utsumi et al.



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(3) Imaging technique

Experimental set-up for viewing small engine

Neutron color I.I.

Small engine

Permalloy

Neutron

Motor

Reference
By courtesy of Dr. Nittoh, Toshiba Corp.
"Visualization of oil behavior in a small 4-cycle engine with electrical motoring by neutron radiography" by M. Nakamura, K. Sugimoto, H. Asano, H. Murakawa, N. Takenaka, K. Mochiki, Nuclear Instruments and Methods in Physics Research A605(2009)204-207.

Mini-engine (24.5 ml)

(a) (b) (a)-(b)=

Neutron color I.I.: 9 inch
NTSC 3CCD: 30 frame/sec
Shutter speed: 1 / 100 s
Lens Aperture: F3

Reference By courtesy of Dr. Nittoh, Toshiba Corp.
"New feature of the neutron color image intensifier," K.Nittoh,C.Konagai,T.Noji,K.Miyabe,
Nuclear Instruments and Methods in Physics Research A605(2009)107-110.

μPIC neutron imaging detector (prototype)
Kyoto University, Cosmic Ray Group

- Time-projection-chamber
 - Gas mixture: Ar-C₂H₆-³He(30%) at 2 atm.
 - Detection efficiency of ~30% for thermal neutrons.
 - Time resolution ~1 μs for each neutron interaction.
- FPGA program "X-ray mode B".
 - FPGA encoder measures width of pulses.
 - Proton, triton identified from shape of energy deposition.
 - Position resolution < 0.2 mm.
 - Data rates up to ~8 MHz (neutron rate of several hundred kHz).

400 μm pitch.
Gain uniformity: ~4%(σ).
Normal operating gain for neutron detection: ~500.
Can operate for > 1 year on single gas filling.
Sizes up to 40 × 40 cm² with virtually no dead space.

By courtesy of Dr. J. Parker and Prof. T. Tanimori (Cosmic Ray Group; Kyoto Univ.)

μPIC neutron imaging detector (prototype)
Kyoto University, Cosmic Ray Group

Test experiments at NOBORO (JSNS BL10).

Proton ID from energy deposition.

Resolution with PID: $194.2 \pm 3.8 \mu\text{m}$ (Includes effects of beam divergence.)

No PID: Resolution ~1 mm. June 2010

November 2009 Preliminary
Diffraction pattern from 200nm SiO₂ nanoparticles

February 2011 Preliminary
Bragg-edge transmission for Fe powder (211) (110) (200)

Resonance absorption
Preliminary
Preliminary
Indium compared with ENDF/B-VI.0
February 2011

μPIC NID: good position and fine time resolutions, high rates, and large area at low cost.

By courtesy of Dr. J. Parker and Prof. T. Tanimori (Cosmic Ray Group; Kyoto Univ.)

Refining position resolution

- Neutron position determination:
 - Measure track length.
 - Determine proton direction from pulse-width distribution.
 - Make correction from mid-point of track.
- Developed two improved methods for measuring the p-t track length: End-Point Extrapolation (EPE) and Peak Interpolation (PI).

Combining both EPE and PI methods produces best result of $\sigma = 116.3 \pm 0.5 \mu\text{m}$.

By courtesy of Dr. J. Parker and Prof. T. Tanimori (Cosmic Ray Group; Kyoto Univ.)

Neutron-gamma separation

Escape events Fully-contained neutrons Event pile-up, scattered protons

- Gamma rejection studied using 1-MBq ¹³⁷Cs source.
- Data taken over 24 hours at a gas gain of ~600.

Pulse-width sum after track-length cut

Pulse-width sum after PID cut

Both neutrons and gammas are detected (γ efficiency < 10%).
Neutrons selected by cuts in total time-above-threshold and 3σ track length.
Fraction of detected γ's surviving neutron cuts < 10⁻⁶ reflective gamma sensitivity of < 10%.

	Containment fraction (95% CL)
Track length	< 9.3 × 10 ⁻⁶
+ PID	< 3.6 × 10 ⁻⁶

By courtesy of Dr. J. Parker and Prof. T. Tanimori (Cosmic Ray Group; Kyoto Univ.)

磁気イメージング法の開発
Dr. Shinohara et al.

スピン回転子の導入と中性子スピンの3次元制御

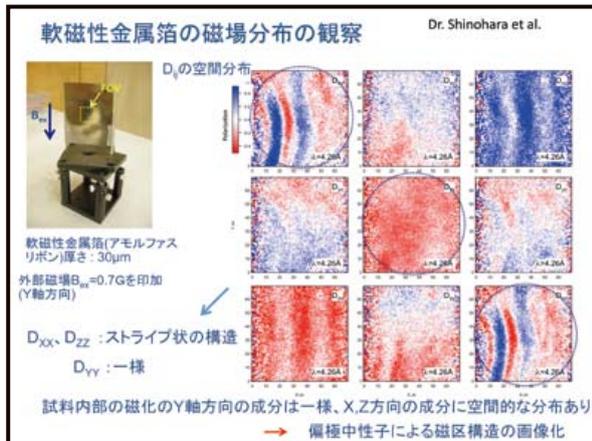
中性子スピンの量子化軸を x,y,zのそれぞれの向きへ制御

試料の前後にスピン回転子を配置 それぞれのコイルがn/2ずつ中性子スピンを回転することで、X,Y,Zの方向へスピンの方向を制御

SRI+SRI2, SR3+SR4: 中性子スピン//X
すべてOFF: 中性子スピン//Y
SRI, SR4: 中性子スピン//Z

スピン回転子の調整
すべての波長についてn/2ずつだけ回転させる
・パルス中性子のタイミングにあわせて1/tで減衰する電流を印加
→ 各回転子について、ピーク電流値を調整

By courtesy of Dr. J. Parker and Prof. T. Tanimori (Cosmic Ray Group; Kyoto Univ.)



Summary

The continuous development efforts on neutron beam fundamentals, such as polarized neutrons, focusing devices, etc. for both the continuous and pulsed source are essential for advanced neutron scattering investigations to realize state-of-the-art experiments such as magnetic structure determination, experiments under novel and extreme sample conditions.

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