19th meeting on high-energy QCD and nucleon structure
2nd September 2022 @ Yamagata university

Polarized targets for spin-contrast-variation neutron scattering

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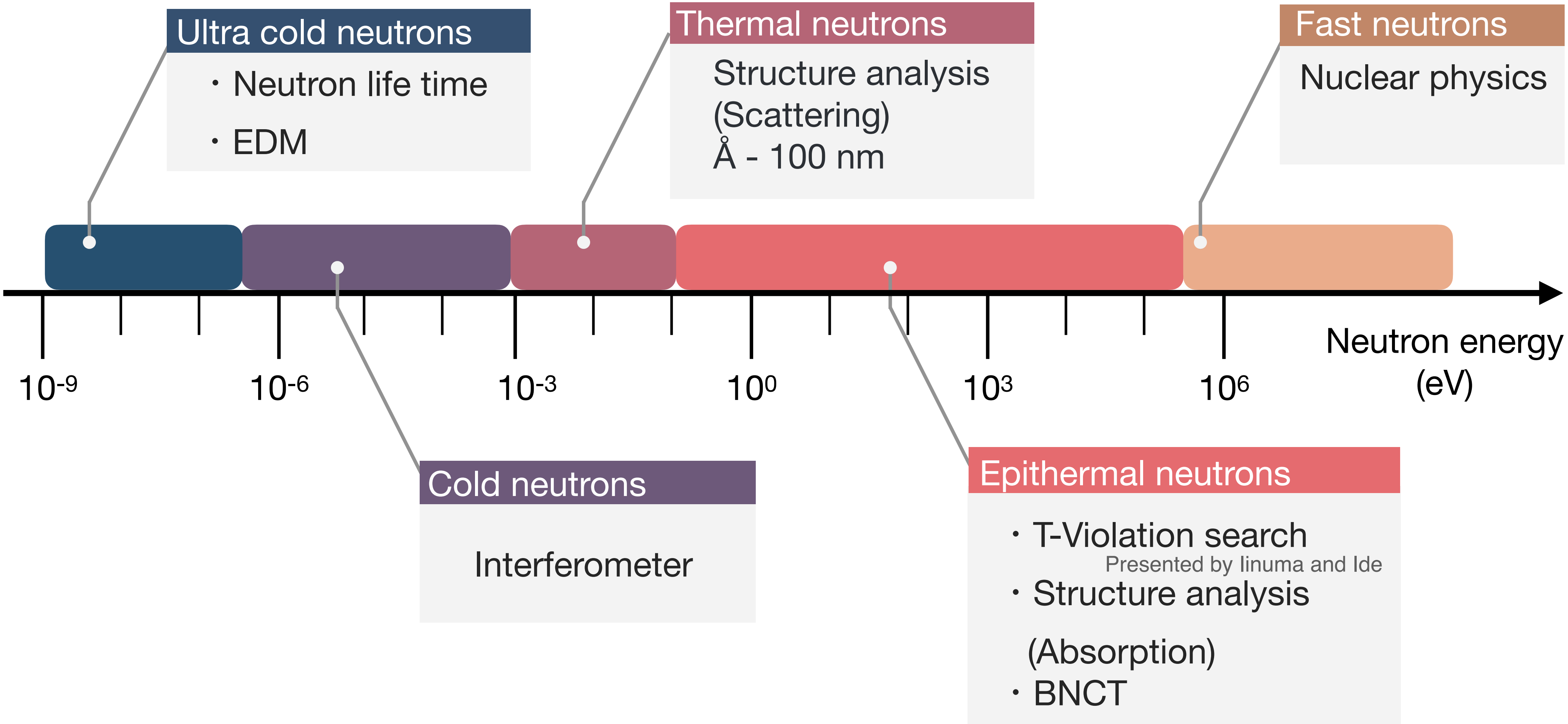
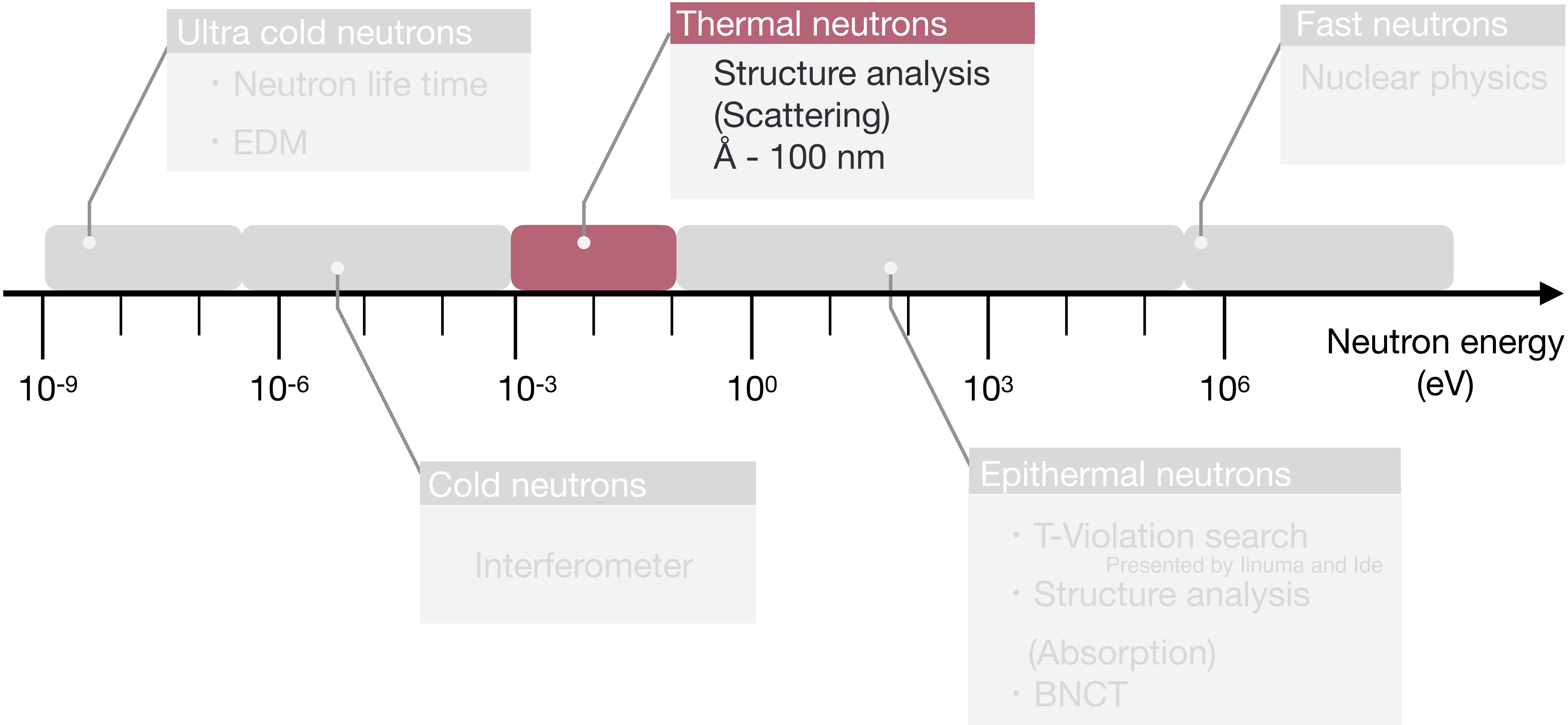


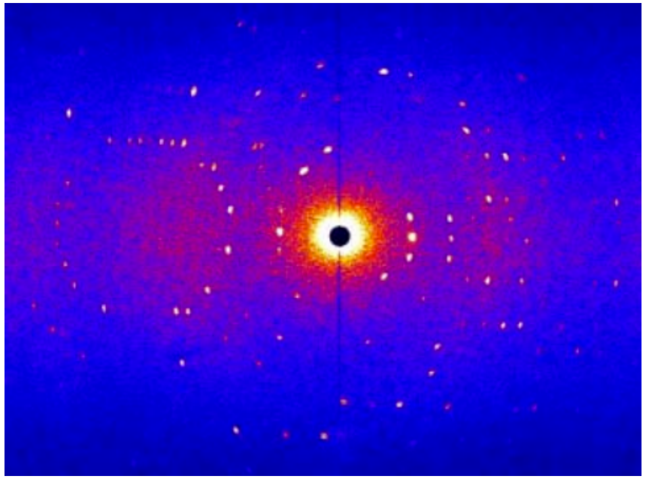
Introduction | Neutron energy



Introduction | Characteristics of neutrons

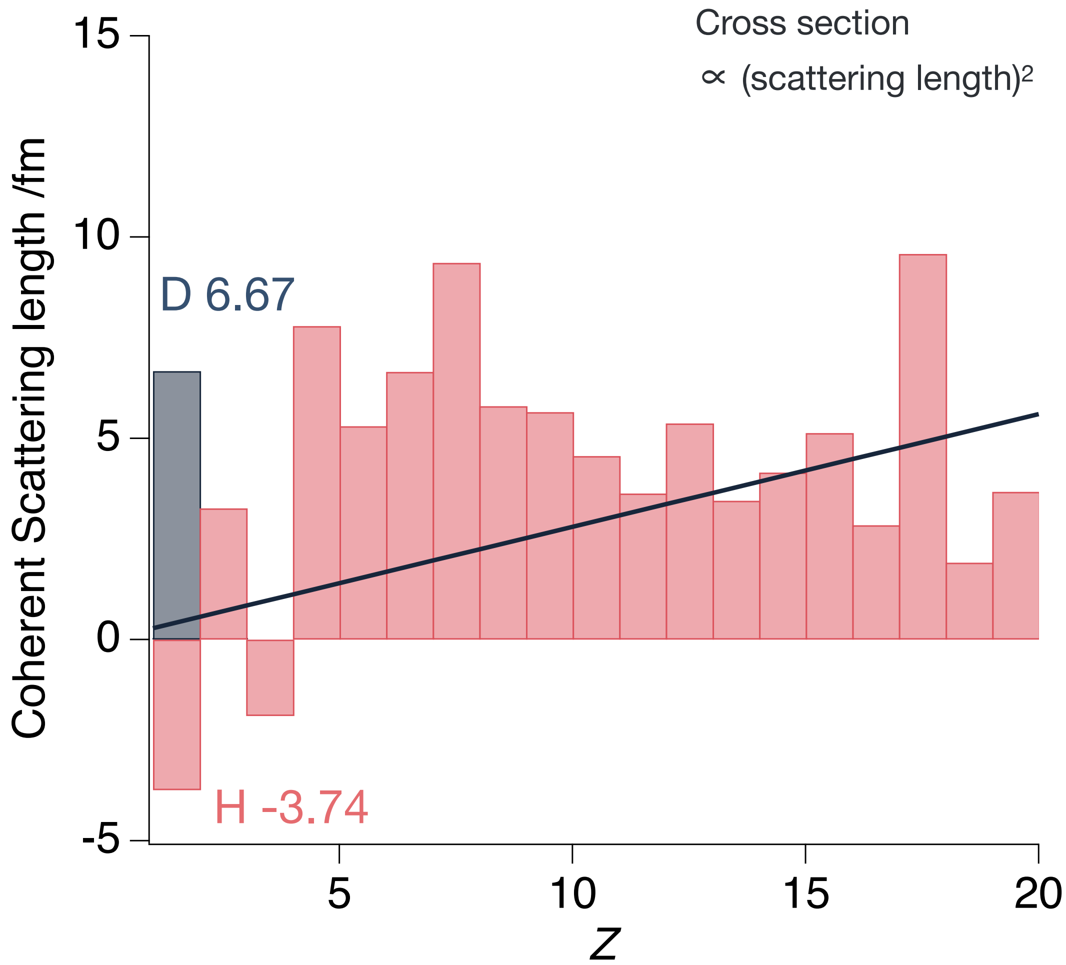
- Sensitivity of the neutron cross section to lighter elements
- Sensitivity to isotopes
- Two types of scattering

Coherent and Incoherent



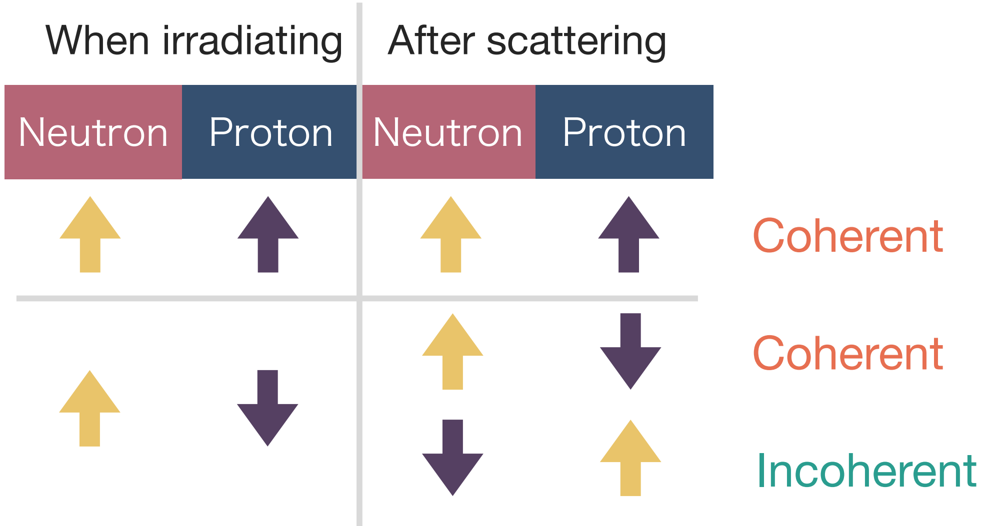
Coherent scattering (Laue pattern)

Incoherent scattering (Isotropically)



Neutron scattering | Spin dependence of proton

Coherent (spin non-flip) and Incoherent (spin flip) scattering



In spin singlet : $b_1 = 10.82$ fm

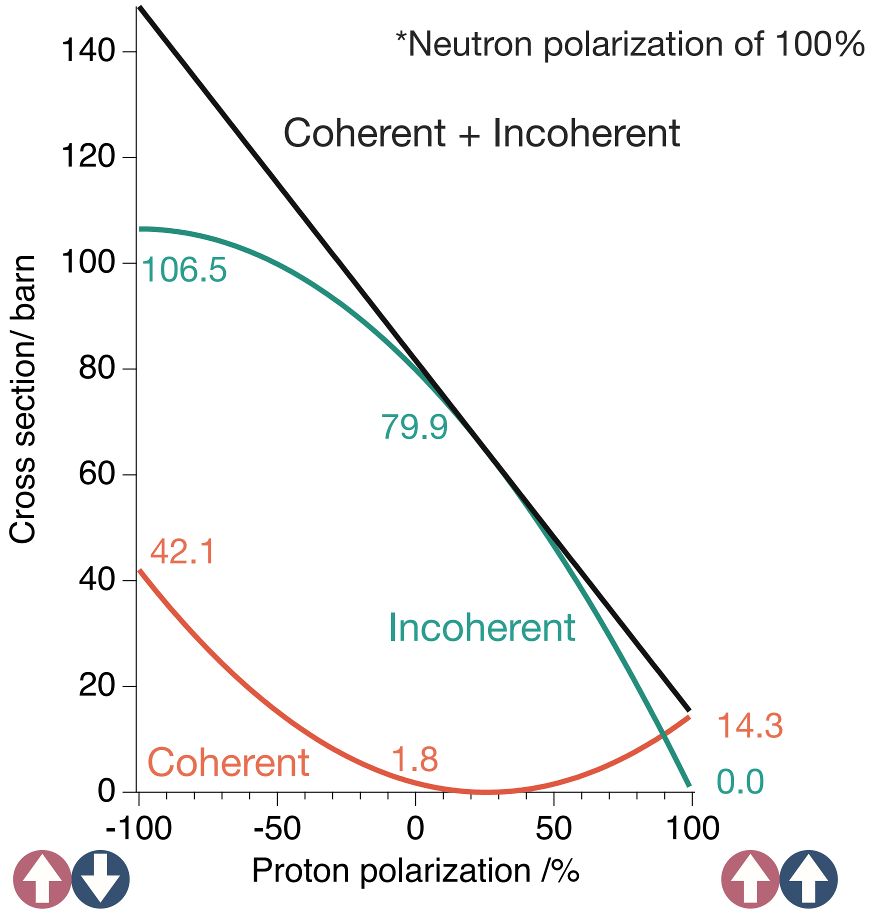
In spin triplet : $b_0 = -47.42$ fm

Coherent scattering length

$$b_c = \frac{3}{4}b_1 + \frac{1}{4}b_0 = -3.74$$

Spin dependence term

$$b_s = \frac{b_1 - b_0}{4} = 14.56$$



Spin-Contrast-Variation (SCV) neutron scattering

Only hydrogen scattering can be extracted.

$$I(\boldsymbol{Q}, P) \propto |F(\boldsymbol{Q}, P)|^2$$
$$= F_{\mathrm{H}}(\boldsymbol{Q}, P) + F_{\mathrm{others}}(\boldsymbol{Q})$$

Structure factor of hydrogens

Structure factor of atoms excluding hydrogens

$$F(\boldsymbol{Q}, P) = \sum_{j=1}^{N} b_{\mathrm{c}}(P) \exp\left(-i\boldsymbol{Q} \cdot \boldsymbol{R}_j\right)$$

N : Number of atoms in crystalline

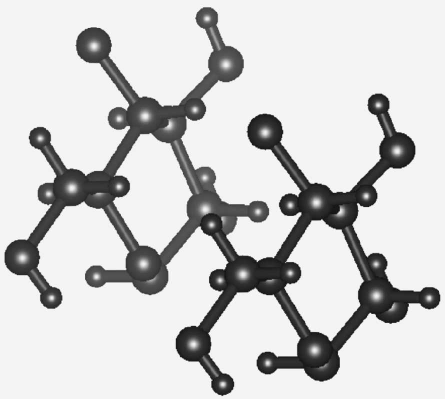
$\boldsymbol{Q}$: Scattering vector

P : Proton polarization

$\boldsymbol{R}_j$: Position vector

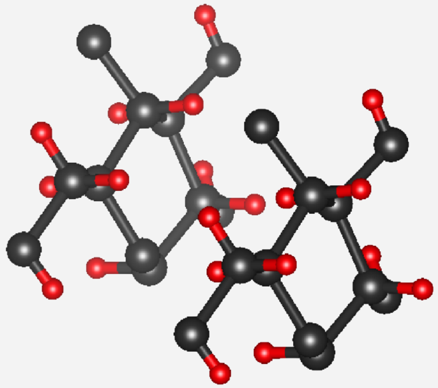
Conventional

Scattering from all atoms



SCV

Change only scattering intensity from hydrogens



Methods to add free electrons for DNP

Glassy samples

Dissolution of free radicals into the sample

ex) alcohol, water+glycerol, etc.

Polymers

Vapor absorption of free radicals

ex) polyethylene, rubbers, etc.

Crystalline samples

- Irradiation of electron-beam to generate latice defect

  ex) NH_3, ^7LiH Niinikoshki,Phys. Lett. A, 1979

  $P_H > 90\%$ (2.5 T, 0.3K)

- Substitution particular nuclei with paramagnetic centers

  ex) $LaAlO_3:Nd^{3+}$ P.Hautle and M. Iinuma, NIMA, 2000.

  $P_{La} > 50\%$ (2.3 T, 0.3K)

- Growth or soak crystals in a matrix with free radicals

  J. Pierce ,PSTP 2017

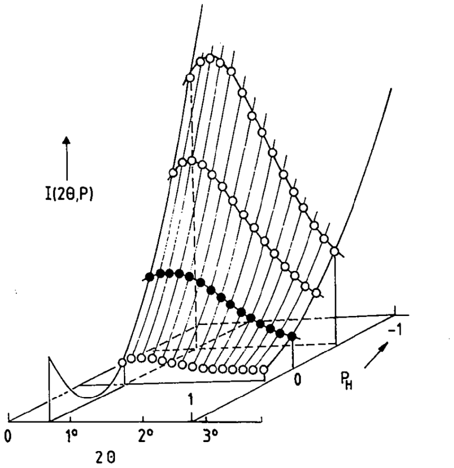
  ex) Lysozyme doped with TEMPOL

  $P_H > 20\%$ (2.5 T, 0.3K)

Examples of SCV neutron scattering

Glassy samples

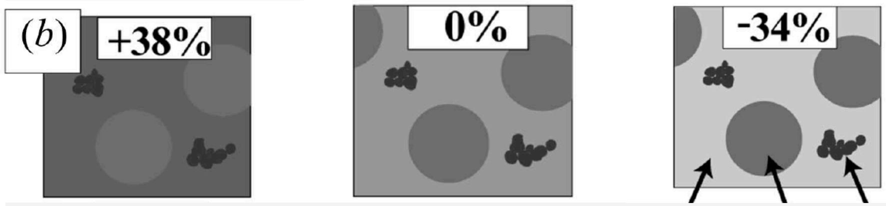
Coherent of proteins in water/glycerol



Polymers

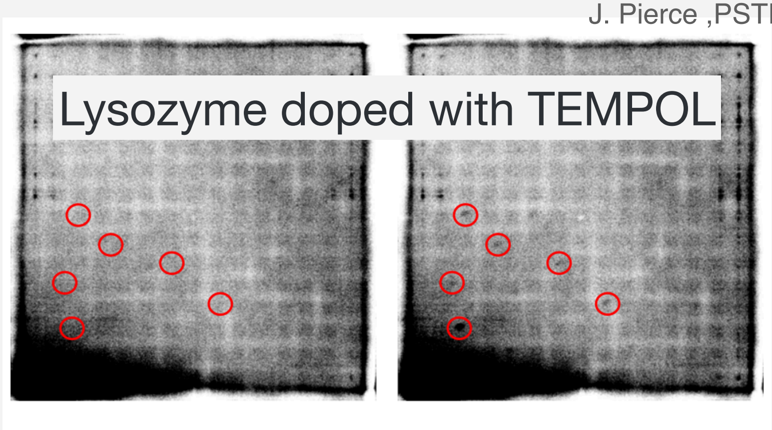
Nanostructure in tires

Y. Noda, *et al.*,J. Appl. Cryst., 2016

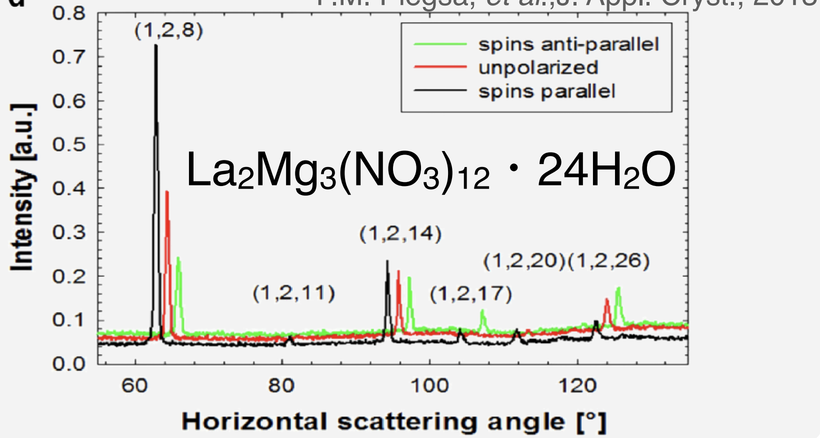


Crystalline samples

J. Pierce ,PSTP 2017



F.M. Piegsa, *et al.*,J. Appl. Cryst., 2013



Examples of SCV neutron scattering

Glassy samples

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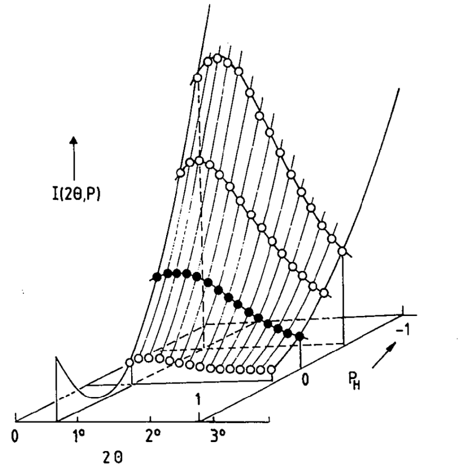

Polymers

Nanostructure in tires

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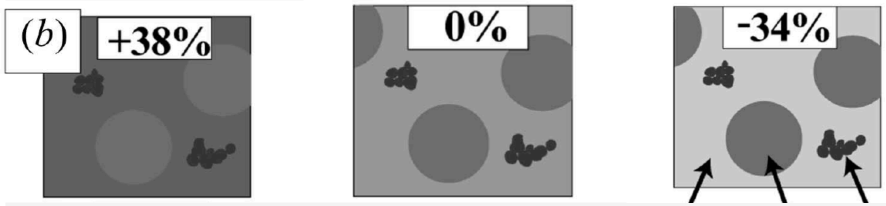
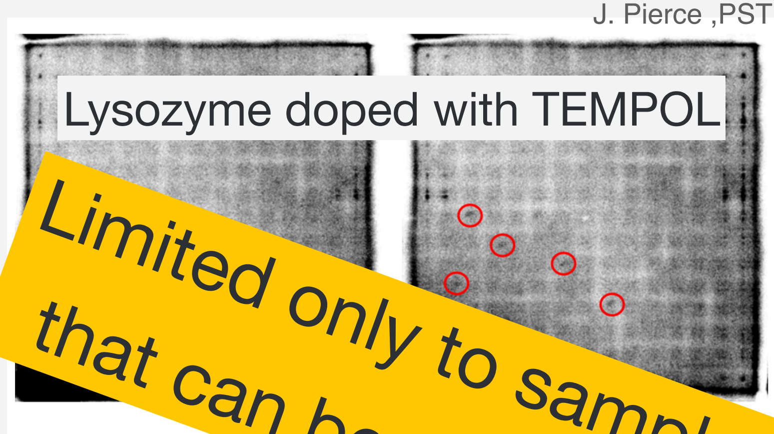


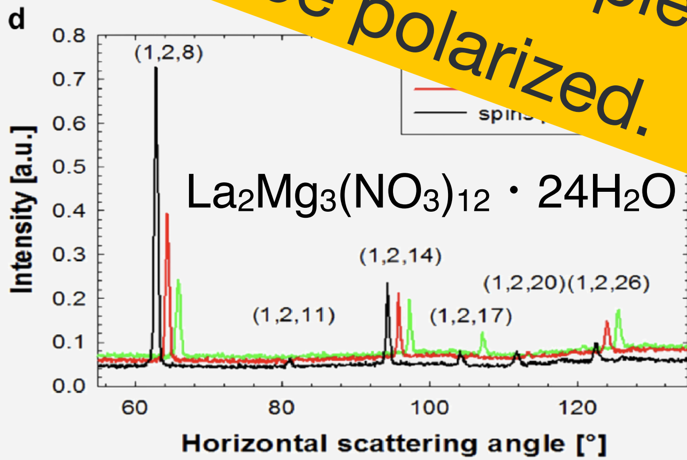
Crystalline samples

J. Pierce ,PSTP 2017



Limited only to samples that can be polarized.

2013



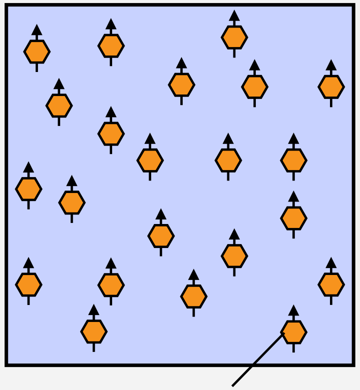
Motivation & Results

Dispersion of micro-size crystalline into DNP matrix

A. Rossini, et al., J. Am. Chem. Soc. ,2012., D. MIura at al., PTEP 2019.

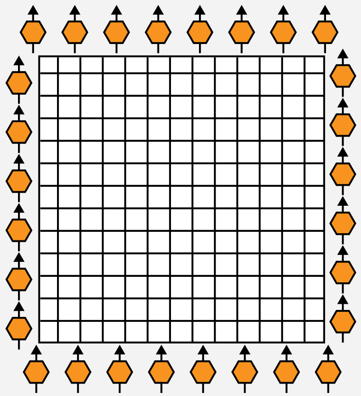
Glassy samples

Free radicals are dispersed in glassy samples.



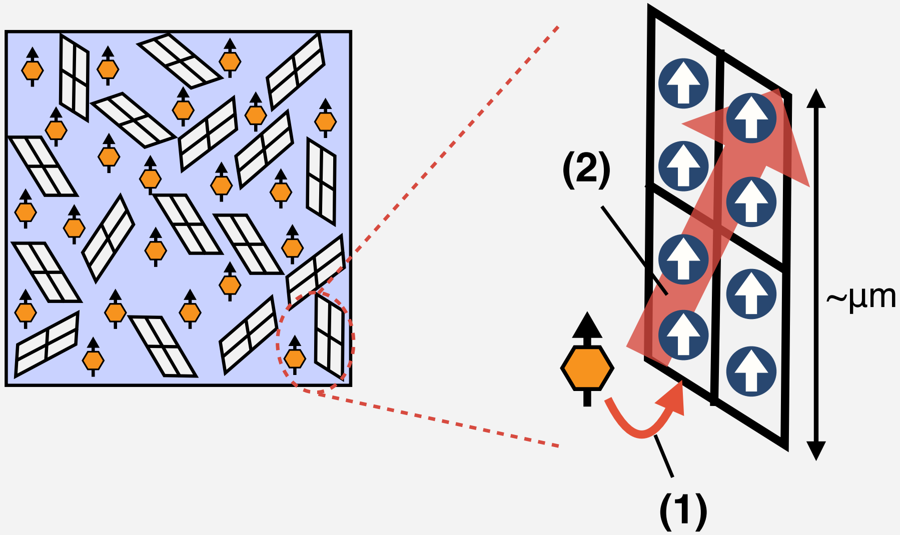
Crystalline samples

Free radicals cannot be penetrated into the crystal



Dispersion crystalline in glassy

Quasi-dissolved state of crystals



(1) Polarization transfers from free-electrons to nucleus in crystal

(2) Nuclear polarize via spin diffusion in crystal

Sample

Crystalline samples

L-glutamic acid 500 mg

- as known structure factor

Matrix

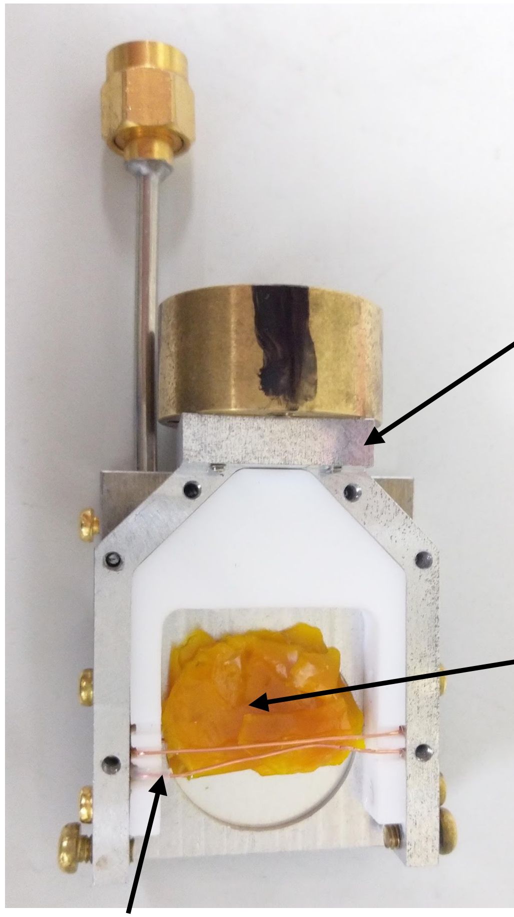
deuterated polystyrene 1000 mg

- deuterated : to observe from scattering from hydrogen in the crystal

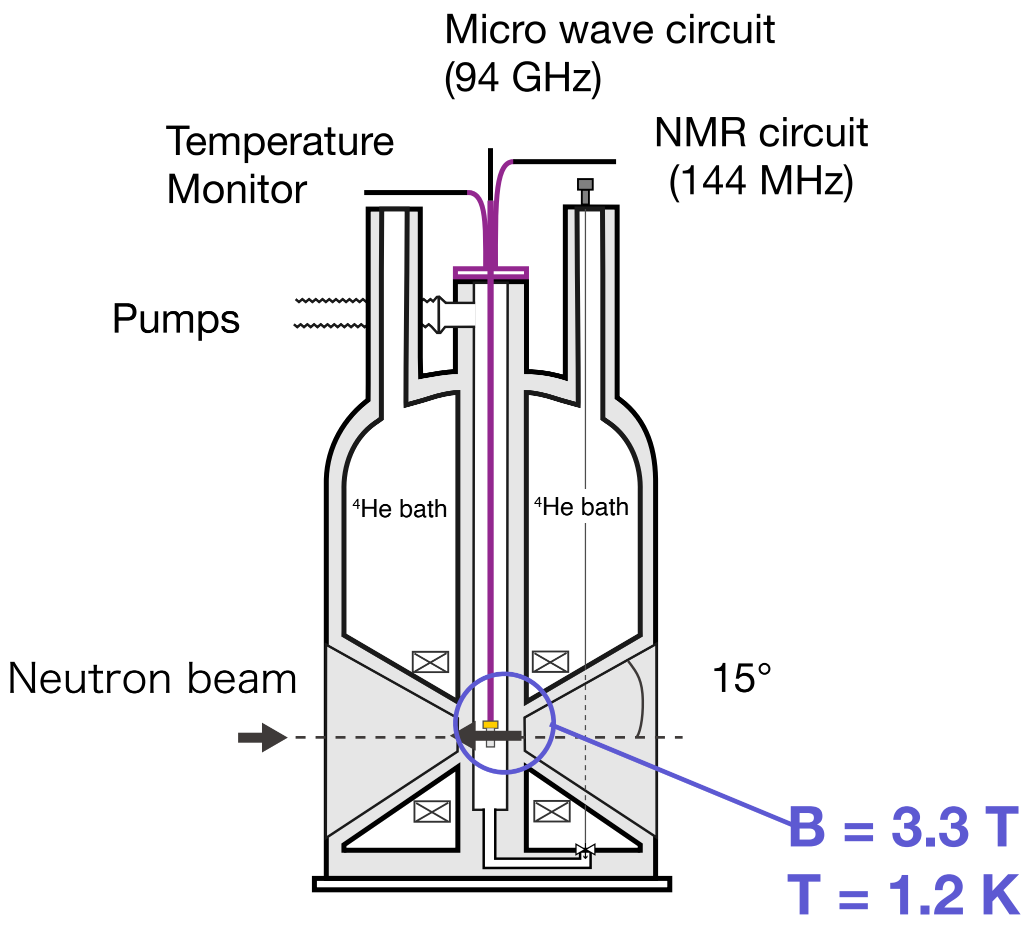
Free radical

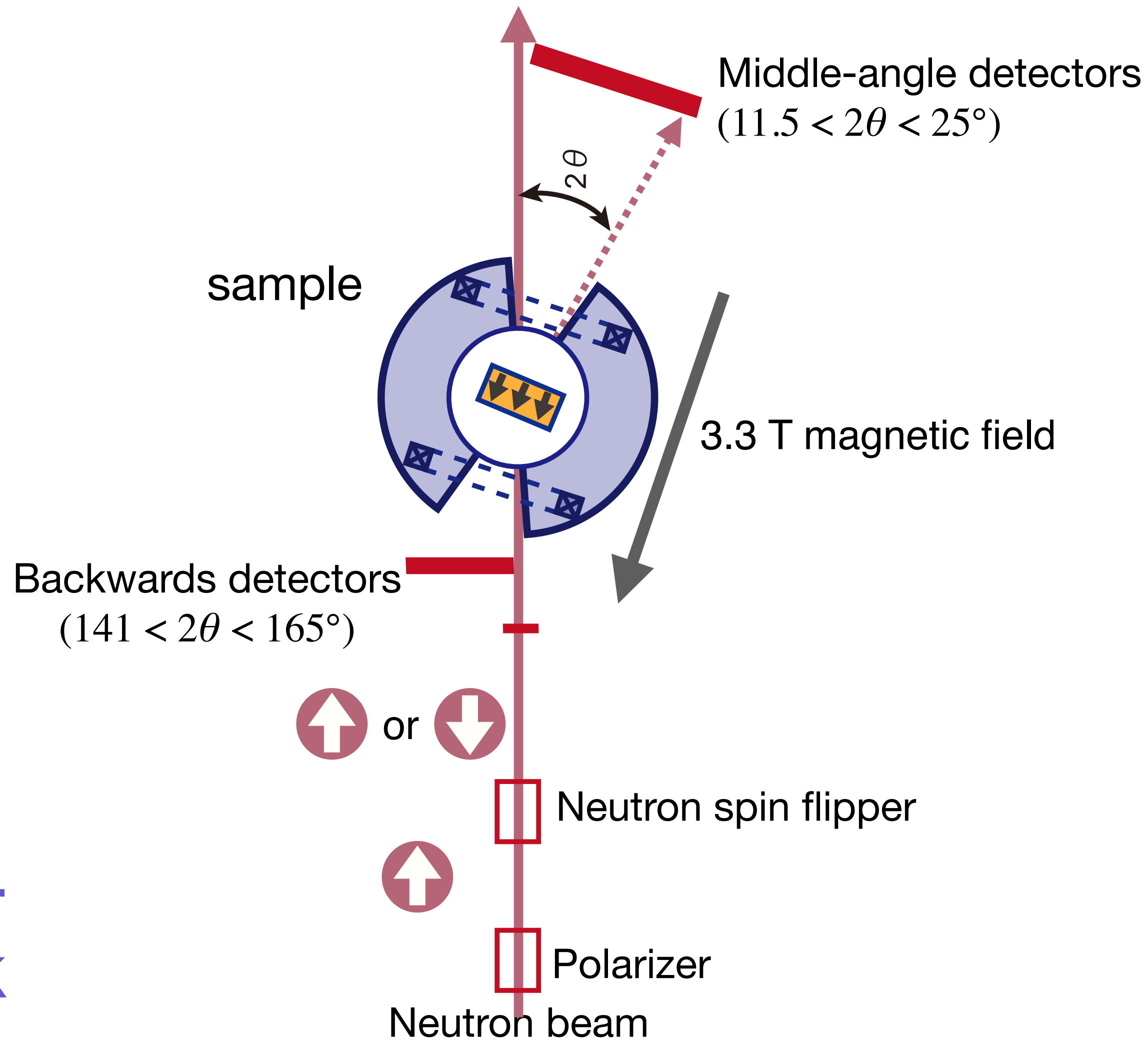
TEMPO methacrylate

2×10^{19} spin/g for deutrerated polystyrene



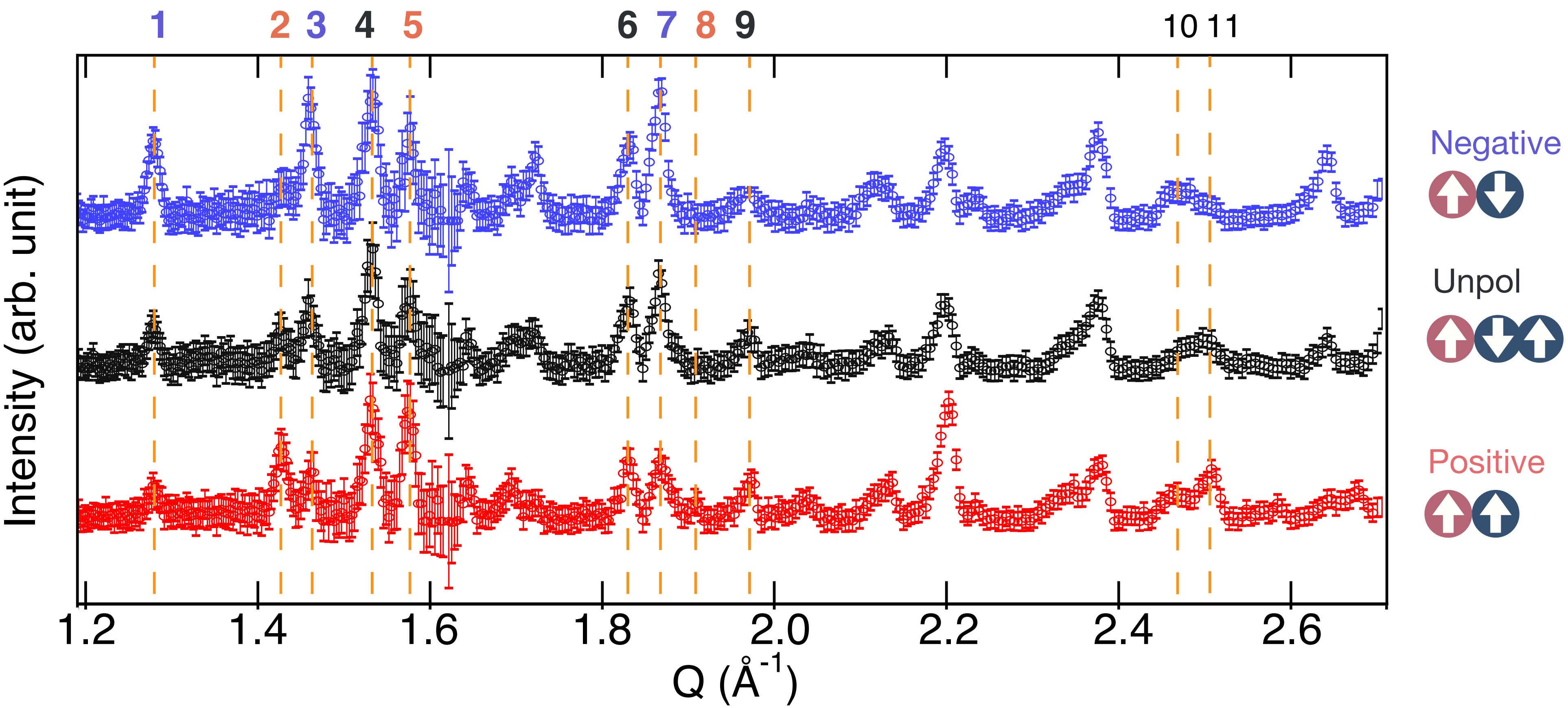
SCV neutron diffractometry @J-PARC MLF (BL15)





Results of SCV neutron diffractometry

Changes in peak intensity with proton polarization at each Q value



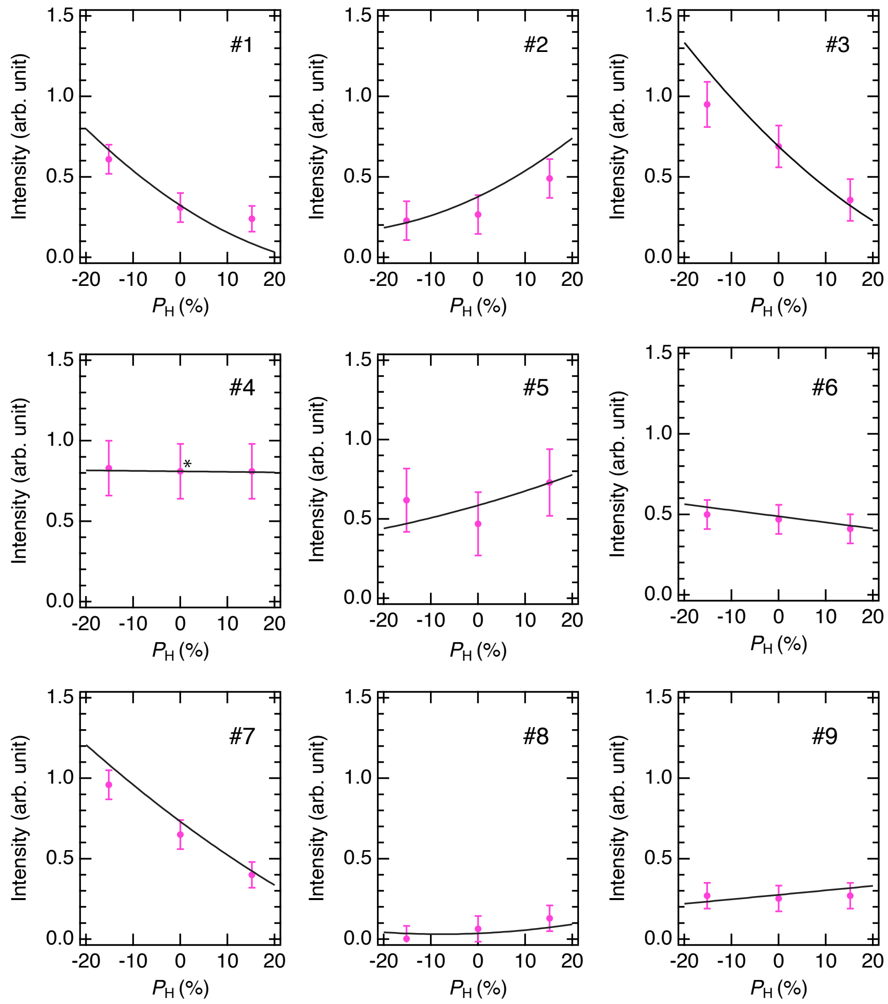
Proton polarization dependence

Calculation based on structure factor

$$I(Q,P) \propto \left| (1+\kappa P)\, F_{\mathrm{H}}(\boldsymbol{Q}) + F_{\mathrm{others}}(\boldsymbol{Q}) \right|^2$$

- Proton polarization was determined +/-15% from incoherent scattering.
- Calculation and measurement results are normalized using peak#4's intensity at polarization.

Good agreement with calculation



Extraction hydrogen structure factor

(a) Positive − Negative

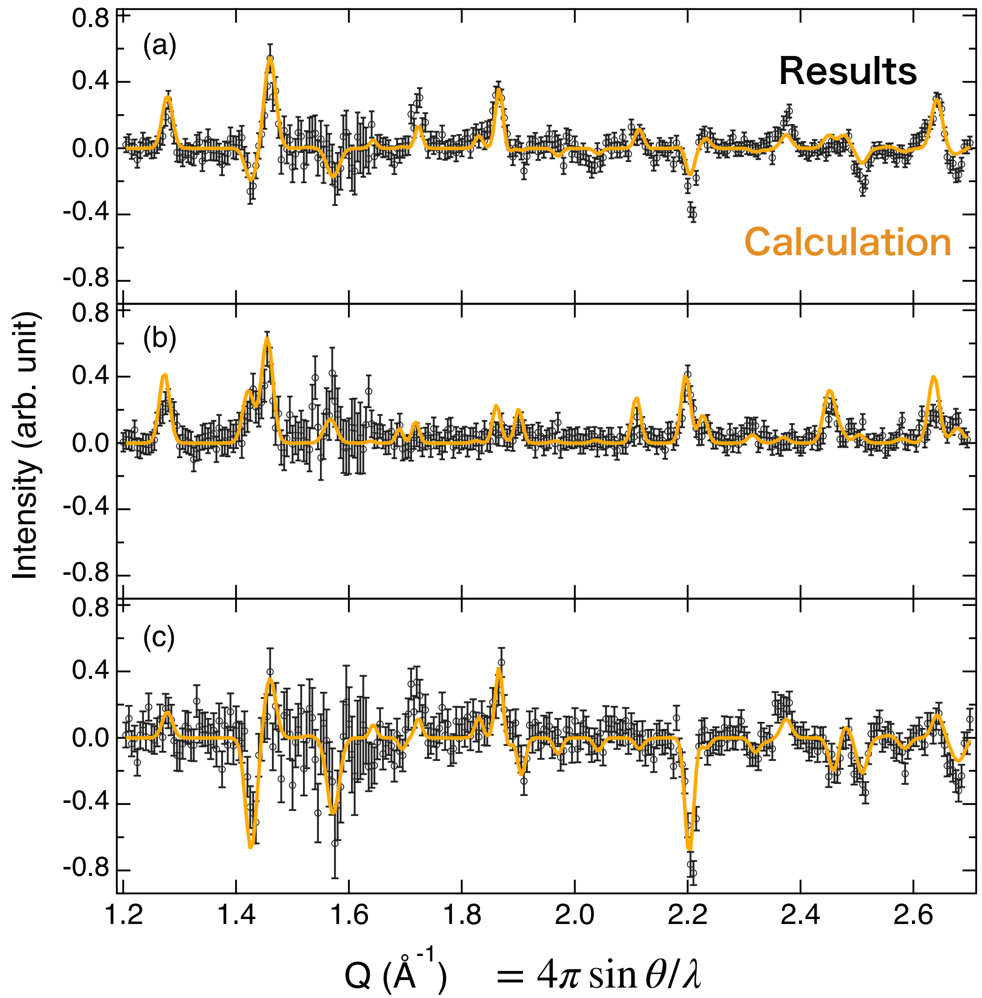
$$\propto |F_{\rm H}(\boldsymbol{Q})|^2 + {\rm Re}[F_{\rm H}(\boldsymbol{Q})F_{\rm others}(\boldsymbol{Q})]$$

(b) Positive + Negative − 2*unpol

$$\propto |F_{\rm H}(\boldsymbol{Q})|^2$$

(c) (a) −(b)

$$\propto {\rm Re}[F_{\rm H}(\boldsymbol{Q})F_{\rm others}(\boldsymbol{Q})]$$



$= 4\pi \sin\theta/\lambda$

○ **Good agreement with calculation**

SCV can extract hydrogen structure factor.

△**Poor S/N**

Apparatus and sample preparation need improvement.

Conclusion

SCV neutron scattering

- Technique that can extract the hydrogen structure factor owing to the cross section for hydrogen changes with the polarization
- Used as a structural analysis tool for non-crystalline samples

Demonstration of SCV neutron diffractometry

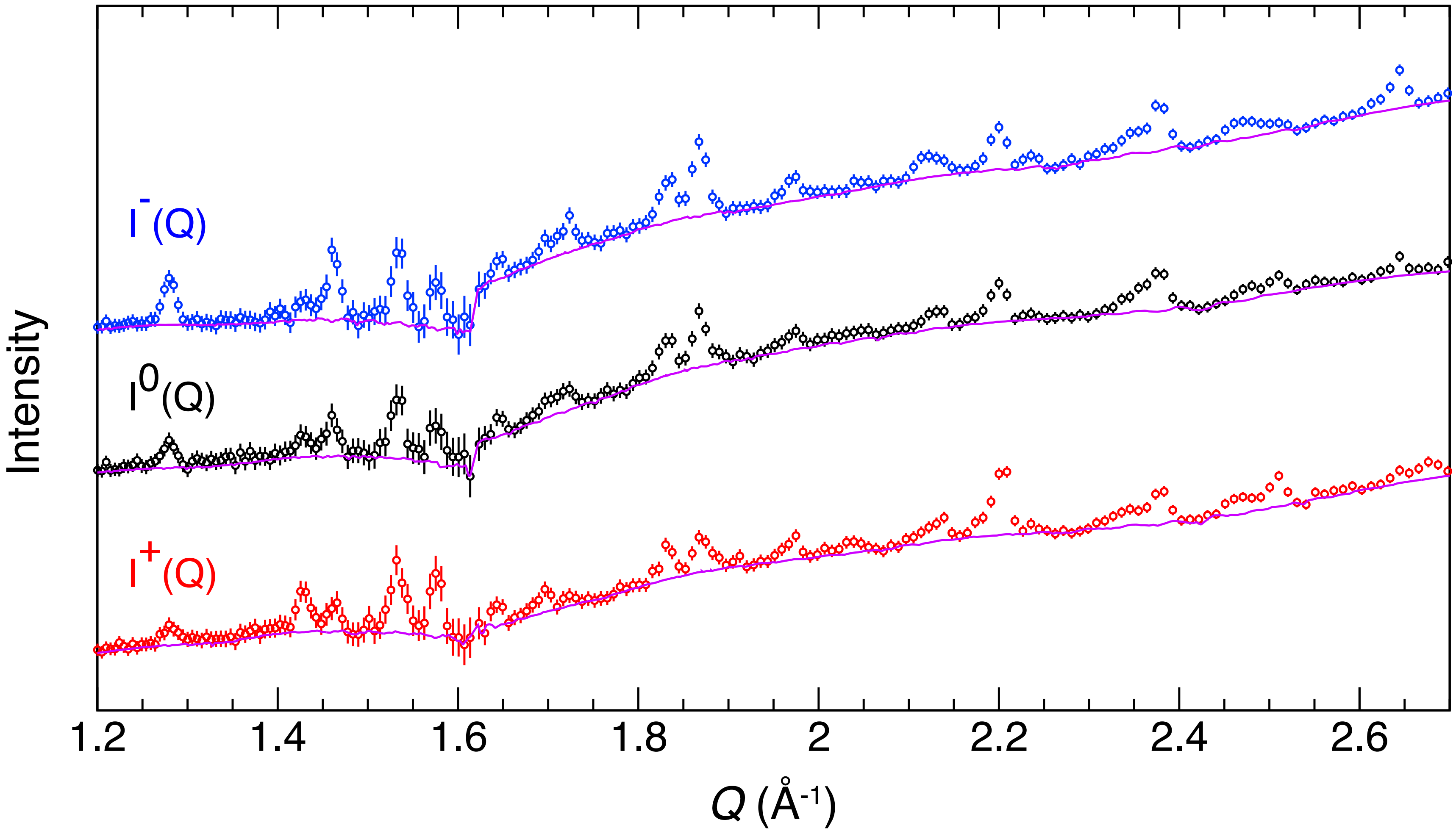
- Glutamic acid dispersed including TEMPO methacrylate (15% proton polarization @3.3T, 1.2 K)
- Change of diffraction peak intensities according to the proton polarization
- Succeeded in separating diffraction peaks into hydrogen structure factors and other atoms structure factors

Future plan

Development of SCV-neutron scattering using triplet-DNP

Backup

Baseline



XRD