Observation of a 96% Proton Polarization in Irradiated Ammonia

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(Received 27 December 1989)

Using dynamic nuclear polarization we obtained proton spin-polarization values of $(+95\pm5)\%$ and $(-97\pm5)\%$ in radiation-doped frozen ammonia. Moreover, the polarization reached 90% in about 25 min. These results were obtained using our new 1-K high cooling power ^4He evaporation refrigerator operating in a 5-T magnetic field with 140-GHz microwaves. This unexpectedly large and rapid polarization coupled with about 1 W of cooling power should allow significant improvements in high-energy spin-physics experiments and may have some physics interest in itself.

PACS numbers: 33.25.Hv, 29.25.Kf, 75.25.+z

The use of dynamic nuclear polarization[1,2] (DNP), to spin polarize the free protons in materials such as alcohols, has allowed detailed studies of spin effects in high-energy physics scattering experiments.[3] A limited number of materials such as butanol, propanediol, and ethanol have been used extensively in polarized proton targets for these scattering experiments. The paramagnetic radicals required for the DNP process were chemically doped into the host materials; this typically allowed a polarization of up to 80% using ^3He evaporation refrigerators operating at 0.5 K in a 2.5-T magnetic field.

A high-intensity particle beam is necessary to precisely study low cross-section processes, such as spin effects in high-$P_\perp^2$ proton-proton scattering. Overcoming the heat load from the intense beam requires a high-powered cryogenic system in the polarized proton target. Moreover, the high-intensity beam causes significant radiation damage to the target material which disrupts the polarization process; the polarizability falls approximately exponentially with radiation dose. The polarizability can be partially recovered by annealing, but eventually the target material must be replaced. Therefore, a search was made for more radiation-resistant materials.

Hwang and Sanders and others[4] suggested that the radicals might be doped in by radiation rather than by chemical techniques. This radiation-doping method was first shown to work in butanol by Court and co-workers.[5] Niinikoski and Rieubland[6] then obtained polarizations above 90% in irradiated ammonia (NH_3) using a dilution refrigerator operating at 0.15 K and 2.5 T. However, with this low temperature and their radiation dose, the polarization growth time was typically about 30 h. Subsequently, NH_3 was used as the target material in many scattering experiments[7,8] with polarized targets operating at 0.5 K and 2.5 T. A polarization of about 70% could be reached in about 2 h. Ammonia was found to be more resistant to radiation damage than the best chemically doped materials; moreover, the polarizability appears to be completely recoverable by annealing.

For many years we did scattering experiments using a polarized proton target with a 2.5-T magnetic field and a ^3He evaporation refrigerator operating at 0.5 K. Unfortunately, the high-intensity proton beam could raise the temperature of the target material considerably above that of the surrounding fluid because of the poor thermal properties of the liquid ^3He. Moreover, it was difficult to increase the cooling power of our ^3He refrigerator above about 140 mW. During recent years we made several improvements to our refrigerator, such as improving the fluid path and changing the evaporating fluid to a mixture of ^3He and ^4He.[9] Nevertheless, we could not operate beyond a limit of about 2×10^{10} beam protons/sec; exceeding this limit reduced the target polarization considerably. In this paper we report on recent studies where we have polarized the protons in irradiated NH_3 at 1 K using a powerful ^4He refrigerator operating in a 5-T magnetic field with 140-GHz microwaves.

Our new polarized proton target was designed to operate with a high-intensity beam by using the superior thermal properties of ^4He. We constructed a high-cooling-power ^4He evaporation refrigerator which operates at 1 K inside a new 5-T magnet. Figure 1 shows the refrigerator and magnet assembly. The split coil superconducting magnet was designed to operate at up to 5.1 T with a field uniformity of $\pm10^{-4}$ inside a 4-cm-diam sphere; it was supplied by Oxford Instruments. The refrigerator is vertical and is inserted along the bore of the magnet. The refrigerator operates by first drawing liquid ^4He, at 4.2 K, from the magnet Dewar into a separator. The cold-^4He vapor from the separator is then pumped away and used to cool three radiation baffles. The liquid ^4He from the separator traverses a heat exchanger and then passes through a control needle valve into a liquid-^4He pool at the bottom of the refrigerator. The liquid ^4He is finally pumped on by a 5900-m^3 per hour Roots pumping system; the cooling power

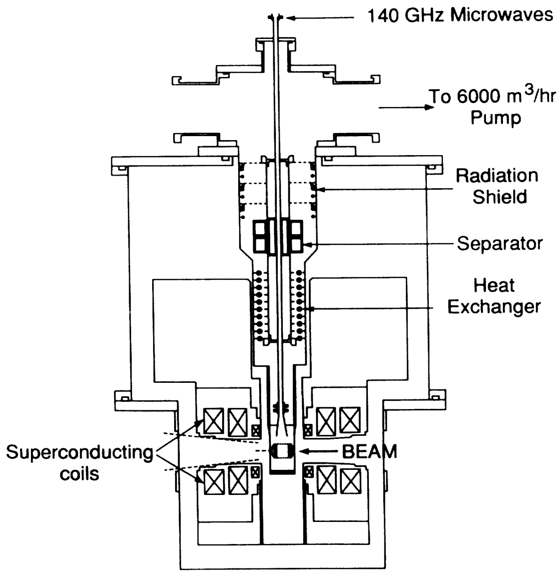

FIG. 1. Diagram of the new polarized-proton-target apparatus. The superconducting magnet produces a highly uniform 5-T field. The ^{4}He cryostat produces about 0.9 W of cooling power at 1 K. The target material is contained in the small cavity at the bottom of the cryostat. The 140-GHz microwaves are fed into the target cavity via the horn. The proton beam will pass through the target cavity.

due to the evaporation at 1 K is about 0.9 W.

The target cylinder, containing the frozen NH_3, is mounted at the bottom end of a long-vertical insert. The cylinder is constructed of Kapton with bronze end caps and is 36 mm long by 20 mm in diameter. The frozen NH_3 in the cylinder is inserted into the helium pool along the vertical refrigerator axis. The insert also contains the temperature and liquid-level sensors, the microwave waveguide, and the NMR coils along with their attached cables. The microwaves are generated by a Varian extended-interaction oscillator tube, which operates in the frequency range 138–142 GHz with a maximum power output of 21.8 W. For these studies, the total microwave power delivered to the NH_3 was about 0.3 W as measured by the ^{4}He evaporation rate. The optimum frequencies were found to be 140.35 GHz for the positive enhancement and 140.66 GHz for the negative enhancement.

The polarization inside the target cavity was measured using an NMR coil which is part of a series *LCR Q* meter.[10] The NMR coil is a straight wire along the target cylinder axis. The magnet was operated at a field of 5.011 T corresponding to a NMR frequency of 213.3 MHz. The NMR system was calibrated by using it to measure the thermal equilibrium polarization at several temperatures in the range 1.5–1.7 K with the microwave power turned off. Each polarization was then calculated using the standard formula

$$P = \tanh\left(\frac{\mu_p B}{kT}\right), \qquad (1)$$

where μ_p is the proton's magnetic moment, B is the magnetic field, and k is Boltzmann's constant. The temperature T was measured by using both vapor pressure techniques and calibrated resistors. The linearity of the NMR system was tested by varying the rf driving voltage over a factor of 2 and by varying the signal detection gain. All measurements agreed within 5%. With the NH_3 material removed from the target cylinder both the thermal and enhanced signals were less than 2% of the NH_3 signals. We estimate our total error in the polarization measurement to be $\pm 5\%$.

The target material was prepared by slowly freezing liquid NH_3 and then crushing it and selecting fragments of about 2 mm in size. The frozen NH_3 material was then irradiated under liquid argon at a temperature of about 89 K using a 260-MeV electron beam at the MIT Bates Linac. The average beam current during the irradiation was about 10^{13} electrons/sec. Typically the NH_3 in the Dewar was irradiated by a total of about 10^{17} electrons with a spot size of about 3 cm^2. Subsequently, the material was stored at 4.2 K under liquid ^{4}He for three months before being polarized in our old polarized proton target at 0.5 K and 2.5 T. The resulting polarization growth curves shown in Fig. 2 are consistent with our data from previous NH_3 irradiations.[11]

The same NH_3 fragments were then stored under liquid nitrogen at 77 K for about ten months and then

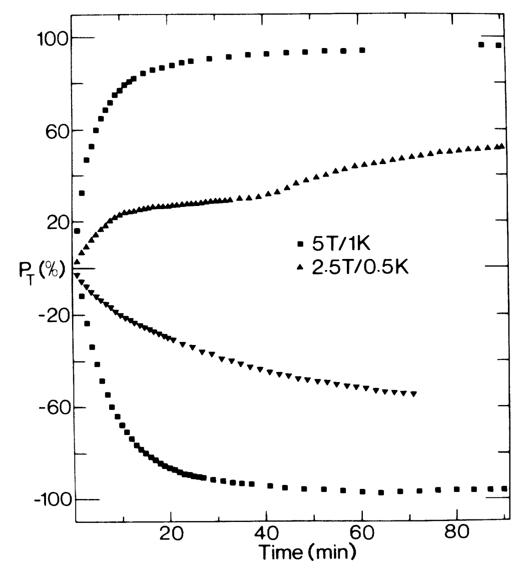

FIG. 2. The spin polarization of the free protons in NH_3 is plotted against the time of microwave irradiation. The new data at 5 T and 1 K are shown as squares; the earlier data at 2.5 T and 0.5 K are shown as triangles.

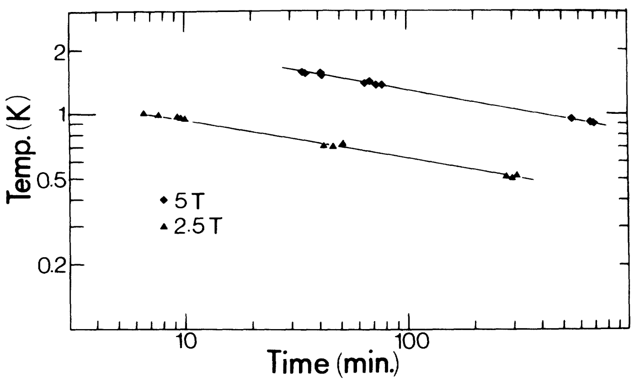

FIG. 3. The spin-relaxation time for the protons in NH_3 is plotted against temperature. The new data at 5 T are shown as diamonds; the earlier data at 2.5 T are shown as triangles.

used for the measurements with the new polarized target at 1 K and 5 T. The results obtained at 5 T and 1 K are also shown in Fig. 2. Notice that for both signs of polarization there is now a rapid growth of polarization to 90% in about 25 min. The ultimate polarizations achieved were $(+95\pm 5)\%$ and $(-97\pm 5)\%$.

The only other polarization data for NH_3 at 5 T is from Seely *et al.*,[12] who obtained a polarization of about 75%. However, their irradiations were made *in situ* at 1 K with a lower dose; thus it is not obvious that their results can be compared directly with ours. It appears that NH_2 is the dominant radical in the DNP process after the irradiation of NH_3 at 90 K; while in 1-K irradiations other radicals certainly can contribute to the polarization process. Their 1-K irradiations,[12] with a dose of about 10^{15} electrons/cm^2, produced a proton spin-relaxation time of about 300 min. We have about the same proton spin-relaxation time with our dose of about 3×10^{16} electrons/cm^2 at 89 K. Our new 5-T measurements of the proton spin-relaxation times are shown in Fig. 3 along with our earlier 2.5-T measurements for comparison. Seely *et al.* indicated that much higher electron doses were needed to obtain reasonable polarization when they irradiated at 20 to 80 K. They also noted that they were limited in the amount of microwave power which they could apply to their NH_3 samples.

In summary, we have shown that irradiated NH_3 can have a proton spin polarization of about $(96\pm 5)\%$ with a fast polarization growth. Apparently, this high polarization requires operation near 5 T and 1 K with high microwave power and considerable cryogenic cooling power. Using this new polarized proton target should allow considerably improved studies of spin effects in high-energy experiments such as proton-proton scattering at high $P_\perp^2$. This unexpectedly high polarization at this temperature and magnetic field may also have some physics interest in itself. The clear improvements in polarization compared to 2.5 T and 0.5 K could be due to the longer proton spin-relaxation time at 5 T and 1 K. It could also be due to the better separation of the positive and negative enhancement peaks at 5 T.[13]

We would like to thank C. E. Arzt and J. M. Caraher for their help with this experiment and the staff of Brookhaven National Laboratory for their hospitality. We also thank the staff of the MIT Bates Laboratory for their help with the irradiation. This research was supported by a grant from the U.S. Department of Energy. C.R. acknowledges early work on this project was done while on leave at the University of Michigan and at MIT.

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