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TITLE: ADVANCED RESEARCH CAPABILITIES FOR NEUTRON SCIENCE AND TECHNOLOGY: NEUTRON POLARIZERS FOR NEUTRON SCATTERING

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Advanced Research Capabilities for Neutron Science and Technology: Neutron Polarizers for Neutron Scattering

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Abstract

We describe work on the development of polarized gaseous ^{3}He cells, which are intended for use as neutron polarizers. Laser diode arrays polarise Rb vapor in a sample cell and the ^{3}He is polarised via collisions. We describe development and tests of such a system at LANSCE.

Background and Research Objectives

Broad-band neutron polarizers are an essential technology for high powered spallation neutron sources like LANSCE, especially in the thermal energy range. Spin-polarized ^{3}He is a very attractive technology. Although it has been used for many years in nuclear and particle physics, it has not yet reached maturity as a tool for neutron-scattering studies of condensed-matter system, and the intent of this LDRD project has been to transfer this technology and apply it to neutron scattering at LANSCE. Key targets are to get ^{3}He polarisations in the 65% range within the environment of a neutron spectrometer, and obstacles to this task include available laser power at the right wavelength, depolarising processes with the cell walls, mechanical integrity of the cells themselves, the magnetic-field environment and so on. 65% is state of the art, and has been claimed by the Institut Laue Langevin Grenoble using a meta-stable ^{3}He polarization method quite different from ours. But to date there have been no successful condensed-matter experiments using ^{3}He polarizing filters, anywhere in the world.

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Importance to LANL's Science and Technology Base and National R&D Needs

The Neutron Laboratory at LANSCE is one of the key scientific facilities in the Laboratory portfolio, and it is now clear that any new national neutron sources will be accelerator-driven neutron sources rather than reactors. Key components in the success of these sources, including LANSCE, will be the availability of intense broad-band polarized neutron beams in the thermal energy range, and the ability to analyze scattered neutrons for their polarization over very large detector areas. This will impact work on magnetic materials in particular, as well as correlated-electron metals with enhanced magnetic susceptibilities. Further in the future there is the possibility of separating coherent from incoherent scattering, using polarized hydrogen to change its contrast in biological molecules and polymers, and the use of spin-precession methods for measurement of neutron energies.

Scientific Approach and Accomplishments

1. Introduction

Applications of polarized ^{3}He technology range from high energy physics to biomedical science as can be learned from papers of the HELION97 workshop at Japan in 1997 [1]. The method for optical polarization of ^{3}He gas was discovered in the 1960s [2], but applications were initially limited, because of a lack of suitable light sources. Later powerful solid state lasers made it practical to polarize large quantities of ^{3}He. Because of ^{3}He has a very large spin-dependent neutron cross section polarized ^{3}He gas can be used to absorb one of the two neutron spin states, allowing the other spin state to be transmitted through with moderate attenuation. In this report we discuss the ^{3}He neutron spin filter. We note that polarized protons can be also used to filter the neutron spin [3].

The goals of this project were to develop the ^{3}He technology which then can be used to build the neutron beam polarizer for neutron scattering experiments. Such a technique is of great importance to many areas of basic and applied research. In particular, they promise to open up broad-band polarized thermal-neutron applications at spallation sources like LANSCE. The work consisted of development of ^{3}He sample cells and loading techniques, construction of the apparatus to polarize and test ^{3}He, development of optics and laser technology, and a test of the ^{3}He spin filter in a neutron beam.

The requirements for the neutron spin filter are:

1) It should be a broad band, providing highly polarized neutrons from the longest wavelength (about 20 Å) to the “hot” neutron energies, 0.4-1 Å.

2) The polarizer should provide the maximum flux of neutrons (per unit wavelength) at the detector and therefore transport the neutron beam from the source to a sample with the maximum efficiency.

3) In some experiments it is important that the polarizer produces negligible or easily shielded gamma backgrounds and any residual background should be constant.

4) The polarizer may provide an additional reversal of neutron spin without changing any magnetic fields which helps to control systematical errors of an experiment.

The conventional method to polarize neutron beams is to use polarizing supermirrors [4,5]. The polarizing supermirror is a well developed technology which in many neutron scattering experiments, at thermal and cold energies, gives better results than the ^{3}He spin filters. But the supermirror polarizer works only with neutron wavelengths of 2Å and greater, and is limited to narrow-divergence applications. In principle, the ^{3}He spin filter has a broader band, can handle arbitrarily large angular divergences, and can be used to polarize neutrons from the long wavelengths down to approximately 0.05 Å.

2. Optically-polarized ^{3}He; a neutron spin filter for neutron scattering

In recent years, neutron beam polarization by transmission through a sample of laser-polarized gaseous ^{3}He has rapidly advanced toward practicality. Two methods have been developed in parallel to polarize ^{3}He nuclei: exchange of polarization from optically-pumped metastable ^{3}He and exchange from optically-pumped Rb. The metastability exchange technique is currently used at Grenoble and Mainz [6]. Since the Rb spin-exchange technique is straightforward and capable of producing thick (high-pressure) polarized ^{3}He targets, it was our method of choice. During the project the Rb spin-exchange technique has benefited from Industry's production of high powered laser diode arrays (LDA) [7].

3. Neutron transmission through polarized ^{3}He gas

Polarized ^{3}He can be used as a neutron spin filter [8] since ^{3}He has a large neutron absorption cross section for the capture reaction $^3\text{He} + \text{n} \rightarrow \text{p} + \text{t}$:

$\sigma_a = \frac{1}{\sqrt{E}}\sqrt{E_0}\sigma_0$, where $\sigma_0 = 5327$ barn at neutron energy of E_0=0.025 eV. The cross section is highly spin dependent due to a broad, unbound resonance in ^{4}He of $J^\pi = 0^+$. In order to conserve angular momentum for this *s*-wave resonance, only the component of neutron spin anti-parallel to the ^{3}He spin is resonantly absorbed. When a unpolarized neutron beam passes through polarized ^3He, beam polarization P_n and transmission T_n are given by

(1)
$$T_\pm = \exp(-n_3\sigma_a l(1 \mp P_3))$$
$$P_n = \frac{T_+ - T_-}{T_+ + T_-} = \tanh(n_3\sigma_a l P_3)$$
$$T_n = \frac{T_+ + T_-}{2} = \exp(-n_3\sigma_a l)\cosh(n_3\sigma_a l P_3) = T_n^0 \cosh(n_3\sigma_a l P_3),$$

where T_+ (T_-) is the transmission when the neutron spin is parallel (antiparallel) with the ^3He spin, n_3 is the number density of the ^3He, l is the length of the ^3He gas and, P_3 is the polarization of the ^3He and T_n^0 is transmission through the ^3He cell when ^3He polarization is zero . With simple algebra we get from eq. (1) for beam polarization

(2)
$$P_n^2 = 1 - (\frac{T_n^0}{T_n})^2.$$

This relationship and eq. (1) show that ^3He polarization will enhance the transmission. According to equation (2) in order to determine the neutron beam polarization we need only to measure the transmission enhancement (T_n / T_n^0). We do not need to know the thickness of the ^3He or its cross section. Because the neutron - ^3He cross section depends upon the neutron energy, the transmission and beam polarization have energy dependences which are not indicated in eqs (1) and (2). A ^3He spin filter can be built to optimize a certain wavelength range as illustrated in figures 1 and 2. Figure 1 shows the calculated figure-of-merit, $P_n^2 T_n$, as a function of neutron wavelength for ^3He polarizations of 40%, 50%, and 60% and a ^3He thickness of 8 atm·cm. The figure-of-merit (FOM) defines how long the experiment must run to achieve a certain statistical accuracy. Figure 2 shows P_n, T_n, and $P_n^2 T_n$ as a function of neutron energy for the filter cell with 50% ^3He polarization and thicknesses of 6 atm-cm and 8 atm-cm. The goal of the design of the spin filter cell and its construction is to optimize figure-of-merit and this includes also practical considerations as well. The shape of the FOM curve depends on the ^3He target thickness, but is almost independent of ^3He polarization. Therefore, the optimal thickness does not change appreciably with ^3He polarization.

4. Spin-exchange optical pumping

Spin-exchange from optically pumped Rb vapor via the hyperfine interaction of the Rb valence electrons with the ^{3}He nucleus was discovered in 1960 by Bouchiat [2]. In this method circulary polarized laser light polarizes the Rb atoms and then through the spin-exchange process the ^{3}He nuclei will become polarized. The polarization of ^{3}He produced by the spin exchange evolves with time as

$$P_3 = P_{\text{Rb}} \frac{\gamma_{\text{SE}}}{\gamma_{\text{SE}} + \Gamma}(1 - e^{(\gamma_{\text{SE}} + \Gamma)t}), \tag{3}$$

where P_{Rb} is the steady state Rb polarization produced by laser pumping, and $\gamma_{\text{SE}} = k_{\text{SE}}[\text{Rb}]$ is the rate of spin exchange from the Rb to the ^{3}He nucleus. Γ is the total ^{3}He spin relaxation rate, in general dominated by interactions with paramagnetic impurities on the wall of the cell and in the gas. Due to the weakness of spin exchange, $\gamma_{SE} = (0.6 - 1.2) \cdot 10^{-1}$ cm^3/s, maximum ^{3}He polarization would require both densities of 10^{14}-10^{15} Rb/cm^3 and ^{3}He relaxation times, $1/\Gamma$, of many tens of hours [9]. These relaxation times can only be achieved by keeping Γmuch smaller than the spin exchange rate. This sets strict requirements for the cell material and preparations of the cells [10]. Studies showed that densities of Rb greater than 10^{14} could be effectively polarized when N_2 is included to suppress radiation trapping, Corning 1720 alumino-silicate and other low-iron and/or low-helium permeability glasses are typically used to suppress the wall relaxation, and sufficient laser power was absorbed by the Rb atoms to balance Rb spin destruction. The large laser power is required to keep the Rb atoms highly polarized.

5. Cell construction

The state of the art of the cell construction has been explained in ref. [10]. The cells must be fabricated from glass material which allows circularly-polarized laser light enter the cell, and the cell strength must be sufficient to tolerate the ^{3}He pressures, which can be as high as several atmospheres. The neutron entry and exit windows of the cells should not absorb too many neutrons from the beam. The only parameters which can be influenced in equation (3) are P_{Rb} , rubidium polarization, which strongly depends upon the laser power, and Γ, the polarization relaxation rate. Γ depends upon the content of paramagnetic impurities in the glass and gas. In order to achieve the high ^{3}He polarization Γ has to kept as small as possible. Therefore the inside wall surfaces of the cells have to be treated carefully to remove paramagnetic impurities [10] and the glass material itself has to have as low iron content as possible [11]. In this project our choice of glass was Corning 1720 alumino-silicate glass which has very low helium permeability. This glass contains 5%

B_2O_3. ^{10}B abundance in natural boron is 20%. The ^{10}B isotope has a very large neutron absorption cross section at thermal energies and therefore should be replaced in glass with ^{11}B. The parts of the cell which interact with the neutron beam, such as the beam windows, should be made from the ^{10}B-free glass. The typical cell geometry is cylindrical with the diameter of about 3.5 cm and the length of 4-10 cm with 2-3 mm thick flat end caps made from ^{11}B-enriched Corning 1720 glass. The end caps were sealed to the cylindrical body with a H_2 flame [12]. With flat ends every neutron has the same interaction path in the ^3He gas. The use of the flat end caps made the cells weak, and in particular the joints between ends and the body of the cell were weak spots. The strength of the cells were studied by pressure tests. With the proper sealing of the ends the rupture pressure was measured to be about 15 atm. This pressure was high enough that the cells could be loaded up to 10 atm. Loading techniques of the cells were developed in the collaboration with different institutes. At the beginning of the project the loading of the cells were done at TRIUMF, NIST, Princeton and the University of New Hampshire. At the end of the project we built an ultra-high vacuum system that allows the loading to be done at Los Alamos. The quality of the cells and the transport properties of polarized ^3He gas was studied using the magnetic resonance imaging (MRI) technique as described in publication 1. Figure 3 shows a profile picture of a polarized cell done with the MRI technique. In the middle of the cell the ^3He polarization has been destroyed and the evolution of the polarization in the cell is shown.

6. Lasers

At present, the most common lasers in use for the Rb optical pumping in high density ^3He gas are high powered arrays of laser diodes (LDA) and Ti:sapphire lasers [7,13]. Due to price considerations and ease of use compared to Ti:sapphire lasers, LDAs dominate. Sufficient laser power is required to polarize the Rb atoms and to balance Rb spin destruction [14]. The bulk Rb spin destruction rate at high Rb densities and ^3He densities below about 5 atm is dominated by Rb-Rb collisions [15] and is given by the total spin destruction rate

$$\Gamma = k_{Rb-^3He}[^3He] + k_{Rb-N_2}[N_2] + k_{Rb-Rb}[Rb], \qquad (4)$$

where k represents the rate constants for the spin destruction, due to collisions of the Rb atoms with the other species and with themselves. For a typical application, $\Gamma \sim 500\ s^{-1}$. Recently these rates have been re-measured at Princeton, and a strong temperature dependence was observed [16,17]. The disadvantage of the LDA is its broad laser linewidth of about 2 nm. This linewidth does not mach well with the D1 absorption linewidth of the Rb atom . The width of this line depends on the ^{3}He pressure and at 6 atm pressure is about 0.5 nm. This means that a main part of the laser light is not absorbed by the Rb atoms. Therefore further development of the LDA is still necessary. The use of LDAs in the ^{3}He neutron spin filter is described in publication 2.

7. Apparatus

Figure 4 shows a schematic of a typical spin-exchange optical pumping apparatus. The cell is in an oven which regulates the Rb density, at temperature of about 175 °C. ^{3}He polarization is held by a 30-gauss homogeneous magnetic field. The field homogeneity has to be better than about 1mG/cm. This gradient minimizes the polarization losses during the NMR measurements or spin reversal. ^{3}He polarization is measured with the adiabatic fast passage (AFP) NMR [18]. AFP can also be used to reverse the polarization direction of the filter. ^{3}He polarization can also be measured with a neutron beam as discussed in the next section.

8. Test of a ^{3}He spin filter in a neutron beam

Use of polarized ^{3}He gas to polarize neutron beams was proposed by Postma [19]. The first experiment with a ^{3}He spin filter was done at LANSCE in 1989 [20]. Similar techniques have also been used at ILL, KEK, NIST, and the University of Michigan. In our experiment the ^{3}He spin filter was used to measure the absolute neutron beam polarization. Using the time-of-flight method, the energy of the neutrons and beam backgrounds can be determined very accurately which then allows accurate transmission measurements. With equations (1) and (2) and results of the transmission measurements the absolute neutron beam polarization can be calculated. Figure 5 shows main components of the experiment performed at MLNSC. Downstream of the bulk shield, a thin ^{3}He ion chamber was used to normalized the beam flux with the accuracy of 10^{-4}. The neutrons were then collimated and transported to the polarized ^{3}He cell located 9 m from the source. In fact two such cells, one polarized and the other unpolarized, were mounted on a common moveable table within the same oven, so that they could be alternatec within the beam. Neutrons were counted with a ^{3}He scintillation detector after traversing the 55-m flight path. More detailed description of the experiment and results can be found from the publication 2. In figure 6, the beam polarization has been plotted as a function of neutron energy from 20 meV to 10 eV. ^{3}He polarization during the measurement shown in figure 6 was 15%. The maximum ^{3}He polarization achieved during the experiments was 55% [21].The measured neutron yield has been corrected for the beam background, detector dead time and length of the flight path. The results indicate that we have determined beam polarization with the accuracy of 0.2-0.3%.

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Figure Captions

Figure 1: Figure-of-merit, $P_n^2 T_n$, for a neutron spin filter with helium polarizations of 40%, 50%, and 60%, and with a target thickness of 8 atm·cm.

Figure 2: Neutron transmission, T_n, polarization, P_n, and figure-of-merit, $P_n^2 T_n$ for a neutron spin filter with 50% helium polarization and a target thickness of 6 atm·cm and 8 atm·cm.

Figure 3: Polarization profile of a polarized ^{3}He cell measured with MRI techniques. The polarization has been destroyed in a small volume in the middle of the cell and figures show the evolution of the polarization.

Figure 4: Schematic diagram of the Rb spin-exchange ^{3}He spin filter. Shown are the laser, optics, Helmholtz coils of the holding field, RF coils for NMR measurement of ^{3}He polarization, oven, pick-up coils for NMR and signal processing electronics.

Figure 5: Conceptual layout of the absolute neutron beam polarization experiment at MLNSC. The most important elements and their distances from the source are indicated.

Figure 6: Neutron beam polarization as a function of neutron energy from 20 meV to 10 eV. The ^{3}He thickness is 50 atm·cm and ^{3}He polarization is 15%.

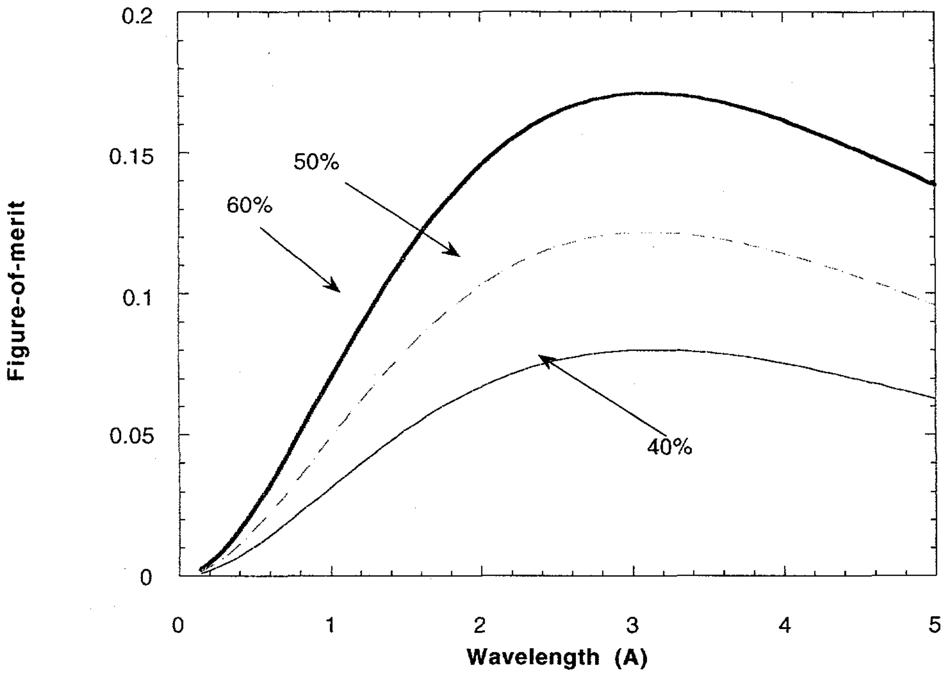
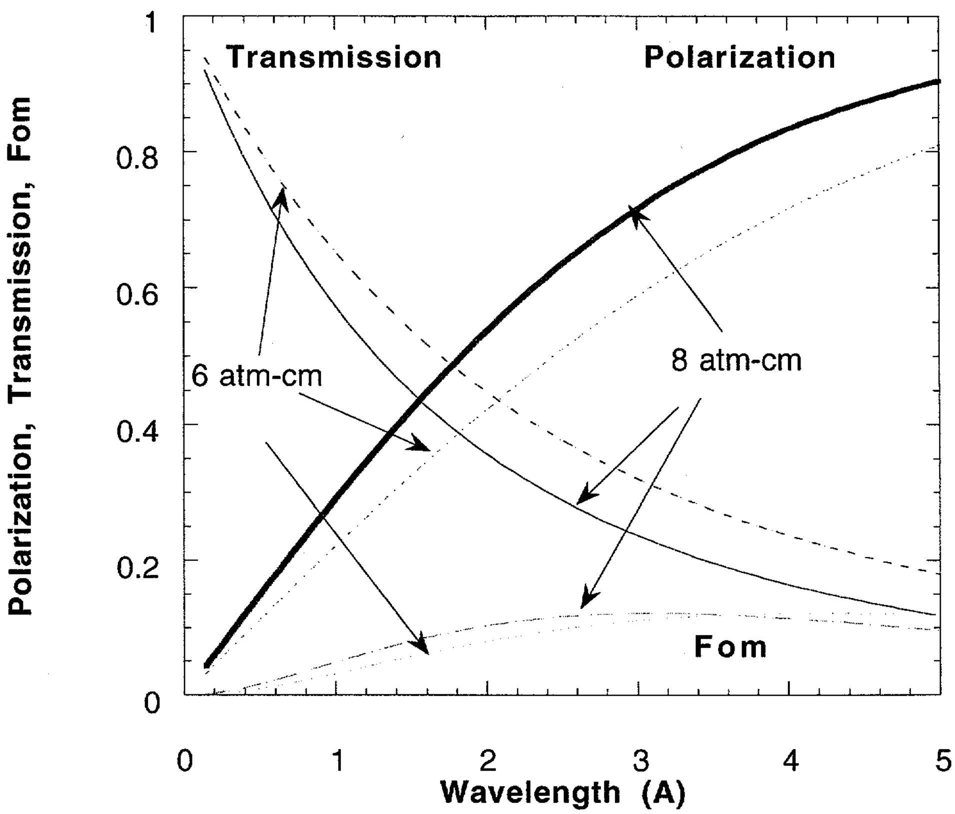
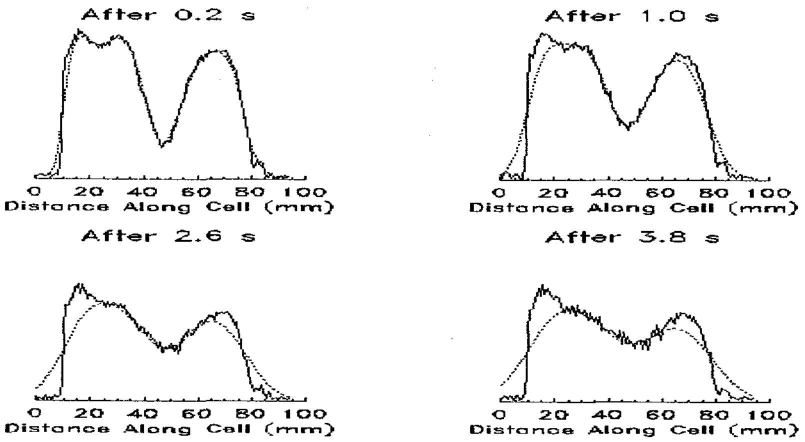


Figure 1



Transmission
Polarization
6 atm-cm
8 atm-cm
Fom
Polarization, Transmission, Fom
Wavelength (A)
0
0.2
0.4
0.6
0.8
1
0
1
2
3
4
5

Figure 2


After 0.2 s
0 20 40 60 80 100
Distance Along Cell (mm)
After 1.0 s
0 20 40 60 80 100
Distance Along Cell (mm)
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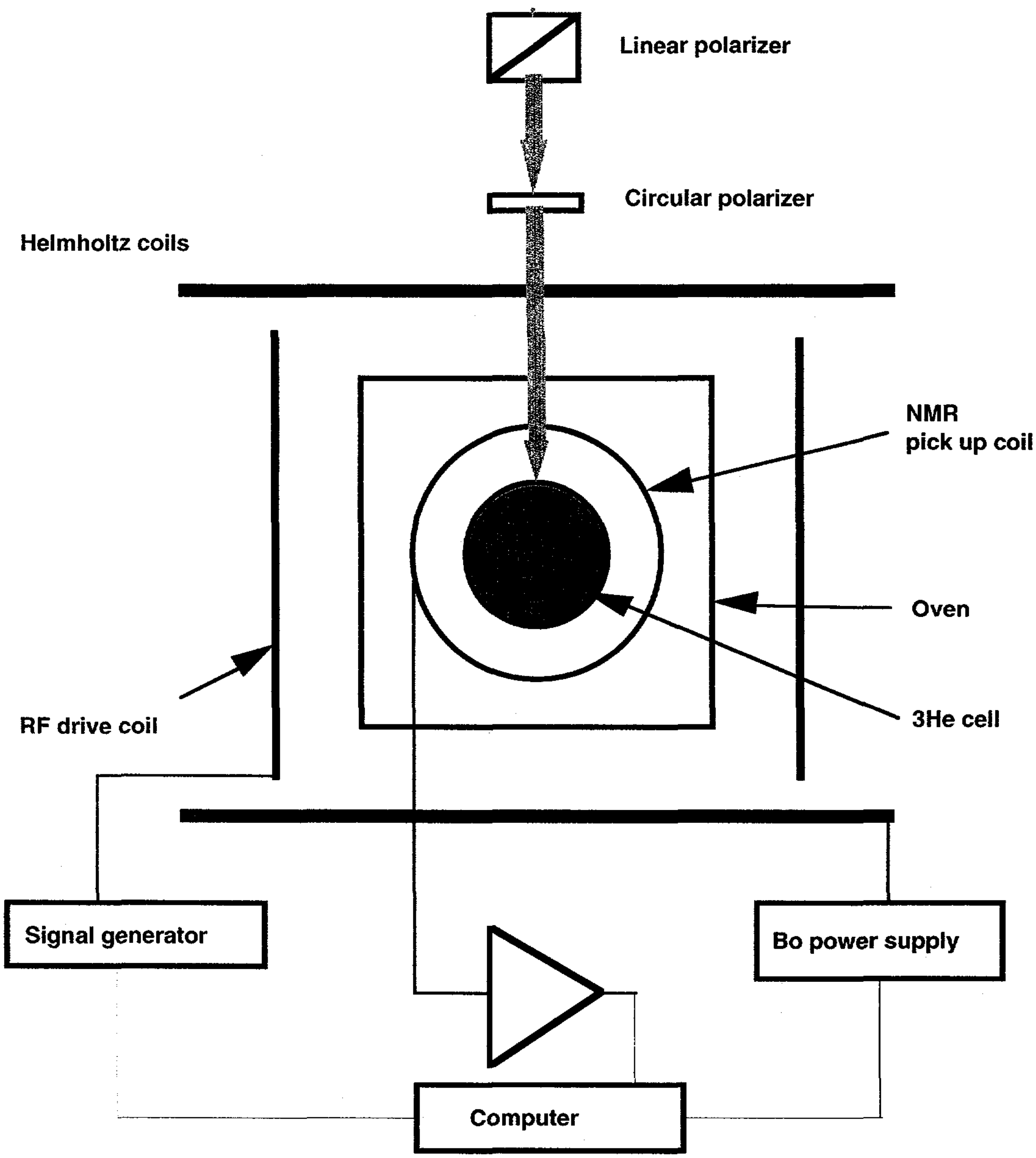
Distance Along Cell (mm)
After 3.8 s
0 20 40 60 80 100
Distance Along Cell (mm)

Figure 3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Circular polarizer
Helmholtz coils
NMR
pick up coil
Oven
RF drive coil
3He cell
Signal generator
Bo power supply
Computer

Figure 4

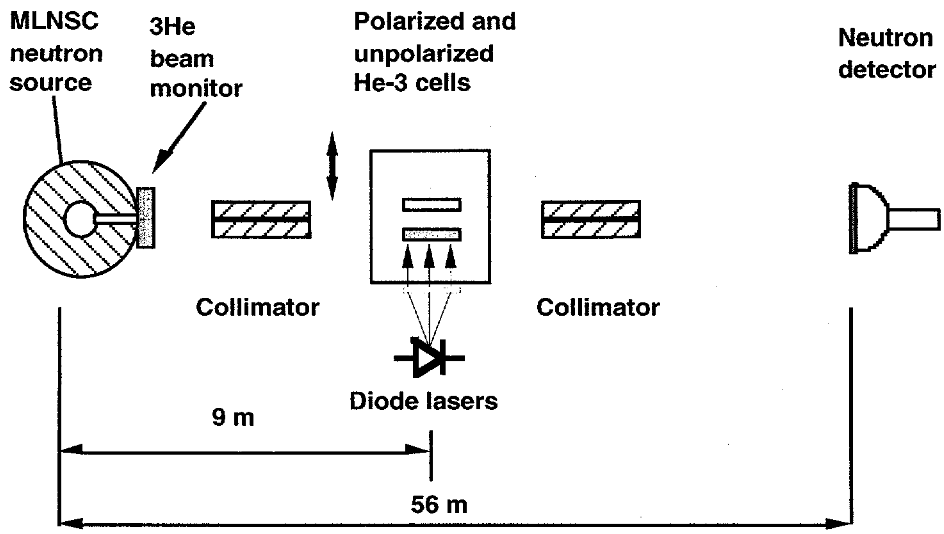
MLNSC
neutron
source
3He
beam
monitor
Polarized and
unpolarized
He-3 cells
Neutron
detector
Collimator
Collimator
Diode lasers
9 m
56 m

Figure 5

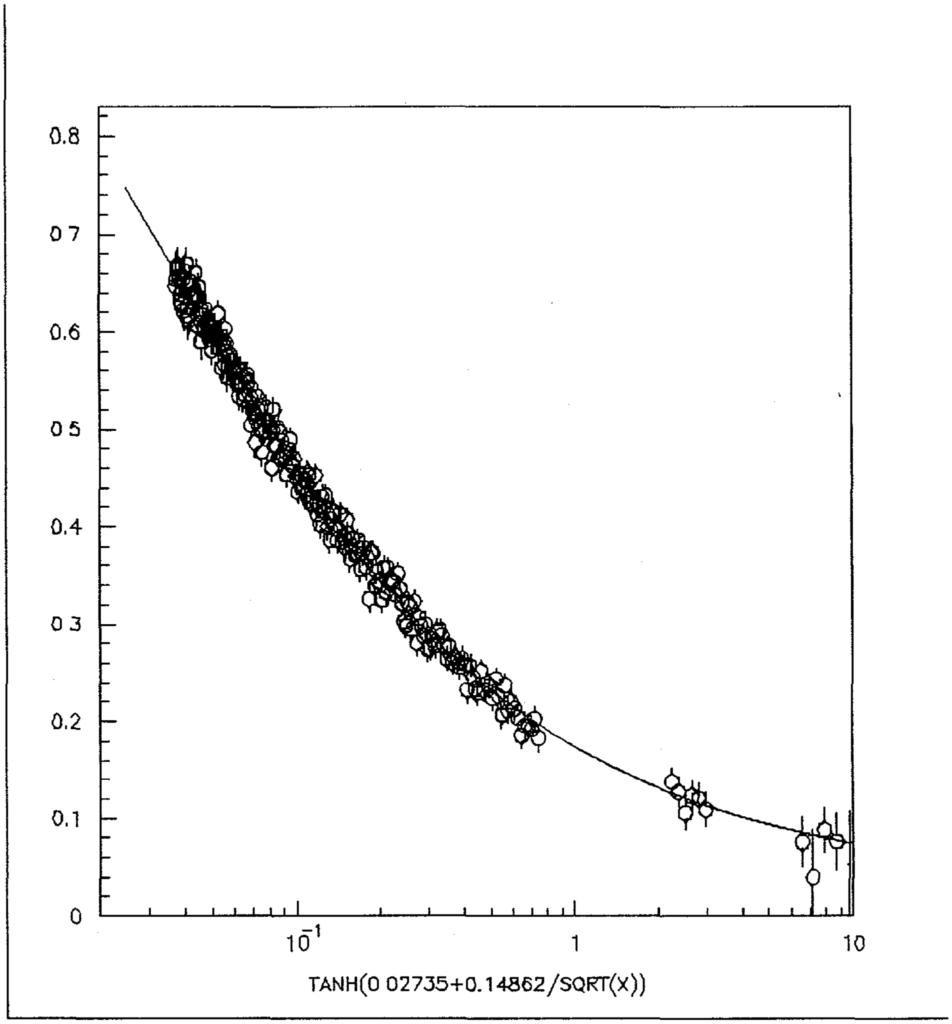

Figure 6