Status of the He-3 spin filter development and application at J-PARC MLF

J-PARC Center, JAEA
Takayuki Oku

Contents

J-PARC, MLF

- Overview of the neutron beamlines of MLF
- Developments of neutron polarizers at MLF
  - SEOP based ^{3}He spin filters
  - Magnetic supermirrors
- How polarized neutrons are used at each neutron beamline of MLF.

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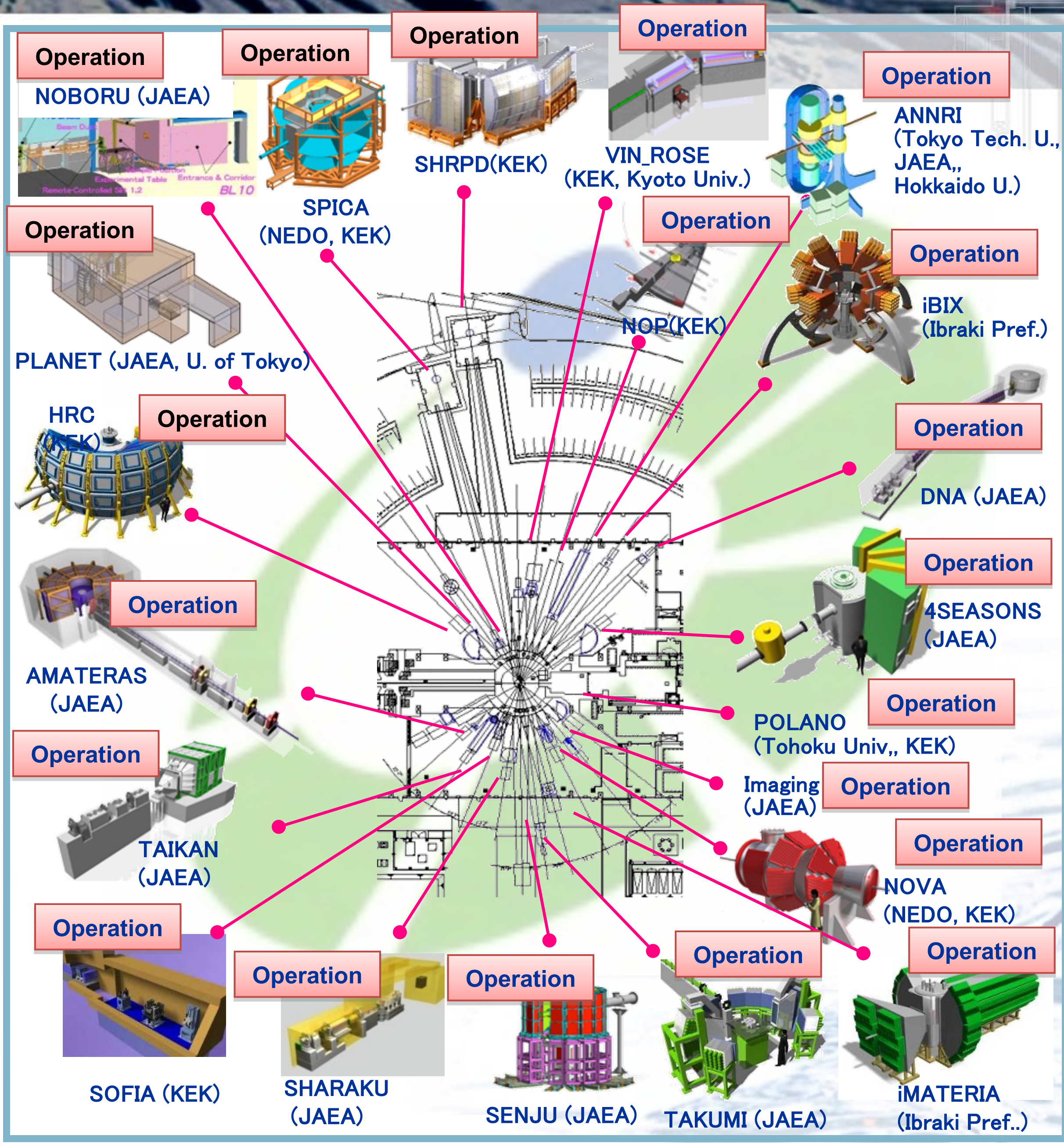
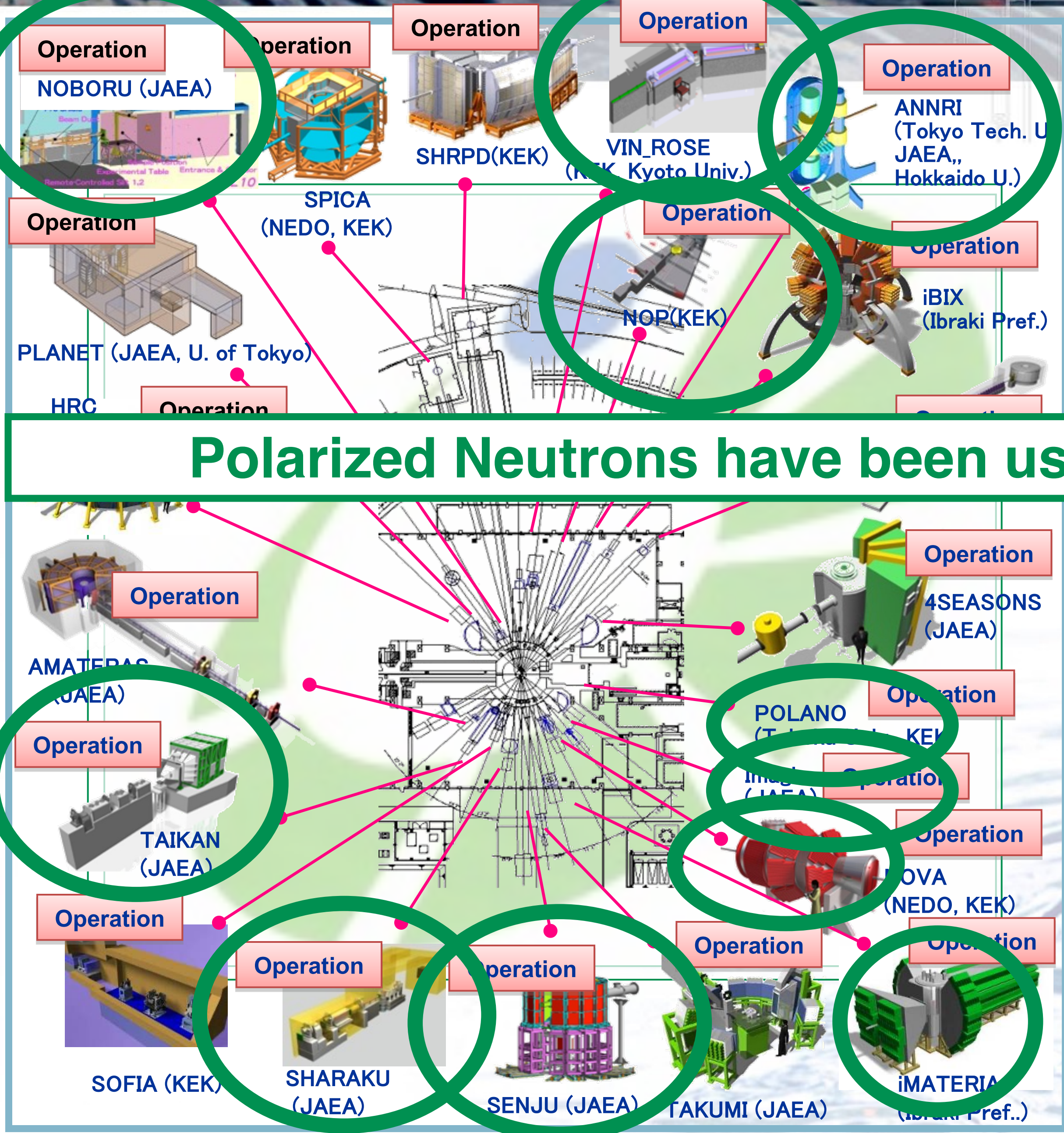
Status of Neutron Instruments at MLF



- The first neutron in May, 2008
- 23 Neutron Beamlines
- In operation: 21
- Operated by
  - ・JAEA
  - ・KEK
  - ・CROSS
  - ・Ibaraki Prefecture
- From Fundamental Physics to Industrial Uses
- Operation days/Year ~180days/year
- Staffs and out-sourcing 150+70 (relevant organ. Altogether)

Status of Neutron Instruments at MLF



Polarized Neutrons have been used at 11 BLs

- The first neutron in May, 2008
- 23 Neutron Beamlines
- In operation: 21
- Operated by
  - KEK
  - JAEA
  - Ibaraki Prefecture (2)
  - ...ling)
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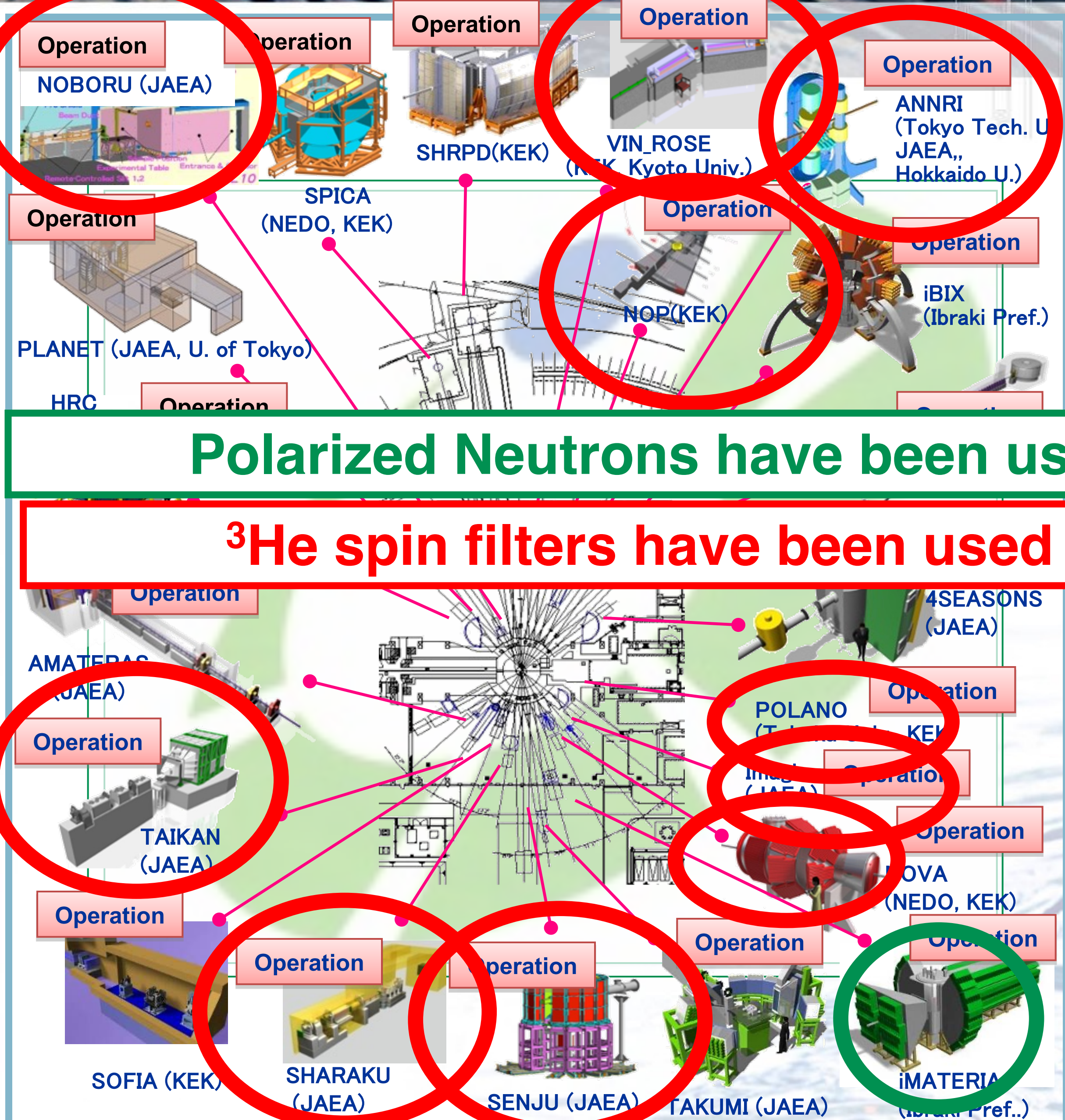
Status of Neutron Instruments at MLF



Polarized Neutrons have been used at 11 BLs

^{3}He spin filters have been used at 9 BLs

- The first neutron in May, 2008
- 23 Neutron Beamlines
- In operation: 21
- Operated by
  - ・KEK
  - ・JAEA
  - ・Ibaraki Prefecture (2)

ling)

Physics to

Industrial Uses

- Operation days/Year
  ~200days/year (Goal)
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J-PARC, MLF

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Developments of neutron polarizers at J-PARC MLF

- SEOP based ^{3}He spin filters

i) Development & Application

T. Okudaira[1), 2)], S. Takada[1), 3)], S. Takahashi[1), 4)], R. Kobayashi[1), 4)], M. Okuizumi[1), 2)], T. Ino[5)], T. Oku[1), 4)]

Application

R. Kiyangi[1], K. Hiroi[1)], T. Honda[5)], T. Yokoo[5)] , M. Fujita[3)]

- Magnetic supermirror polarizers development

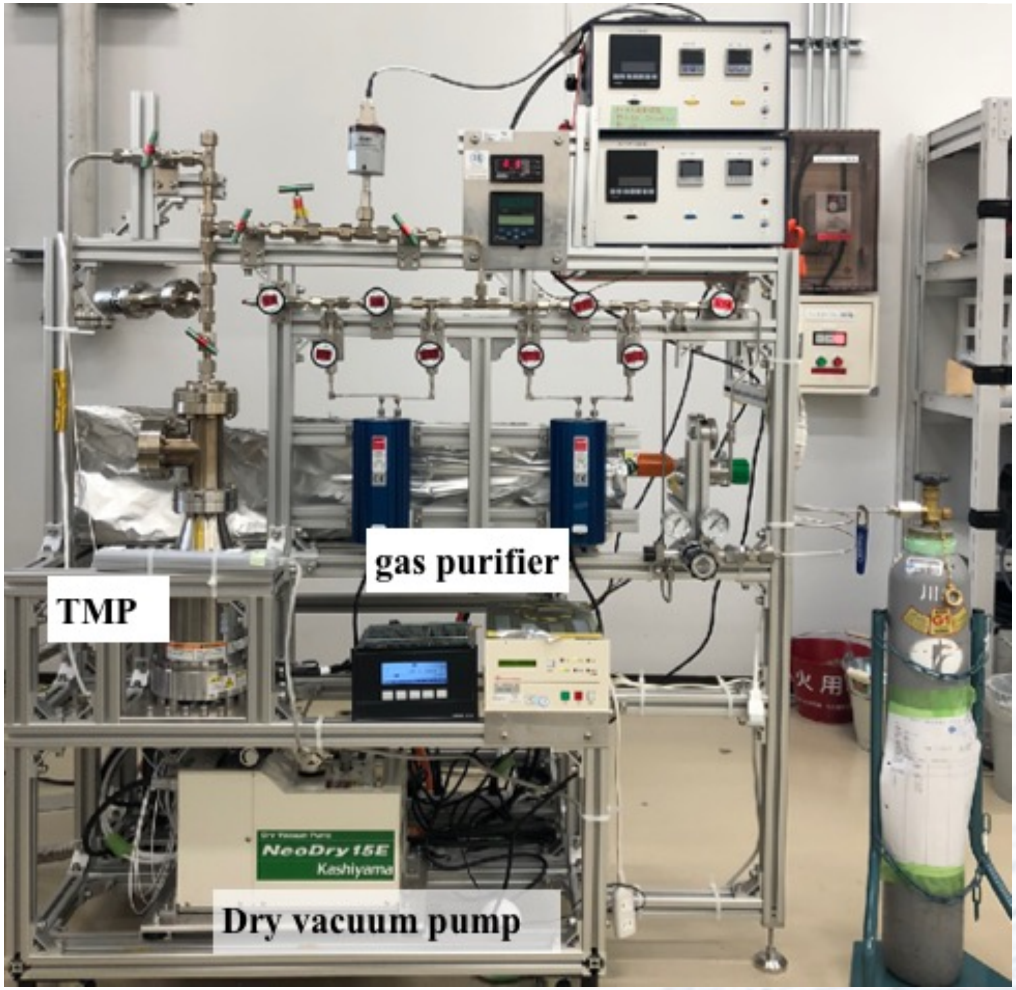
Ryuji Maruyama[1)], Dai Yamazaki[1)]

1)J-PARC Center, JAEA, 2) Nagoya Univ., 3)Tohoku Univ., 4)Ibadraki Univ., 5) J-PARC Center KEK

Developments of the SEOP based ^3He spin filters at J-PARC MLF

^{3}He gas filling system for the ^{3}He spin filter

T. Okudaira, T. Oku, H. Kira, K. Sakai, T. Ino (2018)



Ultra-high vacuum system. Achievable vacuum pressure is < 1×10^{-7} Pa.

The ^{3}He polarization is very sensitive to impurities such as magnetic impurity, Oxygen, Hydrogen.

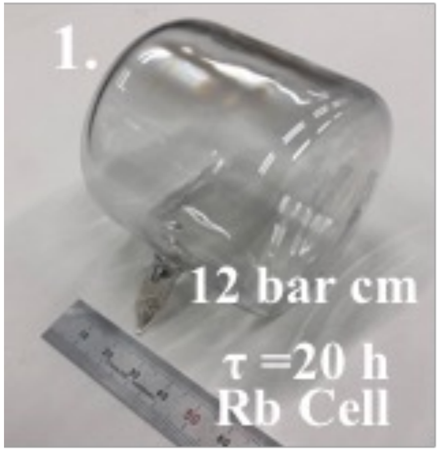
An ultra-high vacuum system is necessary to fill ^{3}He and alkali metal to the glass cell without impurities.

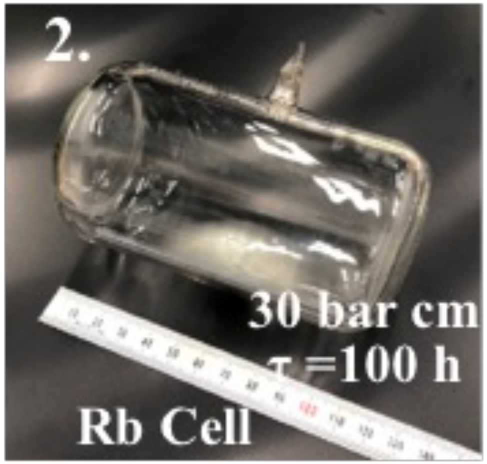
The cleaning is important for the higher ^{3}He polarization and the longer relaxation time.

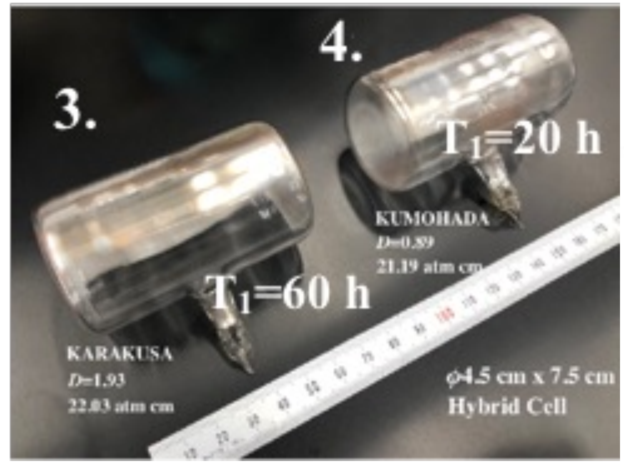
The glass and stainless tubes are baked out over 1 weeks at 400°C and 200°C, respectively.

Alkali metal is distilled for 2 times and encapsulated to the glass cell.

Fabricated ^{3}He cells







We have succeeded in fabricating good quality cells with a long relaxation time of τ ~200 hrs for the 3 atm gas-pressure cell.

Here, the theoretical limit is τ_{dipole} < 250 hrs for a 3 atm cell.

Relaxation of ^{3}He polarization

The ^{3}He polarization decays with relaxation time τ

$$P(t) = P_0 \exp(-t/\tau)$$

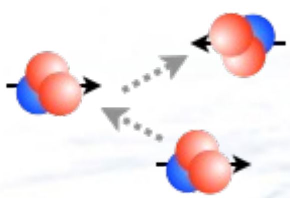
$$\frac{1}{\tau} = \frac{1}{\tau_{B_0}} + \frac{1}{\tau_{\text{dipole}}} + \frac{1}{\tau_{\text{impurities}}} + \frac{1}{\tau_{\text{wall}}} \cdots$$

Quality of Spin Filter

Ununiform magnetic field

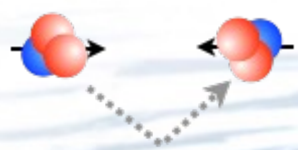
$$\tau_{B_0} = \frac{p}{6700} \frac{B_0}{|\nabla B_x|^2 + |\nabla B_y|^2} \; [h]$$

Collisions with ^{3}He

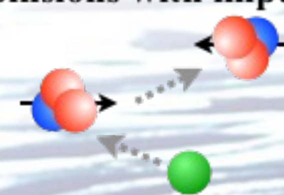


$$\tau_{\text{dipole}} = \frac{810}{p} [h]$$

Collisions with wall



Collisions with impurities

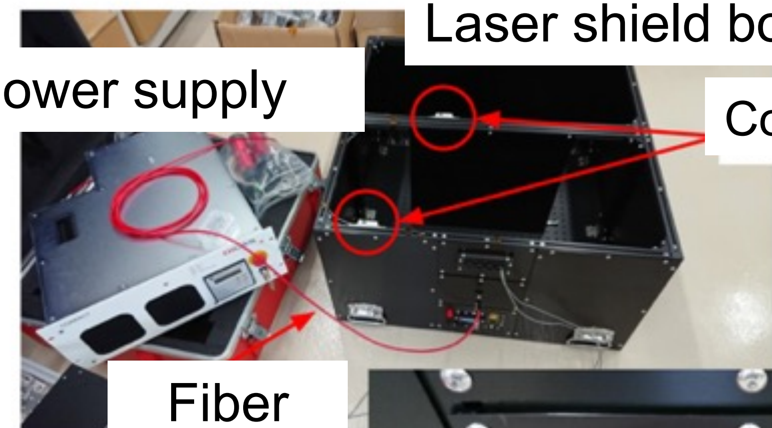


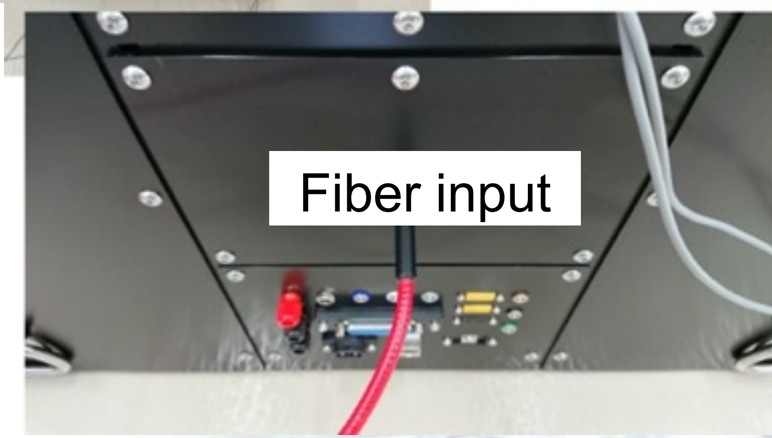
$$\tau_{dipole} < 250 \; [h] \; \text{for 3.2 atm cell}$$

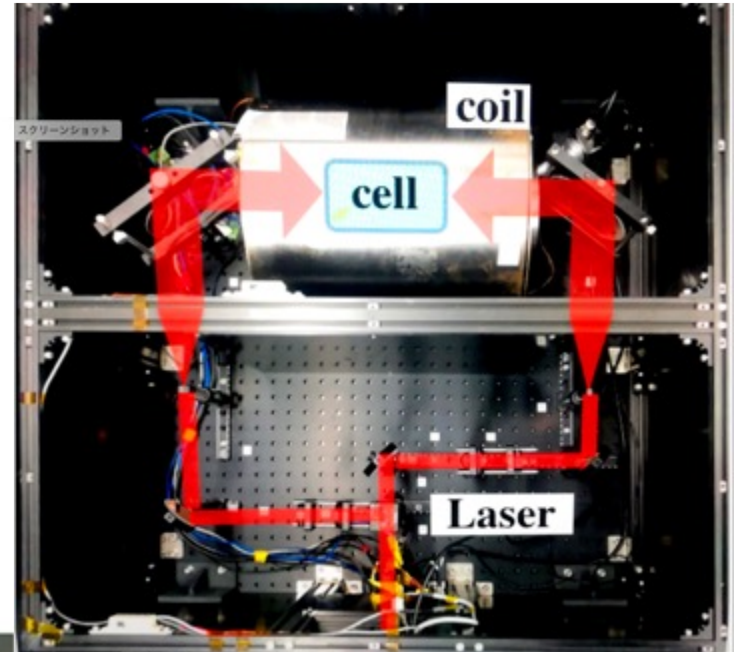
High power fiber-coupled laser setup

H. Hayashida, T. Okudaira et al. (2019)

- Laser power = **110W**
- Continuous mode
- Fiber coupled
- The cell is irradiated from both side





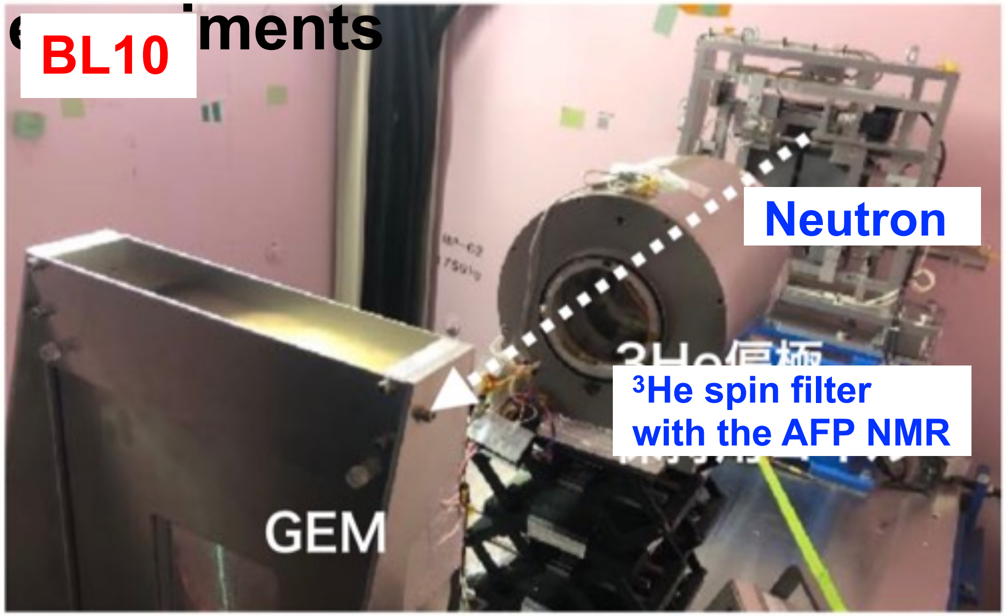


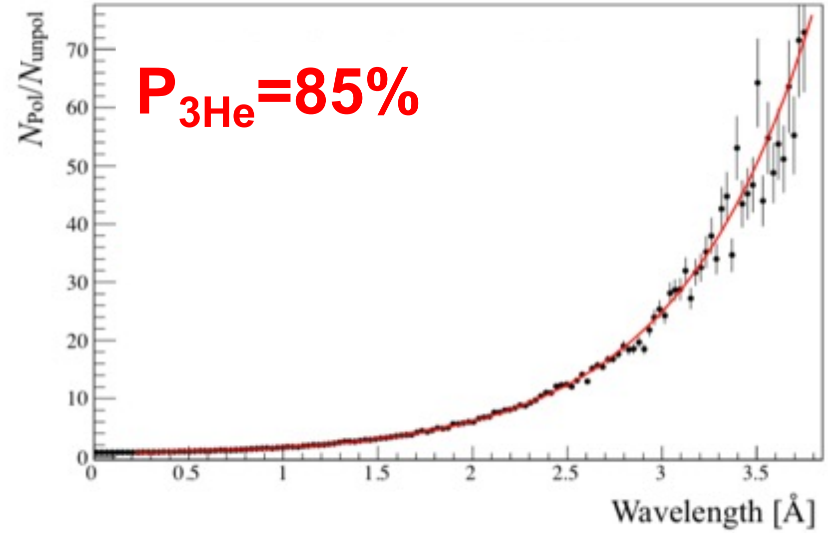
Top view of the SEOP setup

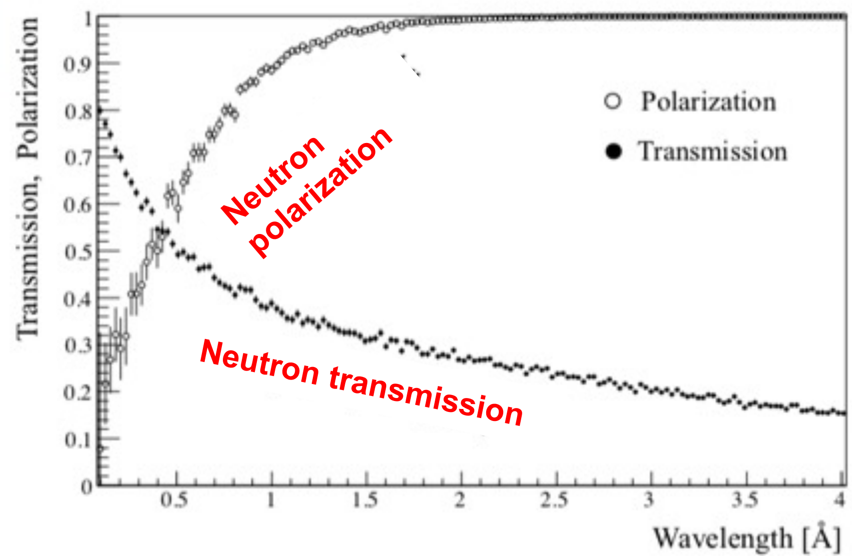
Neutron transmission experiments

T. Okudaira et al., NIM-A 977 (2020)

P_{3He}=85% was determined with **ex-situ setup by the neutron transmission experiments**







Relaxation time T_1 = 174 hrs with ^{3}He gas pressure length of **20 atm cm on the beamline**

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How polarized neutrons are used at each BL.

J-PARC, MLF

SANS and Neutron Reflectometry

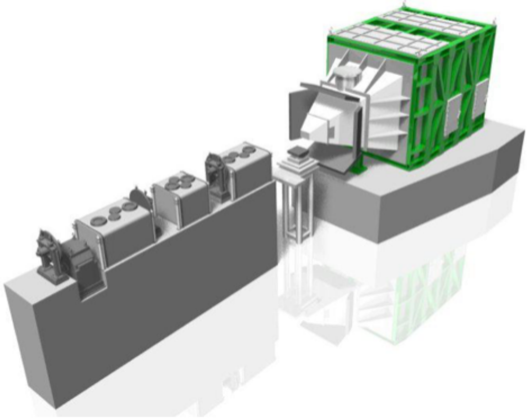
J-PARC, MLF

How polarized neutrons are used at BL15.

BL15 TAIKAN Small and Wide Angle Neutron Scattering Instrument

Analyze the structure of materials over a wide range of spatial scales, from sub-nanometers to microns.

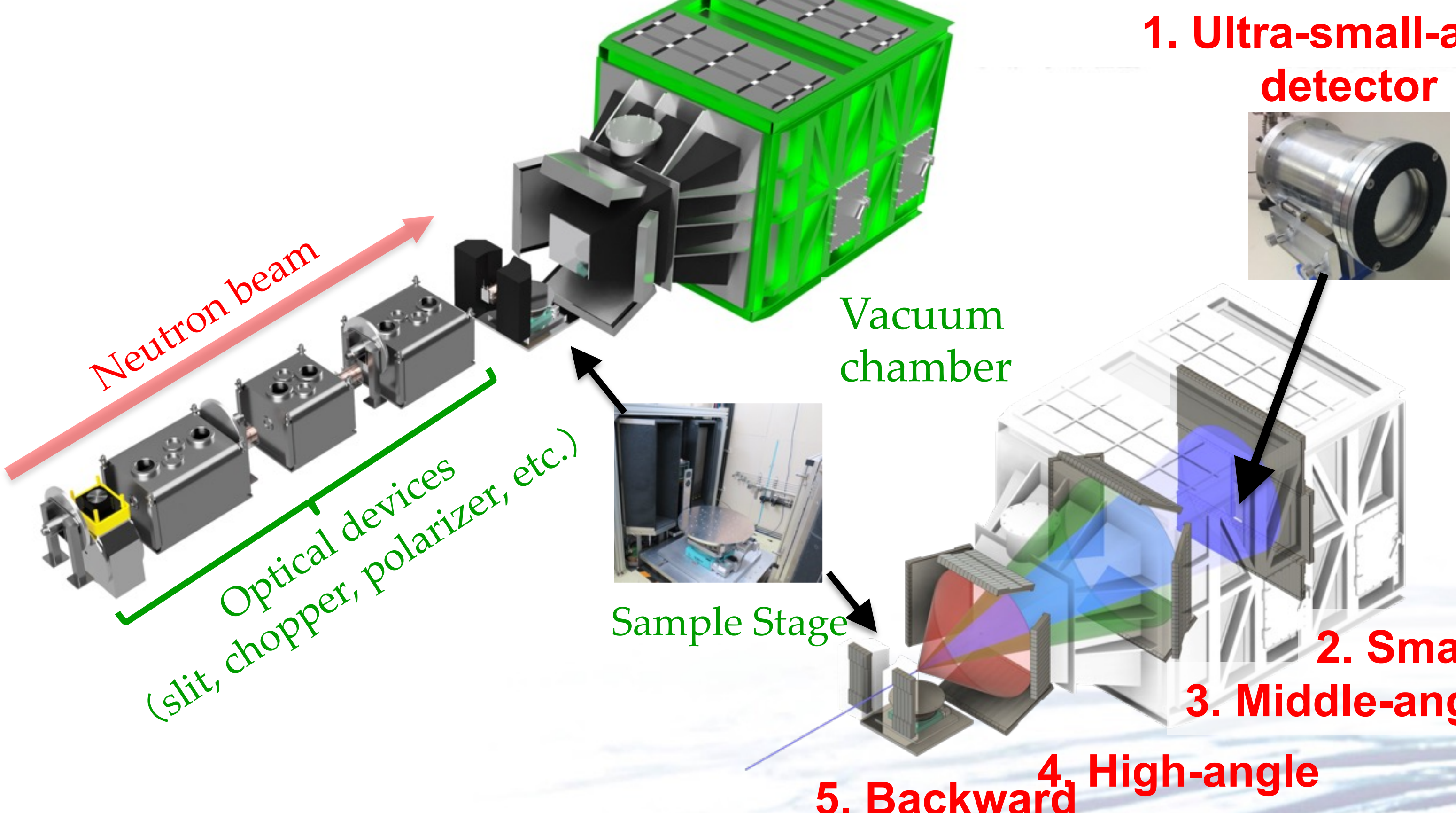
Research	Structural and non-equilibrium phenomena of metals, magnetic materials, superconductors, soft matter, biomacromolecules and their composites
Features	Highly efficient small- and wide-angle scattering measurements with high spatial and temporal resolutions are realized using state-of-the-art neutron optics and detection techniques to elucidate structures on a wide range of spatial scales.



The small and wide angle neutron scattering

BL15 “TAIKAN”

5 Detector Banks!!



Detector arrangement

Broad bandwidth of neutron wavelength

+

Detector banks covering wide angle

Efficient measurement in wide-*q* range

5.0×10^{-3} Å$^{-1} \leq q \leq 17$ Å^{-1}

Setup for polarization analysis with ^{3}He spin filter

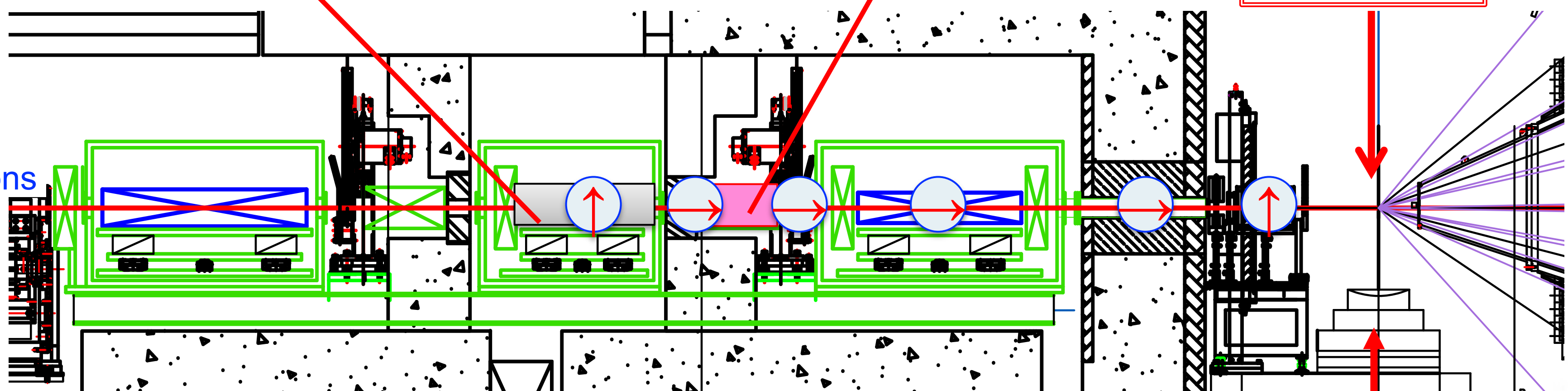
BL15 "TAIKAN"

Magnetic Supermirror polarizer

Spin Flipper

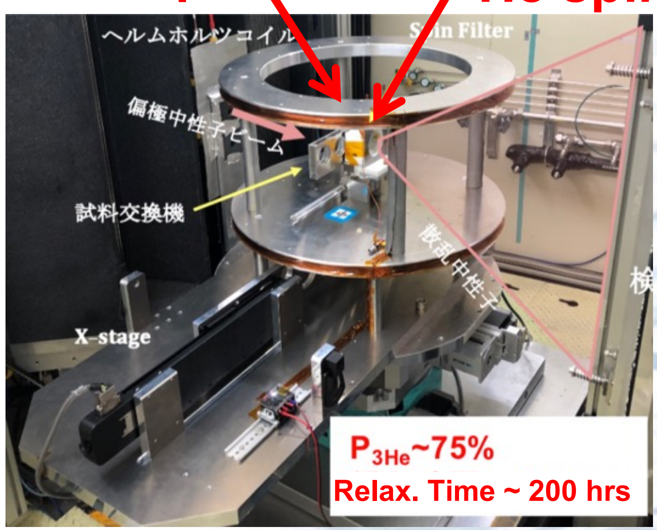
Sample

Neutrons



Sample

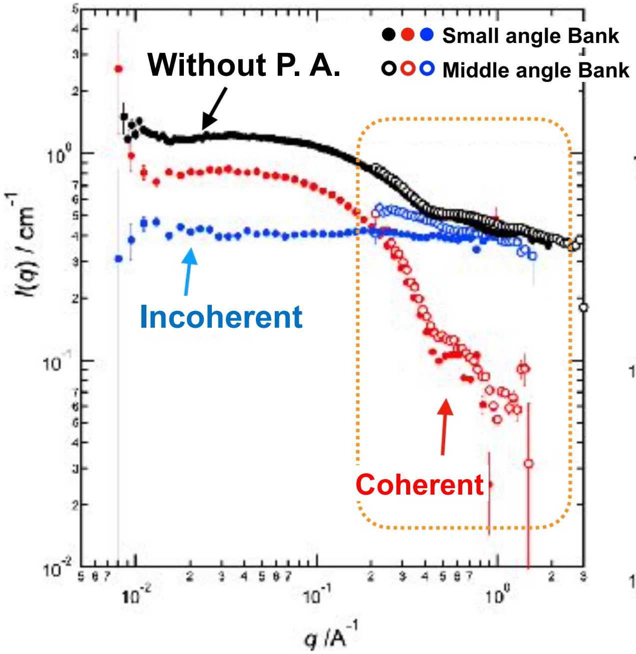
^{3}He spin filter



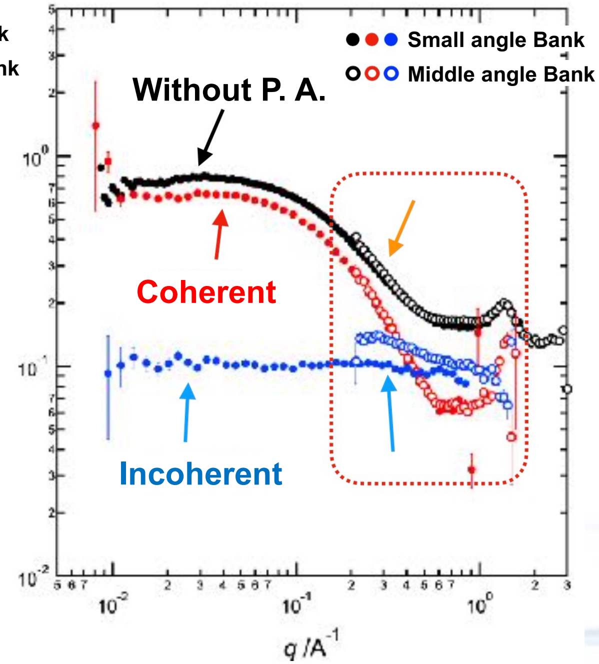
Examples of polarization analysis exp. @ BL15 of Hydrogen contained colloidal samples

T. Okudaira et al., J. Appl. Cryst. (2021)

TBP $(HNO_3)_x$



$PtCl_6$ $(BEHU\text{-}H)_2$

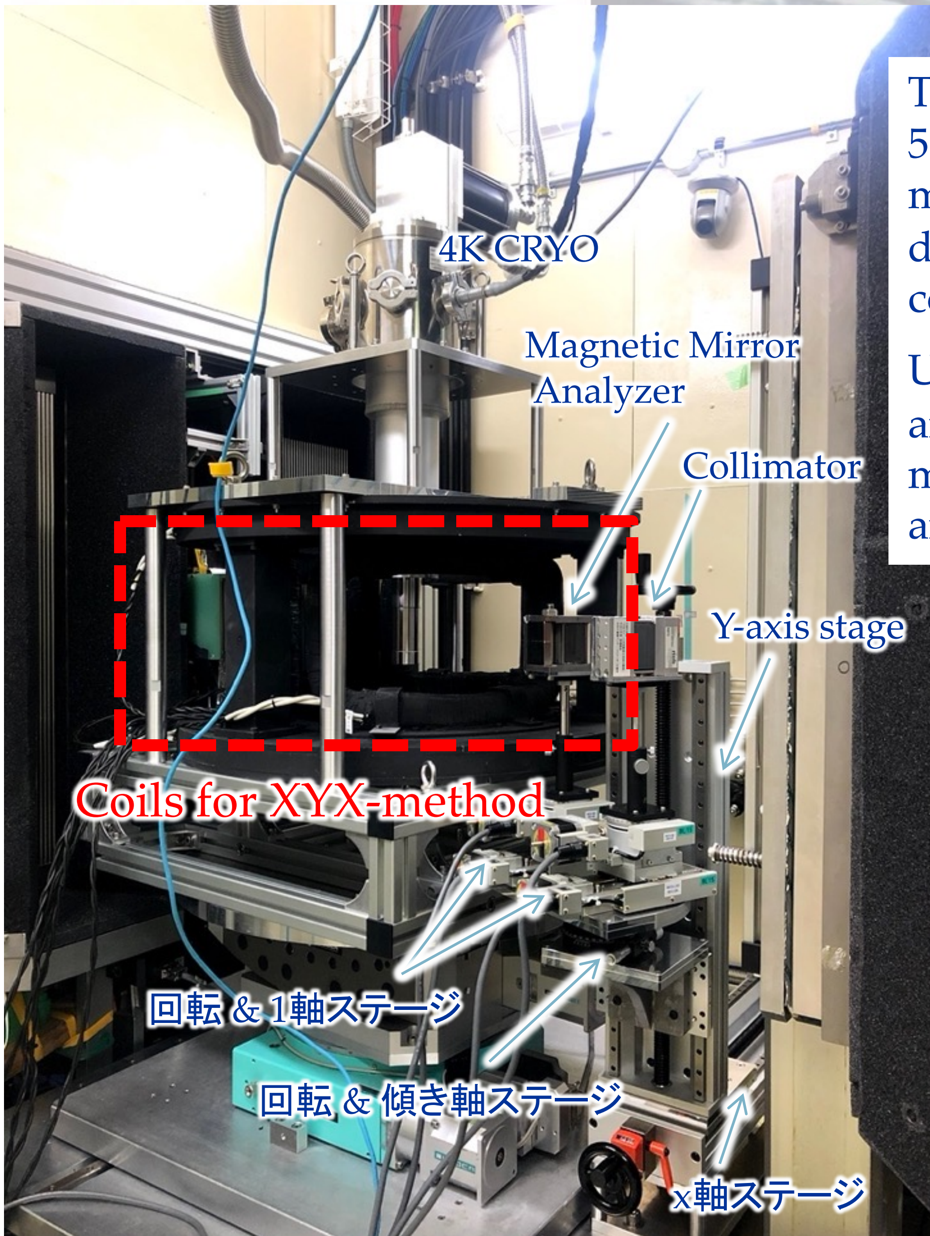


○(open circles) are the data obtained with the ***small-angle*** detector bank.

●(filled circles) are the data obtained with the ***middle-angle*** detector bank.

The coherent and incoherent scattering components were distinguished by the polarization analysis.
the coherent components obtained with the low angle detector bank and the middle angle detector bank are smoothly connected.
On the other hand, there was a gap in the intensities of the incoherent components between the low angle detector bank and the middle angle detector bank

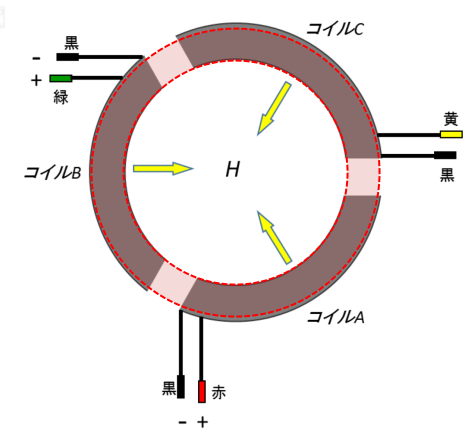
XYZ-method (Uniaxial polarization analysis)



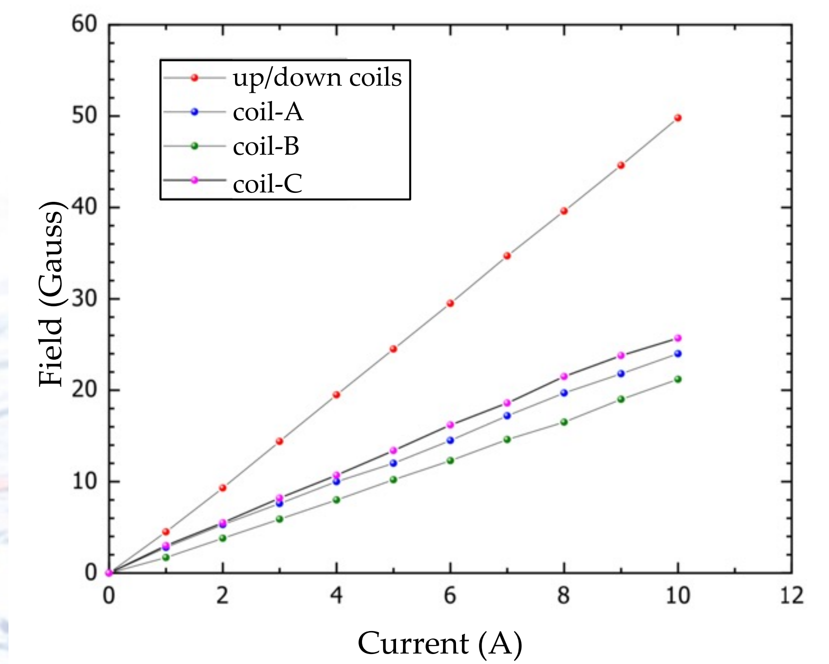
The coils is composed of 5 coils. It can apply a magnetic field in any direction like the PASTIS coil for XYZ-method.

Uniaxial polarization analysis can be done by magnetic mirror analyzer.

Top view of the coils



The coils belong to Tohoku Univ.

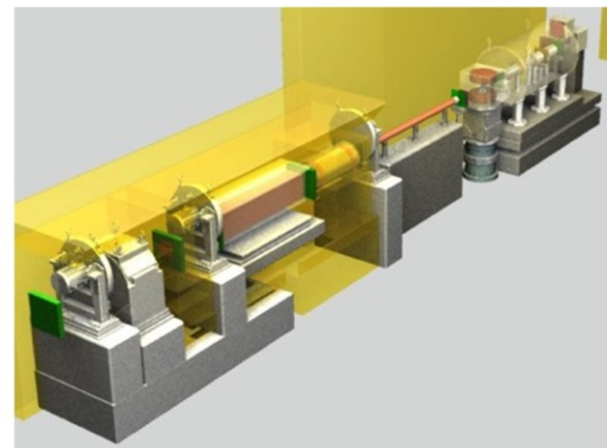


How polarized neutrons are used at BL17.

BL17 SHARAKU Polarized Neutron Reflectometer

Investigation of microscopic structures of interfaces using microscopic neutron spins

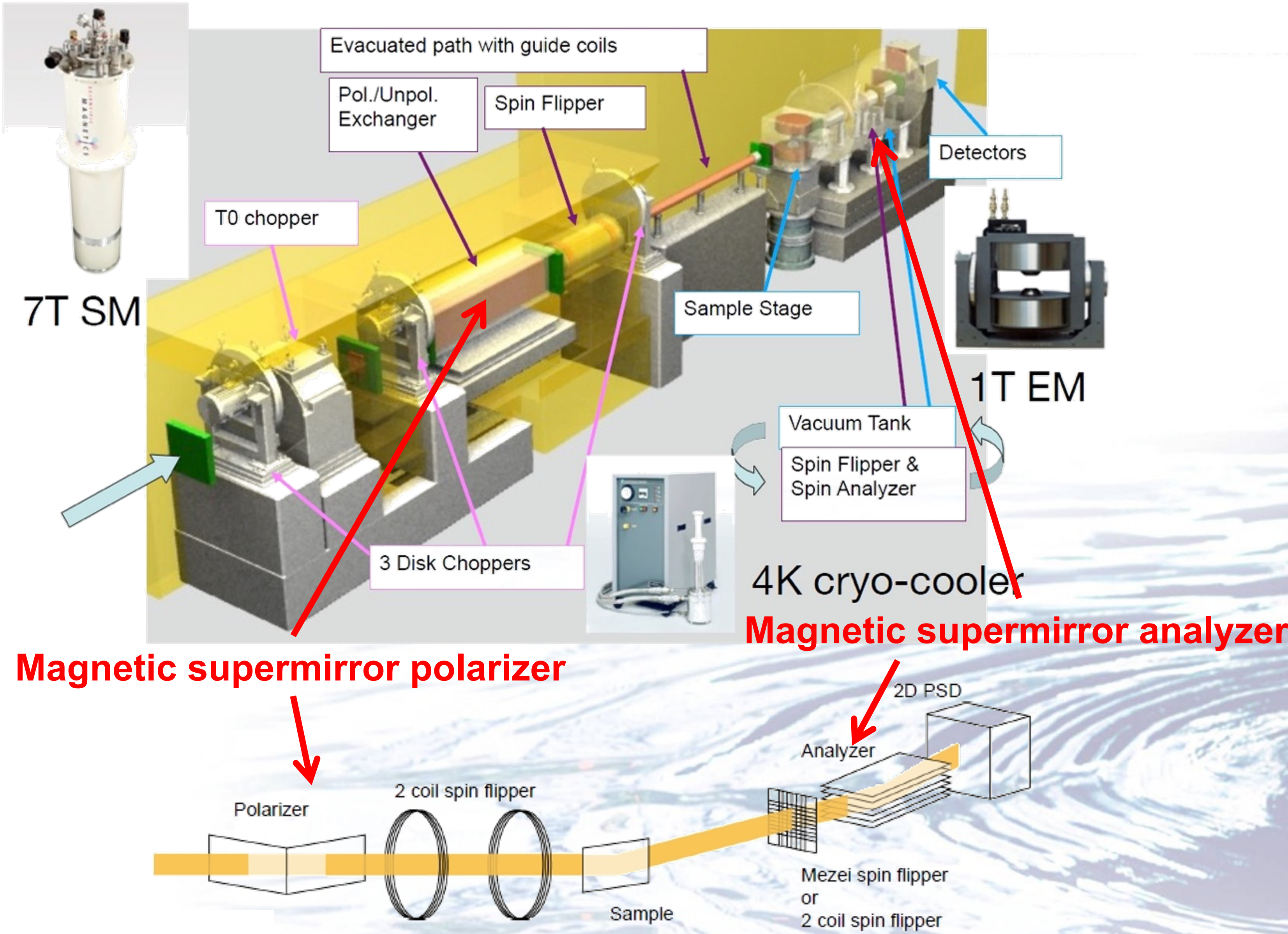
Research	Revealing mechanisms of functions of thin film devices by studying structures of surface and buried interfaces.
Features	Enhanced sensitivity of magnetic moments using polarized neutrons, and a high-precision neutron reflectivity measurement of a wide variety of thin films such as magnetic devices, nonmagnetic films, metallic films, polymer films, regardless of materials.



Application 1

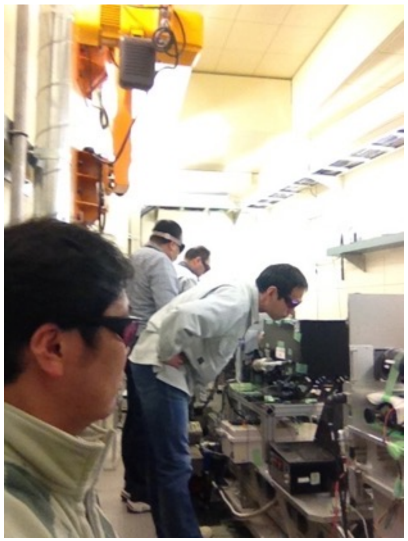
Polarization Analysis in polarized neutron reflectometry

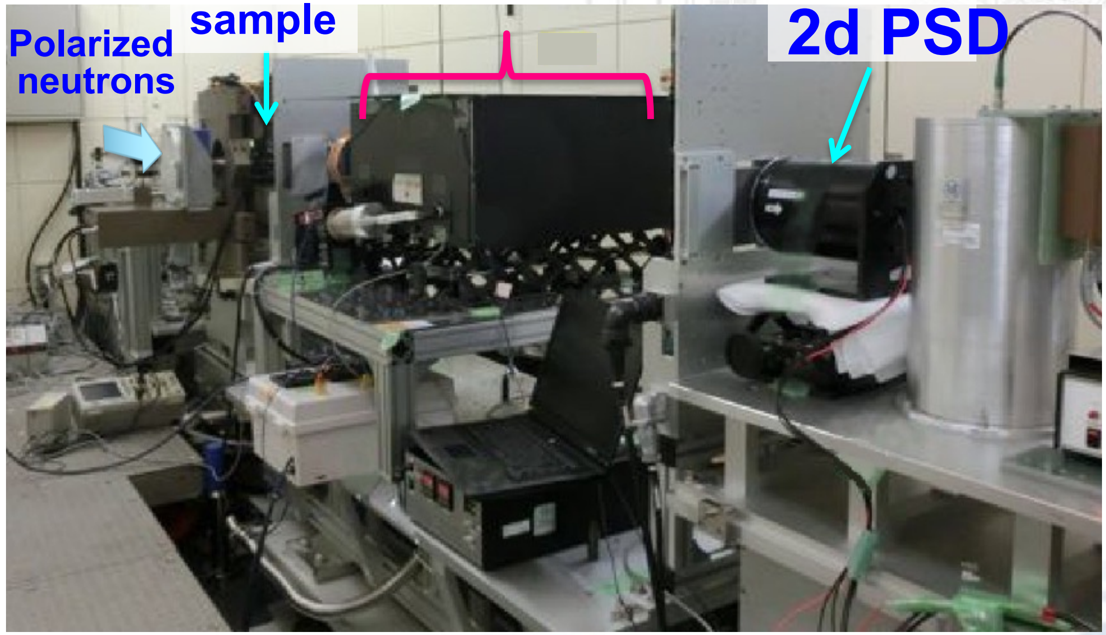
J-PARC/MLF BL17 : Polarized Neutron Reflectometer SHARAKU



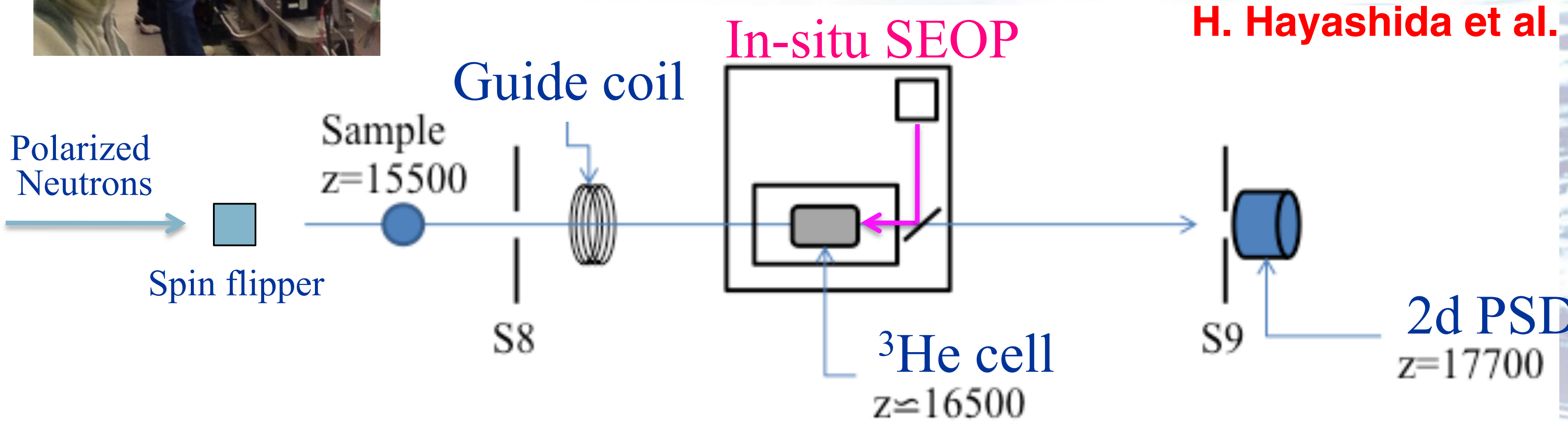
Polarization Analysis in polarized neutron reflectometry

J-PARC/MLF BL17 : Polarized Neutron Reflectometer SHARAKU





H. Hayashida et al.



How polarized neutrons are used at each BL.

Neutron Diffraction

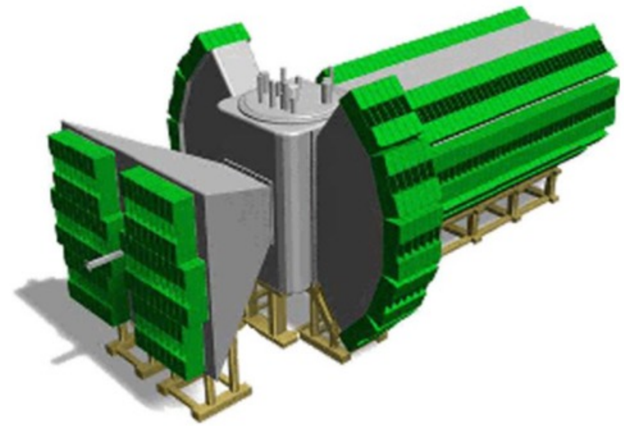
How polarized neutrons are used at BL20.

BL20 iMATERIA IBARAKI Materials Design Diffractometer

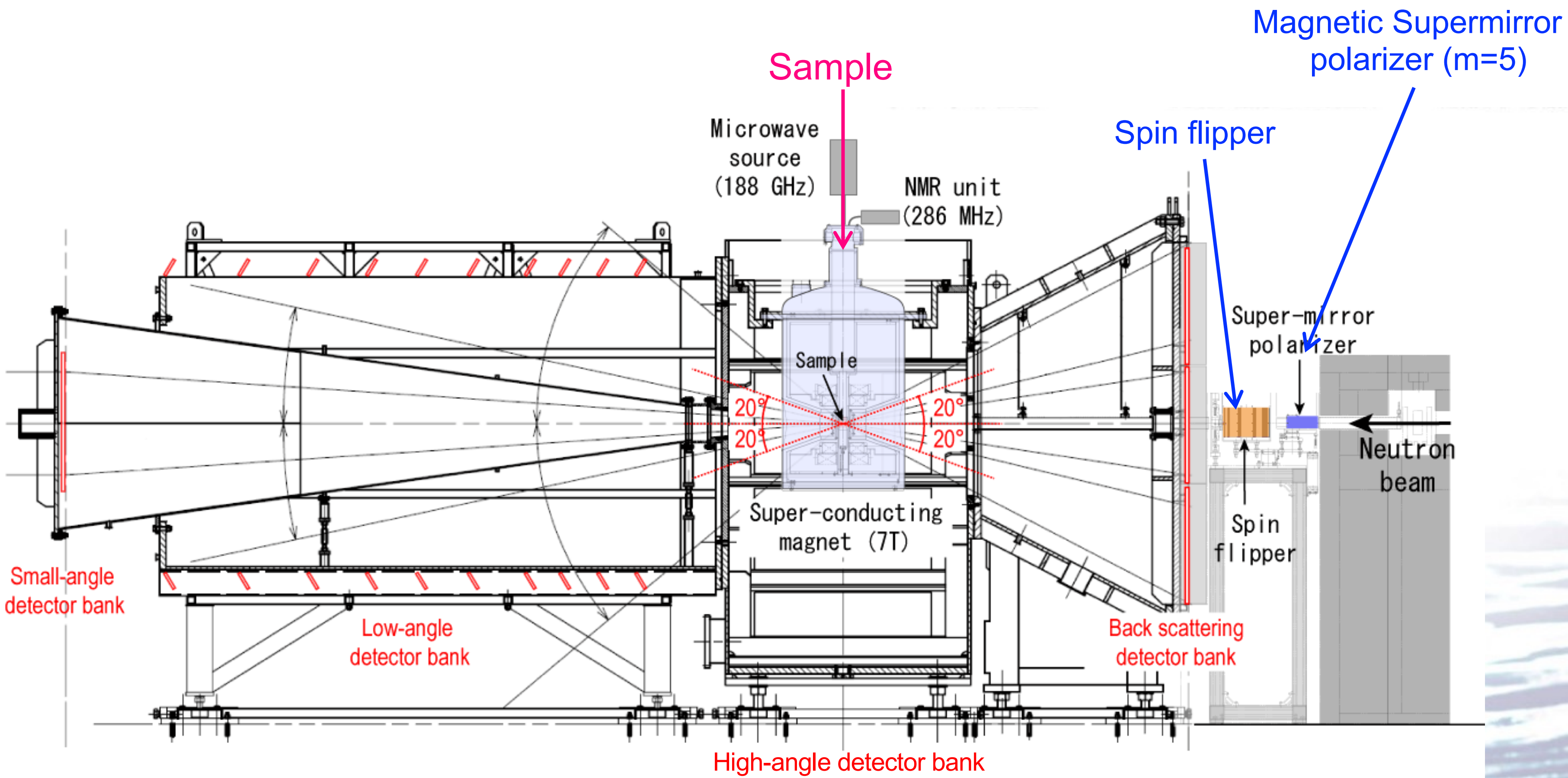
~Diffractometer for the industrial use~

High efficiency and high resolution crystal structure refinements for wide d region

Research	Crystal structure and atomic arrangement refiments for energy device materials, such as Li battery and fuel battery materials.
Features	100 times higher intensity from previous instruments. Various measurement modes using 4 detector banks. Wide d range measurement. Promotion for industrial use.



Schematic layout of iMateria BL20



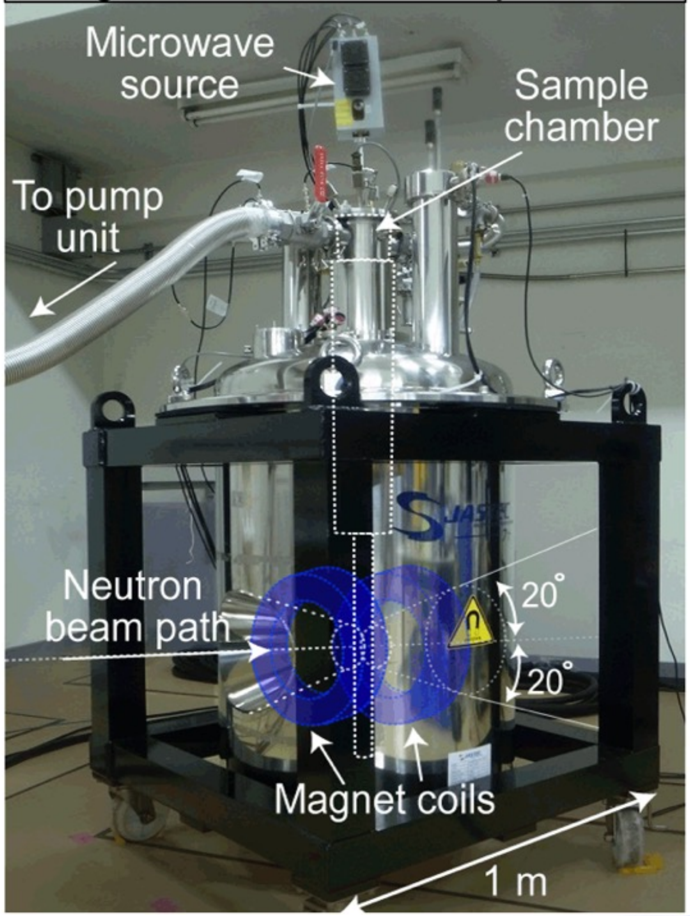
It has 4 detector banks including a small angle detector bank, so that it can cover wide q-range.

DNP-SANS instrument dedicated to industrial use at BL20 iMATERIA

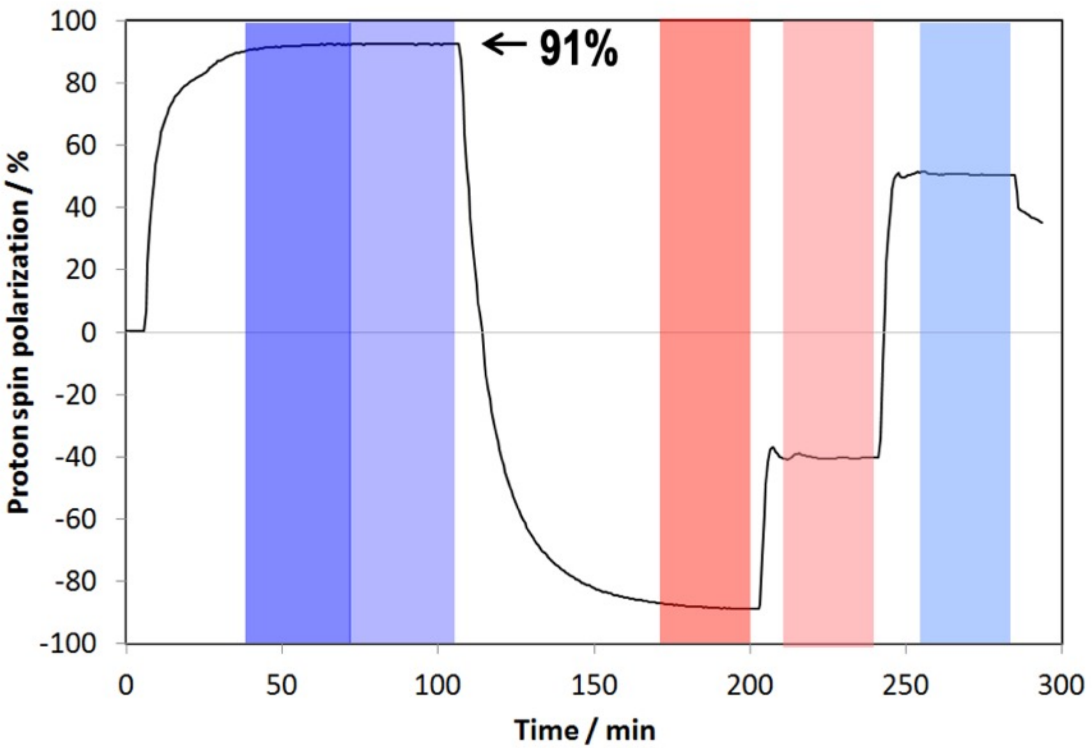
Yohei Noda, Tomoki Maeda, Takayuki Oku, Satoshi Koizumi (Ibaraki Univ.)

The first DNP-SANS instrument satisfying quick sample exchange and contrast variation wider than deuterium substitution.

Magnetic field : **7T**, Temp. : 1.2K



Proton spin polarization **higher than 90%** was achieved for vulcanized rubber sample



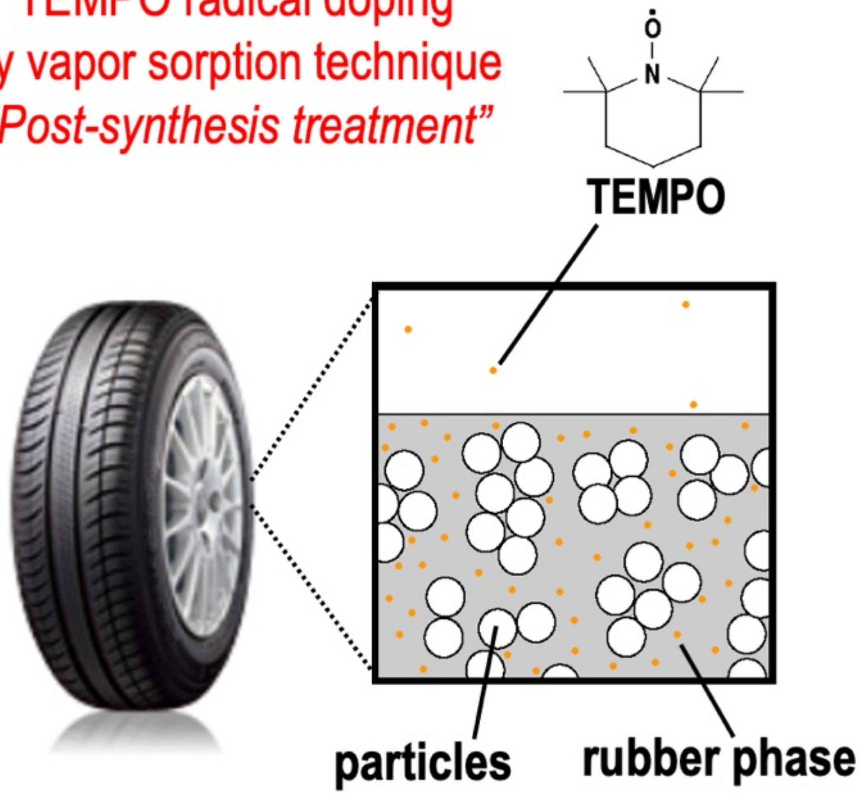
Y.Noda & S. Koizumi, NIMA (2019) 127-133

DNP-SANS instrument dedicated to industrial use at BL20 iMATERIA

茨城大学
Ibaraki University

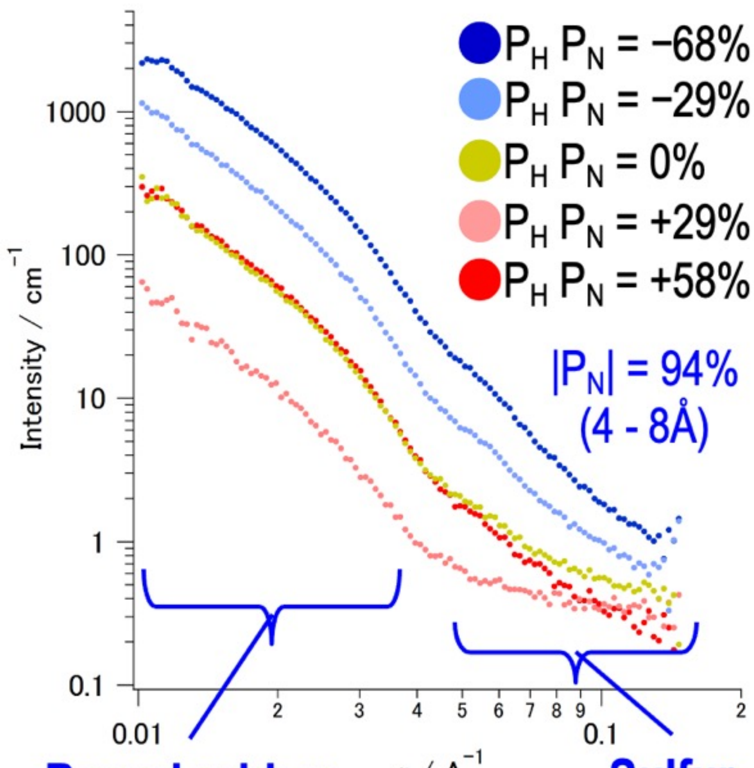
Contrast variation study of industrial rubber samples

TEMPO radical doping
by vapor sorption technique
"Post-synthesis treatment"



Contrast variation of "Product itself"

DNP-SANS of silica-filled rubber (2019 April)
Collaboration with Sumitomo Rubber Industries



Bound rubber around filler particle

Sulfur crosslink

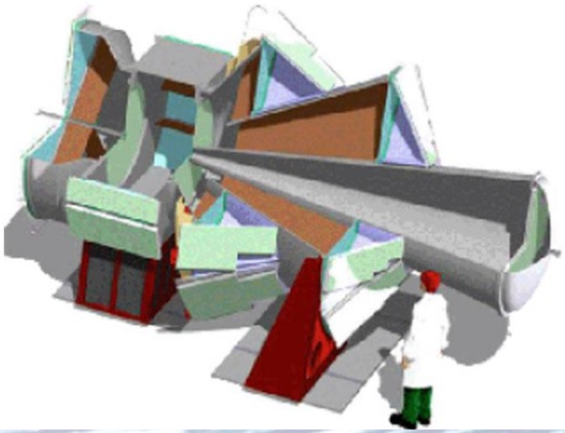
Y.Noda, T.Maeda, T.Oku, S. Koizumi et al., QuBS (2020) 4, 33

How polarized neutrons are used at BL21.

BL21 NOVA High Intensity Total Diffractometer

Investigation of origin of hydrogen-induced physical properties by analysis of atomic position, and hydrogen correlations, and approach from pair correlation in disordered system

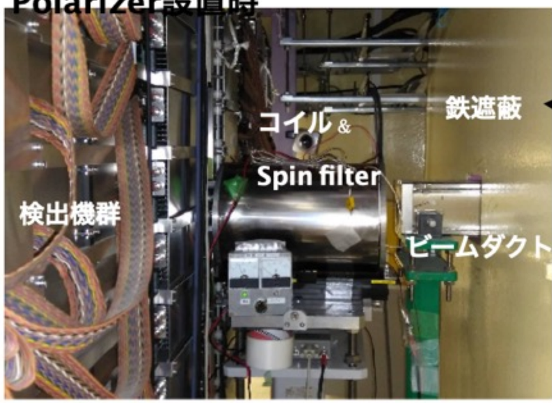
Research	Observation of (magnetic) structural changes or pair correlation in hydrogen-induced physical properties, hydrogen storage materials or amorphous compounds.
Features	Wide range of structural changes can be observed: from nearest atomic distance to nm order distance. The throughput efficiency of one measurement is world's highest as a total diffractometer.

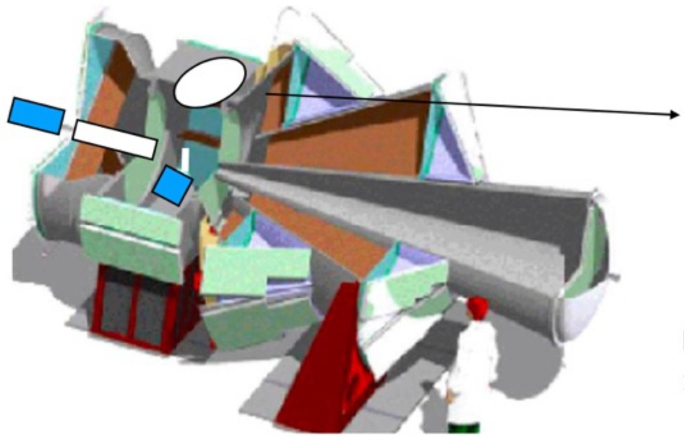


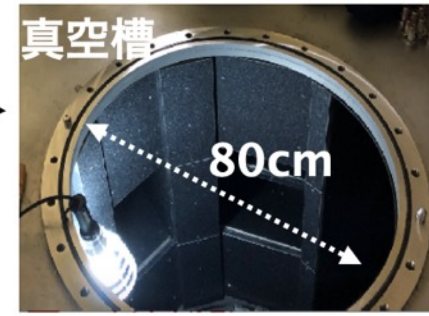
Polarizer and analyzer is ^{3}He spin filter

BL21へのSpin Filterの導入

Polarizer設置時

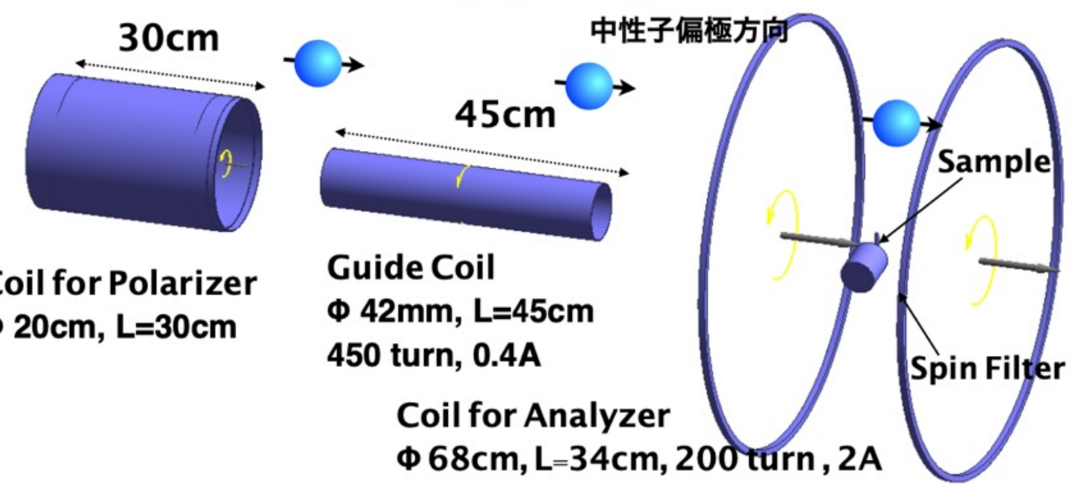






BL21にはサンプルを入れるための真空槽があり、からanalyzer用コイルを入れる。

^{3}He緩和時間60h程度



How polarized neutrons are used at each BL.

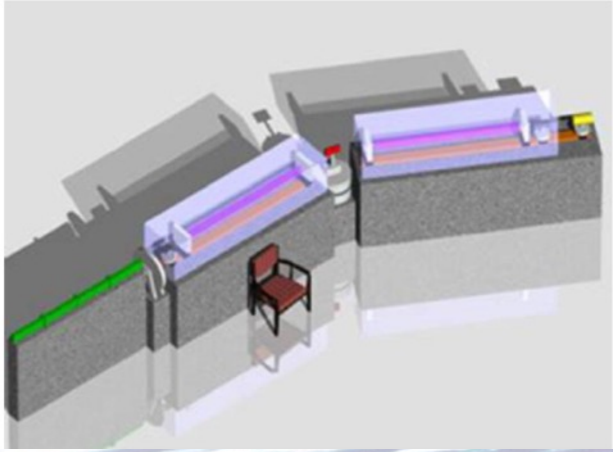
Inelastic Neutron Scattering

How polarized neutrons are used at BL06.

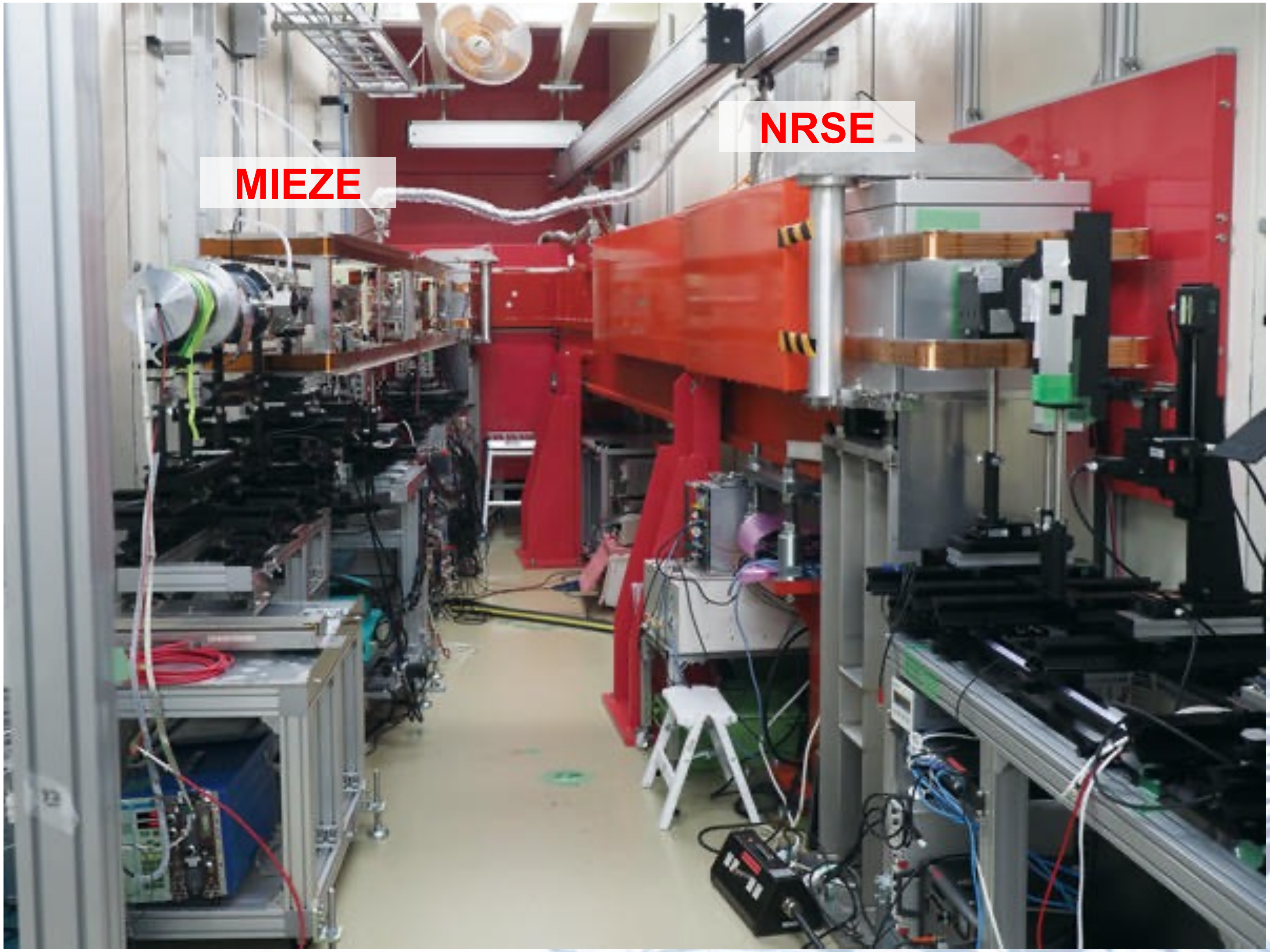
BL06 VIN ROSE Village of Neutron Resonance Spin Echo Spectrometers (VIN ROSE)

Dynamical behaviors from ps to sub-μs

Research	Relaxation properties of small molecules such as water, etc. Diffusion of liquid molecules and assembly. Slow dynamics of glass and large scale structures of magnetic materials and ferroelectrics.
Features	Intermediate structure factor I(Q, t) could be observed directly. The highest energy resolution without losing intensity.



BL06 VIN ROSE



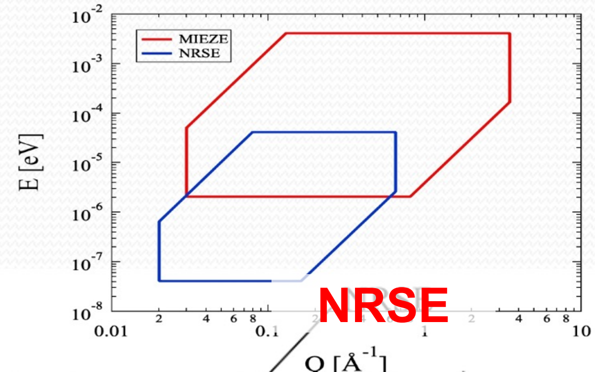
The neutron beamline is divided into two branches; one is for MIEZE and another one is NRSE(Neutron Resonance SE).

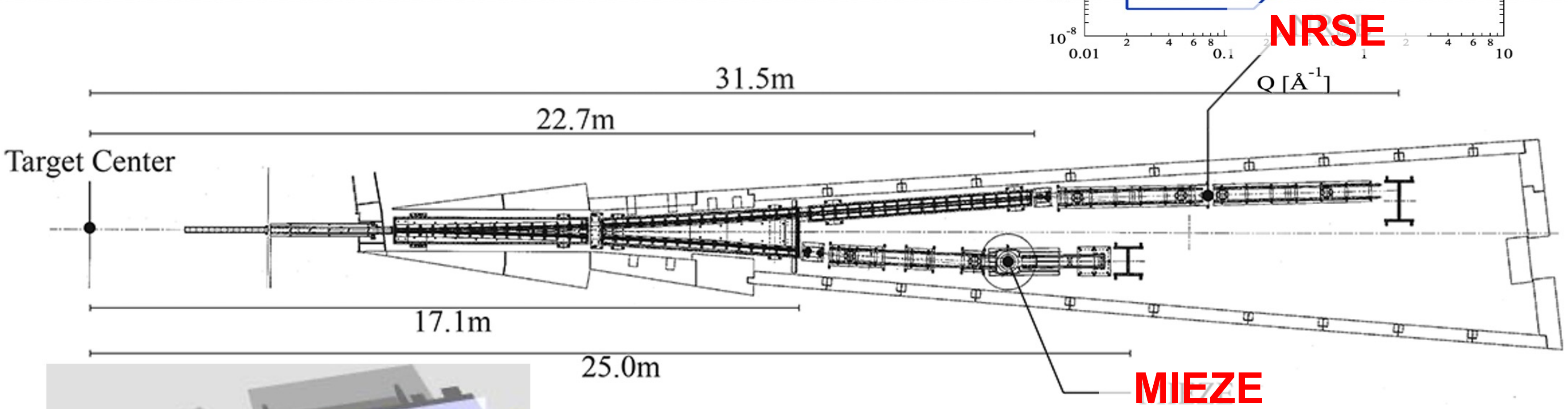
Application of Polarized Neutrons at MLF

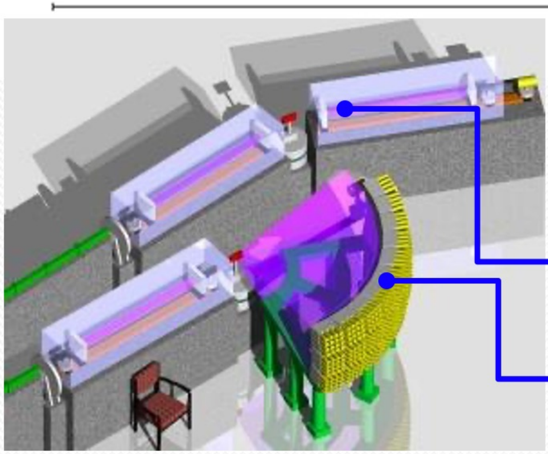
BL06 VIN-ROSE

Village of Neutron Spin Echo Spectrometers

Installing ***2 NSE spectrometers*** with resonance spin flippers (NRSE + MIEZE).



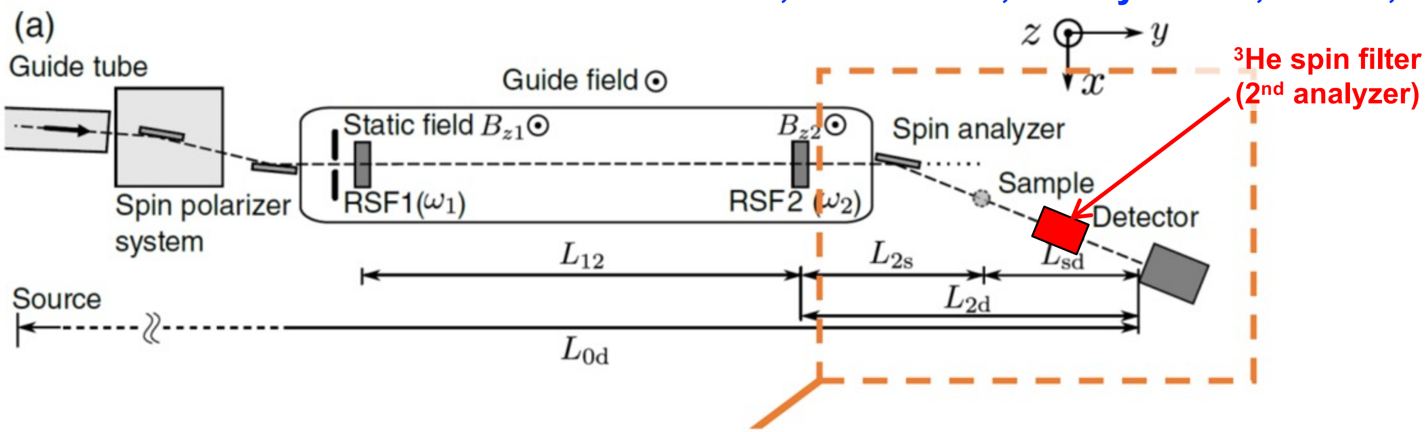




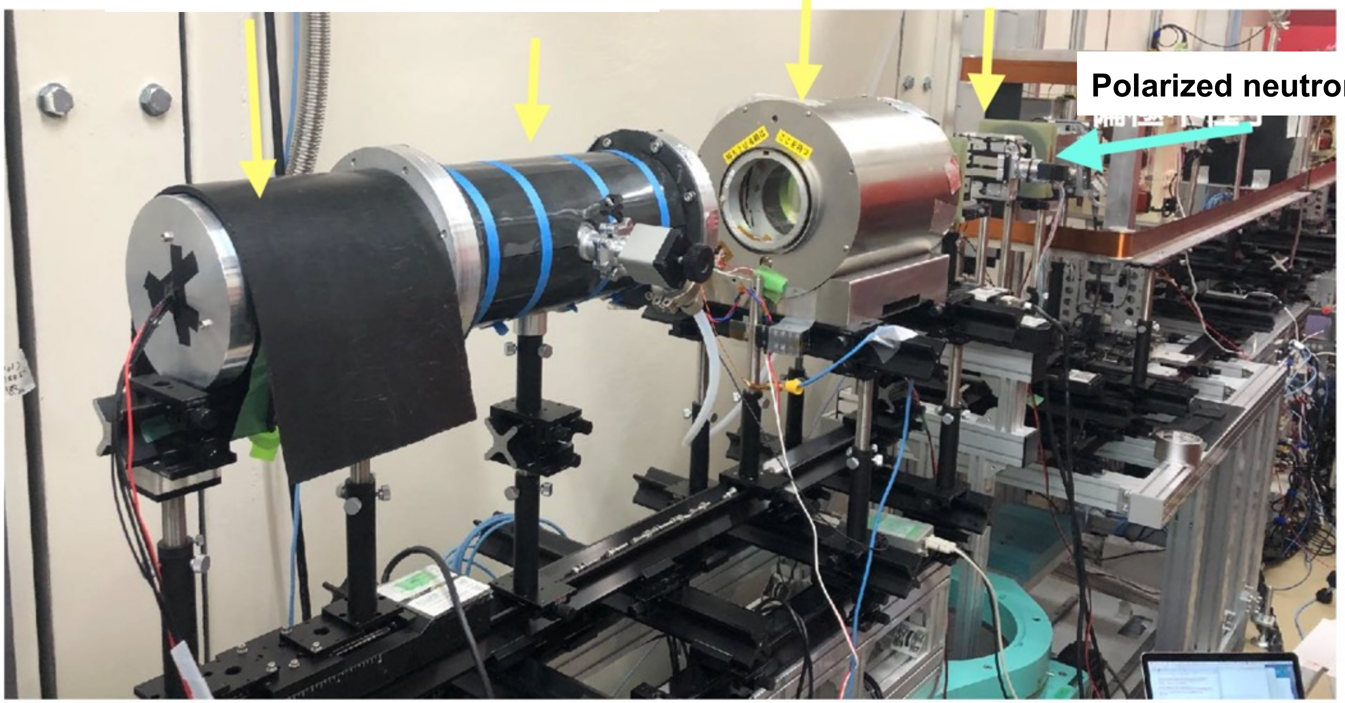
Spectro-meter	Wave-length [Å]	Q range [Å^{-1}]	Fourier Time
NRSE	$5 < \lambda < 20$	$0.02 < Q < 0.65$	0.1 [ns] < t < 0.1 [μs]
MIEZE	$3 < \lambda < 13$	$0.03 < Q < 3.5$	1 [ps] < t < 2 [ns]

^{3}He spin filter as 2nd analyzer for MIEZE

T. Oda, T. Okudaira, H. Hayashida, T. Oku, M. Hino, and H. Endo



2d-detector
Vacuum tube
^{3}He spin filter
Sample



Sample:
Ferro-fluid

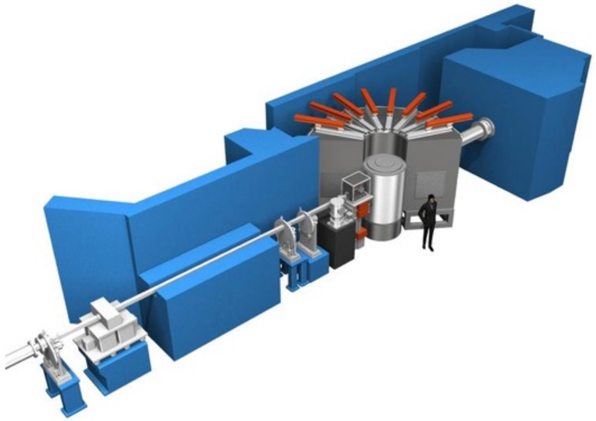
How polarized neutrons are used at BL23.

J-PARC, MLF

BL23 POLANO Polarized Neutron Spectrometer

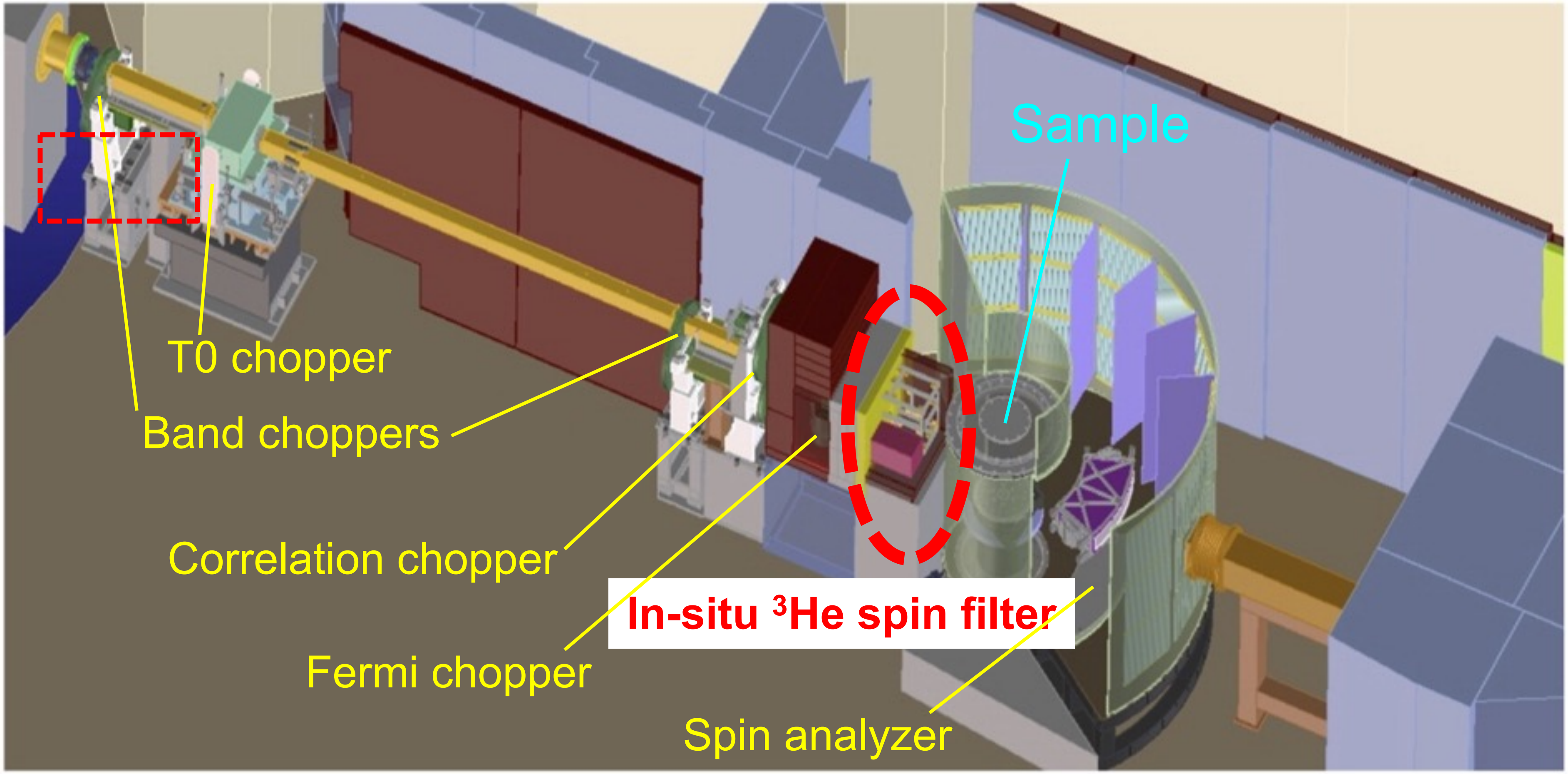
POLANO enables polarization analysis of inelastic scattering in the dynamics of multiple degrees of freedom

Research	Dynamics of spin, orbital, charge, and lattice in magnetic, superconducting, functional materials
Features	POLANO is a direct geometry spectrometer with middle energy and spatial resolution. Combination of SEOP ^{3}He gas spin filter and 5.5 Qc bending mirror analyzer makes it possible higher-energy polarization experiment.



First chopper spectrometer dedicated to polarized neutrons

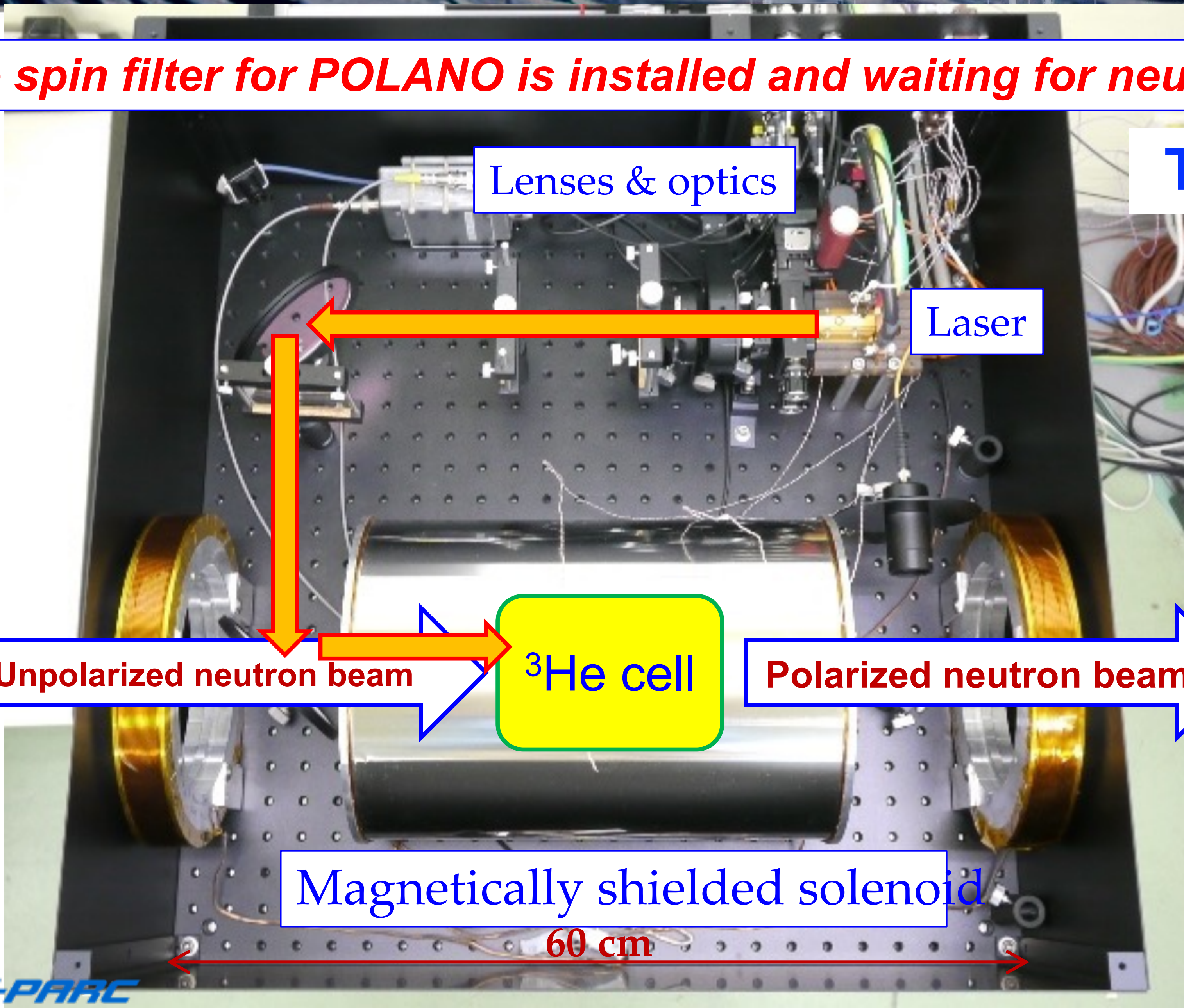
T. Yokoo, T. Ino et al.



In-situ polarized ^{3}He spin filter for POLANO

^{3}He spin filter for POLANO is installed and waiting for neutrons.

T. Ino



Polarized neutron capability of POLANO

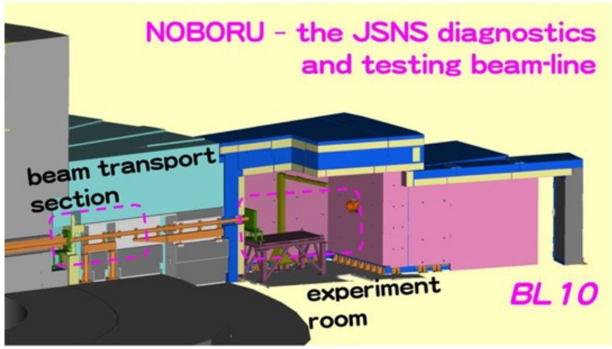
- Incident neutron beam polarization by using *in-situ* SEOP ^{3}He spin filter whose gas pressure length:10 to 30 bar·cm which cover neutrons whose energy from10 to 100 meV
- AFP-NMR is equipped to the ^{3}He spin filter to flip ^{3}He spin polarity.
- The scattered neutrons are analyzed by
  - 1st phase : magnetic supermirrors, E_f~40meV
  - 2nd phase : ^{3}He spin filter, E_f~100meV

How polarized neutrons are used at BL10.

BL10 NOBORU NeutrOn Beam-line for Observation and Research Use

Serving a versatile neutron field for characterizing the neutron source as well as for R&D on various devices, irradiation and analysis of materials, etc

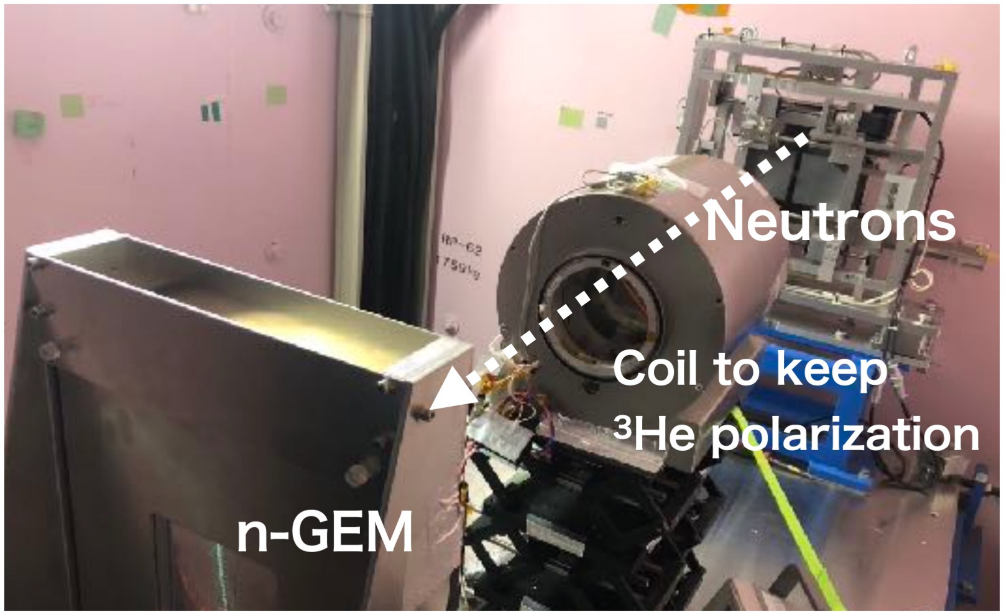
Research	Development of novel experimental methods using pulsed neutrons. Performance evaluation of neutron optics, detectors, etc. Evaluation of radiation resistance of devices using high energy neutrons.

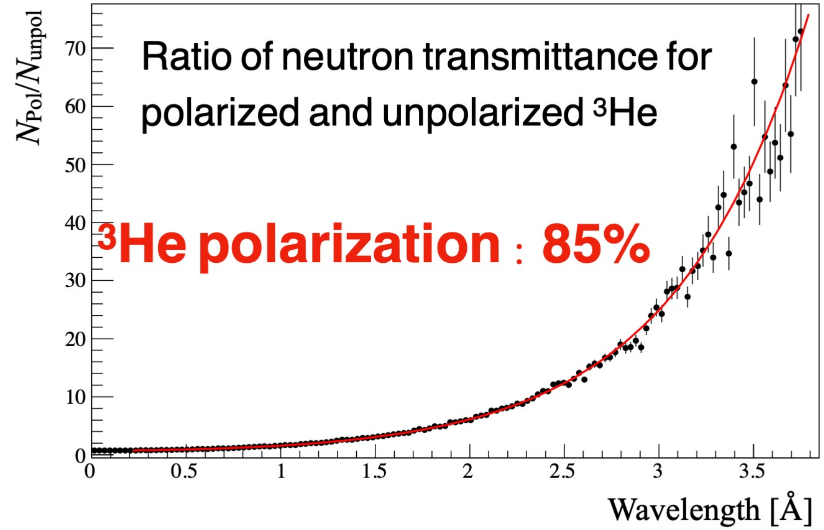


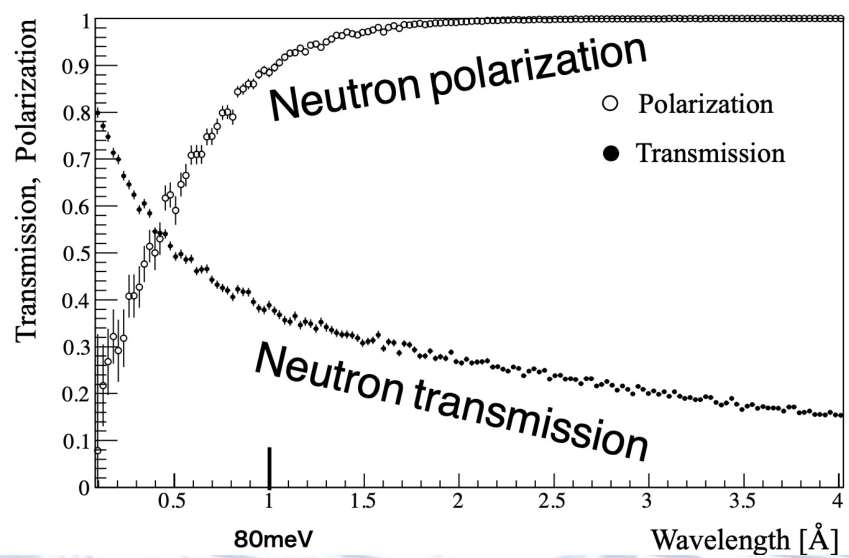
Neutron polarization measurement

^{3}He cell fabricated at J-PARC was polarized by the laser system

After laser irradiation, ^{3}He cell was transported to beamline with a magnetic field applied by battery.





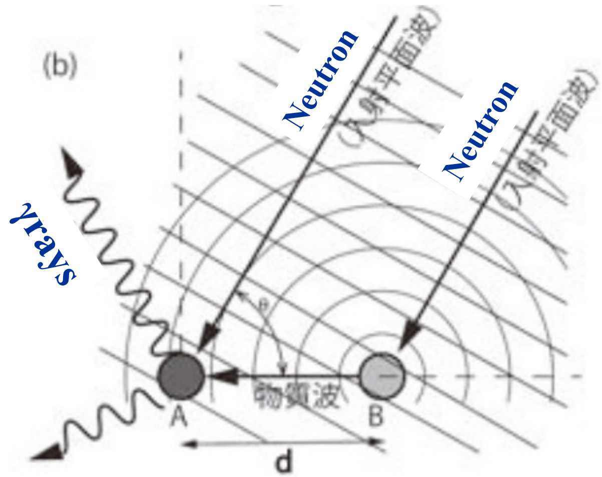


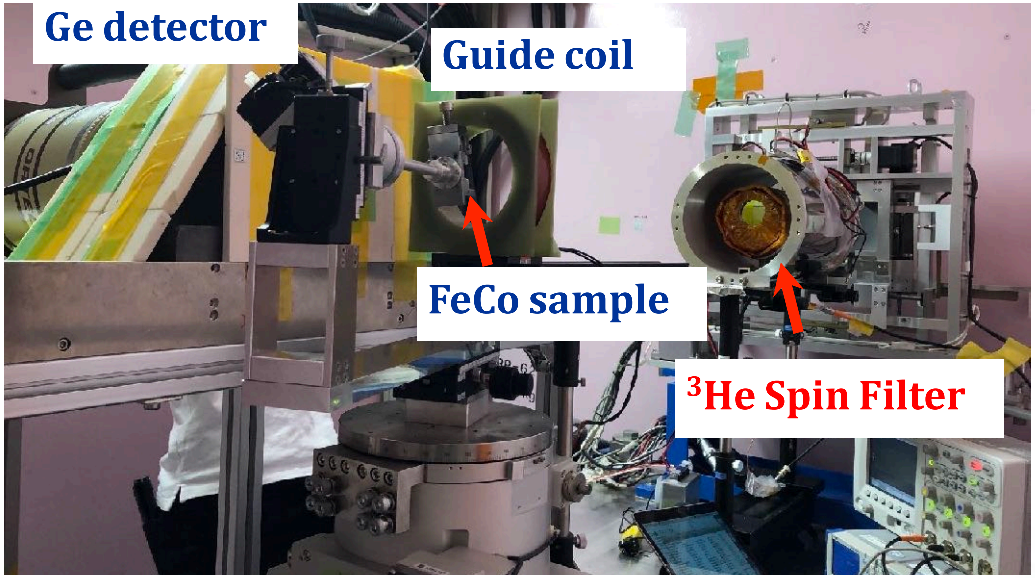
^{3}He relaxation at beamline: 174h

^{3}He thickness : 20atm cm

High ^{3}He polarization was achieved!

Polarized Neutron holography experiments at BL10





Ohoyama (Irabaki Univ.)
Hayashi (Nagoya Institute of Tech.)
Harada, Oikawa, Inamura et al. (JAEA)

They have performed neutron holography experiments to study structure around the dopants.

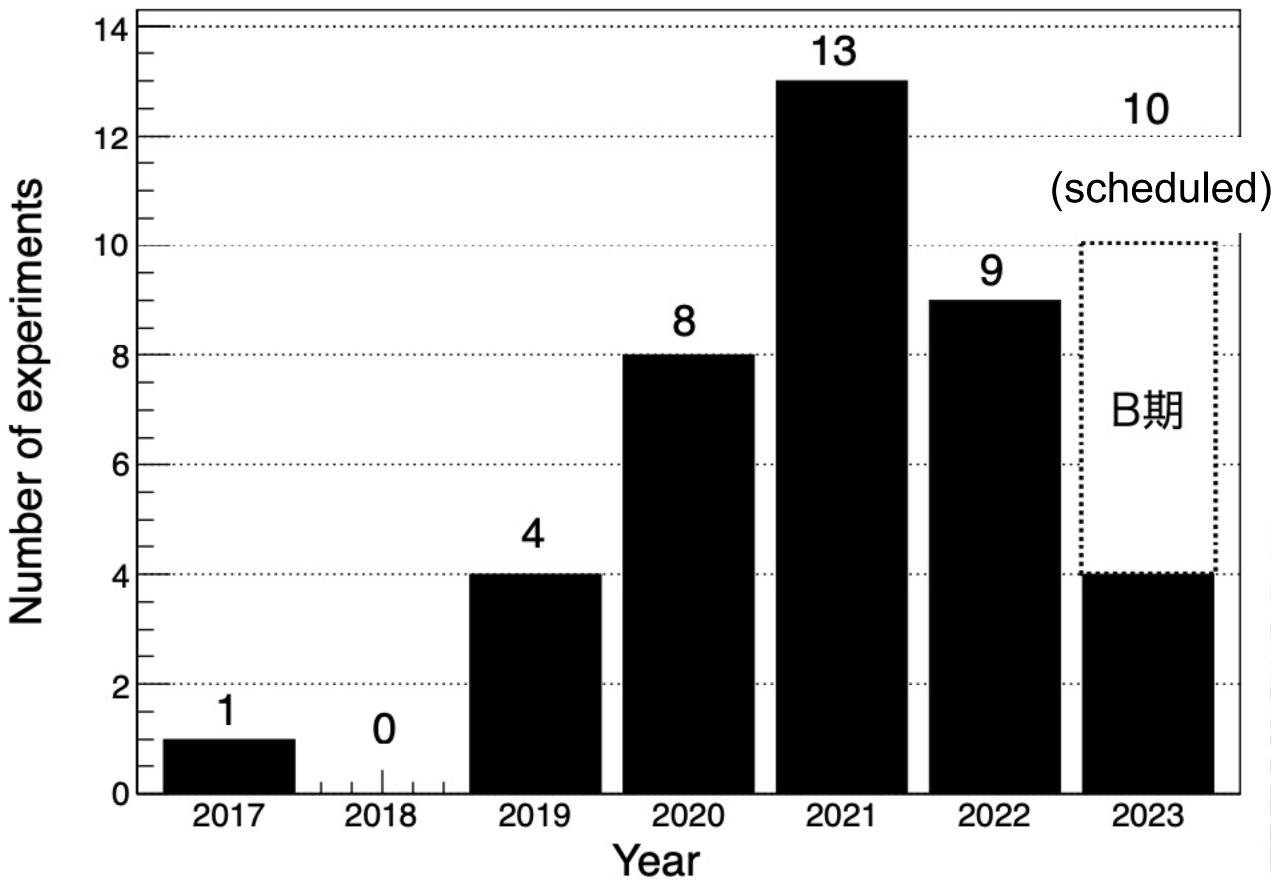
Ohoyama, Hayashi, Harada, Oikawa, Inamura, Okudaira, Oku et al.

We have started to try polarized neutron holography experiments, but we have not gotten reasonable results so far.

^{3}He spin filter

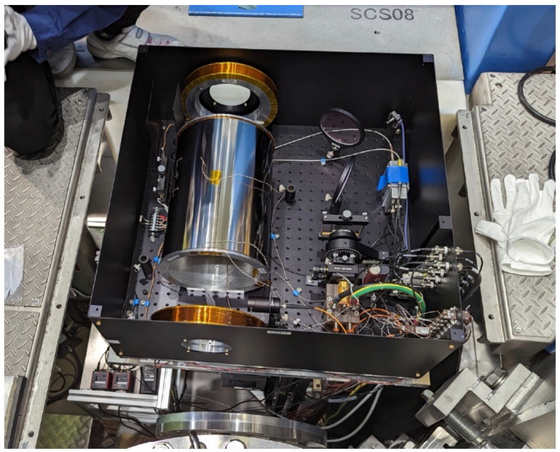
- Gas pressure length:19atm-cm
- Cell size: φ3.5 cm
- P_{3He} ~ 85%
- Relaxation time ~ 160hrs

Numbers of experiments with ^3He spin filter



To make the most use of polarized neutron

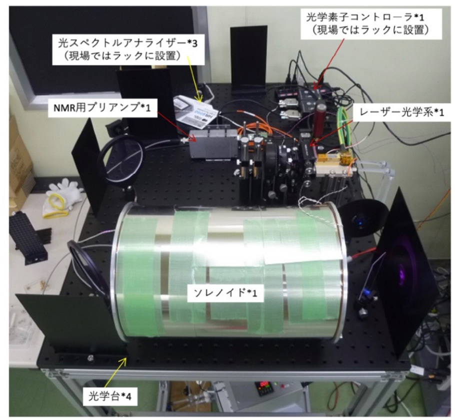
Some setups of In-situ SEOP are going to be used.



J-PARC MLF BL23
Under commissioning
Almost ready for user experiments

T. Yokoo, T. Ino et al.

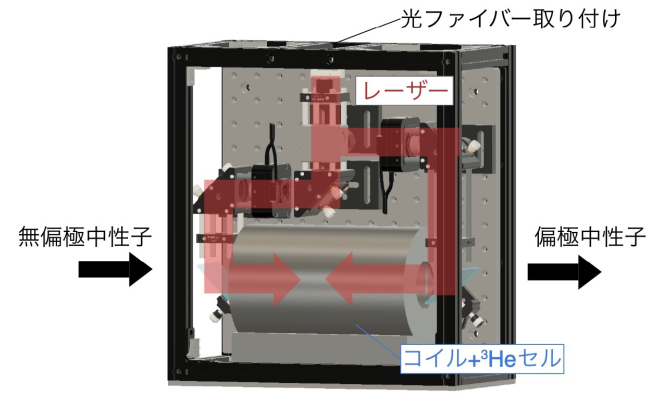
6G TOPAN用装置



JRR-3 6G TOPAN
Installation started

Y. Ikeda, T. Ino, S. Takada, M. Fujita et al.

BL18 SENJU用装置



J-PARC MLF BL18
Under preparation

S. Takahashi, T. Oku
R. Kiyanagi et al.

Current status and perspective

	Current status			Future (expected)	
BL	Polarizesd neutrons	How	What	BL	Polarizesd neutrons
BL01				BL01	
BL02				BL02	✓
BL03				BL03	
BL04	✓	optional	3He	BL04	✓
BL05	✓	regular use	Mirror (3He is optional)	BL05	✓
BL06	✓	regular use	Mirror (3He is optional)	BL06	✓
BL08				BL08	
BL09				BL09	
BL10	✓	optional	Mirror and 3He	BL10	✓
BL11				BL11	
BL12				BL12	
BL14				BL14	✓
BL15	✓	regular use	Mirror and 3He	BL15	✓
BL16				BL16	
BL17	✓	regular use	Mirror	BL17	✓
BL18	✓	Some tests done. Under preparation for regular use	3He	BL18	✓
BL19				BL19	
BL20	✓	optional	Mirror	BL20	✓
BL21	✓	Some tests done. Under preparation for regular use	3He	BL21	✓
BL22	✓	regular use	Mirror	BL22	✓
BL23	✓	regular use	Mirror and 3He	BL23	✓

At MLF, polarized neutrons are used at 11 beamlines now, and are expected used at 13 beamlines in a future.

Thank you for your attention!

Developments of neutron polarizers at J-PARC MLF

- SEOP based ^{3}He spin filters

  T. Okudaira, T. Ino, K. Sakai, H. Kira, H. Hayashida

  S. Takahashi, K. Hiroi, T. Oku

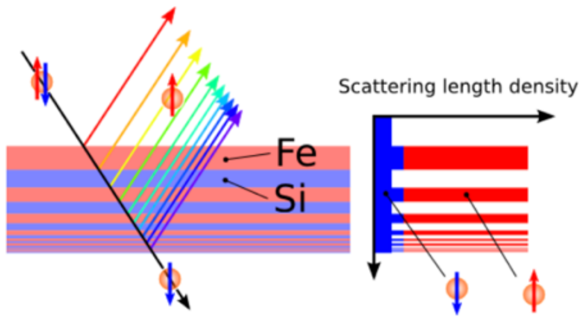
- Magnetic supermirror polarizers

  Ryuji Maruyama

Polarizing supermirror

How to polarize?

- A stack of alternating layers of ferromagnetic and non-magnetic materials
- A neutron beam is polarized by making use of the dependence of the scattering length density (SLD) on the neutron-spin



$$\text{SLD} = N(b_n \pm b_m)$$

N : Atomic number density

$b_{n(m)}$: Nuclear (Magnetic) scattering length

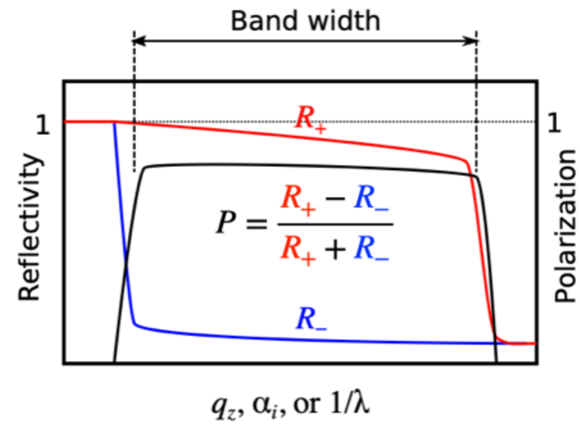
Advantages

- High polarization without loss of intensity
- Uniform polarization over a wide bandwidth
- Maintenance free

For higher performance…

- High polarization → SLD contrast matching
- High performance under low field → Magnetically soft layers
- **Wide bandwidth → Thinner and more layers**

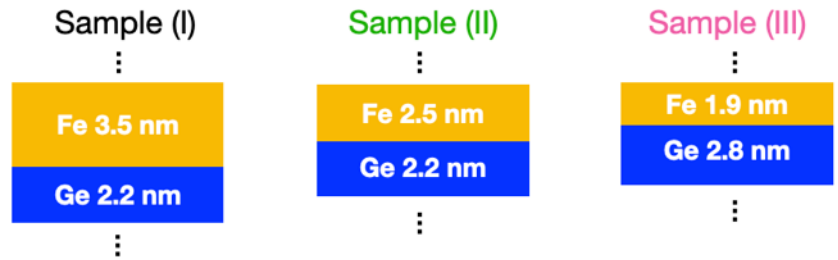
Spontaneous magnetization of Fe layer disappears at a thickness less than 3 nm…

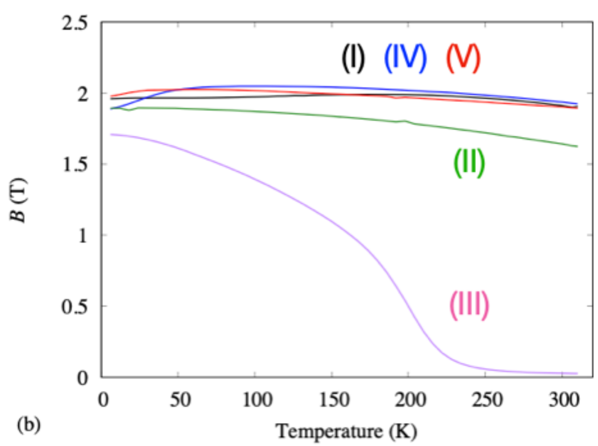


Magnetization measurement

Fe/Ge multilayers (50 bilayers)

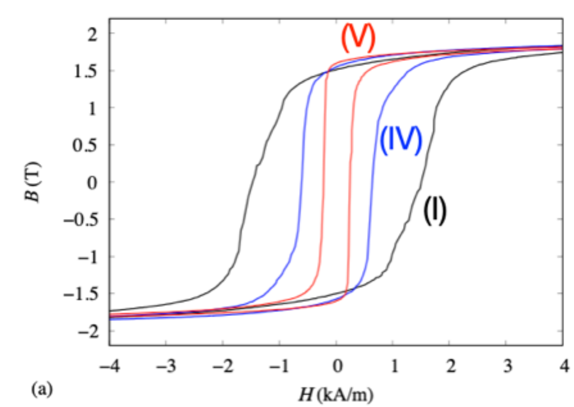
When the Fe thickness is reduced to less than 3nm…





The Curie point : lower than room temperature

Instead, when we keep the Fe thickness and reduce the Ge thickness…



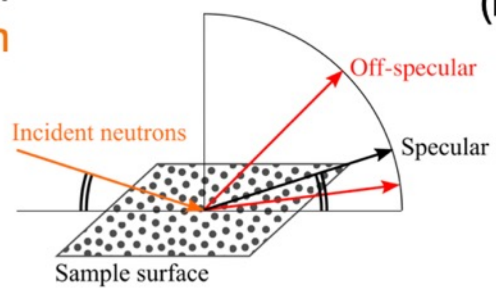
- The magnetization is kept at a value close to (I).
- The sample becomes soft with decreasing Ge thickness.

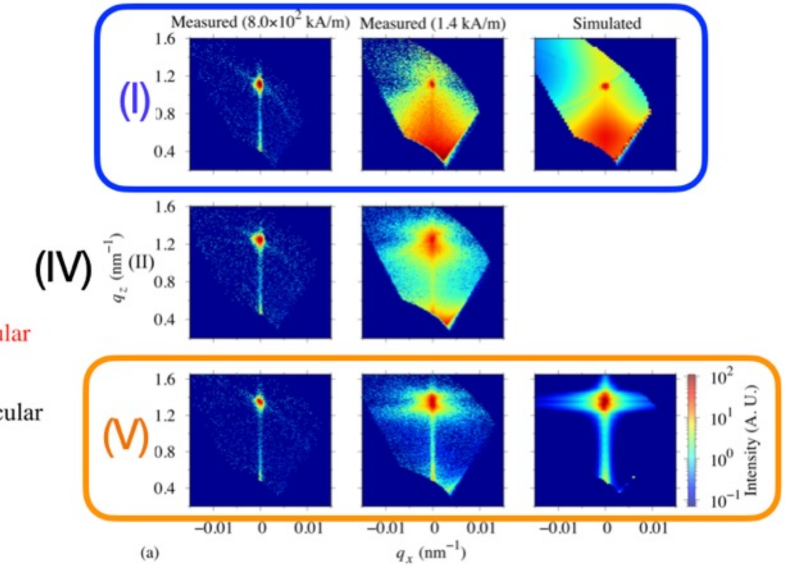
Polarized neutron off-specular scattering measurement

BL17 at MLF

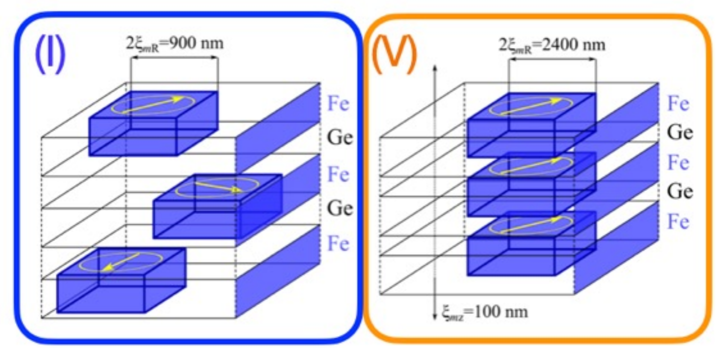
Samples: Fe/Ge multilayers (50 periods)

- (I) Fe: 3.5 nm, Ge: 2.2 nm
- (IV) Fe: 3.5 nm, Ge: 1.7 nm
- (V) Fe: 3.5 nm, Ge: 1.2 nm



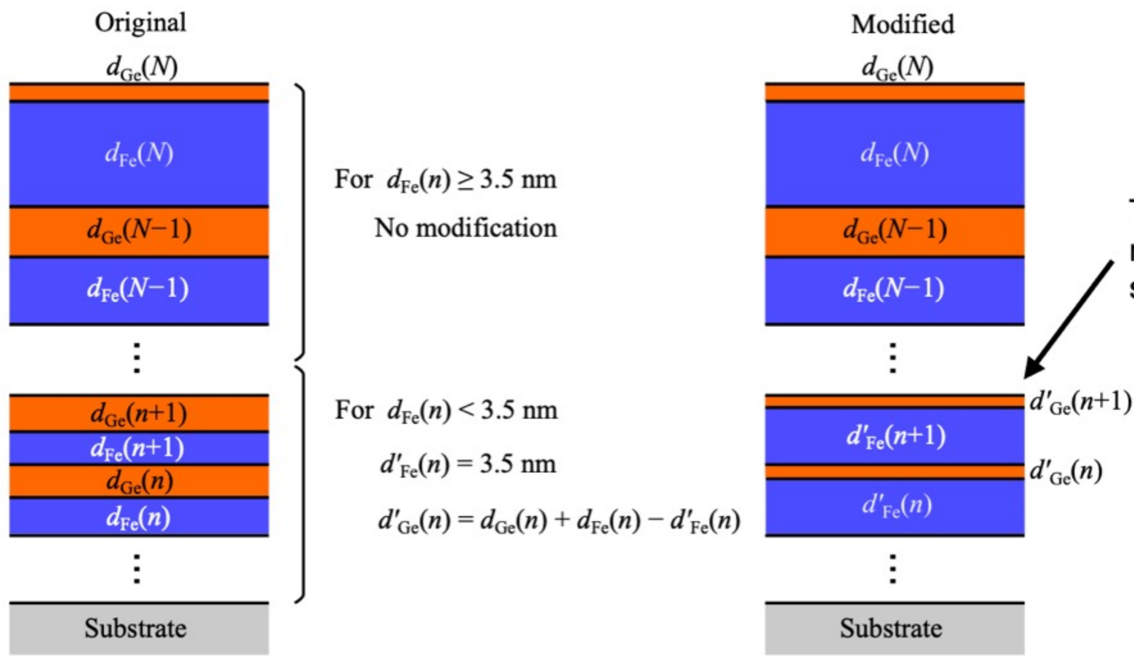


- **Ferromagnetic (FM) interlayer exchange coupling** is seen when the Ge layer thickness is less than 2 nm.
- FM interlayer exchange coupling contributes to the stable spontaneous magnetization and soft magnetic properties.
- This result offers a possibility to extend the bandwidth of the polarizing supermirror.



Design of layer thickness sequence

Aimed to a larger *m*-value



To stabilize the spontaneous magnetization and to obtain soft magnetic properties…

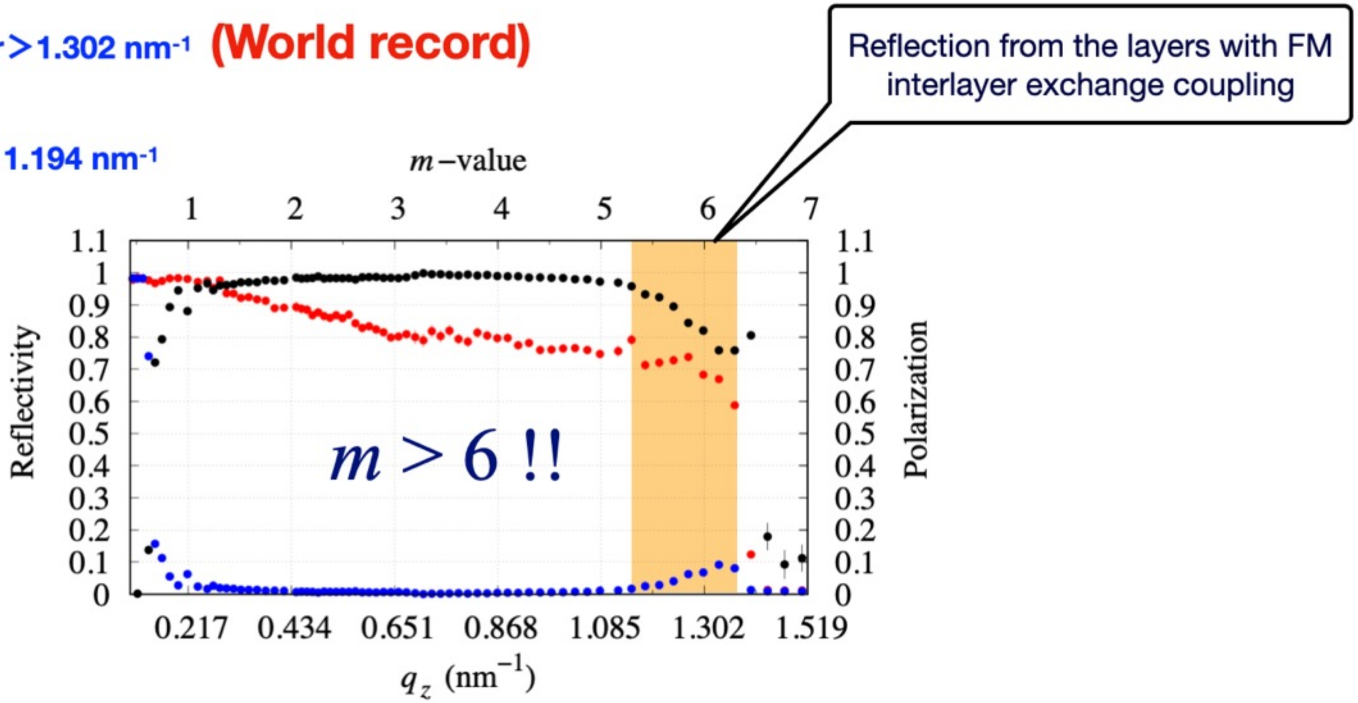
Hayter & Mook algorithm
Both thicknesses (Fe and Ge) are reduced.

Minimum Fe thickness: 3.5 nm
Instead, the thickness of Ge layer is reduced.

Performance test of polarizing supermirror

Measured at BL17

- Total number of layers: 10436
- Total thickness: 32μm
- **Critical momentum transfer＞1.302 nm^{-1}** **(World record)**
- Spin-up reflectivity ＞ 0.70
- **Polarization：＞0.93 for q＜1.194 nm^{-1}**



R. Maruyama *et al*., J. Appl. Phys. **130**, 083904 (2021)