Neutron Polarization

NXS 2023, August 8, 2023

Barry Winn

With Dean Myles & Josh Pierce

& Poster presenters Fankang Li, Jon Leiner…

Scope of this lesson

Yes! Utilizing polarized neutrons to *distinguish* different aspects or dimensions (contrast and character) of the “unpolarized” neutron scattering

Just a taste…*Enhance resolution* via Larmor precession of the polarized neutron before and/or after sample

Not in this school: Using polarized neutrons to better understand the *physics of the neutron* itself

Connections to previous and upcoming lessons & experiments:

Watch for these…

Connections to previous and upcoming lessons:

Watch for these…

A “neutron” by any other name…

Neutrons are NOT neutral

- Doesn’t the term “neutron” imply neutral everything?
  - Electric neutrality, yes
    - Neutrons ignore charge of electrons and protons in atoms
    - Penetrating power!
  - Pesky quarks
    - Neutron **spin**
      - Affects “weak force” interaction
    - Neutron **magnetic dipole moment**
      - Affects magnetic interaction
- Scattering polarized neutrons from materials changes scattered neutron intensity and *sometimes* *neutron polarization state*

Spin:

$\sigma_n = ½$

Dipole moment:

$\mu_n = -1.913\ \mu_N$

Nuclear magneton:

$\mu_N = 5.051E-27\ \mathrm{JT}^{-1}$

Which way is ‘up’?
Depends on what you’re using…
Did we really put these on the same diagram with totally different units?
Yes, yes we did…

Now in 3D!

Key Questions you need to be asking

Q1

Can polarized neutrons help my science?

Not easy to answer. Not just a single Application!

In this presentation we provide a framework for figuring this out

Q2

Are polarized neutrons needed?

Alternative, unpolarized ways to answer same thing?

Q3

Reality check?

A. Available with scattering technique?
B. Time and statistics due to (usually) reduced signal and (often) multiple measurements?
    i. Problematic on high throughput instruments
C. Complications of polarized optics (compatibility with sample conditions, corrections to data, etc.)
D. Ease of reaching results (software tailored for polarization)

Depends on state-of-art, and what's available where

Q1 Clues to how experimentalists utilize polarized neutrons

Somewhere hidden in each publication is a statement that answers this question

- Accounts for the system / material being studied
- Identifies a specific ‘capability’ leveraging polarized neutron scattering

How to find this statement?

- Find the polarized neutron figure, backtrack to the the text where that figure is referenced, and *voila*!

Sometimes even more context

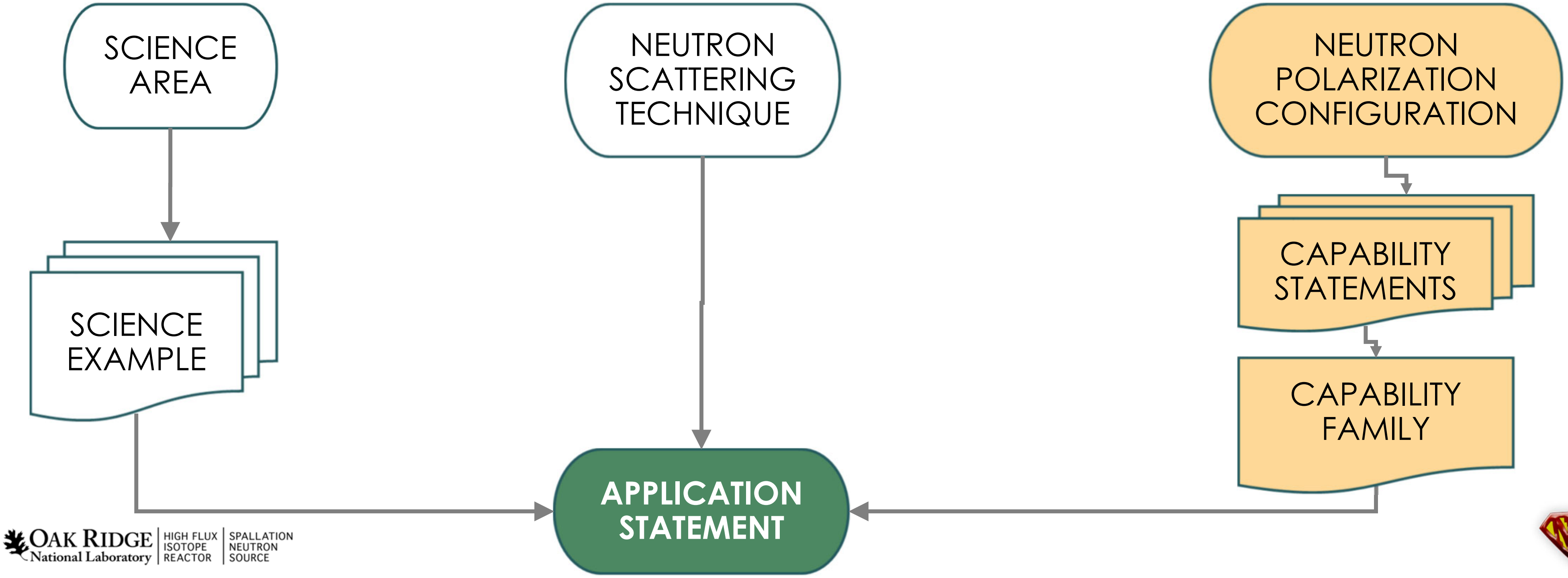
- Often find introductory / explanatory text about polarized neutrons, despite 60-year history

Examples of papers and application statements in mini poster session!

Polarization application statement

Q1 <u>Ingredients</u> for your ‘polarized’ application statement

As a [***SCIENCE AREA***] neutron scattering experimentalist, I want to
[***NEUTRON SCATTERING TECHNIQUE***]
so I can [***APPLICATION statement***] for [***SCIENCE EXAMPLE***]



SCIENCE AREA

SCIENCE EXAMPLE

NEUTRON SCATTERING TECHNIQUE

NEUTRON POLARIZATION CONFIGURATION

CAPABILITY STATEMENTS

CAPABILITY FAMILY

APPLICATION STATEMENT

Drill-down on ingredients for your polarization application statement

Science areas (no, really! Yours, too)

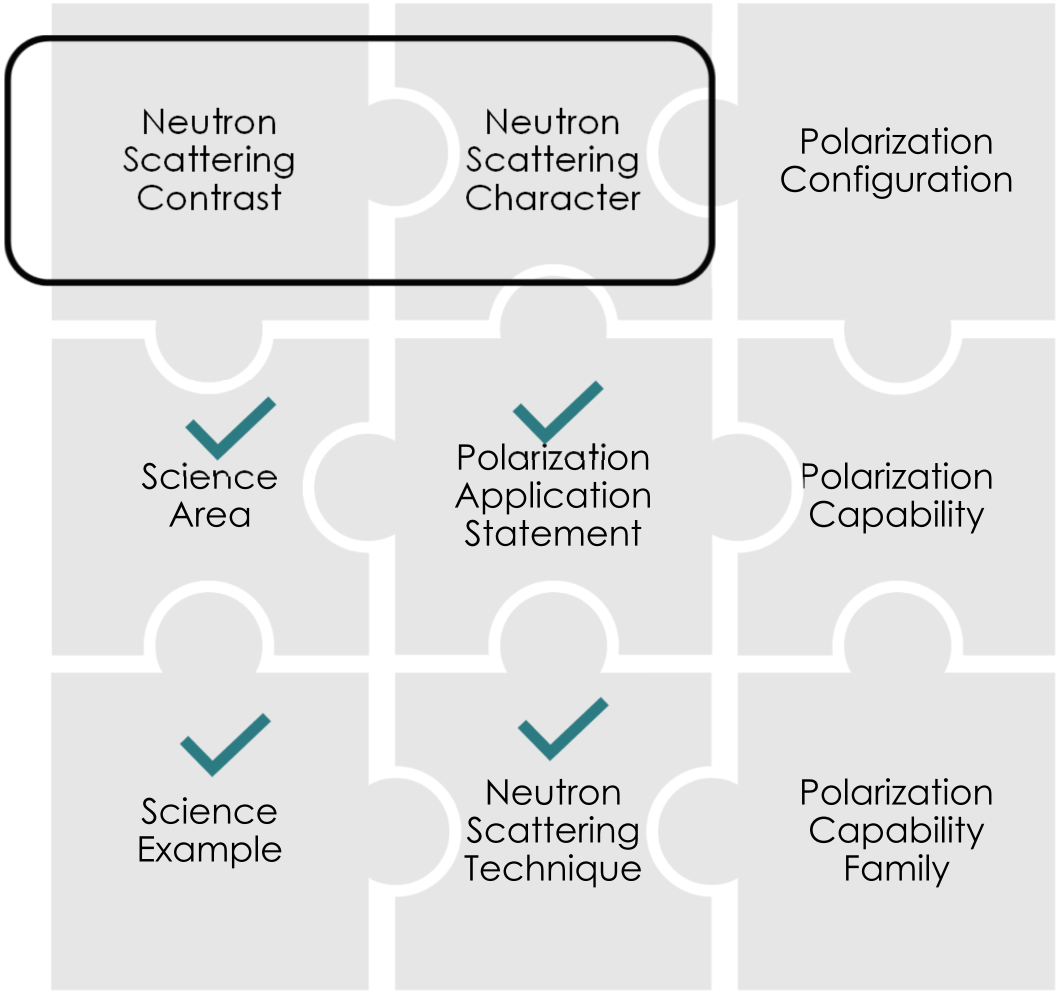
- Biology
- Soft matter & Polymers
- Materials & Engineering
- Condensed matter & Quantum materials
- Chemistry
- Geology
- Environmental Science

Examples in
mini poster session!

Neutron Scattering Technique (Instrument)

- Structure vs dynamics
  - Length / Q scales
  - Time / energy scales
- Real space vs reciprocal space
  - Imaging / microscopy / tomography
  - Reciprocal space
  - Sometimes both
- Meso-structure
  - Layered systems with reflectometry
  - Microscopy

Q1 Can distinguishing different aspects of neutron scattering help my science? Piecing together the puzzle!



Neutron Scattering Contrast

Neutron Scattering Character

Polarization Configuration

✓ Science Area

✓ Polarization Application Statement

Polarization Capability

✓ Science Example

✓ Neutron Scattering Technique

Polarization Capability Family

Neutron Scattering Contrast

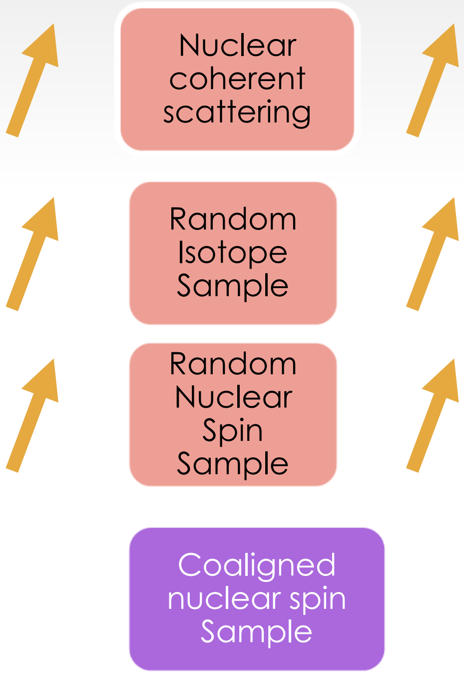
Contrast / scattering length / cross sectional area

New Dimension to Weak Nuclear Force $\vec{\sigma}_n$

- Scattering from nuclei of atoms
- Structure factor
  - Real-space:
    - Highly localized
    - Think “Dot” or point scattering
  - Reciprocal-space:
    - (think Fourier transformed space)
    - “Everything Everywhere All at Once”
    - Most visible at high momentum transfer Q

Old news…

- Nuclear Coherent scattering
  - *Never changes neutron spin state*
- Isotope incoherent scattering
  - *Never changes neutron spin state*
- Nuclear-spin incoherent
  - A random nuclei spin interacting with a neutron spin
  - *When random nuclear spin orientations, inverts neutron spin state 2/3 of the time…*
- When nuclear spins are aligned, some of the ‘incoherent’ scattering from polarized neutrons becomes ‘coherent’



Nuclear coherent scattering

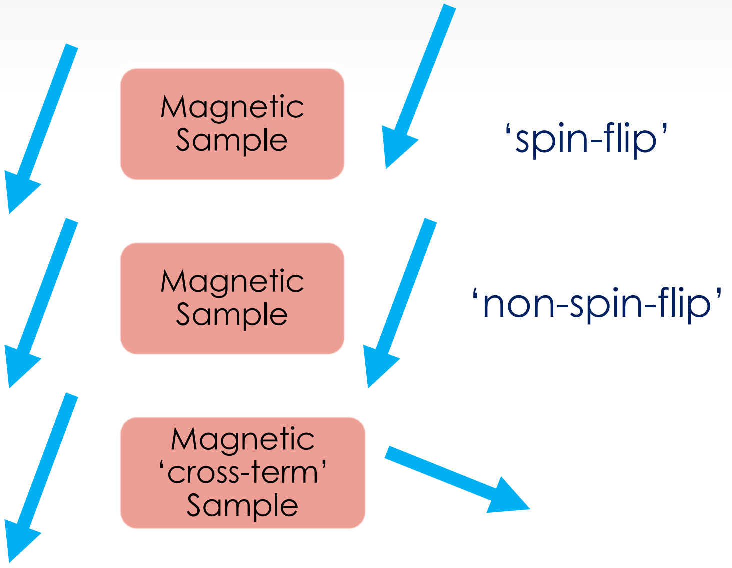
Random Isotope Sample

Random Nuclear Spin Sample

Coaligned nuclear spin Sample

Contrast / scattering length / cross sectional area

Old news…



New Dimension to Magnetism $\vec{\mu}_n$

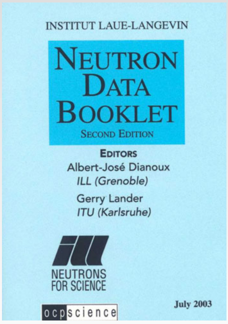
- Structure factor
  - Real-space
    - Magnetism distributed through material
    - Subtle structural effects (e.g. unmatched electron orbits with S,P,D,F shapes)
  - Reciprocal space
    - Most visible at low momentum transfer Q
- Scattering from magnetism within materials
  - *Whether a moment (and therefore spin) state changes is predictable, and depends on momentum transfer and vector algebra*
    - *Usually either inverts or preserves spin state ("spin-flip" or "non-spin flip")*
    - *Sometimes reorients differently (cross terms in Maleev-Blume equations)*
- Multi-dimensional!
  - Both the neutron and different parts of the material structure have different spin / moment directions
  - Momentum transfer plays a role via "Q-perp"

New Dimension to Character of interaction

Neutron
Scattering
Character

Incoherent

- Individual atom behavior
  - Great for movement like diffusion or vibrational modes
  - Not-so-great for position determination
- Incoherence via collective randomness*
- Isotope incoherent
  - Typically, doesn't exist when only one isotope per element
  - Can surprise you when coherent scattering lengths have opposite sign
  - *Never changes neutron spin state*
- Spin incoherent
  - Implies random orientation of spins of atomic nuclei
  - *When random spin orientations, changes neutron spin state 2/3 of the time…*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NEUTRON DATA BOOKLET

SECOND EDITION

EDITORS
Albert-José Dianoux
ILL (Grenoble)
Gerry Lander
ITU (Karlsruhe)

NEUTRONS FOR SCIENCE

July 2003

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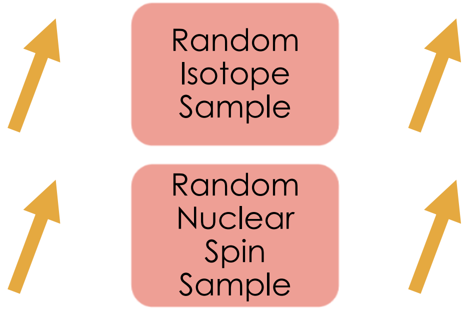
For elements,

$$\sigma_{inc} = \sigma_{isotope} + \sigma_{spin}$$

For isotopes it's just σ_{spin}


Random Isotope Sample

Random Nuclear Spin Sample

*Incoherent scattering can also arise when the coherent scattering is suppressed via destructive interference. In this case there is also no-spin-flip during scattering.

One of our favorite alloys where this occurs is TiZr, where opposite scattering lengths for the two elements lead to only isotope incoherent scattering. At HYSPEC we use this alloy as a standard, similar to Vanadium but with no-spin-flip, to measure both uniformity at our detector array, and flipping ratio.

Oak Ridge
National Laboratory

New Dimension to Character of interaction

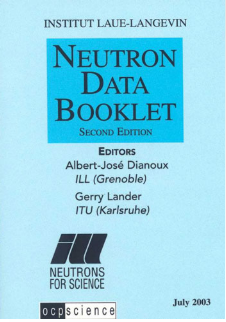
Neutron
Scattering
Character

Coherent

- Cooperative atoms
  - Lattice-like structure
  - Coordinated motion or reorientations
- Think “scattering length”
  - Instead of just
    “cross section area”
  - Negative “lengths” possible with weak force / isotope specific scattering
  - Index of refraction &
    critical reflection angle
  - Bragg’s law
    (you should have a counting game for this by now…)
- *Neutron orientation*
  - *Weak / nuclear changes spin state in predictable ways*
  - *Magnetic, vector algebra…*

New Dimension to Character of interaction

Neutron
Scattering
Character



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😢

σ_{abs} assumes randomly oriented neutron spin and/or nuclear spin

Absorption

- **Weak-force only**
- Nuclear reaction
  - Differs by isotope
  - **Differs by nuclear (nucleus) spin state**
- Leveraged for
  - Neutron detection
  - Shielding
  - Polarization filtering (sometimes)
- Not explicitly in Maleev-Blume equations

OAK RIDGE
National Laboratory

New Dimension to Character of interaction

Neutron
Scattering
Character

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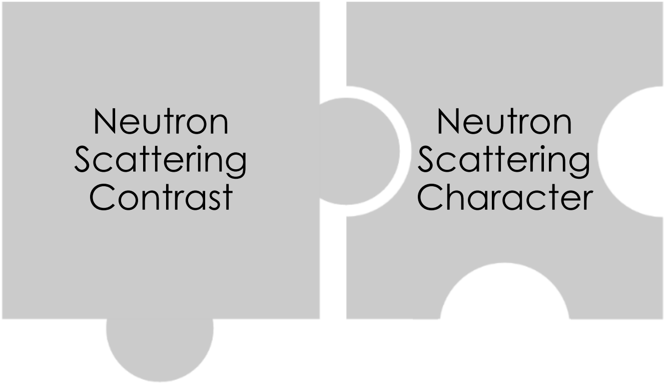
Changing spin state of atomic nuclei enables sloshing between

Combine neutron scattering *contrast* and *character*



Neutron Scattering Contrast

Neutron Scattering Character

Reciprocal space ↔ Real space

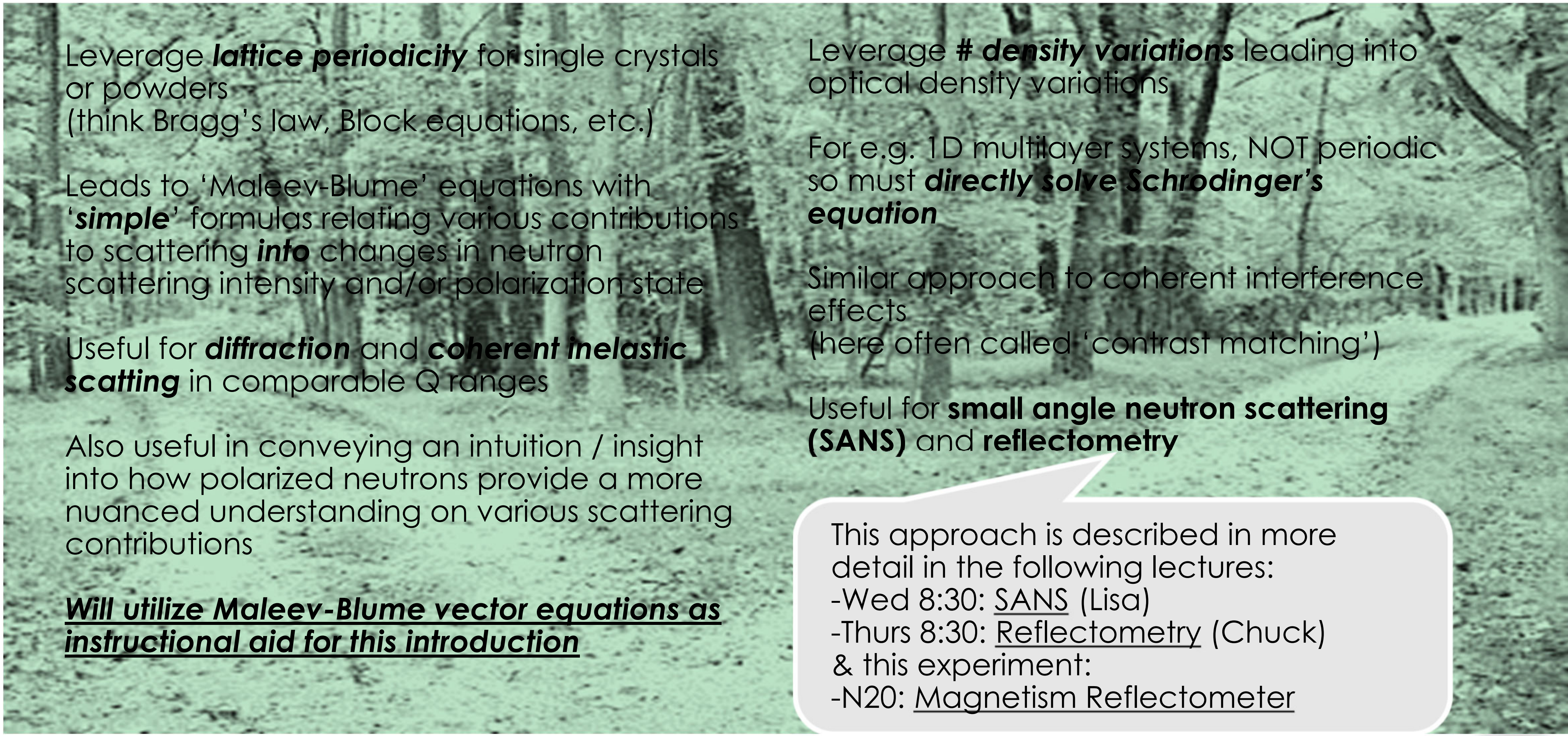
$N(\boldsymbol{Q}) = \sum_n b_n e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$	Nuclear structure factor
$\boldsymbol{M}(\boldsymbol{Q}) = \sum_n \boldsymbol{M}_n e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$	Fourier transform of magnetic moments / magnetic structure factor

	Incoherent	Coherent	Absorption
Weak / Nuclear	I_N & I_S ⟺	$N(\boldsymbol{Q}, E)$	σ_{abs}
Magnetic / Spin	☹	$\boldsymbol{M}(\boldsymbol{Q}, E)$	☹

Vector convention: boldface $\boldsymbol{Q}$ instead of $\vec{Q}$

Absorption never measured in scattered signal

Coherent scattering: "*Two roads diverged…*"



Leverage ***lattice periodicity*** for single crystals or powders
(think Bragg's law, Block equations, etc.)

Leads to 'Maleev-Blume' equations with '***simple***' formulas relating various contributions to scattering ***into*** changes in neutron scattering intensity and/or polarization state

Useful for ***diffraction*** and ***coherent inelastic scatting*** in comparable Q ranges

Also useful in conveying an intuition / insight into how polarized neutrons provide a more nuanced understanding on various scattering contributions

<u>Will utilize Maleev-Blume vector equations as instructional aid for this introduction</u>

Leverage ***# density variations*** leading into optical density variations

For e.g. 1D multilayer systems, NOT periodic so must ***directly solve Schrodinger's equation***

Similar approach to coherent interference effects
(here often called 'contrast matching')

Useful for **small angle neutron scattering (SANS)** and **reflectometry**

This approach is described in more detail in the following lectures:
-Wed 8:30: <u>SANS</u> (Lisa)
-Thurs 8:30: <u>Reflectometry</u> (Chuck)
& this experiment:
-N20: <u>Magnetism Reflectometer</u>

Thankfully, capability statements and families are same for both! (whew)

Q2 Are polarized neutrons needed?

Overlapping information?

- Structure & dynamics in many size and energy scales
  - Sometimes, both at once!
- Nuclei-specific
- Isotope-specific
- Magnetism
- Coherent interference *between* nuclear and magnetic terms!?!

One person's trash is another's treasure

- The lack of coherence of hydrogen scattering makes finding some Bragg peaks *near-impossible*… but that same incoherent scattering is *perfect* for measuring energies of various modes for chemistry

Unclear on directional aspects of your material?

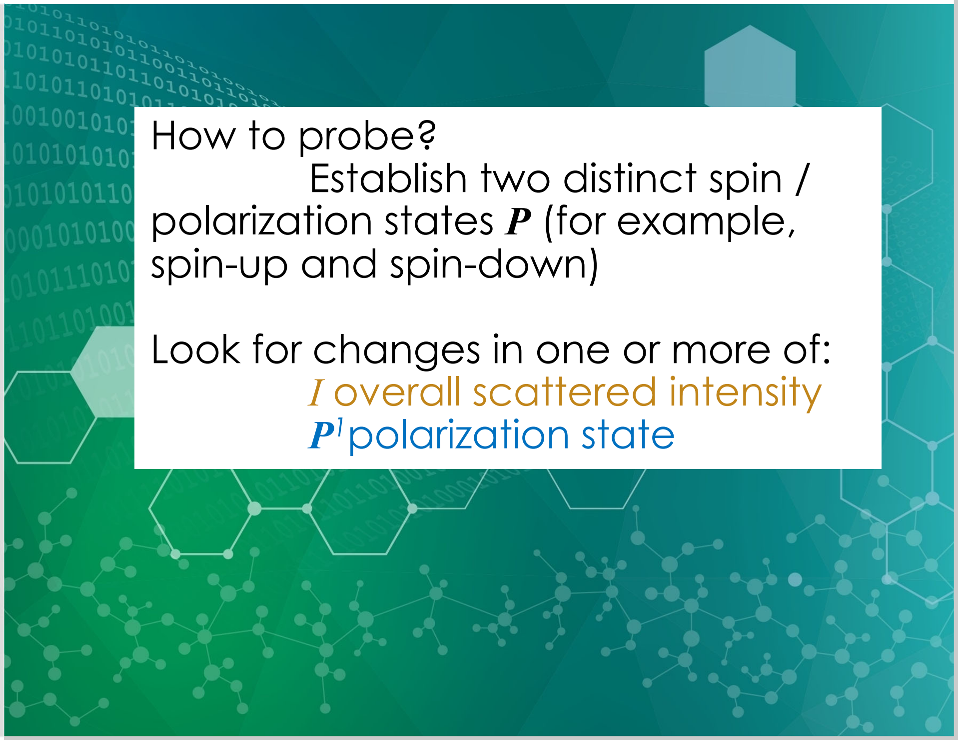
- Even if some kind of scattering is already isolated, the directionality of your materials moments may still be unclear, motivating the use of polarized neutrons to figure out just what kind of scattering you're looking at.

(usually) distinguish even with 'unpolarized' scattering

- Expecting only one kind of scattering
  - Based on system studied
  - Based on where scattering is observed
    - Low momentum transfer Q: magnetic
    - High momentum transfer Q: weak
  - Based on thermodynamic conditions
    - Phase changes (magnetism below T_n…)
    - Compare / contrast / subtract strategies
- MOST neutron scattering experiments leverage unpolarized neutrons!
- But, sometimes, we need to tease them apart

Q2 When polarized neutrons really are needed:

- When there’s no other/better way to distinguish different aspects or dimensions (contrast and character) of the “unpolarized” neutron scattering



How to probe?
Establish two distinct spin / polarization states $\boldsymbol{P}$ (for example, spin-up and spin-down)

Look for changes in one or more of:
I overall scattered intensity
$\boldsymbol{P}'$ polarization state

But how?

Polarization Configuration

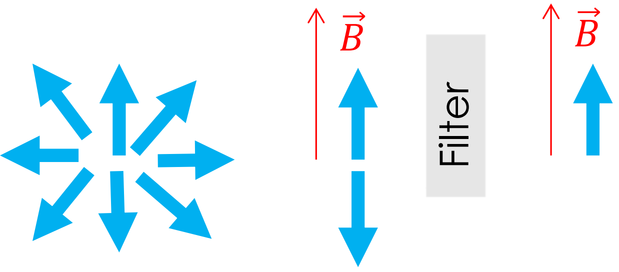
- Polarization Optics
  - Filters: select one neutron spin / polarization state
    - Up to 50% transmission
  - Spin manipulation
    - Flippers, guide fields, nutators, zero field regions, Larmor precession regions
- Polarization Configuration
  - Kinds and locations of optics, before and/or after sample
- Linear algebra
  - Contributions from different kinds of scattering simply add up!
  - “Two equations, two unknowns”
  - Sometimes more than two…

Polarization optics: ingredients for P. Configurations

Polarization Configuration

Filters

- 'Quantum' has its advantages…
  - Unpolarized classical has arrows pointing everywhere
  - In ambient field, though, a quantum superposition of 'up' & 'down'
  - A filter can achieve up to* 50% transmission



*Actual transmission varies widely…

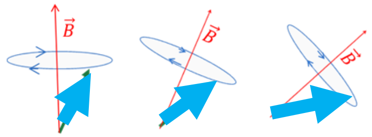
Guide fields and nutators

- Larmor precession, via torque $\vec{\tau}$ on neutron magnetic moment $\vec{\mu}$ by applied magnetic field $\vec{B}$

$$\vec{\tau} = \vec{\mu} \times \vec{B},\ \omega = -\gamma B$$

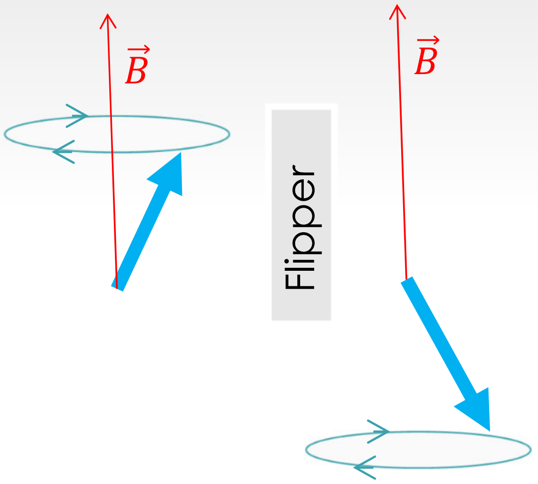
$$\gamma = \text{-1.833E4 rad/Gauss-sec}$$

- Frequency ω is INDEPENDENT of polar angle φ between applied field and moment
- Magnetic 'guide' fields keep $\vec{\mu}$ either aligned or anti-aligned with respect to $\vec{B}$
  - Keeps ω fast while changing direction of $\vec{B}$ slowly



Flippers

- Optionally invert the neutron spin-state with respect to the ambient guide field



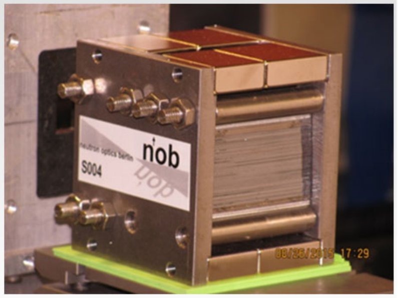
Polarization optics: ingredients for P. Configurations

Polarization Configuration

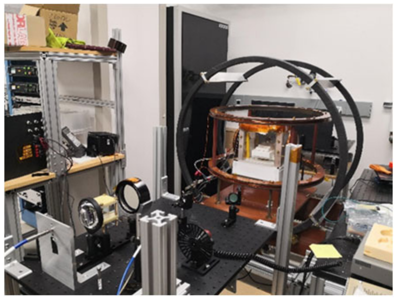
Filters

Heusler crystal

Polarizing Supermirror

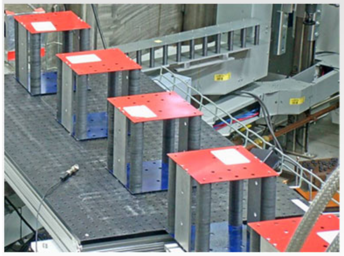


Nuclear-polarized ^{3}He

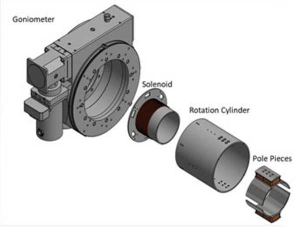


Guide fields and nutators

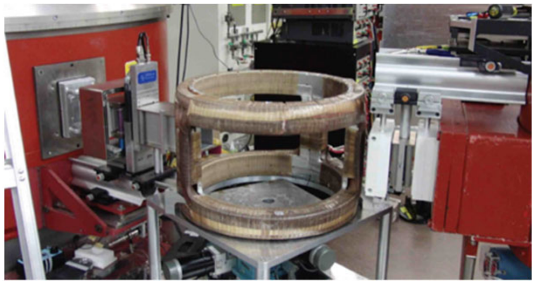
Permanent Magnet Yoked Assemblies



Rotatable nutator

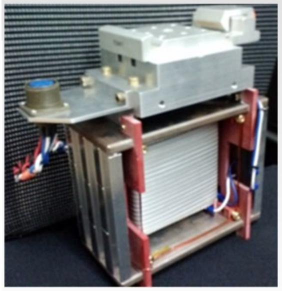


3D Coils



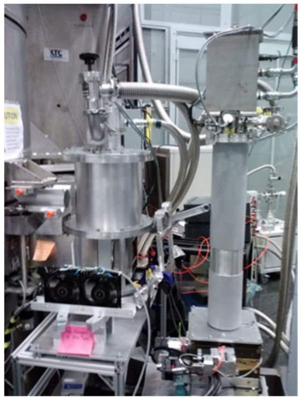
Flippers

Mezei



Radio-Frequency

Cryogenic (Meissner screen)

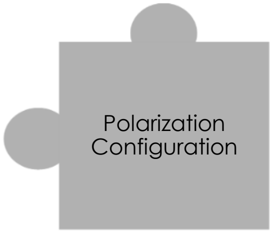


Adiabatic Fast Passage /w ^{3}He

Meet the "Vector" family

$\overrightarrow{R_n}$		Coordinates of one atom in unit cell for crystal	Real space
$\overrightarrow{M_n}$		Which way and how strong a magnetic moment of an *ATOM* points	
$\overrightarrow{P}$	Polarization	A measure of how 'polarized' the incident beam is, and average orientation of those neutrons' spin (or magnetic moment) at sample position	
$\overrightarrow{P^1}$		The new polarization of the scattered neutrons	
$\overrightarrow{Q}$	Momentum transfer	Incident neutron momentum minus final neutron momentum $\overrightarrow{Q_{lab}} = \overrightarrow{k_i} - \overrightarrow{k_f}$	Reciprocal space
$\overrightarrow{M(Q)}$	Magnetic structure factor	Fourier transform of $\overrightarrow{M_n}$	
$\overrightarrow{M_\perp}$	"M perp"	The component of the Magnetic structure factor perpendicular to the momentum transfer $\overrightarrow{Q}$	

Polarization Configurations access Intensity and/or *Polarization State*



$N(\boldsymbol{Q}) = \sum_n b_n e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$	Nuclear structure factor
$\boldsymbol{M}_\perp = \boldsymbol{e_Q} \times \boldsymbol{M}(\boldsymbol{Q}) \times \boldsymbol{e_Q}$	"M perpendicular"
$\boldsymbol{M}(\boldsymbol{Q}) = \sum_n \boldsymbol{M}_n e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$	Fourier transform of magnetic moments / magnetic structure factor
$\boldsymbol{e_Q} = \boldsymbol{Q}/\lvert\boldsymbol{Q}\rvert$	Unit vector along momentum transfer **Q**
I_{si}	Spin incoherent scattered intensity
$\boldsymbol{P}, \boldsymbol{P}^1$	Initial and final polarization

POLARIZATION CONFIGURATION	Impacts the scattered neutron	Optics
Half Polarized Dynamic Nuclear Polarization Solve Phase Problem	Intensity	1 filter 1 flipper
Longitudinal Analysis I Larmor	Polarization State	2 filters 1 flipper
Longitudinal Analysis II Spherical Neutron Polarimetry	Both	2 filters 2 flippers

$$I = I_n + N^\dagger N + I_{si} + \boldsymbol{M}_\perp^\dagger \boldsymbol{M}_\perp + \boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger N + \boldsymbol{P}\cdot\boldsymbol{M}_\perp N^\dagger + i\boldsymbol{P}\cdot\left(\boldsymbol{M}_\perp^\dagger \times \boldsymbol{M}_\perp\right)$$

$$\boldsymbol{P}^1 I = \boldsymbol{P}\left(I_n + N^\dagger N - \frac{1}{3} I_{si}\right) + \left(\boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger\right)\boldsymbol{M}_\perp + \left(\boldsymbol{P}\cdot\boldsymbol{M}_\perp\right)\boldsymbol{M}_\perp^\dagger - \boldsymbol{P}\left(\boldsymbol{M}_\perp^\dagger \boldsymbol{M}_\perp\right) + iN\left(\boldsymbol{P}\times\boldsymbol{M}_\perp^\dagger\right) - iN^\dagger\left(\boldsymbol{P}\times\boldsymbol{M}_\perp\right) + N\boldsymbol{M}_\perp^\dagger + N^\dagger\boldsymbol{M}_\perp - i\left(\boldsymbol{M}_\perp^\dagger \times \boldsymbol{M}_\perp\right)$$

[1] S. V. **Maleev**, V. G. Bar'yaktar, and R. A. Suris, The scattering of slow neutrons by complex magnetic structures Sov. Phys. Solid State 4, 2533 (1963)
[2] M. **Blume**, Polarization effects in the magnetic elastic scattering of slow neutrons, Phys. Rev. 130, 1670 (1963).

Yes, the Maleev-Blume equations are VERY busy

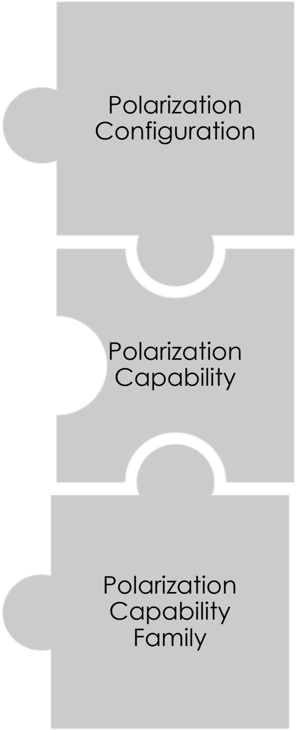
😢 😮 😀

- Leverage *personality flaw* found in some scientists
  A. *Make assumptions* about the system you are studying
  B. Eliminate terms
  C. Simplify / Streamline the math
- Linear algebra
  - *N* equations & *N* unknowns → solvable problem
- Let's call the solutions to the streamlined equations "Capabilities"
  - Think word problems in reverse…

Polarization
Capability

Configurations, Capabilities and Capability Families

- "Configurations" are specific combinations of polarization optics enabling access to different Maleev-Blume equations
    - Will show examples of configurations in upcoming slides!
- "Capabilities" are specific solutions to streamlined Linear algebra problems
    - Can only be solved utilizing a subset of "Configurations"
    - Assume certain terms in Maleev-Blume equations aren't present
    - Polarization-state 'equation' is actually several equations
- "Capability families" are intuitive (?) groupings of those specific solutions


Polarization Configuration

Polarization Capability

Polarization Capability Family

Capability Families

Isolate nuclear scattering	N & I_N
Isolate spin-incoherent scattering	I_{si}
Leverage dynamic nuclear polarization	$N \leftrightarrow I_{si}$
Solve Phase Problem	N & $\boldsymbol{M}_\perp$
Explore magnetic scattering	$\boldsymbol{M}_\perp$
Explore coinciding of nuclear and magnetic scattering	N with $\boldsymbol{M}_\perp$
Explore magnetic chirality	$\boldsymbol{M}_\perp$ cross terms

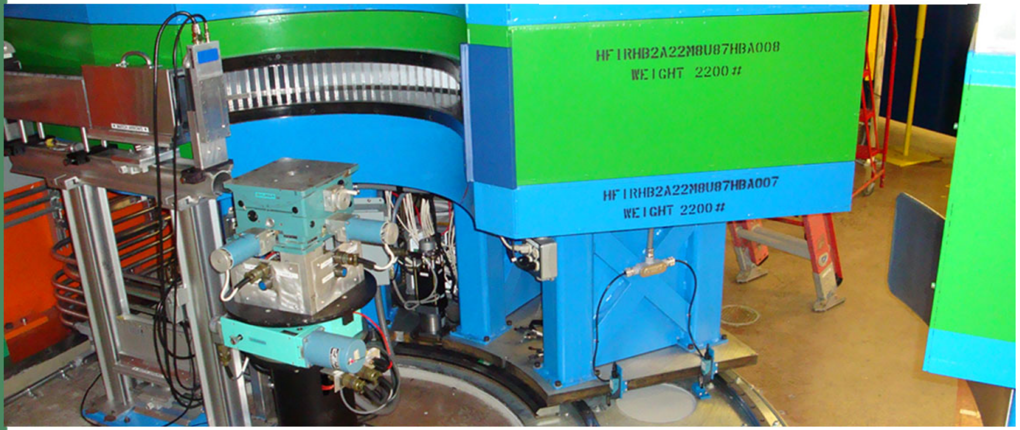
Color Key

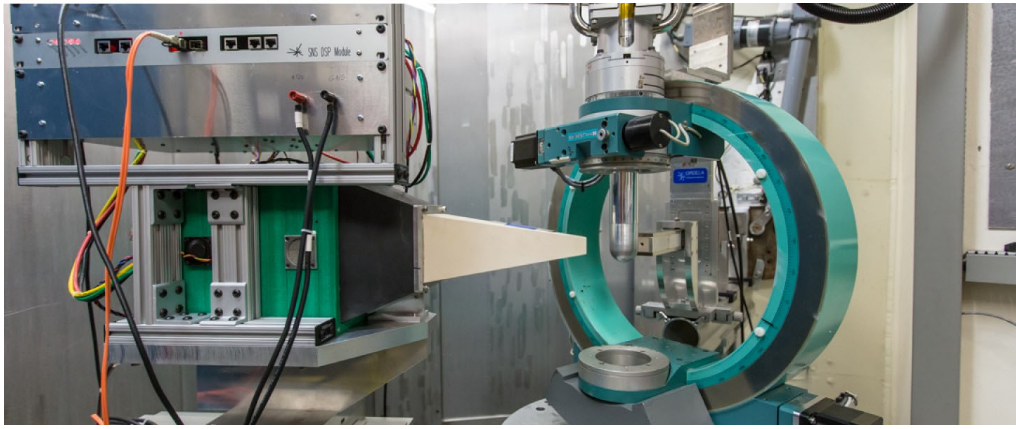
Nuclear Scattering (coherent & isotope-incoherent)
Spin-Incoherent Scattering
Magnetic Scattering
Dynamic Nuclear Polarization
Other

Instruments with half polarized configuration

Demonstration experiment N7

Demonstration experiment (lucky!) N13



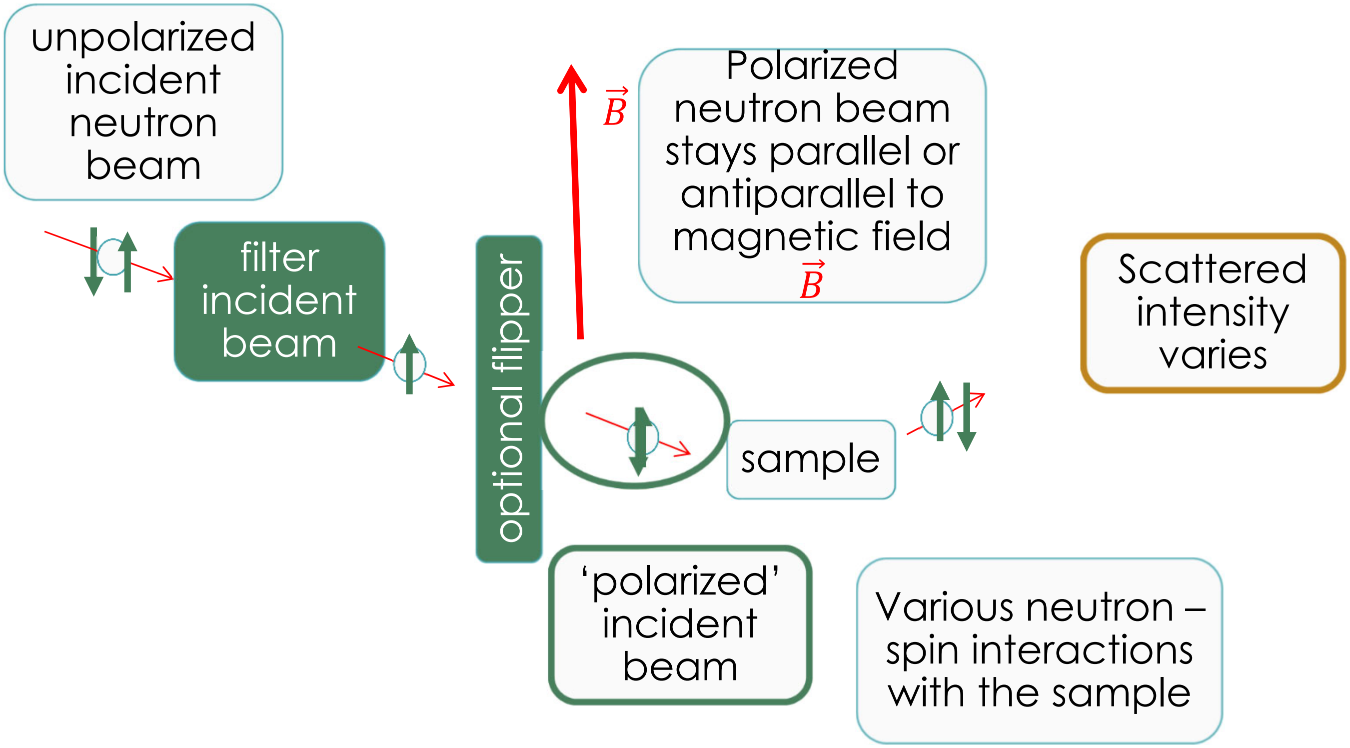


“Half Polarized” Configuration: 1filter, 1 flipper

$$I = N^{\dagger}N + I_{si} + \boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp} + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger} + i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)$$

Polarization Configuration

2 measurements
Doesn’t care about
neutron spin
reorientation



unpolarized incident neutron beam

filter incident beam

optional flipper

$\vec{B}$

Polarized neutron beam stays parallel or antiparallel to magnetic field $\vec{B}$

sample

Scattered intensity varies

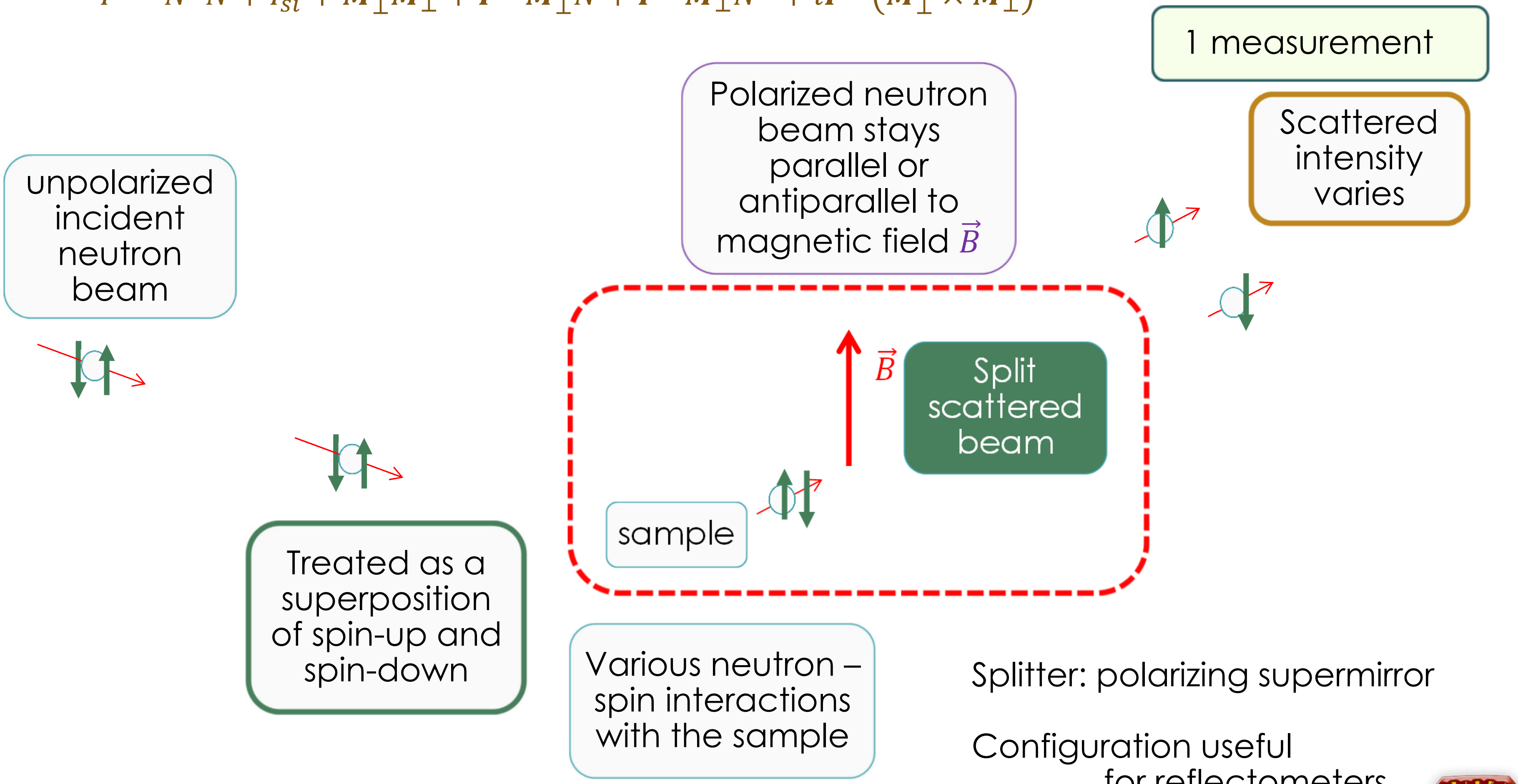
‘polarized’ incident beam

Various neutron – spin interactions with the sample

Polarization Configuration

Equivalent “Half Polarized” Configuration

$$I = N^{\dagger}N + I_{si} + \boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp} + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger} + i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)$$



Splitter: polarizing supermirror

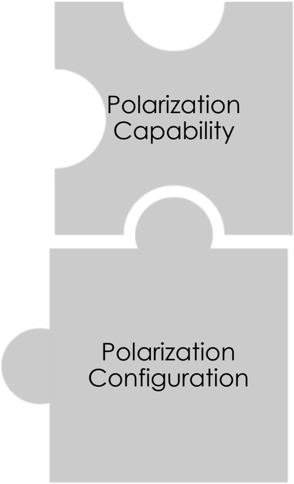
Configuration useful for reflectometers

“Half Polarized” Capabilities

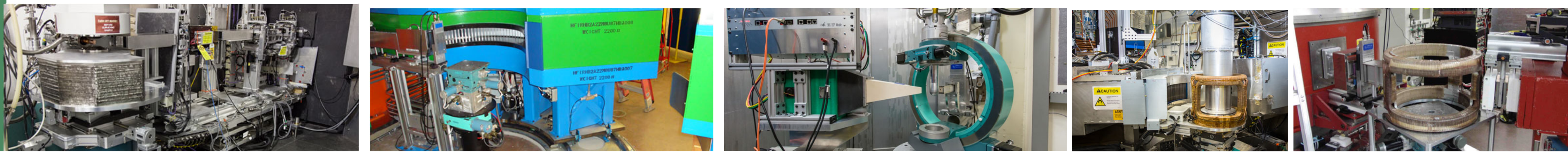
$$I = \underline{N^{\dagger}N + I_{si}} + \boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp} + \underline{\underline{\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N}} + \underline{\underline{\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger}}} + \underline{i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)}$$

“Ways around the phase problem”
slide 32 of SANS talk

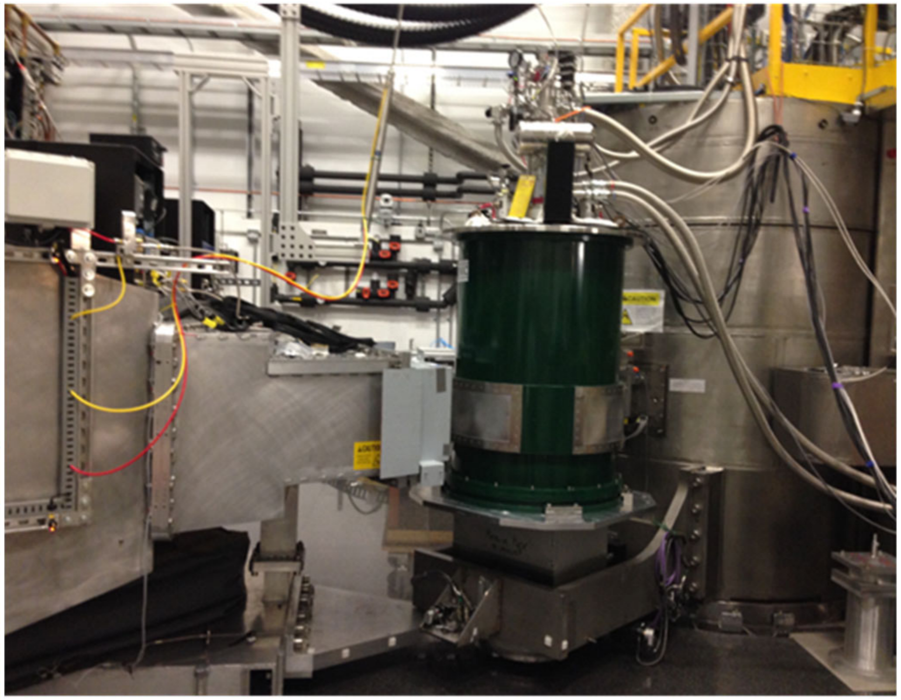
★	Separate magnetic susceptibility from nuclear and spin-incoherent signal
★	Solve the phase problem using an additional magnetic layer
☆	Identify coinciding of nuclear and magnetic coherent scattering in reciprocal space
★	Identify presence but not direction of a magnetic chiral structure
★	Magnon energy gain OR energy loss
★	Enhance coherent scattering contrast and S:N for hydrogen



Capabilities are ‘word problems’ in reverse!



For successful Half-polarized measurements, you must magnetically saturate your sample!



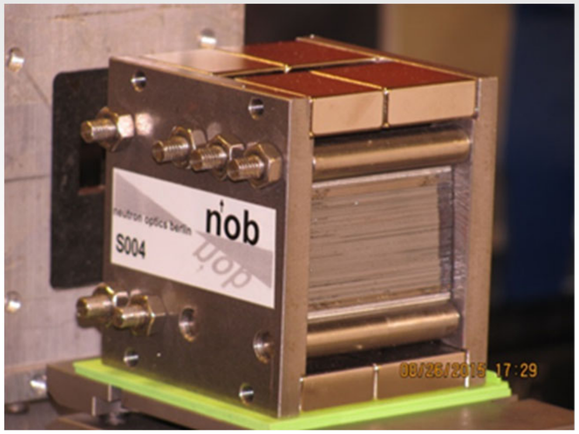
‘Extreme’ half-polarized samples make the best polarization filters

Polarization Configuration

Polarizing supermirrors

- $\boldsymbol{M}_\perp^\dagger \boldsymbol{M}_\perp + \boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger N + \boldsymbol{P}\cdot\boldsymbol{M}_\perp N^\dagger$

  (but using index of refraction)

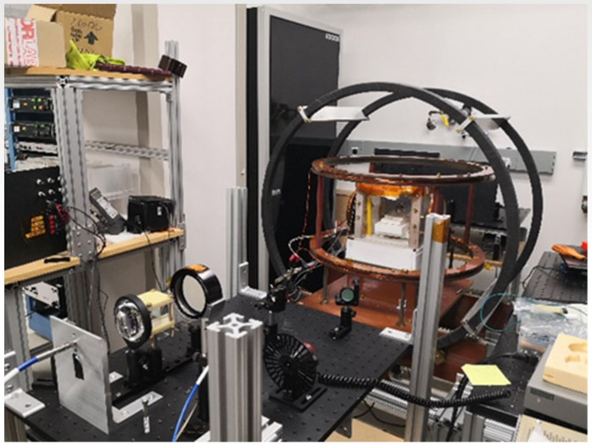


Heusler Bragg Optics

- $\boldsymbol{M}_\perp^\dagger \boldsymbol{M}_\perp + \boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger N + \boldsymbol{P}\cdot\boldsymbol{M}_\perp N^\dagger$

^{3}He filters

- $\sigma^{u}_{abs} \Longrightarrow \sigma^{\uparrow\downarrow}_{abs} + \sigma^{\downarrow\downarrow}_{abs} \Rightarrow \sigma^{\uparrow\downarrow}_{abs}$



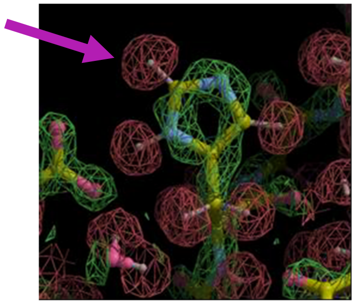
These also demonstrate different ways (Capabilities) to access changes in the Maleev-Blume intensity equation

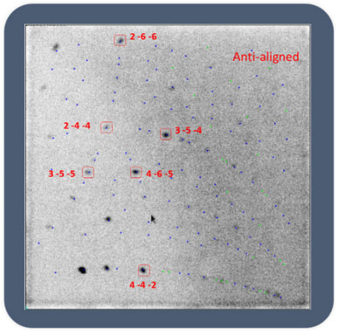
Polarization Capability

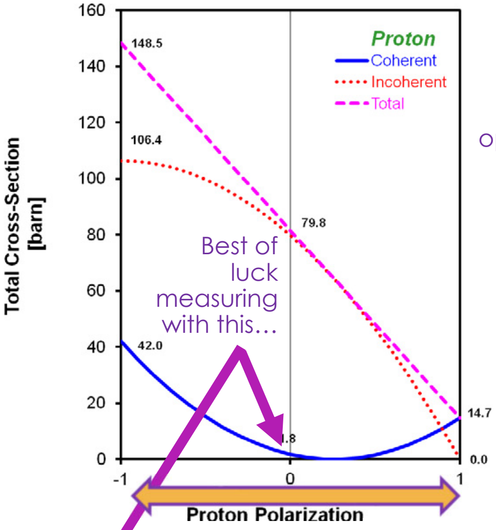
Dynamic Nuclear Polarization Configuration

Anti-aligned

Better



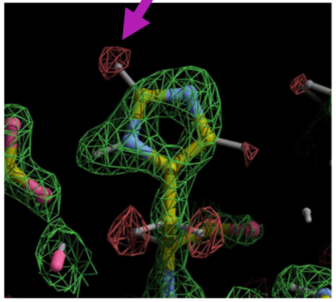


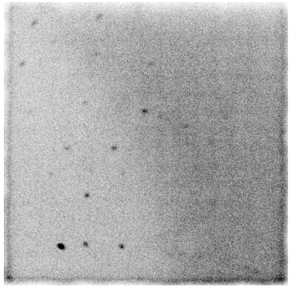


Best of luck measuring with this…

Hydrogen

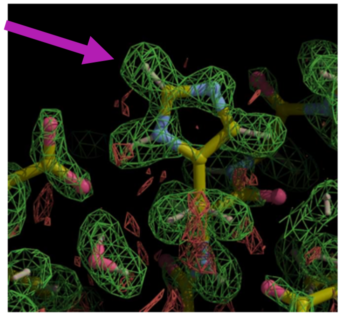
Unpolarized

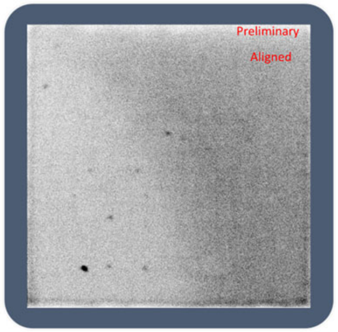




Better and opposite sign

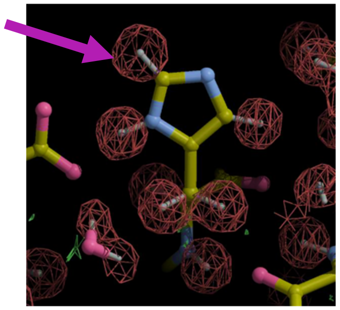
Aligned





Anti-aligned - Aligned

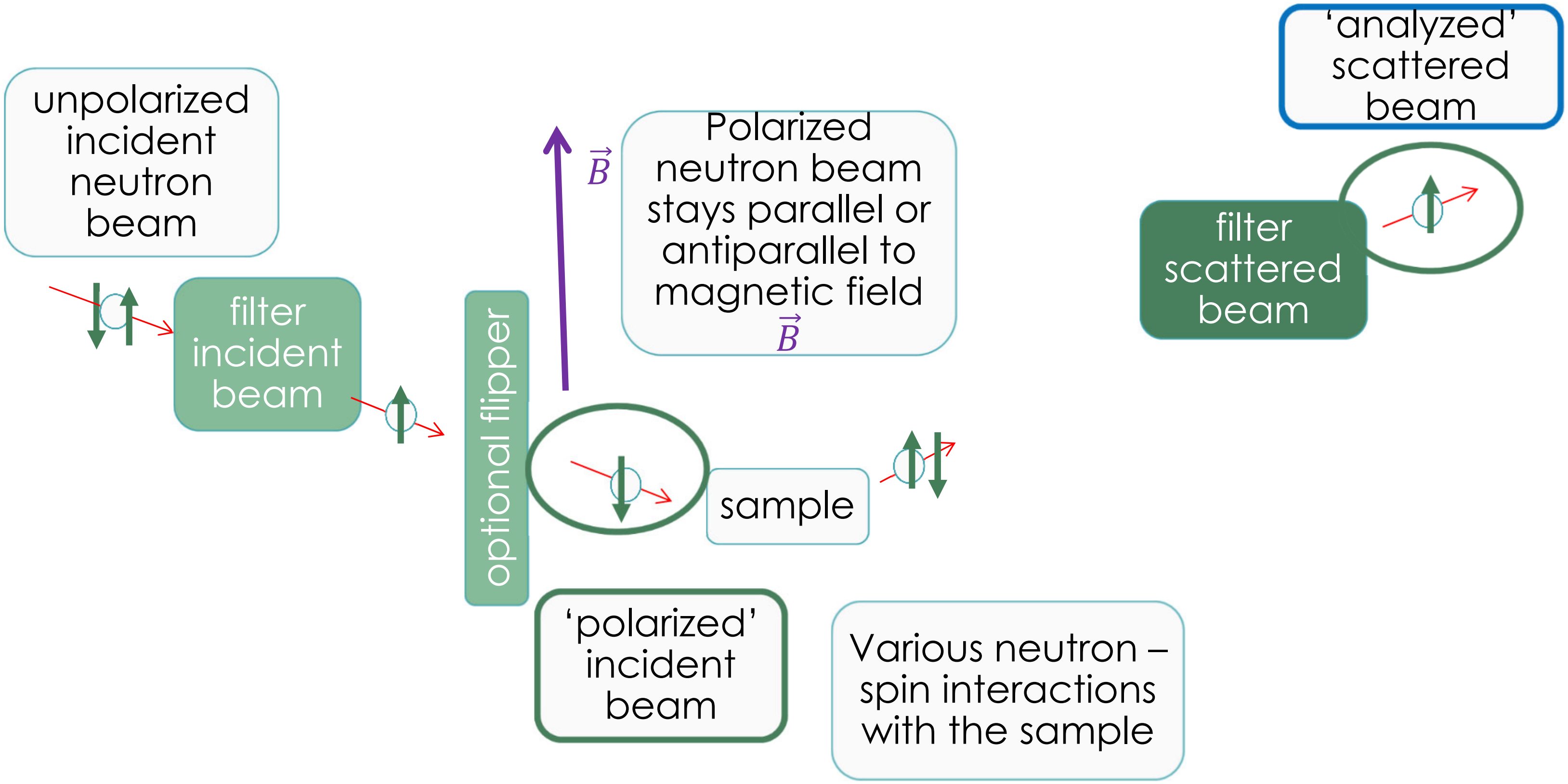
Nailed the hydrogen location



Longitudinal Analysis 1 Configuration

2 measurements
Assumes $I^{\uparrow\downarrow} = I^{\downarrow\uparrow}$

$$\boldsymbol{P}^{1}I = \boldsymbol{P}\left(N^{\dagger}N - \frac{1}{3}I_{si}\right) + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger})\boldsymbol{M}_{\perp} + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp})\boldsymbol{M}_{\perp}^{\dagger} - \boldsymbol{P}(\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp}) + iN(\boldsymbol{P}\times\boldsymbol{M}_{\perp}^{\dagger}) - iN^{\dagger}(\boldsymbol{P}\times\boldsymbol{M}_{\perp}) + N\boldsymbol{M}_{\perp}^{\dagger} + N^{\dagger}\boldsymbol{M}_{\perp} - i(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp})$$



unpolarized incident neutron beam

filter incident beam

optional flipper

$\vec{B}$

Polarized neutron beam stays parallel or antiparallel to magnetic field $\vec{B}$

'polarized' incident beam

sample

Various neutron – spin interactions with the sample

'analyzed' scattered beam

filter scattered beam

distinguish between spin flip and non-flip at sample

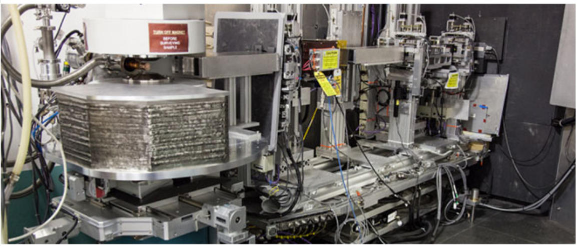
Longitudinal Analysis 1 Capabilities

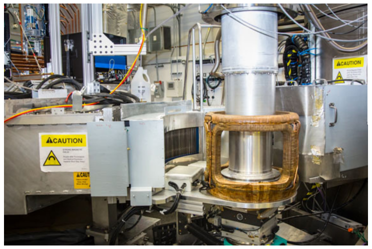
$$\boldsymbol{P}^{1}I = \boldsymbol{P}\left(N^{\dagger}N - \frac{1}{3}I_{si}\right) + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger})\boldsymbol{M}_{\perp} + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp})\boldsymbol{M}_{\perp}^{\dagger} - \boldsymbol{P}(\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp}) + iN(\boldsymbol{P}\times\boldsymbol{M}_{\perp}^{\dagger}) - iN^{\dagger}(\boldsymbol{P}\times\boldsymbol{M}_{\perp}) + N\boldsymbol{M}_{\perp}^{\dagger} + N^{\dagger}\boldsymbol{M}_{\perp} - i(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp})$$

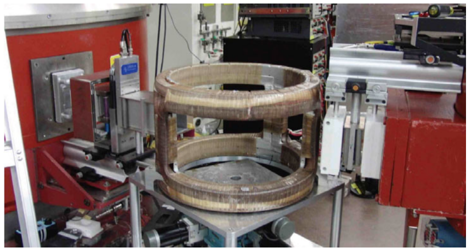
	☆	Separate nuclear scattering from spin-incoherent scattering
	★	Separate spin-incoherent scattering from nuclear scattering
	★	Determine quickly whether a signal is magnetic in origin
	★	Track an order parameter for ferromagnetism of a powder via depolarization of thru beam
3D	☆	Separate nuclear scattering from both spin-incoherent scattering and magnetic scattering
	★	Separate spin-incoherent scattering from both both nuclear scattering and magnetic scattering
	★	Quantify the isotropic magnetic moment magnitude via separation from nuclear and spin-incoherent scattering
	★	Quantify the magnetic moment magnitude and direction and separate it form both nuclear and spin-incoherent scattering
	★	Enhance coherent scattering contrast and S:N for hydrogen and Separate from spin-incoherent scattering

Polarization Capability

Polarization Configuration



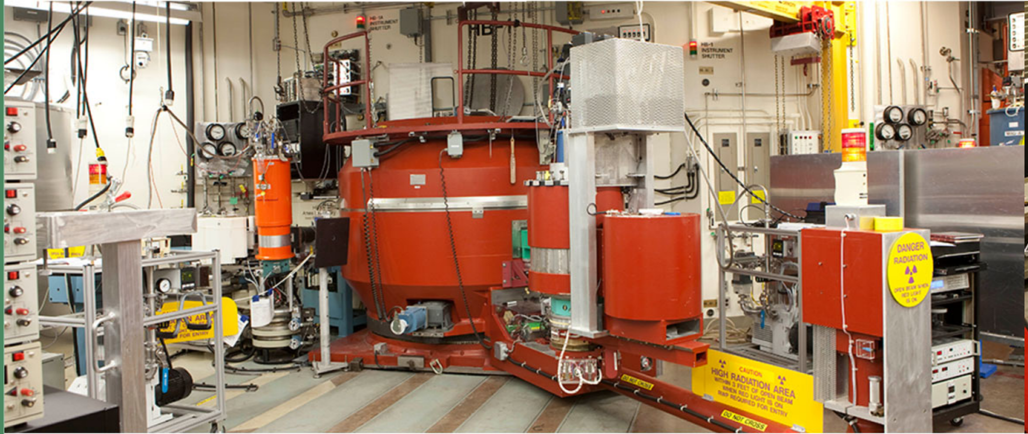


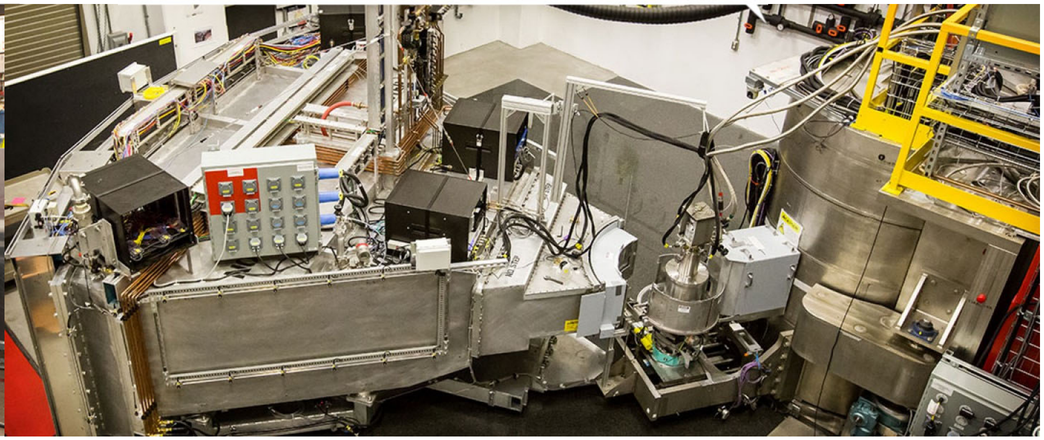


Both Half Polarized and Longitudinal 1configurations

Demonstration experiment N1-a

Demonstration experiment **N2** leverages the **Longitudinal 1** configuration



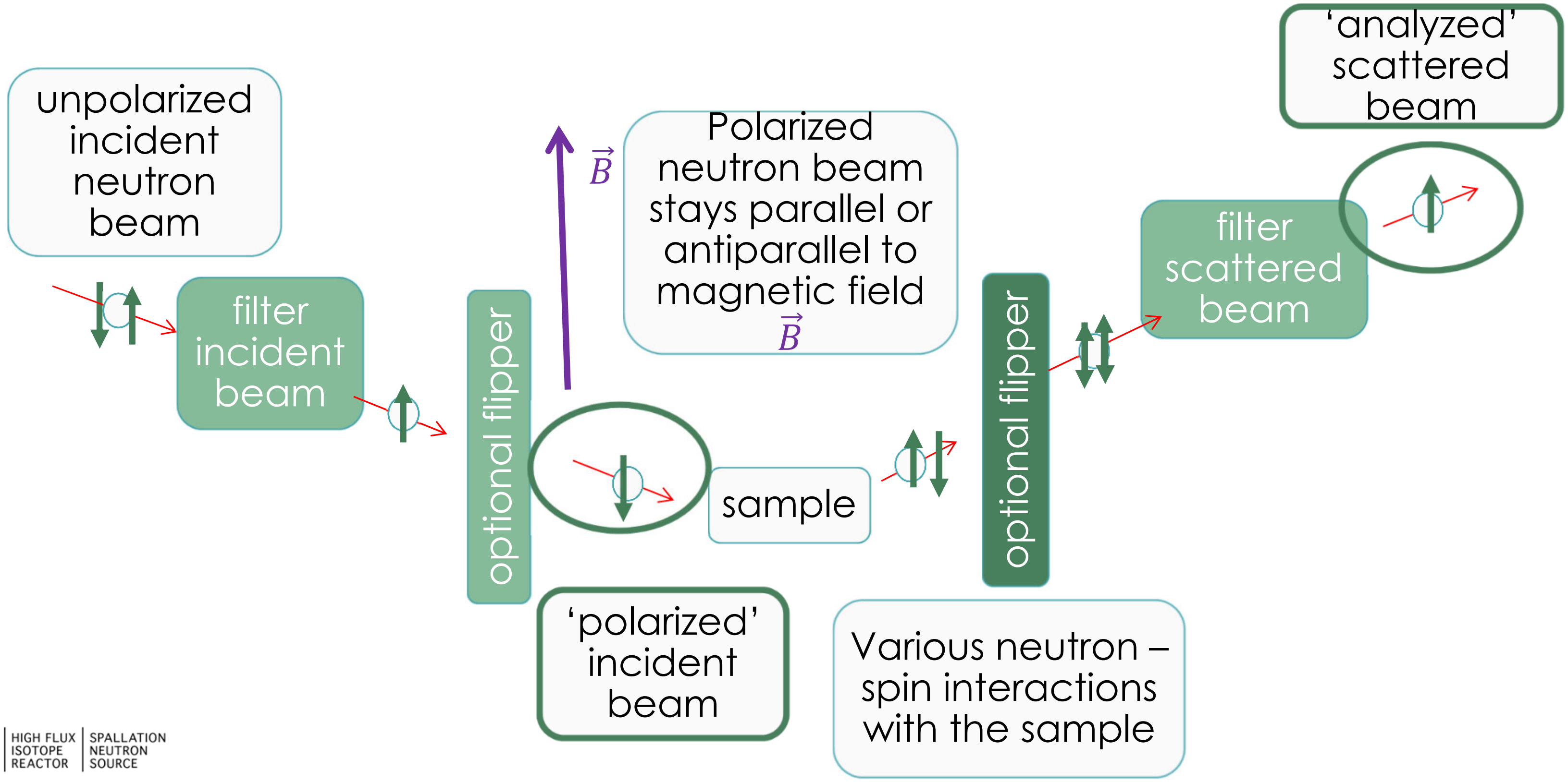


Longitudinal Analysis 2 Configuration

4 measurements
12 for 3D
Allows $I^{\uparrow\downarrow} \neq I^{\downarrow\uparrow}$

$$I = N^\dagger N + I_{si} + \boldsymbol{M}_\perp^\dagger \boldsymbol{M}_\perp + \boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger N + \boldsymbol{P}\cdot\boldsymbol{M}_\perp N^\dagger + i\boldsymbol{P}\cdot\left(\boldsymbol{M}_\perp^\dagger \times \boldsymbol{M}_\perp\right)$$

$$\boldsymbol{P}^1 I = \boldsymbol{P}\left(N^\dagger N - \frac{1}{3}I_{si}\right) + (\boldsymbol{P}\cdot\boldsymbol{M}_\perp^\dagger)\boldsymbol{M}_\perp + (\boldsymbol{P}\cdot\boldsymbol{M}_\perp)\boldsymbol{M}_\perp^\dagger - \boldsymbol{P}\left(\boldsymbol{M}_\perp^\dagger\boldsymbol{M}_\perp\right) + iN\left(\boldsymbol{P}\times\boldsymbol{M}_\perp^\dagger\right) - iN^\dagger(\boldsymbol{P}\times\boldsymbol{M}_\perp) + N\boldsymbol{M}_\perp^\dagger + N^\dagger\boldsymbol{M}_\perp - i\left(\boldsymbol{M}_\perp^\dagger\times\boldsymbol{M}_\perp\right)$$



unpolarized incident neutron beam

filter incident beam

optional flipper

$\vec{B}$

Polarized neutron beam stays parallel or antiparallel to magnetic field $\vec{B}$

‘polarized’ incident beam

sample

Various neutron – spin interactions with the sample

optional flipper

filter scattered beam

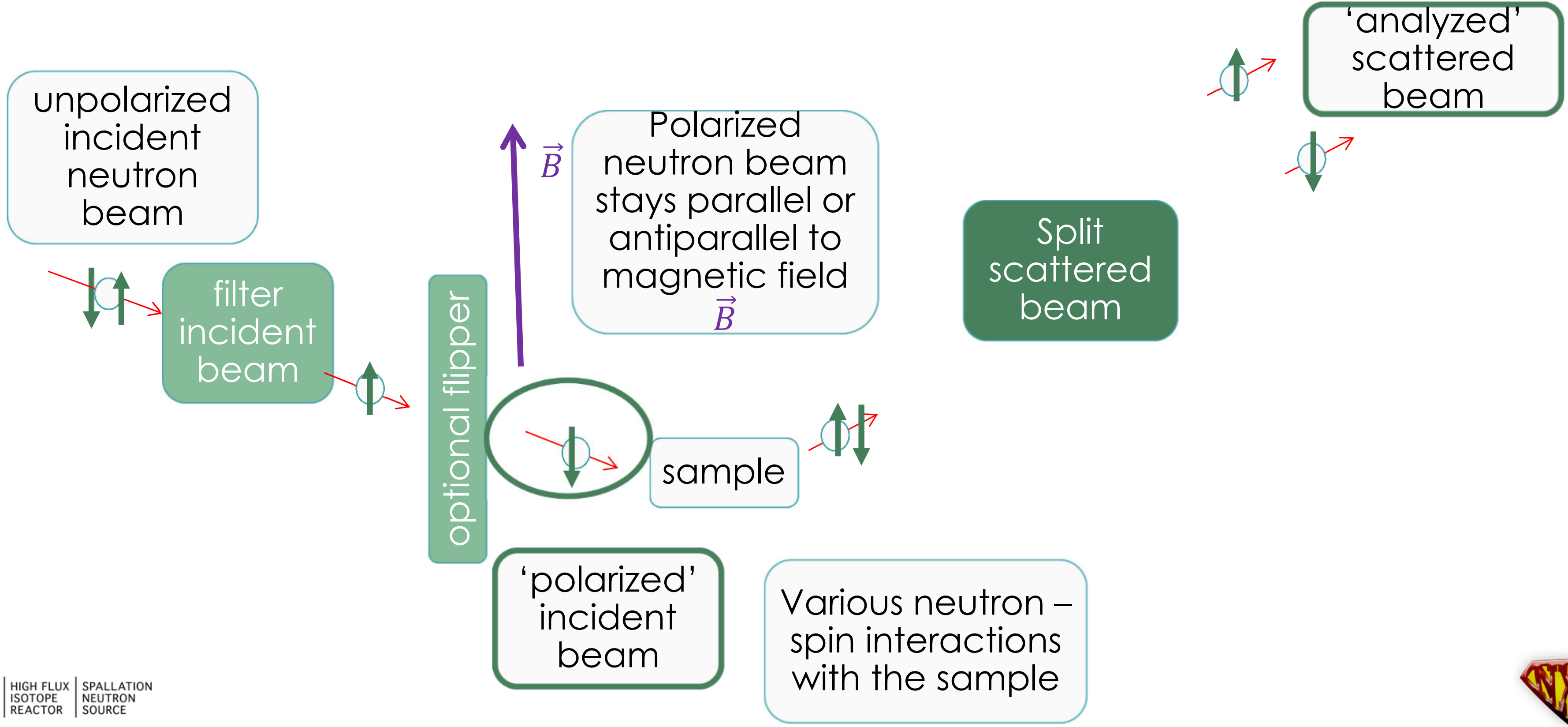
‘analyzed’ scattered beam

Equivalent Longitudinal Analysis 2 Configuration

2 measurements for MAGREF 1D

$$I = N^{\dagger}N + I_{si} + \boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp} + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger} + i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)$$

$$\boldsymbol{P}^{1}I = \boldsymbol{P}\left(N^{\dagger}N - \frac{1}{3}I_{si}\right) + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger})\boldsymbol{M}_{\perp} + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp})\boldsymbol{M}_{\perp}^{\dagger} - \boldsymbol{P}\left(\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp}\right) + iN\left(\boldsymbol{P}\times\boldsymbol{M}_{\perp}^{\dagger}\right) - iN^{\dagger}(\boldsymbol{P}\times\boldsymbol{M}_{\perp}) + N\boldsymbol{M}_{\perp}^{\dagger} + N^{\dagger}\boldsymbol{M}_{\perp} - i\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)$$



unpolarized incident neutron beam

filter incident beam

optional flipper

$\vec{B}$

Polarized neutron beam stays parallel or antiparallel to magnetic field $\vec{B}$

sample

Split scattered beam

'analyzed' scattered beam

'polarized' incident beam

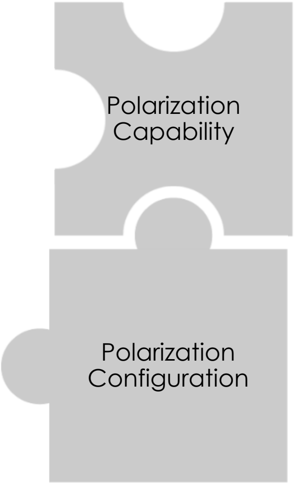
Various neutron – spin interactions with the sample

Longitudinal Analysis 2 Capabilities

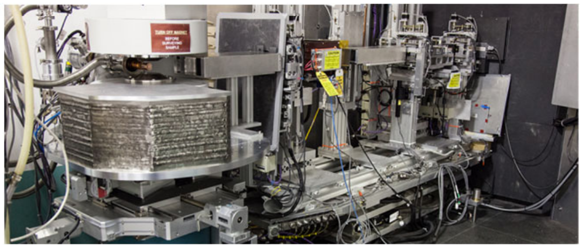
$$I = N^{\dagger}N + I_{si} + \boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp} + \underline{\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger}} + \underline{i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)}$$

$$\boldsymbol{P}^{\prime}I = \boldsymbol{P}\left(N^{\dagger}N - \frac{1}{3}I_{si}\right) + \underline{(\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger})\boldsymbol{M}_{\perp} + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp})\boldsymbol{M}_{\perp}^{\dagger} - \boldsymbol{P}\left(\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp}\right)} + iN\left(\boldsymbol{P}\times\boldsymbol{M}_{\perp}^{\dagger}\right) - iN^{\dagger}(\boldsymbol{P}\times\boldsymbol{M}_{\perp}) + \underline{N\boldsymbol{M}_{\perp}^{\dagger} + N^{\dagger}\boldsymbol{M}_{\perp}} - i\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)$$

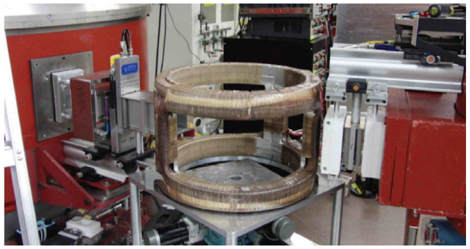
★	Quantify magnetic moment and direction
★	Quantify 3D aspect of the coinciding of nuclear and magnetic coherent scattering in reciprocal space
★	Partly quantify chiral magnetic structure or dynamics



Demonstration experiment **N20** leverages the **Longitudinal 2** configuration



Rarely used.
Impossible with magnet due to size restrictions



Coming at neutron spin from another angle…

- Enhanced resolution via Larmor techniques
- In a *sick and twisted* sense, this is a variation of Longitudinal 2…

One person’s Trash is another’s Treasure

We NEVER want $\vec{n} \perp \vec{B}$ for polarization analysis

- Magnetic Guide Fields
  - Intended to preserve neutron spin state
  - Either aligned or anti-aligned with magnetic field
  - $\vec{n} \parallel \vec{B}$
  - Larmor precession used to maintain this relationship
- OK… One exception was pitched*

Depolarization via Larmor precession in a macroscopic system is ANOTHER magnetic contrast mechanism

And yet, that’s EXACTLY what’s done for ‘Larmor’ techniques

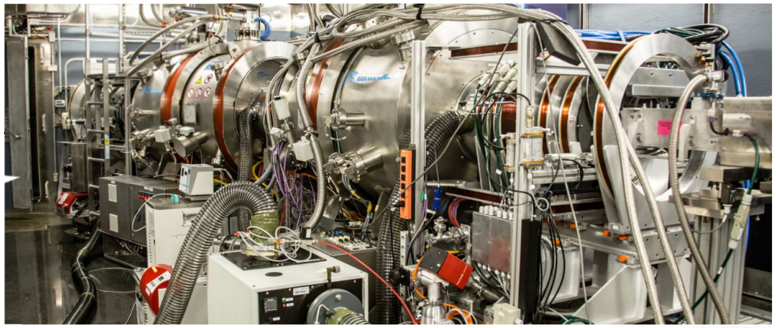
- More how we manipulate the neutron before and after sample
- Leverage well defined precession frequency to “clock in, clock out”
- Utilizes $\pi/2$ “flippers”, from aligned with guide field to $\perp$

See Poster on Wollaston prisms during 2nd half of this lecture

See Lecture
Thurs 11:30: NSE (Laura)
& Experiment
N6: Neutron Spin Echo

Utilize Larmor precession to reveal subtle phase changes

- Less about your sample, contrast & character
- Enhanced resolution for
  - Angle (diffraction, small angle scattering)
  - Energy / time scales



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*Not entirely true. One published demonstration combined SNP & Larmor technique. Never repeated to my knowledge.
W. Schweika, “Time-of-flight and vector polarization analysis for diffuse neutron scattering” *Physica B* **335** 157 (2003)

Prelude to the final polarization configuration: <u>3 more components</u>

Zero field region

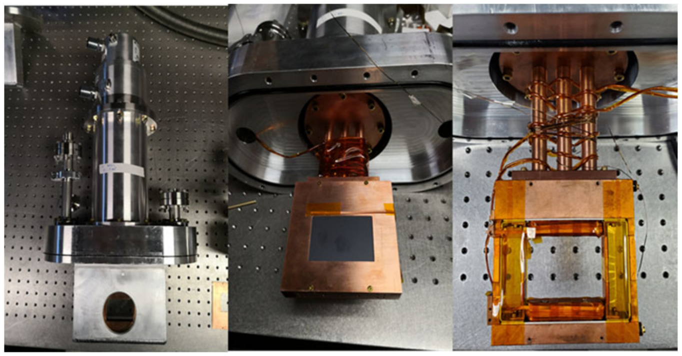
- *Instead of* preserving neutron moment direction with ‘guide field’ & Larmor precession
- Only way* to move beyond ‘spin-flip’ and ‘non-spin-flip’ for scattering at sample

Zero-field chamber established in part with m-metal enclosure

Meissner (superconducting) barrier

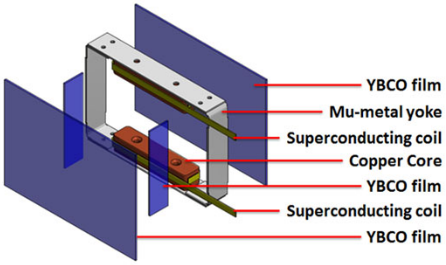
- $\vec{B}$ at interface is either $\|$ surface or 0

One screen on this precession chamber couples with external guide field, & the opposite screen couples with zero-field chamber



Precession chamber

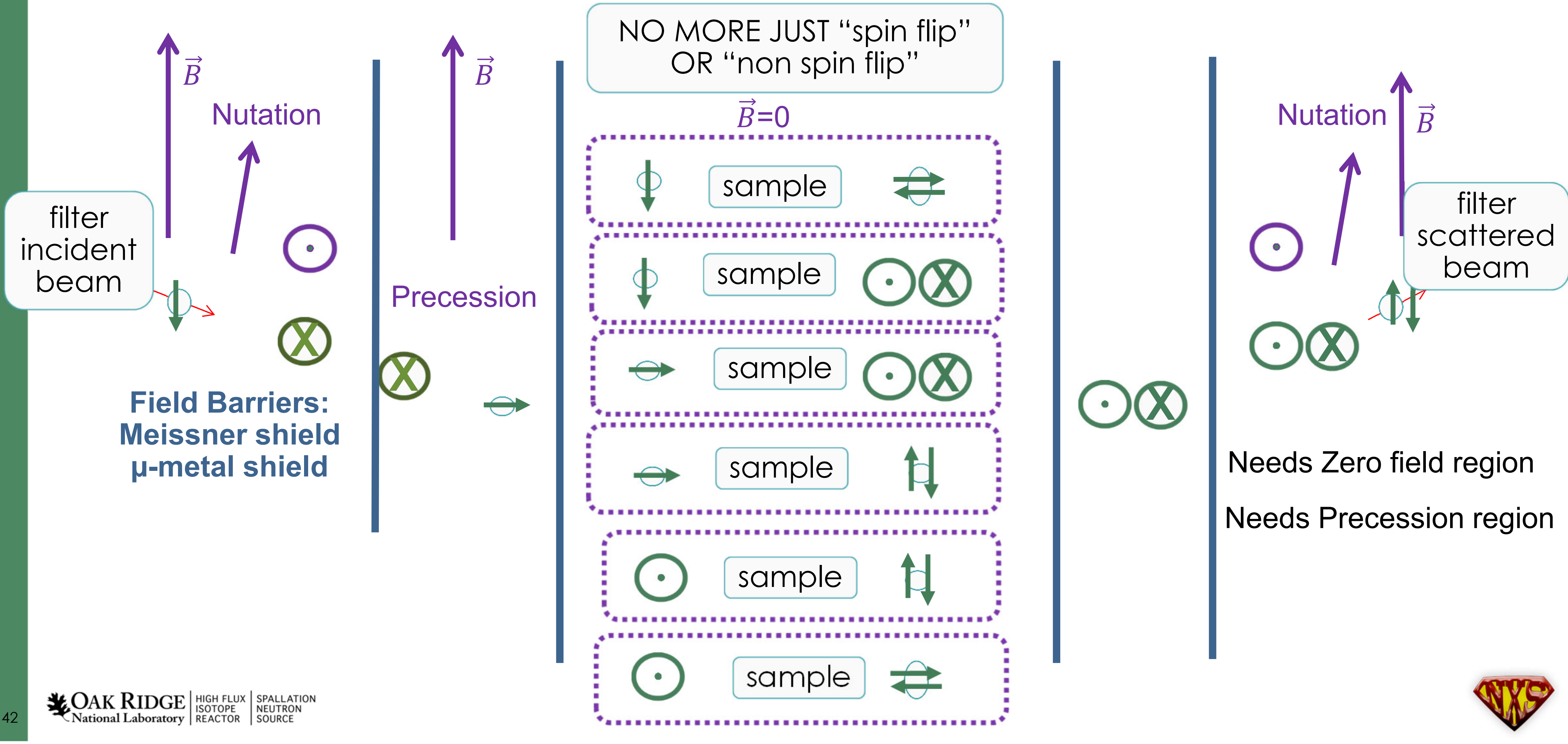
- Like Mezei flipper, but rotates to arbitrary angle, not just 180°
- Coupled with nutator or another precession chamber, enables arbitrary orientation for neutron moment
  - Established before sample
  - Extracted after sample



*Not entirely true. One published demonstration combined SNP & Larmor technique. Never repeated to my knowledge.
W. Schweika, “Time-of-flight and vector polarization analysis for diffuse neutron scattering” *Physica B* **335** 157 (2003)

Spherical Polarimetry Configuration

many measurements



NO MORE JUST “spin flip” OR “non spin flip”

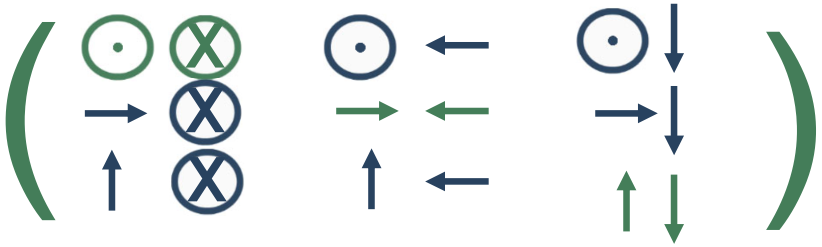
Field Barriers:
Meissner shield
μ-metal shield

Needs Zero field region

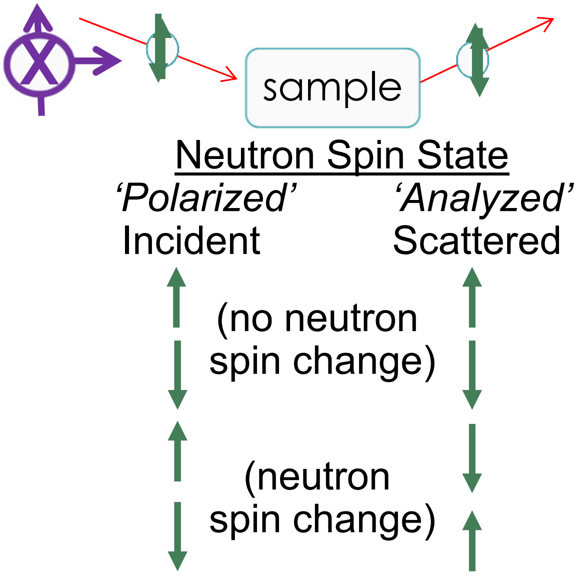
Needs Precession region

$S_{ab}(\vec{Q},\omega)$ Full Magnetic Structure Factor Tensor

$$\begin{pmatrix} S_{xx}(\vec{Q},\omega) & S_{xy}(\vec{Q},\omega) & S_{xz}(\vec{Q},\omega) \\ S_{yx}(\vec{Q},\omega) & S_{yy}(\vec{Q},\omega) & S_{yz}(\vec{Q},\omega) \\ S_{zx}(\vec{Q},\omega) & S_{zy}(\vec{Q},\omega) & S_{zz}(\vec{Q},\omega) \end{pmatrix}$$



Neutron spin parallel or antiparallel to $\vec{B}$



Neutron Spin State

'Polarized' Incident

'Analyzed' Scattered

(no neutron spin change)

(neutron spin change)

Off-Diagonal elements

needs 0 field at sample

Beyond Spin-Flip

Need for more exotic systems (chiral, spin-lattice coupling[1])

Diagonal elements

Multiferroics

Antiferromagnets

Diffuse scattering

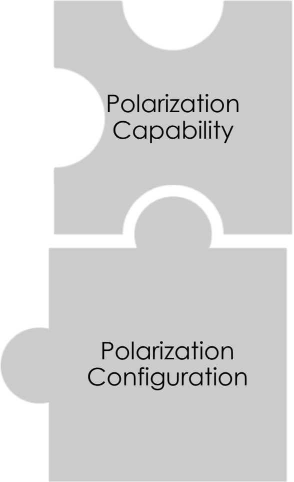
[1] J. Phys.: Condens. Matter **9**, 4729 (1997)

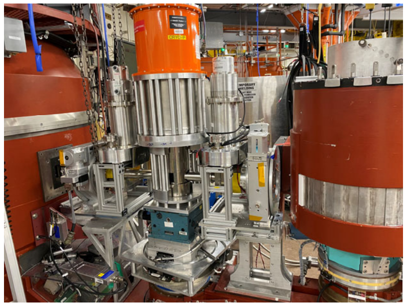
Spherical Polarimetry Capabilities

$$I = N^{\dagger}N + I_{si} + \underline{\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp}} + \underline{\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger}N + \boldsymbol{P}\cdot\boldsymbol{M}_{\perp}N^{\dagger}} + \underline{i\boldsymbol{P}\cdot\left(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp}\right)}$$

$$\boldsymbol{P}'I = \boldsymbol{P}\left(N^{\dagger}N - \frac{1}{3}I_{si}\right) + \underline{(\boldsymbol{P}\cdot\boldsymbol{M}_{\perp}^{\dagger})\boldsymbol{M}_{\perp} + (\boldsymbol{P}\cdot\boldsymbol{M}_{\perp})\boldsymbol{M}_{\perp}^{\dagger} - \boldsymbol{P}(\boldsymbol{M}_{\perp}^{\dagger}\boldsymbol{M}_{\perp})} + \underline{iN(\boldsymbol{P}\times\boldsymbol{M}_{\perp}^{\dagger}) - iN^{\dagger}(\boldsymbol{P}\times\boldsymbol{M}_{\perp}) + N\boldsymbol{M}_{\perp}^{\dagger} + N^{\dagger}\boldsymbol{M}_{\perp}} - \underline{i(\boldsymbol{M}_{\perp}^{\dagger}\times\boldsymbol{M}_{\perp})}$$

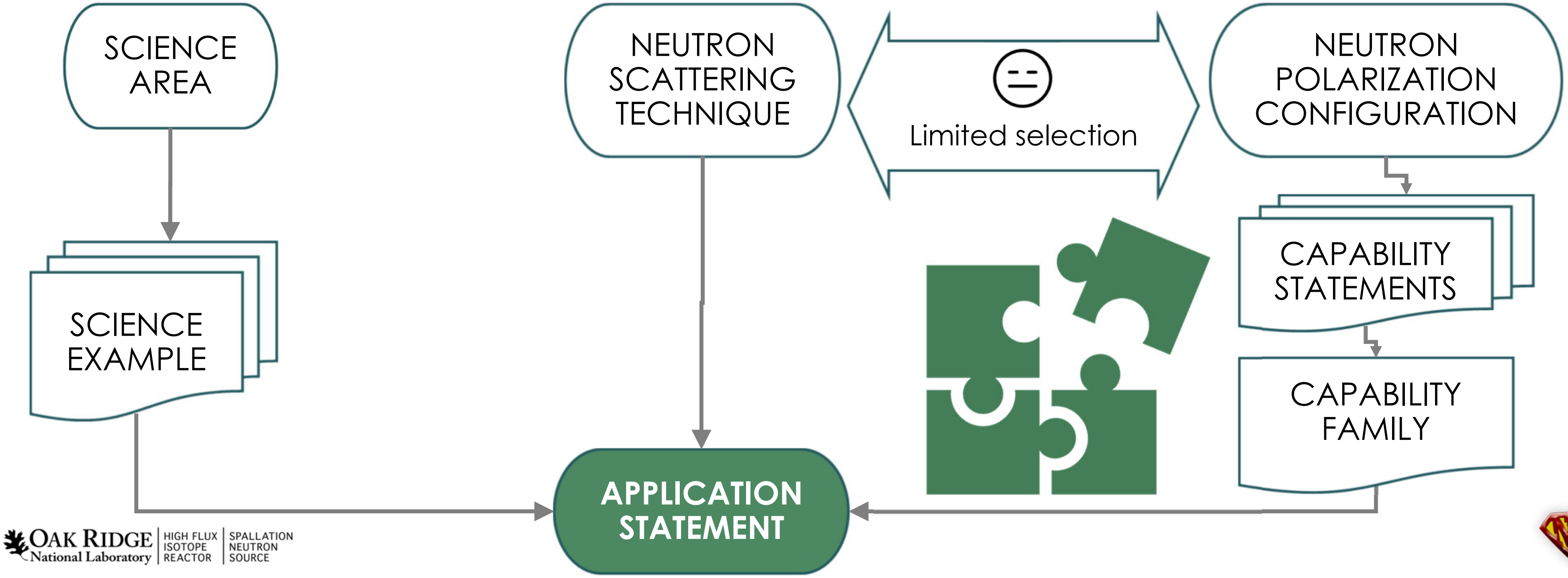
★	Quantify magnetic moment and direction
☆	Quantify 3D aspect of the coinciding of nuclear and magnetic coherent scattering in reciprocal space
★	Quantify magnitude and direction of chiral magnetic structure or dynamics
☆	Insight into magnetic domains
	Best approach for polarization analysis of any superconducting sample of unusual shape





Q1 Ingredients for your ‘polarized’ experimental question

As a [***SCIENCE AREA***] neutron scattering experimentalist, I want to [***CAPABILITY FAMILY***] so that when using [***NEUTRON SCATTERING TECHNIQUE***] with [***NEUTRON POLARIZATION CONFIGURATION***] I can [***APPLICATION statement***] for [***SCIENCE EXAMPLE***]



SCIENCE AREA

SCIENCE EXAMPLE

NEUTRON SCATTERING TECHNIQUE

Limited selection

NEUTRON POLARIZATION CONFIGURATION

CAPABILITY STATEMENTS

CAPABILITY FAMILY

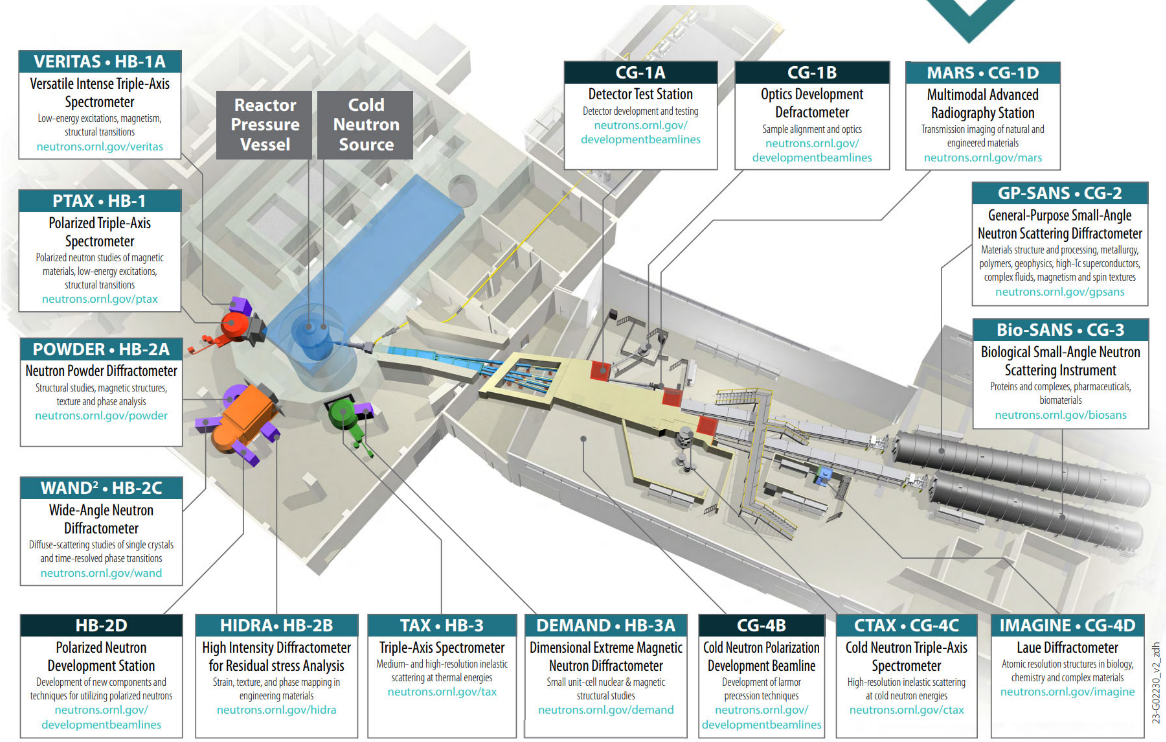
APPLICATION STATEMENT

Q3 Reality Check?

Limitations of Neutron Polarization Configurations

-or-

“Why aren’t we doing this ALL the time!?!”



Dev

Dev

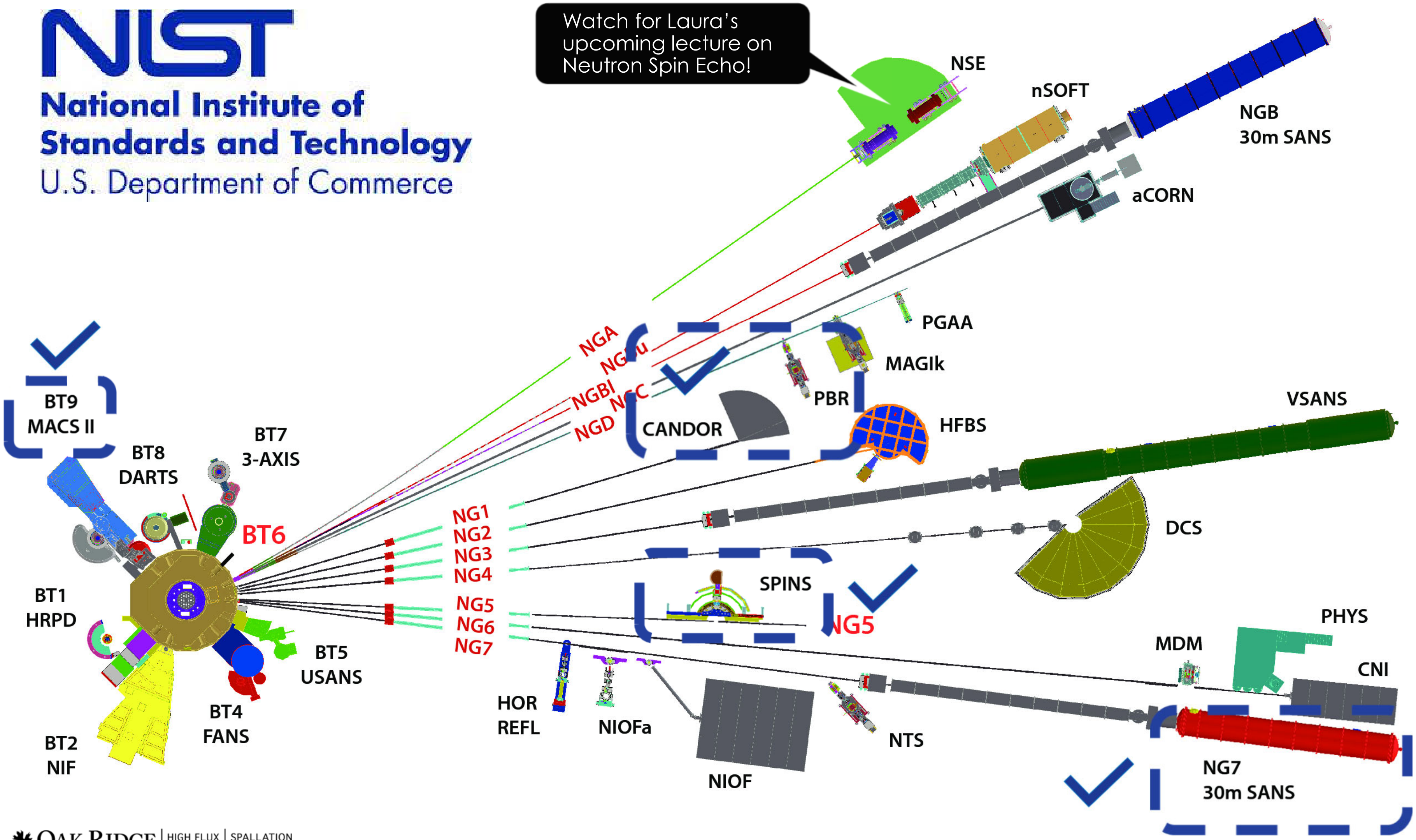
Reactor-based instrument advantages for polarized neutrons:

- More easily configurable about the sample
- Often monochromatic incident beam very compatible with ALL polarization optics
- Smaller detector arrays,

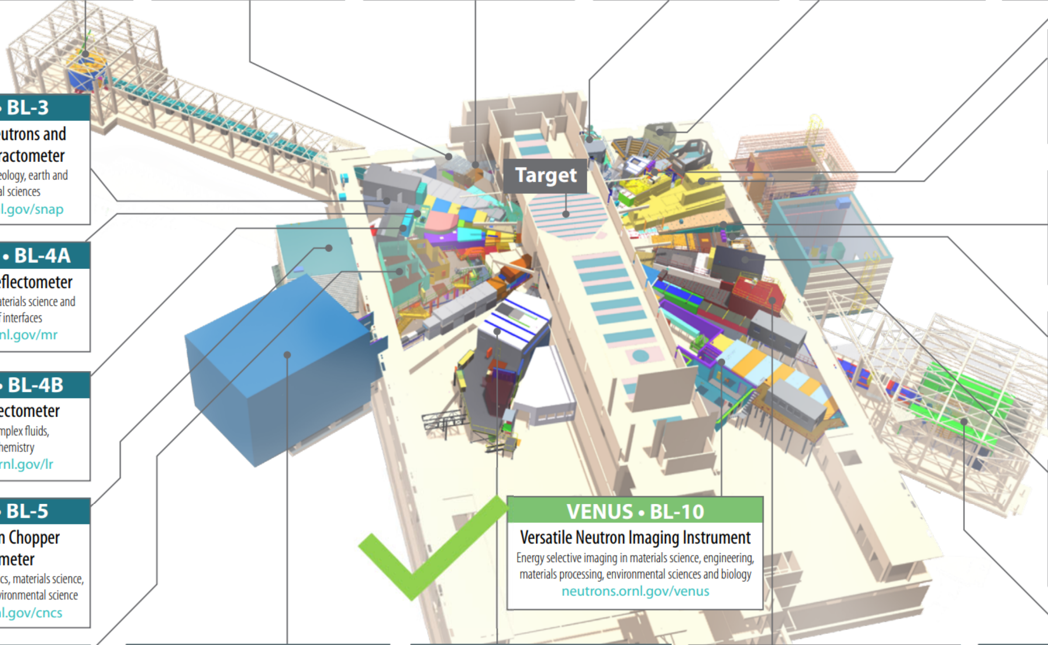
→ **better** for polarization development

National Institute of Standards and Technology
U.S. Department of Commerce

Watch for Laura's upcoming lecture on Neutron Spin Echo!



See Lecture
Thurs 11:30: NSE (Laura)
& Experiment
N6: Neutron Spin Echo



BASIS • BL-2
Backscattering Spectrometer
Dynamics of macromolecules, constrained molecular systems, polymers, biology, chemistry, materials science
neutrons.ornl.gov/basis

NOMAD • BL-1B
Nanoscale-Ordered Materials Diffractometer
Liquids, solutions, glasses, polymers, nanocrystalline and partially ordered complex materials
neutrons.ornl.gov/nomad

USANS • BL-1A
Ultra-Small-Angle Neutron Scattering Instrument
Life sciences, polymers, materials science, earth and environmental sciences
neutrons.ornl.gov/usans

ARCS • BL-18
Wide Angular-Range Chopper Spectrometer
Atomic-level dynamics in materials science, chemistry, condensed matter sciences
neutrons.ornl.gov/arcs

SEQUOIA • BL-17
Fine-Resolution Fermi Chopper Spectrometer
Dynamics of complex fluids, quantum fluids, magnetism, condensed matter, materials science
neutrons.ornl.gov/sequoia

VISION • BL-16B
Vibrational Spectrometer
Vibrational dynamics in molecular systems, chemistry
neutrons.ornl.gov/vision

NSE • BL-15
Neutron Spin Echo Spectrometer
High-resolution dynamics of slow processes, polymers, biological macromolecules
neutrons.ornl.gov/nse

HYSPEC • BL-14B
Hybrid Spectrometer
Dynamics of quantum materials with optional polarization analysis
neutrons.ornl.gov/hyspec

FNP • BL-13
Fundamental Neutron Physics Beam Line
Fundamental properties of neutrons
neutrons.ornl.gov/fnpb

Polarized neutrons enable deeper understanding of 'fundamental physics'

TOPAZ • BL-12
Single-Crystal Diffractometer
Atomic-level structures in chemistry, biology, earth science, materials science, condensed matter physics
neutrons.ornl.gov/topaz

SNAP • BL-3
Spallation Neutrons and Pressure Diffractometer
Materials science, geology, earth and environmental sciences
neutrons.ornl.gov/snap

MAGREF • BL-4A
Magnetism Reflectometer
Condensed matter, materials science and magnetism of interfaces
neutrons.ornl.gov/mr

LIQREF • BL-4B
Liquids Reflectometer
Interfaces in complex fluids, polymers, chemistry
neutrons.ornl.gov/lr

CNCS • BL-5
Cold Neutron Chopper Spectrometer
Condensed matter physics, materials science, chemistry, biology, environmental science
neutrons.ornl.gov/cncs

EQ-SANS • BL-6
Extended Q-Range Small-Angle Neutron Scattering Diffractometer
Polymers, soft materials and colloidal systems, materials science, life science, earth and environmental sciences
neutrons.ornl.gov/eqsans

VENUS • BL-10
Versatile Neutron Imaging Instrument
Energy selective imaging in materials science, engineering, materials processing, environmental sciences and biology
neutrons.ornl.gov/venus

VULCAN • BL-7
Engineering Materials Diffractometer
Mechanical behaviors, materials science, materials processing
neutrons.ornl.gov/vulcan

CORELLI • BL-9
Elastic Diffuse Scattering Spectrometer
Detailed studies of disorder in crystalline materials
neutrons.ornl.gov/corelli

MANDI • BL-11B
Macromolecular Neutron Diffractometer
Atomic level structures of proteins, macromolecules and DNA
neutrons.ornl.gov/mandi

POWGEN • BL-11A
Powder Diffractometer
Atomic-level structures in chemistry, materials science, and condensed matter physics including magnetic structure
neutrons.ornl.gov/powgen

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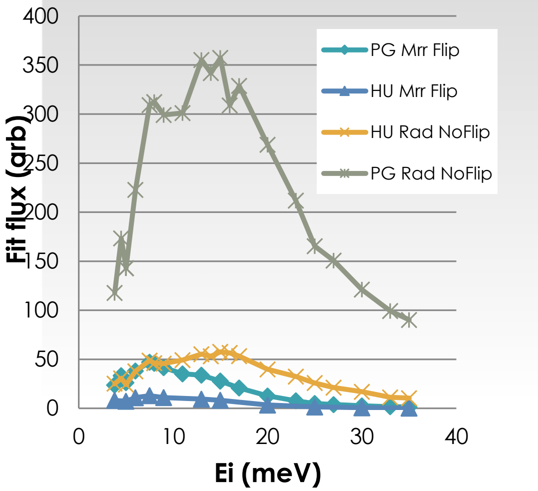
Q3 Reality Check

Low Transmission, Spectra



- Weak signal from low moment samples (for magnetism)
  - Try to minimize background to enhance S:N
  - Recover statistics via flux-resolution trade-off

Imperfect Polarization

- Polarization P

$$P = \frac{N^{\uparrow} - N^{\downarrow}}{N^{\uparrow} + N^{\downarrow}}$$

$$-1 < P < 1$$

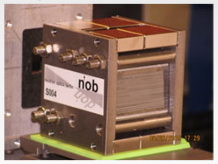
- Flipping ratio F

$$F = \frac{N^{\uparrow}}{N^{\downarrow}}$$

- Due to imperfect
  - Filters
  - Guide fields
    - Modeling precession can help…
  - Flippers
  - Even *samples* can introduce depolarization effects

Neutron beam trajectory & Sample conditions

- Not wide angle
- OK magnetized sample



- OK wide angle
- Not OK magnetized sample

Thanks to NCNR for providing wide angle 'banana' 3He cell for ORNL

Many measurements

- N x measurements
- Errors pile up
  - And with low transmission statistics can be bad to begin with…
- Recommend modeling to the measurement instead of working back to the model

For many polarized neutron applications, you benefit from completing an unpolarized neutron scattering experiment in advance

<u>Complementarity</u> between polarized neutron scattering -&- X-ray magnetic circular dichroism

- In addition to the other pro's & con's comparing neutrons & x-rays…

Polarized neutrons	X-ray dichroism
polarized diffraction goes beyond element specificity to lattice-site-specific moment measurements	Element specificity near absorption edges, & small sample sensitivity due to resonant enhancement
Not element specific in general when it comes to magnetism	Access to magnetism when x-rays excite an electron that contributes to magnetism (transition metals –d electrons →L x-ray edges)
Relatively direct measurement of magnetism	Calculation gymnastics
Indirectly could leverage neutron isotope specificity, possibly with coherent interference effects	Potentially separate magnetism into orbital and spin moments

See lecture NEXT week:
-Monday Aug 14 11:00:
<u>X-Ray dichroism</u>
(Jian Liu)

See Poster after this lecture showing example research which leveraged
BOTH polarized neutrons AND x-ray dicrhoism!

How to prepare for a polarized neutron experiment

- Reach out to polarized instrument staff
  - For help preparing your ‘polarization application statement’
  - For identifying configuration
  - For differences compared to unpolarized experiments (longer, increased # of measurements, etc.)
  - For preparing your proposal
  - For sample preparation (may need a smaller sample, well centered, etc.)

Your invitation to take the red pill

Explore further during NXS

Mini-poster session

Demonstration experiment **N2** leverages the **Longitudinal 1** configuration at HYSPEC

Demonstration experiment **N20** leverages the **Longitudinal 2** configuration at the Magnetism Reflectometer

For future reading

- Several dissertations
  - See instrument-specific publication lists
- Various online slide decks and tutorials
  - Kathryn Krycka, ‘Neutron Polarization’ slides & video at https://neutrons.ornl.gov/nxs/2021/lectures
  - Ross Stewart, https://www.oxfordneutronschool.org/2011/lectures/osns_stewart_polarised_2011.pdf
  - Werner Schweika, https://juser.fz-juelich.de/record/20415/files/C6_Schweika.pdf
- Books / chapters
  - Tapan Chatterji (ed.), *Neutron Scattering from Magnetic Materials* (2006) / several chapters
  - Stephen W Lovesey, *Theory of Neutorn Scatteirng from Condensed Matter V2* (1984) / ch 10
  - G. Shirane, SM Shapiro, JM Tranquada, *Neutron Scattering with a Triple Axis Spectrometer* (2002) / ch 8

Active development and community

- Semi annual meetings / proceedings of PNCMI (polarized neutrons for condensed matter investigations)
  - Proceedings from 2016: https://iopscience.iop.org/issue/1742-6596/862/1
  - Proceedings from 2018: https://iopscience.iop.org/issue/1742-6596/1316/1
  - Proceedings from 2022: https://iopscience.iop.org/issue/1742-6596/2481/1
- Aspirations & new directions at ORNL & NCNR
  - Just ask!
- Actively building user community via training workshops

Conclusion

Questions?

Now go out to the poster session and discover how your specific research might benefit!