The Development of Polarized $^3$He Neutron Spin Filters at the Oak Ridge National Laboratory

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Outline:

• Neutron Scattering in general
• Polarized Neutron Scattering
• Polarized $^3$He in general
• *ex situ* polarized $^3$He system
• *in situ* polarized $^3$He system
• Polarized $^3$He filling station
Neutron Properties

- Neutral
- Penetrating, probe nuclei
- Non-destructive

- Spin 1/2
- Magnetic moment
- Can be polarized

- Energy similar to excitations in solids
  - Molecular vibrations
  - Atomic motion
  - Lattice modes

- Wavelengths similar to interatomic spacings
  - Crystal structures
  - Huge probe range from $10^{-13}$ to $10^{-4}$ cm
The 1994 Nobel Prize in Physics – Shull & Brockhouse

When the neutrons collide with atoms in the sample material, they change direction (are scattered) – elastic scattering.

Detectors record the directions of the neutrons and a diffraction pattern is obtained. The pattern shows the positions of the atoms relative to one another.

Crystal that sorts and forwards neutrons of a certain wavelength (energy) – monochromatized neutrons

When the neutrons penetrate the sample they start or cancel oscillations in the atoms. If the neutrons create phonons or magnons they themselves lose the energy these absorb – inelastic scattering

Neutron Sources – how do we get neutrons

High Flux Isotope Reactor (HFIR)

Spallation Neutron Source (SNS)
A Typical Neutron Scattering Experiment

- Elastic scattering
- Inelastic scattering

\[
\left( \frac{d^2 \sigma}{d\Omega dE_1} \right)_{\lambda \rightarrow \lambda_1} = \left( \frac{m}{2\pi \hbar^2} \right)^2 \frac{k_1}{k_0} \left| \langle k_1 | V | k_0 \rangle \right|^2 \delta (\hbar \omega + E_0 - E_1)
\]

- Lots of neutrons – neutron source
- Determine incident wavevector \( k_0 \)
- Sample
- Determine scattered wavevector \( k_1 \)
- Neutron detector

\[
k = \frac{2\pi}{\lambda} = \frac{2\pi}{h/mv}
\]

\[
E = \frac{1}{2} mv^2
\]
Polarized Neutron Scattering

• Determination of magnetic structures and spin densities
• Identification of magnetic fluctuations and excitations
• Separation of coherent scattering from incoherent scattering
• Separation of magnetic scattering from nuclear scattering
• Improve the energy resolution of spin echo spectrometer
A Typical Polarized Neutron Scattering Experiment

Need to polarize neutrons

- Polarizer
- Analyzer
- Guide field
- Flipper

\[
\frac{d^2 \sigma}{d\Omega dE_1} \bigg|_{\lambda \rightarrow \lambda_1} = \left( \frac{m}{2\pi\hbar^2} \right)^2 \frac{k_1}{k_0} |\langle k_1 S_1 | V | k_0 S_0 \rangle|^2 \delta (\hbar \omega + E_0 - E_1)
\]

\[
k = \frac{2\pi}{\lambda} = \frac{2\pi}{h/mv}
\]

\[
E = \frac{1}{2} mv^2
\]
How to polarize neutrons

- Polarizing monochromators
  - Usually Heusler alloy (Cu2MnAl)
  - Using preferential **Bragg diffraction**

- Supermirrors
  - Very efficient polarizers
  - Using **total reflection**
  - Disadvantage is that small angular beam divergence required

- Polarizing filters
  - Usually polarized $^3$He
  - Using **preferential absorption cross section**
  - Good for polarizing large, divergent neutron beams
  - broadband

$^3$He is like a honey badger, it doesn’t care, it just polarizes
Production of Polarized $^3$He

- Spin-exchange Optical Pumping (SEOP)
- Use high power diode lasers to polarize

- GE180 glass
- $^3$He
- $N_2$
- Rb & K
**Polarized $^3$He as neutron spin filter**

$P_{3\text{He}}$: $^3$He polarization 70%-75%

$T_e$: Glass transmission

Cell thickness:

$$A = n\sigma_0 l \propto \text{pressure} \times \text{length}$$

where:

- $n$: number density
- $\sigma_0$: absorption cross section for $\lambda = 1\text{Å}$
- $l$: cell length

$$P_{\text{neutron}}(\lambda) = \tanh(A\lambda P_{3\text{He}})$$

$$T_{\text{neutron}}(\lambda) = T_e \exp(-A\lambda) \cosh(A\lambda P_{3\text{He}})$$

- **Compromise between polarization and transmission**
- **Improving $^3$He polarization can increase both neutron polarization and transmission**
Lab-based Filling Station
**ex situ Optical Pumping Station**

- 2 pumping stations at SNS lab C-241
- 2 pumping stations at HFIR (being built)
- 1-2 days of pumping time

Monitor $^3\text{He}$ polarization
- FID NMR
- AFP NMR
- EPR
ex situ Optical Pumping Station – HB2A, as polarizer

ex situ Optical Pumping Station –HB1, as analyzer

\[ P_{He} \approx 70\% \]

<table>
<thead>
<tr>
<th>Energy (meV)</th>
<th>Heusler flipping ratio</th>
<th>Polarized 3He FR</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.5</td>
<td>20.3</td>
<td>67.4</td>
</tr>
<tr>
<td>30.5</td>
<td>6.65</td>
<td>16.7</td>
</tr>
<tr>
<td>50</td>
<td>2.52</td>
<td>8.93</td>
</tr>
</tbody>
</table>
**ex situ Optical Pumping Station – disadvantages**

- The $^3$He polarization relaxes once removed from the laser beam

\[ P(t) = P_i \exp \left(-\frac{t}{T_1}\right) \]

- May need to repolarize 3He for a lengthy experiment
- Data analysis may be complicated
in situ Polarized $^3$He Pumping station

**Why in situ?**

- Constant Polarization
- Lower field gradient requirement
- User friendly

**Difficulties?**

- Tight space confinement
- Laser safety
- Over heating issue
- Ambient field invasion
in situ $^3$He Analyzer for Magnetism Reflectometer at SNS

39 in L x 15 in W x 29 in H

- Two 250W laser pumping
- System enclosed with laser interlock
- Remote operating and controlling
- Built-in $^3$He spin flipper
- High $^3$He polarization achieved

The measurements

**Unpolarized neutron measurement**
- $^3\text{He}$ polarization
- $^3\text{He}$ cell thickness
- Neutron transmission through $^3\text{He}$

**Polarized neutron measurement**
- $^3\text{He}$ as analyzer
- Measured supermirror bender polarization

**Reference sample measurement**
- Full polarization analysis on a reference sample
- Four cross-sections measured
Unpolarized neutron measurements

\[ T_n(\lambda) = T_e \exp(-n\sigma_0 l\lambda) \cosh(n\sigma_0 l\lambda P_{He}) \]

\[ T_0(\lambda) = T_e \exp(-n\sigma_0 l\lambda) \]

\[ T_e : \text{empty transmission through glass wall} \]
\[ n : \text{number density of } ^3\text{He gas} \]
\[ \sigma_0 : \text{absorption cross section per wavelength} \]
\[ l : \text{cell length} \]
\[ \lambda : \text{neutron wavelength} \]
Unpolarized neutron measurements -- results

\[ A = n\sigma_0 l = 1.270 \pm 0.013 \]
\[ T_e \sim 82\% \]
\[ P_{3He} = 76\% \pm 1\% \]

\[ T_0(\lambda) = T_e \exp(-A\lambda) \]

\[ P_{neutron}(\lambda) = \tanh(A\lambda P_{3He}) \]

\[ T_{neutron}(\lambda) = T_e \exp(-A\lambda) \cosh(A\lambda P_{3He}) \]
Polarized neutron measurements – Flipping Ratios

Supermirror bender  Spin Flipper  $^3$He Cell  Detector  $T_+(\lambda)$

Supermirror bender  Spin Flipper  $^3$He Cell  Detector  $T_-(\lambda)$

$$F(\lambda) = \frac{T_+(\lambda)}{T_-(\lambda)}$$

$$P_{SM}P_{3He}P_{ST}P_{SF} = \frac{F - 1}{F + 1}$$

SM: Supermirror
ST: Spin transport
SF: Spin flipper
Polarized neutron measurements -- results

\[ P_{SM} P_{3He} = P_{FR} = \frac{F-1}{F+1} \], assume perfect spin transport and spin flip efficiency.
Sample measurements

Supermirror  Spin Flipper  Sample  $^3$He Cell  Detector

$\sigma_{++}$  $\sigma_{+-}$  $\sigma_{-+}$  $\sigma_{--}$

$^{57}$Fe (6.7 nm)/Cr(-1.2 nm)

anti-ferromagnetic inter-layer exchange coupling and an in-plane magnetic domain structure

Sample measurements -- results

The wavelength dependence of the specularly reflected, off-specularly scattered and transmitted intensities for the four spin-states.

In the coordinates of the PSD and being integrated over the TOF.
Summary for BL4A’s in-situ setup

Performance
- 76% $^3$He polarization achieved
- 98% neutron polarization for wavelength 2.5 Å and above (96% at 2 Å)
- 25% average transmission (40% at 2 Å and 18% at 4.5 Å)

Uniqueness
- Motor controlled, can be moved in and out of the beam
- All $^3$He setup computer controlled
- Class 1 laser certification, user friendly

Stability
- 3 weeks of running without interruption

It’s so easy that [face] can operate it!
Compact in situ Analyzer for HB1 at HFIR

24 in L x 22 in W x 18 in H

- One 70W fiber-coupled laser pumping
- System enclosed with laser interlock
- Built-in $^3$He spin flipper
- $^3$He polarization (54%) achieved
Future Development

• Super compact *in situ* system
  - 1 ft long in the direction of neutron beams
  - can fit into most neutron beam lines at SNS and HFIR
The Hybrid Spetrometer - HYSPEC

- High flux by trick of tall guide & vertical focusing with crystals (~6x)
- Located in external building – space around beamline, very flexible, low background
- Curved guide & vertical axis T0 chopper – background reduction
- \(3.6 < E_i < 90 \text{ meV}, 0.9 \AA < \lambda < 4.7 \AA\)
- \(10^5 - 10^7 \text{ n/cm}^2/\text{s} \quad \Delta E / E_i - 2\% \rightarrow 10\%\)

Huge neutron wavelength/energy span!
Choosing the Right 3He Pressure – Max Figure of Merit

\[ FOM(\lambda) = P_{\text{neutron}}(\lambda)^2 T_{\text{neutron}}(\lambda) \]

Assume 70% \( P_{3\text{He}} \)
Example: Max. $^3$He polar. = 72%, optimized at 90 meV. $T_1$ = 100 hours, exchange all the gas every 5 hours. (Plot: for 50 meV neutrons)
Polarized 3He Filling Station - Blueprint
Polarized $^3$He Filling Station – Prototype Assembled

first of its kind in the world

- A 1045 turn rectangular solenoid was built to create the magnetic field necessary to keep the polarization of the 3He cell aligned

- This is designed to slide over the laser system, oven, and gas syringe cylinder on the upper left-hand side of the cart

- The gradient of the field was measured to be $4.42 \times 10^{-4} \text{ cm}^{-1}$ which leads to reasonable polarization relaxation times for this application
The gas syringe was manufactured out of non-magnetic material (Brass, Aluminum, and Titanium)

This syringe has four air-controlled valves that connect it to the vacuum pump, gas supply system, pump-up cell, and analyzer cell

A linear actuating motor controls a piston within the syringe, moving the gas.
Pump Up Station and Cell

- The pump-up cell is made of GE180 glass.
- The end flange is made of brass.
- In the middle is pyrex capillary tubing connecting the cell and the end flange.
- Electrical heating
Wide-angle Cell & Coils

Wide-angle analyzer cell

- Analyzer sits in the center of the coils
- Analyzer converges to the center of the sample stage
- Wall thickness is about 4mm, pressure tested to 35 psi (2.4 bar)
- The cell was filled with 50 Torr of N$_2$ and 300 Torr of $^3$He. T1=70.1 hr +/- 0.6 hr
**Tests and Results**

- **Saturated Polarization Measurement**

- **Left,** the saturated 3He polarization slowly decayed with time.

- **Right,** after we drove Rb to the capillary tubing to separate the optical pumping cell from the valve, the 3He polarization stayed constant for more than a week, indicating a small gas leakage from the valve.
Tests and Results

Cell Lifetime Measurement

- Left, the OPC decay measurement, the fit gives $8.7 \pm 0.4$ hours of lifetime
- Right, the OPC is blocked from capillary using Rb, the fit gives $10.5 \pm 1.6$ hours of lifetime
Tests and Results

- Unpolarized gas transfer between the optical pumping cell and the recycle tank shows a negligible loss.

- Polarized $^3$He transfer between the optical pumping cell and the syringe shows ~15% polarization loss per transfer.

- EPR measurements show ~20% $^3$He polarization achieved.

Showed proof of principle
Next version is underway
Future Improvements

- A Higher power fiber-coupled laser (200W)
- A completely redesigned cart, more compact
• A titanium bellow system to replace the current syringe
Summary

- **3**He cells of long lifetime and high polarization
  - 70%+ 3He polarization
  - 100+ hours of decay time

- **drop-in** 3He cells as polarizer or analyzer
  - 95%+ neutron polarization (wavelength dependent)
  - 40% neutron transmission (wavelength dependent)
  - Fast turn-around time

- **in situ** 3He pumping systems
  - Maintains steady 3He polarization
  - Permanent installation with minimum maintenance required

- **online** 3He filling system for HYSPEC
  - The world’s first polarized gas transfer system using SEOP
Thanks
Where do we get the neutrons to do the scattering?
Spallation Neutron Source at Oak Ridge National Laboratory

The world’s most intense pulsed, accelerator-based neutron source

**Backscattering Spectrometer (BASIS) • BL-2**
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