

## Development of Polarized $^3\text{He}$ Neutron Spin Filters at Oak Ridge National Laboratory

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**Abstract:** Nuclear spin-polarized  $^3\text{He}$  is widely used in many scientific areas. One of the applications is used as a neutron spin filter to polarize neutrons. Among all the neutron polarizing techniques, nuclear-spin-polarized  $^3\text{He}$  neutron spin filters have shown great flexibility and versatility because of its highly spin-dependent neutron absorption cross section, large neutron acceptance angle and working over a broad neutron wavelength band. At the Oak Ridge National Laboratory,  $^3\text{He}$  is routinely polarized via spin-exchange optical pumping (SEOP). Over the last several years, various SEOP-based systems have been developed to suit the needs of different neutron instruments at the Spallation Neutron Source (SNS) and the High Flux Isotope Reactor (HFIR). Particularly, much effort has been put in developing *in situ* systems to address the problem caused by  $^3\text{He}$  polarization decay in the widely used “drop-in cell” setup. Because most instruments at SNS and HFIR have very limited space, the *in situ* system development has been focused on having a compact form factor design tailored to each individual beamline while still achieving high  $^3\text{He}$  polarization. We report the development of polarized  $^3\text{He}$  neutron spin filters at ORNL.

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## 1. Introduction

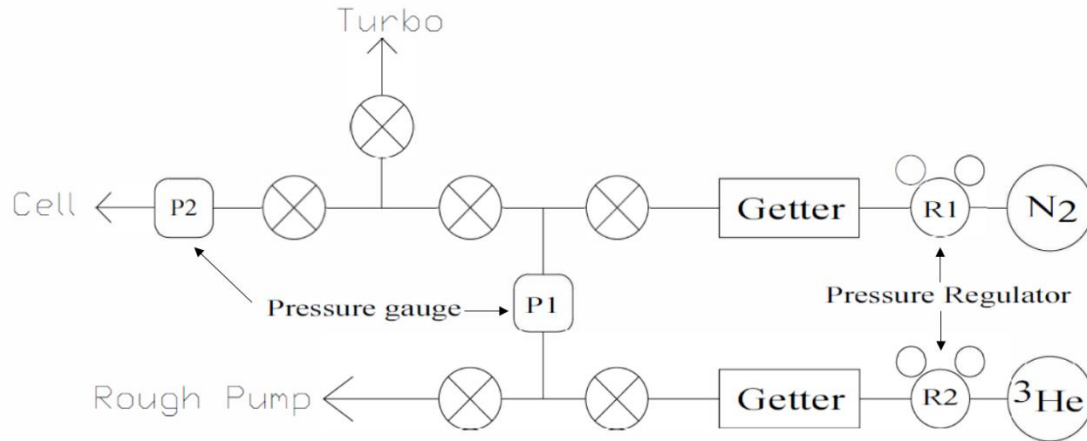
Polarized neutron scattering is a powerful and indispensable tool in today's condensed matter physics, materials science and other fields. Almost every major neutron facility in the world has established a polarized neutron program to keep up with the increasing demand for polarized neutrons. For a polarized neutron experiment, it is vital to have a high-performance neutron polarizer and/or analyzer. Three methods have been routinely used to produce polarized neutrons or analyze neutron polarization: Heusler crystals, polarizing supermirrors and nuclear spin polarized  $^3\text{He}$  neutron spin filters. Among these three methods, polarized  $^3\text{He}$  has several unique advantages. First, polarized  $^3\text{He}$  can accept neutron beams with large divergence while both Heusler crystals and supermirrors do not work well with divergent beams; second, polarized  $^3\text{He}$  does not change the neutron beam direction and adds no additional divergence to the beam; third, polarized  $^3\text{He}$  can work well over a large neutron energy band from epithermal to thermal neutrons, but the supermirror usually has a cut-off wavelength of around 2 Å; lastly, polarized  $^3\text{He}$  can also serve as a neutron spin flipper and thus reduce the additional components needed for polarized neutron experiments.

Oak Ridge National Laboratory (ORNL) houses two large neutron facilities: The High Flux Isotope Reactor (HFIR) and the Spallation Neutron Source (SNS). Polarized neutron scattering has been routinely performed at ORNL, and a polarized  $^3\text{He}$  program has been running for years to support polarized neutron experiments [1]. The ORNL  $^3\text{He}$  program is focused on using spin-exchange optical pumping (SEOP) to produce polarized  $^3\text{He}$  [2]. Traditionally,  $^3\text{He}$  is first polarized on an *ex situ* pumping station and then transferred to a neutron instrument for use [3]. Two pumping stations have been built at ORNL to provide easy access to polarized  $^3\text{He}$ . Although this method is relatively straightforward to implement, it faces a problem, i.e.,  $^3\text{He}$  polarization starts to decay exponentially once it is removed from the pumping station. The decay of  $^3\text{He}$  polarization brings two problems. First, it degrades the performance of a  $^3\text{He}$  neutron spin filter as a neutron polarizer or analyzer. Once the polarization decays to below a certain level, the  $^3\text{He}$  needs to be brought back to get repolarized. Second, the changing  $^3\text{He}$  polarization leads to a changing neutron polarization or analyzing power which brings complication in data analysis. To overcome these problems, great effort has been made at ORNL to develop *in situ* polarized  $^3\text{He}$  systems. An *in situ* system enables continuous pumping of  $^3\text{He}$  on a neutron instrument and keeps  $^3\text{He}$  polarization stable once it is saturated. Several *in situ* polarized  $^3\text{He}$  systems tailored to different neutron instruments have been developed at ORNL to meet the demand for polarized neutrons [4-7]. Both the *ex situ* and *in situ*  $^3\text{He}$  systems at ORNL routinely produce polarized  $^3\text{He}$  with over 70% polarization and provide high-performance neutron polarizers/analyzers for neutron instruments at both HFIR and SNS.

## 2. Cell Preparation

High quality  $^3\text{He}$  cells are vital for producing highly polarized  $^3\text{He}$  gas. Most of the  $^3\text{He}$  cells at ORNL are made of boron-free aluminosilicate GE180 glass which has low  $^3\text{He}$  permeation and neutron absorption. The glassblowers at ORNL usually blow a cell into a cylindrical shape from 25mm GE180 tubing. A typical  $^3\text{He}$  cell for SEOP contains  $^3\text{He}$ , nitrogen and alkali metals (rubidium and/or potassium). A high-vacuum filling station has been built to fill a cell with all

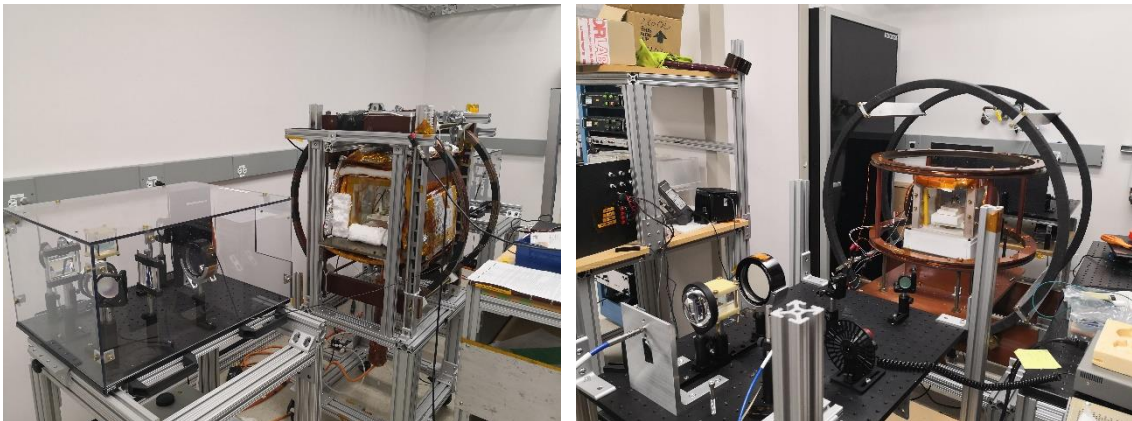
required ingredients (**Fig. 1**). To prepare a  $^3\text{He}$  cell, an empty cell is cleaned thoroughly before it is connected to the filling station. The cell is baked at 400 °C for several days to remove all impurities through a turbo pump. Then alkali metals are driven into the cell using a torch followed by filling of  $^3\text{He}$  and  $\text{N}_2$ . In the end, the cell is sealed via a torch and detached from the filling station. More detailed description can be found in Ref. 2 and 8.



**Figure 1.** The schematic of the filling station

### 3. Ex situ pumping stations

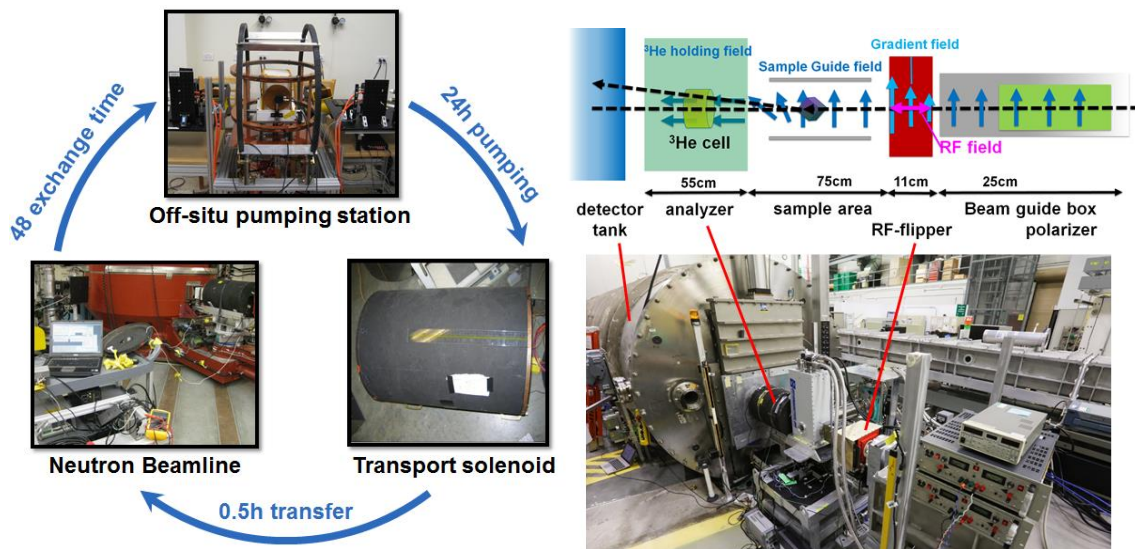
We have set up two optical pumping stations in our lab based on SEOP (**Fig. 2**). One station is equipped with two 100 W 795 nm diode laser bars, each spectrally narrowed by a chirped grating to better match the Rb D1 absorption spectrum. The other station utilizes a 200 W narrowband 795 nm fiber coupled laser to do optical pumping. Each station has a set of Helmholtz coils installed to provide a uniform main field required for SEOP. To monitor the  $^3\text{He}$  polarization during optical pumping, Free Induction Decay (FID) Nuclear Magnetic Resonance (NMR) is employed to give a relative measure of the polarization. Adiabatic Fast Passage (AFP) NMR is also used to flip the  $^3\text{He}$  polarization when necessary. Electronic Paramagnetic Resonance (EPR) is set up on both pumping stations to measure the absolute value of  $^3\text{He}$  polarization. Both stations have provided reliable production of highly polarized  $^3\text{He}$  for use on neutron instruments.



**Figure 2.** Left, the pumping station using diode laser bars and chirped gratings. Right, the station using fiber-coupled laser.

To support a polarized neutron experiment, a  $^3\text{He}$  cell is first polarized in one of the pumping stations. Once the polarization is saturated, the cell is then placed in a transport solenoid and transferred to the place of use. This is referred as the “drop-in cell” method. Usually, after a day or two of use, the  $^3\text{He}$  cell needs to be brought back to the pumping station to get replenished due to the polarization decay. **Fig. 3 left** shows the whole process. To save the turnaround time, two pumping stations are used at the same time, each polarizing a cell of similar size and parameters, to ensure there is always a  $^3\text{He}$  cell ready for use. **Fig. 3 right** shows a recent application of using polarized  $^3\text{He}$  as a neutron polarization analyzer on the General-Purpose Small-Angle Scattering Diffractometer (GP-SANS) at HFIR [9]. The  $^3\text{He}$  cell was placed in a 55 cm long transport solenoid with a double layer mu-metal shielding to shield the  $^3\text{He}$  cell from the stray field of the superconducting magnet at the sample position.

The “drop-in cell” method requires using a  $^3\text{He}$  cell with a long relaxation time ( $T_1$ ).  $T_1$  great than 100 hours on neutron instruments is usually preferred for a  $^3\text{He}$  cell.  $T_1$  is determined by three major relaxation mechanisms: the wall effect, the dipole-dipole interaction and the field gradient [2, 10]. However, once a  $^3\text{He}$  cell is filled and sealed, the field gradient is the only thing that can be manually adjusted to improve  $T_1$ . Thus, it is important to maintain good field uniformity in a transport solenoid in order to optimize  $T_1$ . We have carefully made several transport solenoids. Each of these solenoids is shielded with mu-metal and has an offline transverse field gradient on the order of  $10^{-4}$  /cm.



**Figure 3.** Left, the cycle of using the “drop-in cell” method. Right, polarized  $^3\text{He}$  as the analyzer on a recent SANS experiment

#### 4. In situ $^3\text{He}$ systems

The pumping station and “drop-in cell” method provides an easy and fast way to implementing polarized  $^3\text{He}$  on neutron instruments. However, for a neutron experiment using polarized  $^3\text{He}$  and running for several days, it is almost inevitable to change the  $^3\text{He}$  cell multiple times because of the  $^3\text{He}$  polarization decay. The decay not only lowers the performance of the  $^3\text{He}$  cell, but also complicates the following data analysis. To address this problem, we have developed various *in situ* polarized  $^3\text{He}$  systems at ORNL which enable continuous pumping of

$^3\text{He}$  on a neutron beamline and keep  $^3\text{He}$  polarization steady throughout a polarized neutron experiment once it is saturated.

An *in situ*  $^3\text{He}$  system needs to incorporate all the elements in a pumping station to an enclosed space, and strict safety measures must be implemented to ensure the system's safe running on a neutron instrument. Each ORNL system is enclosed with laser safety panels to make it a Class I laser environment while operating. These panels are also interlocked to the laser power supply to add an extra layer of protection. Fiber-coupled lasers are used in our systems. A photodiode is placed inside each *in situ* box to monitor the laser delivery and serve as another laser interlock in case the laser fiber is broken. Our *in situ* systems are also equipped with NMR and EPR to monitor and flip  $^3\text{He}$  polarization.



**Figure 4.** Left, the large *in situ*  $^3\text{He}$  system installed on MAGREF at SNS as the analyzer; Right, the medium system used as the analyzer on CG-1D at HFIR. Bottom, the compact system set up on HB-3A as the polarizer

At ORNL, we have developed *in situ*  $^3\text{He}$  systems of different sizes (large, medium and compact) to meet the needs of different instruments. On the Magnetism Reflectometer (MAGREF) at SNS, a large *in situ* system has been permanently installed to serve as one of the two neutron polarization analyzers (**Fig. 4 left**), which has a dimension of 69 cm in length, 59 cm in width and 54 cm in height [6]. The medium sized system (50 cm L x 55 cm W x 45 cm H) is portable and has been used on several instruments. **Fig. 4 right** shows using the medium system as the analyzer for polarized imaging experiments on the CG-1D Cold Neutron Imaging Facility beam at HFIR [7,11]. Both the large and medium systems employ double side pumping and use similar optical layout. Recently, a new compact system has been commissioned as the neutron polarizer on the HB-3A Four-circle diffractometer at HFIR (**Fig. 4 bottom**). This compact system

is only 30 cm long along the neutron beam direction and uses one-side pumping to save space. To our knowledge, it is the most compact *in situ* system in the world. All the *in situ* systems at ORNL have shown great stability and reliability in lengthy experiments.

## 5. Conclusions

Over the last decade, ORNL has established a robust polarized  $^3\text{He}$  program to support polarized neutron experiments at both HFIR and SNS. Both the *ex situ* and *in situ* systems can routinely produce highly polarized  $^3\text{He}$  gas through SEOP for use as neutron polarizers or analyzers. As more instruments at ORNL are showing interest in using polarized  $^3\text{He}$ , future development will be focused on further improving  $^3\text{He}$  polarization, developing more user-friendly *in situ* systems and establishing a wide-angle  $^3\text{He}$  cell capability.

## Acknowledgements

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