I. Introduction

Several dynamically polarized targets are available for experiments at intermediate and high-energy accelerators. The advantage over a statically polarized sample is the possibility to reverse the polarization with respect to the magnetic field. The dynamic method is based on the property that the electron spin relaxation of paramagnetic impurities is very short compared to the nuclear spin relaxation time. If nuclear spin flips are induced during microwave irradiation of the electron spin system, the electron reservoir recovers fast, while the nuclear polarization is transported away through spin diffusion. The polarization can be reversed by changing the frequency of the microwaves [1].

High proton polarizations (0.5) were first obtained by Schmugge and Jeffries [2] in lanthanum magnesium nitrate doped with a few percent neodymium. At CERN, polarization of frozen alcohols was developed [3] and the construction of a horizontal He-3/He-4 dilution refrigerator provided a target of the 'frozen spin' type [4]. With this technique a reduced and less homogeneous magnetic field can be applied in the target region during the time that the particle beam is transmitted. For studies of charged particle induced reactions another horizontal frozen spin target was constructed at KEK [5] and vertical refrigerators have been built in Saclay [6] and Los Alamos [7].

After a test of charge symmetry in elastic neutron scattering was approved at TRIUMF, a frozen spin polarized proton target was constructed with the specifications of table 1. The target was operated for about 2000 hours in periods that were up to 3 weeks long. A preliminary report on part of our polarization decay data was presented in ref. [8].

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THE FROZEN SPIN POLARIZED PROTON TARGET AT TRIUMF FOR INTERMEDIATE ENERGY NEUTRON SCATTERING

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Abstract

A large volume (55 cm³) polarized proton target was constructed at TRIUMF to study neutron induced reactions. The polarizable protons were contained in the molecules of 1-butanol that was frozen in the form of beads and placed in the mixing chamber of a vertical dilution refrigerator. The target system was operated about 2000 hours for a high precision neutron scattering experiment to test charge symmetry breaking. The highest polarization achieved was -0.97. The relaxation time was typically 150 hours in a magnetic field of 0.257 T and at a temperature of 60 mK.

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The only other reported relaxation times [11,14] near our temperatures and magnetic field, were measured in Saclay.

2. Apparatus

To produce dynamic polarization a sample is mounted on a refrigerator to obtain the appropriate electronic and nuclear relaxation times. The magnetic field removes the degeneracy of the Zeeman levels. Then, the microwave irradiation can change the occupation numbers of the Zeeman levels. This distribution is measured with a nuclear magnetic resonance (NMR) set-up. Finally, a digital control system is used to collect and store the operational data. These five parts will be described in the following sections.

2.1 The He-3/He-4 dilution refrigerator

In fig. 1 a schematic diagram is drawn of the main refrigerator components and the location of the diagnostic sensors. A photograph of the refrigerator is shown in fig. 2. The He-3 is circulated by a pumpset from Alcatel consisting of two roots pumps (model MIV 3000 and model MIV 350) and a forepump (model 2060 H). From here, the He-3 passes several filters and a flow meter. After entering the cryostat the He-3 is precooled in a tubular counterflow heat exchanger. Further cooling takes place in the separator pot with liquid helium at a temperature of 4.2 K, and in the condenser at a temperature of 1.4 K. As is usual for this type of set-up, the He-4 precooled system is located above the still inside the He-3 pumping tube.

Liquid helium is continuously supplied to the separator from a 100 % storage dewar. The boil off gas is pumped out through the precooled counterflow heat exchanger and, in parallel, through a tube that cools three radiation baffles. This baffle line is open only during the

start-up phase of a run. The condenser is supplied with liquid helium from the separator through a needle valve that is operated from the top of the refrigerator. An automatic controller reads the output of a superconducting level probe. At two preset liquid levels a motor changes the valve setting between two preset positions. All presets are manually adjustable. The liquid helium consumption of the refrigerator is 1.7 % to 2.0 % per hour.

Below the condenser the He-3 tube splits into two branches, in each of which the flow rate is regulated by a needle valve that is operated manually from the top of the cryostat. One branch is a straight tube with a 1 mm inner diameter that leads directly into the mixing chamber. This leg is only used to cool the target initially from a temperature of 100 K to 4 K. The other branch leads to the main restriction of the refrigerator: a valve with a flow impedance that is variable between $1.2 \times 10^8 \text{ cm}^{-3}$ and $170 \times 10^8 \text{ cm}^{-3}$. Below this valve, the He-3 passes through two tubular heat exchangers, that are located in the gas phase (60 cm long) and the liquid phase (240 cm long) of the still, respectively. The space below the still is filled up, down to the mixing chamber, with a tightly fitted phenolic rod (clearance on the diameter less than 0.1 mm at room temperature) in which grooves are machined for the components of the main dilution unit heat exchanger. At the top of the rod a secondary restriction ($3.5 \times 10^8 \text{ cm}^{-3}$) is located, followed by a Cu-Ni tube of 5 m length. The last part consists of two copper tubes with about 1.5 g of copper powder (45 μm particle size) sintered inside and 4 g of copper powder sintered on the outside wall. The stream of concentrated He-3 exits from the sintered copper units to the mixing chamber, where it is diluted, and diffuses through the space between the

phenolic rod and the inner wall of the cryostat into the still where it evaporates and is pumped out. The cryostat is removed to load and unload the target material. The radial thickness of the cryostat is 0.8 g/cm^2 at the height of the neutron beam.

The parts of the refrigerator are mounted on a center tube with a diameter of 19 mm. Two NMR cables and a microwave guide run in this tube through a vacuum chamber below the still to the mixing chamber. Three compartments are contained in the mixing chamber with a volume of approximately 50 cm^3 each. One vacuum chamber is located above and another one below the sample (see fig. 3). A microwave cup, that is formed as a vertical cylinder 4 cm in diameter and 5 cm high from $50 \mu\text{m}$ thick copper foil, is suspended from the upper vacuum compartment with a glue joint. This cup contains a hole near the top with a diameter of 5 mm to accommodate a funnel for loading of the target material. When this is finished, a FEP (teflon) sleeve is pushed down to force the He-3 flow through the sample. The bottom of the cup is a perforated copper foil to which a heater, a copper constantan thermocouple and two resistance thermometers are attached. The foil is held up by four straps that are soft soldered to the cup and the bottom. To unload the target, three straps are cut, after which the bottom pivots to a vertical position and the butanol beads drop into a container with liquid nitrogen.

2.2 The magnet system

A superconducting solenoid, that can produce a magnetic field of 2.7 T in its center, is mounted near the top of a cryostat that has a vertical room temperature bore with a diameter of 12 cm*. The homogeneity of this magnetic field is 5×10^{-5} over a volume of 100 cm^3 and the decay rate in

*This magnet was manufactured by Oxford Instruments Ltd.

persistent mode is 1×10^{-5} per hour. The consumption of liquid helium is 0.5 l/hour in persistent mode. About 15 cm above the magnet cryostat an electromagnet is mounted with a bore of 30 cm and a field of 0.25 T in the center. This magnet is energized with a current of 500 A that is stable within 10^{-4} . The two magnets are rigidly supported within one frame that can be moved in the vertical direction to put the target in the center of the superconducting solenoid or in the middle of the gap where the central field is 0.257 T. The position for beam transmission is accurate to 1 mm in all directions and the magnetic field on the target is reproducible and stable to better than 10^{-3} .

2.3 Microwaves

The microwaves are produced by a Varian klystron model VRE2101B7, with a nominal output power of 600 mW at 70 GHz. The waveguide consists of a K-band transmission line outside the cryostat and a copper plated Cu-Ni tube with a 4 mm inner diameter inside the refrigerator. A microwave power of about 350 mW can be delivered to the mixing chamber. The frequency is measured with an EIP model 548A microwave frequency meter. The forward power and the reflected power are determined with Hewlett Packard model 432A power meters. The process of polarizing the target is started with a microwave power level of 100 mW, which is gradually reduced to 20 mW as the polarization is increased.

2.4 The nuclear magnetic resonance system

To measure the polarization a series resonant constant current NMR system is built. In the mixing chamber the NMR coil (inductance $\approx 0.2 \mu\text{H}$) is embedded in the target material. Three windings with a diameter of 2 cm are formed from copper tube of 2.2 mm outer diameter. A teflon sleeve with a wall thickness of 0.5 mm covers the coiled tube. In the

mixing chamber one end of the NMR coil is soft-soldered to ground and the other side is connected to a semirigid coaxial cable via a capacitor. Through trial and error this capacitor (capacitance ≈ 10 pF) is selected to obtain a resonance frequency equal to 108.5 MHz at 4 K. The capacitance is constant below this temperature. Outside the cryostat a reference chamber with a coil and its capacitor is made as nearly identical to the mixing chamber as possible. The input currents for both coils are delivered by the same source, but they are out of phase by 180 degrees. The magnetic field is adjusted to make the Larmor frequency for the protons equal to 108.5 MHz. The sum of the two output voltages is negligible as a function of frequency, with the exception of the region where the sample resonates and consequently modifies the impedance. By adjusting the input current, the output voltage at resonance is made comparable in size for the calibration at thermal equilibrium and for the measurement of the polarization enhancement due to the microwave irradiation. Calculations show that in the NMR circuit the systematic errors due to non-linearities are reduced to a level below 1 percent.

2.5 The digital control system

Two microprocessors in CAMAC organize the input and output of the measuring equipment. This consists of steering the frequency synthesizer, digitizing the NMR signal, selecting thermometers for the resistance bridge and reading out of the resistance bridge, thermocouples, pressure transducers, vacuum gauges and a flow meter. The collection and storage of the data is supervised by a LSI 11-23 in the same CAMAC crate. This computer controls the following peripherals: A serial link with the data acquisition computer for the particle detectors, a dual floppy disk drive, a printer and a graphics display terminal with keyboard.

3 Results

Two important parameters in the figure of merit for a frozen spin target are the absolute value of the polarization and its relaxation time. The polarization is determined after the sensitivity of the NMR system is calibrated, when the proton spin system is in thermal equilibrium (TE) with the lattice. The polarization can be calculated according to the Maxwell-Boltzmann distribution if the magnetic field and the temperature are determined. A very accurate value for the magnetic field is derived from a measurement of the Larmor frequency. To determine the temperature, two calibrated thermometers were used. A germanium resistor was calibrated from a temperature of 3 K until 50 mK**, and a carbon resistor that was calibrated between 1 K and 40 mK. Both thermometers agree very well between 100 and 800 mK (see fig. 4). It should be noted that germanium resistors are very sensitive to the magnetic field, while this effect is negligible for carbon resistors [9]. Therefore, the carbon resistor was calibrated in the temperature region near 1.6 K with the germanium resistor. After the calibration the germanium resistor was replaced by another carbon resistor for consistency checks.

Special attention was paid to the prevention of thermal gradients between the target material and the thermometers. Before a TE measurement the target was typically at a temperature near 60 mK. Then, the He-3 pump line and condenser pump line were closed. The mixing chamber heater warmed the sample up to a temperature of 2 K. Next, the condenser pump line was opened and the sample cooled in 1 hour to a temperature between 1.6 K and 1.7 K. A state was reached where the temperature drift

**Purchased from Scientific Instruments Inc., West Palm Beach, U.S.A.

was less than 5 mK per hour. Then, the NMR signal was measured several times over a period of one hour. Typically, the TE calibration was carried out once a week during cyclotron maintenance periods. From the neutron scattering data the polarization was determined independently with an accuracy of 4%. On this error level both polarization measurements agree well [10]. In table 2 the results are listed that were obtained during 6 measuring periods on the beam line. Regularly, negative polarizations around 0.9 were reached after 2 hours of microwave irradiation. Irradiating four hours with the microwaves gave the highest polarization -0.97 which shows that in butanol nearly complete polarization can be obtained as was previously reported for propanediol [13]. Table 2 also contains 70 decay times that were collected in a magnetic field of 0.257 T and at temperatures between 50 mK and 90 mK. A measurement with the target polarized lasted about four hours since the polarization and/or the magnetic field direction were reversed daily and the ratio of the beam times with a polarized target and unpolarized target was about 1:4. From fig. 5 it is clear that the data show fluctuations in the decay time of about a factor 2, which corresponds to a change in temperature of approximately 7 mK. This is attributed to a variation of the temperature difference between the center of the target, where the NMR coil is located, and the bottom of the microwave cup to which the thermometer is attached. No large difference of the decay rates for positive and negative polarization is observed as was indicated in ref. 11. Averaging of our six measurements in the temperature region from 53 mK to 57 mK yields a relaxation time of 11 days. In ref. 14 a decay time of about 7 days is indicated for a temperature of 55 mK in a magnetic field of 0.25 T. In view of the systematic uncertainties the

agreement is good. It is obvious in fig. 5 that the relaxation times are not strongly dependent on the concentration C of the EHBA doping [17], that provides the paramagnetic electron centers in the target material.

This is in contrast with the results in lanthanum magnesium nitrate [12] where a concentration dependence $\tau \sim C^{-3}$ was observed in a magnetic field of 0.235 T. A logarithmic plot of the data (fig. 5) shows fairly well a linear dependence on the inverse temperature with the exception of the measurements near 85 mK that seem shifted upwards by about 7 mK. If these data points, that belong to run 3, are omitted the least squares approximation according to

$$\log_{10} \tau = a(1/T) + b$$

yields the values $a = 0.176 \pm 0.010$ and $b = 0.79 \pm 0.15$. Here the decay time τ and the temperature T are expressed in hours and kelvin, respectively. It should be noted that the data are also described well by a power law

$$\log_{10} \tau = a \log_{10} T + b$$

with $a = -6.09 \pm 0.14$ and $b = -5.3 \pm 0.7$. This T^{-6} function was also observed for propanediol near 0.5 K [13]. A similar steep temperature dependence was measured for lanthanum magnesium nitrate [12]. Theoretical estimates explain the relaxation features only qualitatively. (see e.g. [15,16] and references therein).

Summary

A set-up was built for dynamic polarization of a proton target with a volume of 55 cm³. It was operated in frozen spin mode with good reliability for a high precision neutron scattering experiment. Nearly complete polarization $P = -0.97$ was obtained in butanol. From the

scattering data an independent check on the NMR results was derived that showed these measurements to be free of systematic errors to the 4% level. Polarization decay times upto about 20 days in a magnetic field of 0.257 T were measured. In this field the temperature dependence of these data is described by an exponential function or a power law T^{-6} in the temperature region from 50 mK to 90 mK.

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Table 1 Specifications of the frozen spin proton target for the test of charge symmetry in elastic neutron scattering at 480 MeV.

Polarization	0.8
Relaxation time	150 Hours
Magnetic holding field	0.26 T
Holding field reproducibility	$\pm 10^{-3}$
Target volume	55 cm ³
Space around the target center	Horizontal ± 7.5 cm
free from substantial background material	Vertical ± 7.5 cm
Opening angle	Horizontal $\pm 93^\circ$
	Vertical $\pm 11^\circ$

Table 2 Initial polarizations P_I , relaxation times τ and temperatures T during a test of charge symmetry.

RUN	EHBA concentration 4×10^{19} molecules/cm ³			EHBA concentration 6×10^{19} molecules/cm ³		
	P_I (10 ⁻²)	τ (hrs)	T (mK)	P_I (10 ⁻²)	τ (hrs)	T (mK)
1-1	+76.7±0.9	38.4± 0.7	75	5-1	-56.5±0.7	26.2± 0.7
1-2	-88.1±0.9	30.1± 0.4	77	5-2	+87.6±1.2	67.6± 3.6
1-3	+78.5±0.6	47.0± 0.9	70	5-3	-92.4±1.5	23.4± 1.3
1-4	-87.3±0.7	44.8± 0.8	76	5-4	+78.7±1.5	49.9± 2.6
2-1	+79.9±1.6	66.1± 8.3	72	5-5	-87.0±1.8	39.4± 1.0
2-2	-88.8±1.7	52.3± 3.8	72	5-6	+83.6±1.1	84.4± 2.8
2-3	+74.2±1.4	41.6± 2.9	75	5-7	+82.4±0.9	109. ± 5.
2-4	-89.8±1.8	48.5± 3.2	72	5-8	-88.9±0.9	61.2± 2.4
2-5	+82.6±1.0	35.8± 1.0	78	5-9	+81.9±0.9	83.2± 3.7
2-6	-85.3±0.9	36.0± 1.0	77	5-10	-88.6±1.1	63.3± 2.1
2-7	-80.1±0.7	37.1± 1.2	74	5-11	+86.8±1.4	97.3± 2.5
3-1	+76.2±1.0	22.7± 0.7	87	5-12	-90.7±1.3	81.8± 3.6
3-2	-84.7±1.0	23.8± 0.7	81	5-13	+79.7±1.0	88.3± 5.3
3-3	+77.6±1.0	30.7± 1.3	84	5-14	-82.9±0.9	157. ± 9.
3-4	-85.0±1.4	28.7± 1.6	83	5-15	+84.9±0.9	165. ± 9.
3-5	+62.2±1.3	31.7± 1.8	83	5-16	-91.2±1.0	106. ± 5.
3-6	-76.5±3.4	29.6± 8.0	84	5-17	+78.7±1.0	270. ±19.
3-7	+76.4±1.5	30.4± 4.3	86	6-1	-97.2±1.2	25.9± 0.6
3-8	-84.9±1.6	25.3± 3.3	86	6-2	-86.3±1.1	96.8± 3.3
3-9	+77.3±1.3	25.4± 3.1	88	6-3	+79.7±1.2	102. ± 3.
3-10	-81.9±1.4	22.2± 2.5	87	6-4	-91.5±1.6	146. ± 8.
3-11	+74.6±1.3	18.4± 1.6	88	6-5	+82.5±1.6	89.9± 4.1
3-12	-79.6±1.4	21.5± 1.7	87	6-6	+88.6±2.2	90.9± 2.7
4-1	+82.7±1.5	73.5± 2.8	56	6-7	-88.2±1.7	15.1± 0.3
4-2	-91.0±1.6	59.3± 0.8	63	6-8	+79.8±1.4	24.3± 0.9
4-3	+79.0±1.2	134. ± 3.	58	6-9	-90.7±1.6	17.2± 0.7
4-4	+81.0±1.1	166. ± 7.	58	6-10	+82.0±1.7	25.3± 2.0
4-5	+79.9±1.2	119. ± 6.	63	6-11	-92.7±2.1	22.1± 1.4
4-6	+79.9±1.2	91.6± 4.7	65	6-12	+86.6±1.9	114. ± 7.
4-7	-85.4±1.4	152. ±10.	62	6-13	-93.4±1.7	210. ±20.
4-8	+82.1±1.5	266. ±14.	55	6-14	+83.7±1.5	133. ± 6.
4-9	-93.4±1.8	102. ± 5.	61	6-15	-91.0±2.0	117. ± 5.
4-10	+86.4±1.4	123. ± 7.	62			
4-11	-92.2±1.3	286. ±23.	61			
4-12	+86.9±1.2	176. ±17.	62			
4-13	-92.7±1.3	292. ±28.	53			
4-14	+86.0±1.4	259. ±15.	56			
4-15	-91.9±1.6	457. ±43.	53			

Figure Captions

Fig. 1 Schematic lay-out of the gas handling in the target system with the diagnostic sensors, HTR-Heater, R-Resistance thermometer, TC-Thermocouple.

Fig. 2 Photograph of the dilution refrigerator with the target; 1-precooling heat exchanger, 2-separator, 3-condenser, 4-still, 5-sintered copper heat exchanger, 6-vacuum chamber, 7-mixing chamber.

Fig. 3 Schematic diagram of the target chamber.

Fig. 4 Comparison of the temperature calibrations of the germanium resistor and carbon resistor between 70 mK and 800 mK.

Fig. 5 Temperature dependence of the polarization decay time.

Fig. 3

Fig. 4

Fig. 5