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Ammonia as a polarized solid target material—a review

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Abstract

The enormous efforts towards dynamically polarized ammonia and deuterated ammonia (d-ammonia) with their reasonable high polarizable nucleon contents (quality factor) are reviewed. The early attempts are also a story of failures until a radiation doping method for NH_3 was successfully demonstrated in the end of the 1970s. Some years later the outcome of intense studies and new developments (with pitfalls, too) was the availability of the highly polarizable target materials $^{14}\text{NH}_3$, $^{15}\text{NH}_3$, $^{14}\text{ND}_3$ and $^{15}\text{ND}_3$, all of them doped for the dynamic nuclear polarization (DNP) by radiation. These materials have been used in new particle physics experiments which, up to that time, suffered either from the lower quality factors or from the modest polarization resistance against radiation damage of the existing alcohol and diol target materials.

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1. Introduction

The development, in the early 1960s, of the dynamic nuclear polarization (DNP) scheme in diamagnetic materials, doped with paramagnetic radicals, led to the use of solid polarized H and D targets in nuclear and particle physics experiments. A crucial value of any experiment is the optimization of the reaction counting rate. The number of counts is proportional to the luminosity \mathcal{L} —defined as the product of the beam current and the number of target particles per cm^2 —as well as to the running time t . The running time required to achieve a chosen accuracy ΔA in the measured physics asymmetry A is

$$t \approx \frac{1}{f^2 P^2 \mathcal{L} \Delta A^2} \quad (1)$$

where f is the target material quality factor,¹ defined as the ratio of the number of polarizable nucleons to the total number of nucleons in the target material and P is the polarization degree.

A figure of merit FOM_{exp} for the solid polarized targets used in particle physics experiments can then be defined by

$$\text{FOM}_{\text{exp}} = f^2 P^2 \mathcal{L}. \quad (2)$$

The optimization of the FOM_{exp} is a many-step procedure:

- Select a material with a high-quality factor f .
- Find for this material an appropriate radical doping method to make use of the DNP process.

¹In the literature labelled as ‘dilution factor’, which can be misunderstood.

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- Measure the highest polarization values for various magnetic field and temperature conditions.
- Find an optimal radical concentration for a short polarization build-up time or for frozen spin target operation.
- Measure the polarization for additional nuclei (if any) in the material.
- Determine the resistance of the polarization against ionizing radiation (normally done during the particle physics experiments).
- Find optimal temperature conditions to repair radiation damage (annealing procedure).

After the first use of lanthanum magnesium nitrate $\text{La}_2\text{Mg}_3(\text{NO}_3)_{12} \cdot 24\text{H}_2\text{O}$ (LMN)-doped with neodymium (Nd) [1], the above-mentioned requirements were reasonably fulfilled by alcohol- and diol materials doped by chemical methods [2]. Parallel to the developments in the field of cryogenics and superconducting magnet technology polarized solid target research was always focussed on the improvement of the target materials. At the end of the 1960s ammonia was considered as a possible candidate mainly due to its higher-quality factor $f = 0.176$ compared, e.g. to that of butanol $f = 0.135$.

2. Ammonia handling

2.1. Properties of ammonia

Ammonia gas was discovered in 1773 by Priestley. NH_3 is usually produced by the direct combination of nitrogen and hydrogen gases (Haber Bosch procedure). NH_3 is a colorless gas at room temperature and has a pungent smell. The handling of ammonia has to be done extremely carefully. On the one hand it is dangerous for the eyes and on the other it is suffocating. The maximum tolerable concentration is 35 mg/m^3 . The density of NH_3 is 0.817 g/cm^3 at 194 K. Its melting point is 195.5 K and its boiling point is 239.8 K.

2.2. Solidification of ammonia

Compared to the solidification of the alcohol and diol target samples, solidification of ammonia is more complicated and time consuming. First of all, the ammonia gas has to be liquefied, which can be done by a bath of methanol dry-ice at about 193 K. During the condensation process chemical dopants like Cr(V) glycerol complexes [3] or sodium can be dissolved and mixed. Afterwards the production of frozen beads can be done by dripping the liquefied ammonia into a nitrogen bath [4]. However, this solidification process has two major disadvantages: firstly, the mechanical stability of the polycrystalline beads (opaque white) is rather poor, resulting from voids and cracks inside the beads; secondly, the frozen beads possess a hole and their shape is nonspherical, which reduces the filling factor of a target volume to approximately 55% compared to 60% of a butanol target [5].

Very high mechanical stability of frozen ammonia is obtained with (mono) crystalline material. This is produced by slowly freezing ammonia ($t > 1 \text{ h}$) at about 193 K to a solid block, which is crushed in liquid nitrogen to small pieces of about 10 mm^3 [6]. However, the filling factor of crushed ammonia material is also in the order of 55%. To overcome both disadvantages the production of frozen ammonia beads was tried by dripping liquid ammonia into fluid isopentane (C_5H_{12}) [4]. This hydrocarbon substance was cooled to about 150 K. The result of this production method was clear beads (crystalline or glassy?), without holes and with good mechanical stability. However, storing the beads in liquid nitrogen as usual, they are coated with solid isopentane. A procedure to remove the solid isopentane still has to be found.

3. DNP in chemically doped ammonia

First dynamic proton polarization in ammonia was reported by Scheffler in 1970 [3]: “After trying a number of paramagnetic centers such as sodium, porphyrin, porphyrin, diphenylpicrylhydrazil, Ziegler’s radical, di-tert.-butyl-nitroxide, per-chlorotriphenylmethyl and violanthrone, all

of which gave low polarization or none at all, we doped the liquefied ammonia with Cr(V) glycerol complexes.”

Proton polarization values of 40% were reached in NH₃ doped with Cr(V) glycerol complexes at 1 K and 2.5 T. Even better results of 70% were obtained in ammonia doped with ethanediol-Cr(V) [7]. The relatively slow growth of the polarization (5–9 h) was explained with a model based on the formation of clusters of the complex, surrounded by a relatively pure ammonia matrix. However, the high polarization values were not confirmed by later works [8], and ammonia was not introduced as a polarized target material at that time.

The difficulties to obtain reproducible DNP results (if any) in chemically doped ammonia were confirmed by a later work with potassium-doped NH₃ [9]. Only 0.2% proton polarization after 2 h build-up at 1 K and 2.5 T were obtained in samples doped with 7×10^{19} K-atoms/cm³ and 7×10^{20} K-atoms/cm³, respectively.

4. DNP in radiation-doped ammonia

4.1. First results with NH₃

First dynamic proton polarization in irradiated NH₃ is reported by Niinikoski and Rieubland [10]. The irradiation of the ammonia sample was done with the 580 MeV proton beam of the CERN synchrocyclotron. During the irradiation the sample was kept under liquid N₂. The accumulated flux was 0.95×10^{15} protons/cm². Highest proton polarization values of +90.5% and –93.6% were obtained at temperatures below 0.5 K in a ³He/⁴He dilution refrigerator and at 2.5 T. The polarization build-up curve is shown in Fig. 1. The rather long polarization build-up time of 7–8 h is due to a low concentration of the radicals ($\approx 5 \times 10^{18}$ spins/cm³). Further proton irradiation of NH₃ samples at CERN led to explosions. Therefore, the activities were stopped, because there was no firm explanation for the cause of the explosions.

Shortly after this very promising DNP results with irradiated ammonia, this new target material

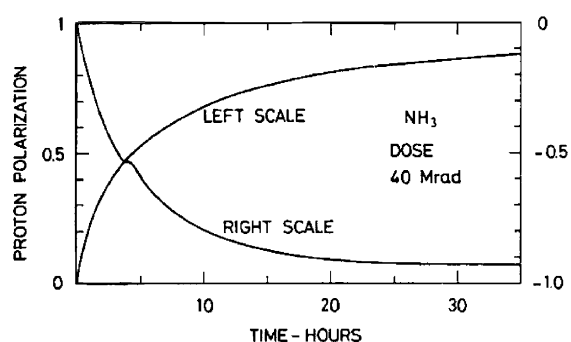


Fig. 1. Growth of the polarization in irradiated solid NH₃ for both signs of the polarization (from Ref. [16]).

was prepared by irradiation at the Bonn University using the 20 MeV injection linac of the electron accelerator [11,12]. Based on the experiences with irradiations of lithium compounds in SACLAY—there were some explosions when samples of lithium hydride immersed in liquid N₂ were irradiated with a 3 MeV electron beam [13]—ammonia samples were kept under liquid argon at ≈ 90 K (high-temperature irradiation) during the irradiation at the Bonn linac. The electron beam current was typically 2×10^{14} e⁻/s. With an accumulated flux of 10^{17} e⁻/cm², which was obtained in 2.5 h, high proton polarization values with short polarization build-up times (Fig. 2) were measured.

Storing the sample at liquid N₂ temperature (77 K) the produced radicals (see Section 5) are reasonably chemically stable so that DNP still worked 1.5 years after the irradiation process in the same sample [12].

Parallel to the measurements in Bonn, DNP studies with ammonia were performed using the Yale-SLAC polarized target system, which operated at 1 K and 5 T [14,15]. The target was setup at a beam line at SLAC, where the target materials were irradiated in situ at 1 K (low-temperature irradiation). In contrast to the high-temperature irradiation, this method allows the DNP measurement simultaneously. The irradiations at SLAC were done with a 6–20 GeV electron beam with a time averaged beam intensity of 3×10^{11} e⁻/s.

The maximum proton polarization for NH₃, 75%, was obtained after an irradiation dose of

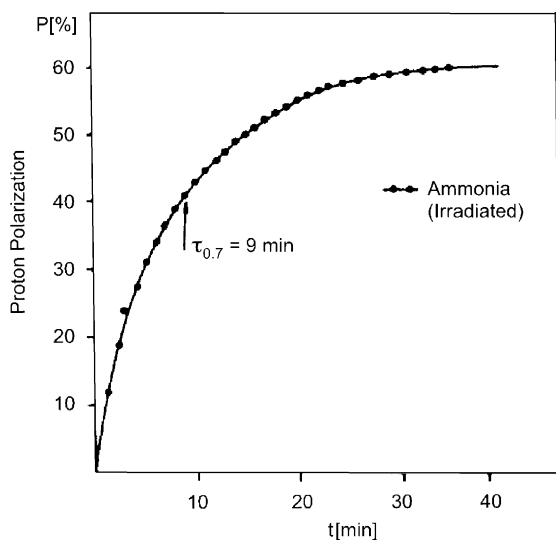


Fig. 2. Build-up of the polarization in NH_3 at 0.5 K.

$\approx 3 \times 10^{15} \text{e}^-/\text{cm}^2$. These first DNP measurements in NH_3 altogether demonstrated the superior radiation hardness—mainly as a result of the radical formation by irradiation—compared to that of the alcohol materials. The depolarizing doses are more than one order of magnitude above the depolarizing doses for the conventional target materials (Table 1). Finally, it should be noted that initial studies of NH_3 at SLAC using rapidly frozen polycrystalline ammonia beads (see Section 2.2) failed. These beads disintegrated to a fine powder when the dose reached $\approx 1 \times 10^{15} \text{e}^-/\text{cm}^2$.

4.2. First results with ND_3

First DNP results with irradiated ND_3 are reported from Bonn [16,17] and SLAC [15]. The preparation of ND_3 samples for the DNP measurements is the same as for NH_3 . In Bonn deuterated (99%) ammonia was irradiated with $10^{17} \text{e}^-/\text{cm}^2$ at 90 K in liquid argon (high-temperature irradiation). As in the case of NH_3 the material shows a dark violet color after the irradiation and is kept under liquid N_2 until the DNP measurements are performed. The following deuteron polarization results at 2.5 T were achieved: 3.5% at 1 K; 11% at 0.5 K and 31% at ≈ 0.2 K.

The polarization calibration was done by the thermal equilibrium method. Due to the microcrystalline target material, even enhanced NMR signals in ND_3 show a more wiggly structure compared to that of the glassy deuterated alcohol materials.

Remarkable in the case of these first measurements was the strong dependence of the deuteron polarization upon temperature. Furthermore, a comparison of the deuteron polarization with the proton polarization values of the unsubstituted protons (1%) in ND_3 indicated that the equal spin temperature (EST) theory is not applicable to irradiated ND_3 (see Section 6).

At SLAC first polarization results were obtained by means of the low-temperature irradiation method. The deuteron polarization of ND_3 increases as the target material is irradiated, leveling off at $\approx 25\%$ (1 K; 5 T) when the dose reaches $\approx 2 \times 10^{15} \text{e}^-/\text{cm}^2$. As the dose is increased, the polarization begins to decrease. As in the case of NH_3 the depolarizing doses are in the order of $\approx 10^{16} \text{e}^-/\text{cm}^2$.

4.3. Low- versus high-temperature-irradiated ammonia

In addition to the activities in Bonn and SLAC in the early 1980s, DNP in irradiated ammonia was also studied in Brookhaven [18,19], Liverpool [20] and later in Kharkov [21], too. In 1982 there were two methods for the preparation of ammonia for the DNP on the market:

1. High-temperature irradiation in liquid argon (87 K).
2. Low-temperature irradiation at 1 K with permanent DNP monitoring.

Due to the different target setups, in which these measurements were done, it was desirable to polarize low- and high-temperature irradiated material in the same polarized target apparatus. In 1983 this was done in dedicated runs at the 2.5 GeV electron synchrotron in Bonn [21]. For the high-temperature irradiation the beam of the above-mentioned 20 MeV injection linac was used. The pre-irradiated samples were stored in liquid

N_2 and transported to the experimental area of the external electron beam, where the polarized target system was setup. DNP measurements were performed at 1 K and 2.5 T, the same conditions, at which the low-temperature irradiation took place with 1.5 GeV electrons. To ensure a homogeneous irradiation the electron beam was scanned over the target front face with an intensity of $<5 \times 10^{10} e^-/cm^2$.

The maximum proton polarization in pre-irradiated NH_3 was 36%. With an additional dose from the high-energy electron beam this polarization degree decreased with a nonexponential decay rate (Fig. 3). The number in Table 1 for the critical doses ϕ_0 is obtained from different exponential fits using $P(\phi) = P_0 \exp(\phi/\phi_0)$. For comparison, the ϕ_0 values for butanol, chemically doped with porphyrin, are also given.

With increasing irradiation at 1 K a change of the frequency curve was also observed, i.e. the distance between the microwave frequencies for optimal positive and negative polarization values increased continuously with the irradiation dose. Furthermore, the polarization build-up and the nucleon relaxation time decreased. The best annealing temperature was found to be between 75 and 80 K.

The polarization dependence for low-temperature irradiated NH_3 is shown in Fig. 4. Missing any radicals the polarization starts at zero. The curve is interrupted by different annealing procedures resulting in a remarkable increase of the proton polarization (e.g. after an annealing procedure at 80 K up to 47%!)

In Fig. 5 the behavior of a virginal sample is compared to that of the pre-irradiated sample.

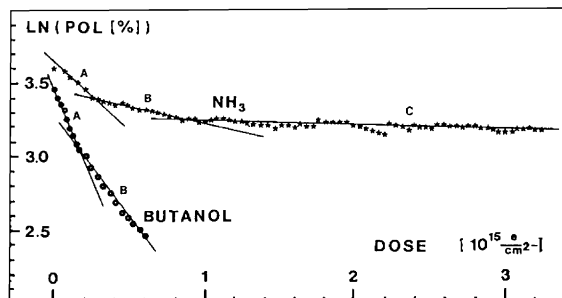


Fig. 3. Radiation damage of butanol and NH_3 (log scale).

Table 1
Critical dose for NH_3 and butanol [22]

	See Fig. 3	$\phi_0 [10^{15} e^-/cm^2]$
NH_3	A	1.0
	B	4.1
	C	30.0
Butanol	A	0.38
	B	0.66

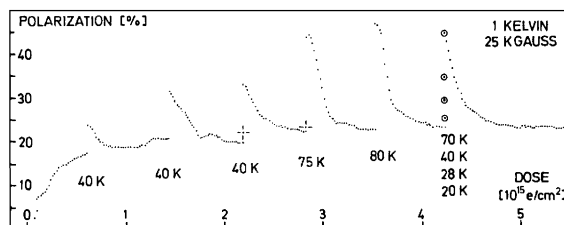


Fig. 4. NH_3 polarization without any pre-irradiation in dependence on the accumulated dose. Annealing procedures are responsible for the step structure.

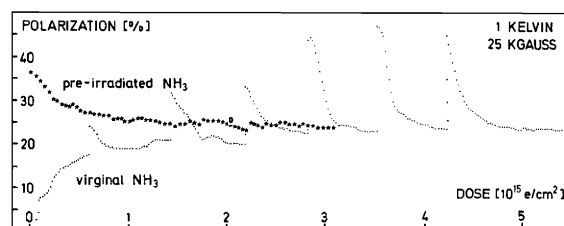


Fig. 5. Polarization dependence of pre-irradiated and virginal NH_3 in dependence on the dose.

Ignoring the annealings both samples reach the same asymptotical polarization value of about 25%.

In summary, low-temperature irradiation means that the radicals necessary for DNP are produced during the particle physics experiment. Since high radical densities are wanted in order to reduce the polarization build-up time, this preparation method is only suitable in combination with high-intensity beams of ionizing particles such as electrons and protons. The results clearly demonstrate one big advantage of the high-temperature irradiation method: DNP preparation of NH_3 and ND_3 (see below) can be done well in advance of the particle physics experiment, no matter whether

electron or proton beams or even low-intensity secondary beams such as muons, pions, kaons or photon beams are used.

As experience shows, the polarization behavior of ND_3 is more complicated than that of NH_3 . Whereas it is sufficient to dope NH_3 via high-temperature irradiation only, in order to create high proton polarization values under all conditions, the preparation of ND_3 demands an additional low-temperature irradiation at experimental conditions (≈ 1 K). As shown in Figs. 6 and 7, the typical beam flux to achieve best deuteron polarization values is about 4×10^{14} [22]– $10^{16} \text{e}^-/\text{cm}^2$ [23].

The steep increase of the deuteron polarization during the initial phase of an irradiation at low temperatures is completely different from that seen for the alcohol materials and for NH_3 . The hypothesis of a change of the dominant DNP-process in ND_3 or at least of the relevant time constants of the electron spin system during the further irradiation is supported by a remarkable shift of the optimum microwave frequency (see Fig. 8).

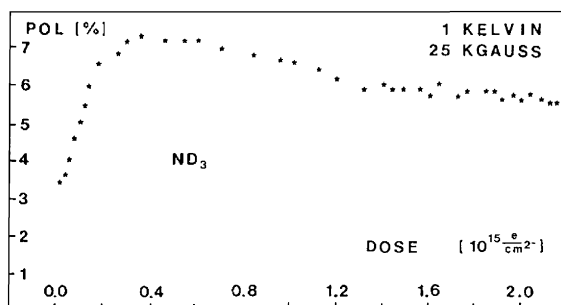


Fig. 6. Irradiation dependence of the ND_3 polarization.

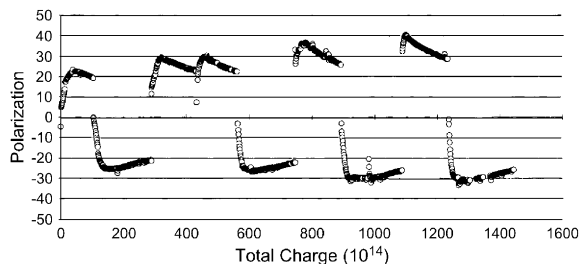


Fig. 7. Polarization history on one $^{15}\text{ND}_3$ target as a function of the received charge [23].

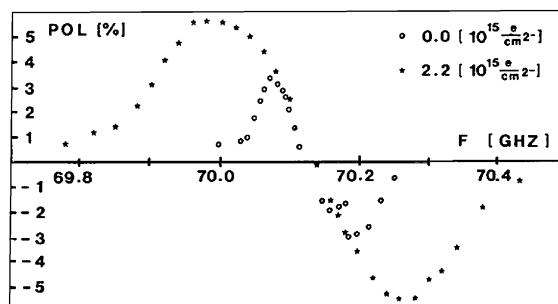


Fig. 8. Enlargement of the frequency curve and increase of the deuteron polarization during the initial phase of an irradiation at low temperatures.

Therefore, to obtain an optimum deuteron polarization the microwave frequency has always to be adjusted during the particle physics experiment. This is facilitated due to a shortening of the polarization build-up time as further radicals are created. Good annealing results in ND_3 are also achieved at temperatures around liquid N_2 .

5. Radicals in irradiated ammonia

The first visible result of an irradiation of ammonia is the violet color of the material. The intensity of this color depends on the irradiation dose and is a scale for the radical density.

Electron spin resonance (ESR) measurements performed with high-temperature-irradiated $^{14}\text{NH}_3$, $^{14}\text{ND}_3$ and $^{15}\text{ND}_3$ have indicated that the radicals $^{14}\dot{\text{N}}\text{H}_2$, $^{14}\dot{\text{N}}\text{D}_2$ and $^{15}\dot{\text{N}}\text{D}_2$, respectively, are responsible for the DNP in ammonia [12,17]. The differential ESR-absorption spectra are shown in Fig. 9, obtained with a conventional spectrometer operating in the X-band, i.e. at a frequency of ≈ 10 GHz corresponding to a magnetic field of some 350 mT. The samples are cooled at liquid N_2 temperatures. The broadening of the ESR-lines is due to hyperfine interactions of the radicals with the surrounding hydrogen (deuteron) and nitrogen (^{14}N and ^{15}N resp.) nuclei. The corresponding integrated spectra are also shown in Fig. 9. More ESR-studies are reported in Ref. [24], where also some stable $\dot{\text{H}}$ -radicals in the ammonia matrix are measured.

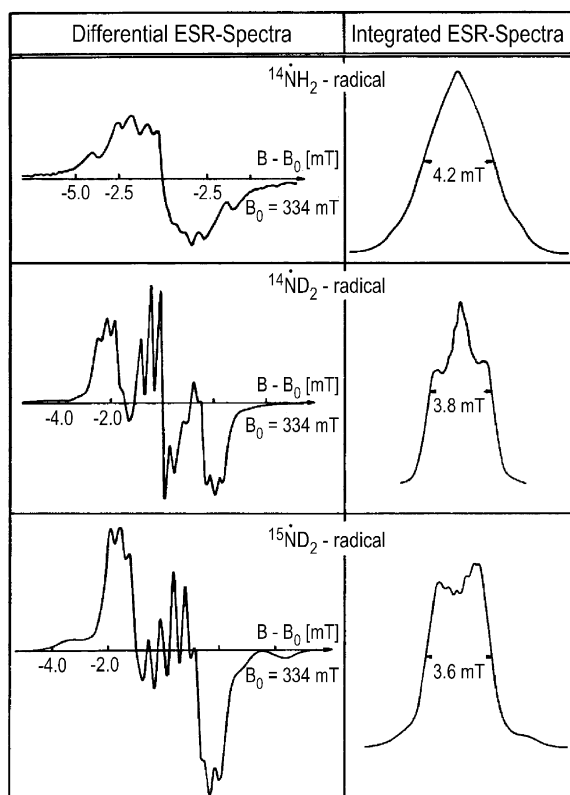


Fig. 9. The differential and integrated ESR-absorption spectra of $^{14}\text{NH}_2$, $^{14}\text{ND}_2$ and $^{15}\text{ND}_2$, taken at liquid N_2 temperatures with an X-band ESR-spectrometer.

At room temperature radicals have only a volatile existence because they react very rapidly either with themselves or with other molecules. However, at liquid N_2 temperatures they are more or less stable. After some weeks the $^{14}\text{NH}_3$ material, kept under liquid N_2 , became paler, thus indicating a loss of radicals. In $^{14}\text{ND}_3$ this effect could not be observed as clearly as in $^{14}\text{NH}_3$. The amine radical $^{14}\text{NH}_2$ completely disappears at temperatures higher than 117 K. The complete loss of the violet color in ND_3 occurs at 134 K. This loss of radicals was checked by measuring the nucleon spin relaxation time T_{1P} or the polarization build-up time T_{07} . To my knowledge, however, it was never tried to polarize material, which shows no color after the bleaching. Also no ESR measurements were performed with low-tempera-

ture-irradiated material, which in the optimal case requires a high field ESR apparatus [25].

6. DNP mechanisms in ammonia

For a precise determination of the cross-section asymmetries in the particle physics experiments, an exact knowledge of the polarization degree of all nuclei, which are present in the target sample, is crucial. In the case of ammonia there are polarized background nuclei ^{14}N (spin-1) or ^{15}N (spin- $\frac{1}{2}$) in $^{14}\text{NH}_3$, $^{15}\text{NH}_3$, $^{14}\text{ND}_3$ and $^{15}\text{ND}_3$, respectively. The ^{15}N polarization is relatively easy to measure, but the ^{14}N nucleus, whose quadrupole moment is about 10 times larger than that of the deuteron, challenges the nuclear magnetic resonance (NMR) measurement. With a combination of different techniques, this problem was solved [26,27].

As pointed out by Borghini and Scheffler, different mechanisms of the DNP can be distinguished by comparing the polarizations of two (or more) kinds of nuclei in the material [28]. In the DNP process of the chemically doped alcohols or diols the equal spin temperature (EST) hypothesis is valid, i.e. all spin species in the material are at the same spin temperature so that the individual polarizations depend only on the individual magnetic moments.

Early activities of the Bonn–Liverpool group [29,30] gave indications that the EST-model works also for high-temperature-irradiated $^{14}\text{NH}_3$ and $^{15}\text{NH}_3$, respectively. However, with the ability to perform more precise nitrogen polarization measurements, it was demonstrated that at proton polarizations higher than 50%, the polarizations of ^{15}N [31,23] and ^{14}N [27] do not follow the EST prediction precisely. An example for this behavior is shown in Fig. 10.

From the comparison of the polarization values of the unsubstituted protons (1%) in $^{14}\text{ND}_3$, and $^{15}\text{ND}_3$, and the deuterons, it is evident that there is no equal spin temperature between these species [29,23]. The plots of the ^{15}N polarization in $^{15}\text{ND}_3$ versus the unsubstituted proton (1%) and deuteron polarization, respectively, show that in neither case the EST model is applicable in this material. This is in contrast to the results from the Liverpool

group obtained in $^{14}\text{ND}_3$ [30]. It should be mentioned, that there is a significant difference depending on whether the microwaves are on or off [23]. As an example this behavior is shown in Fig. 11.

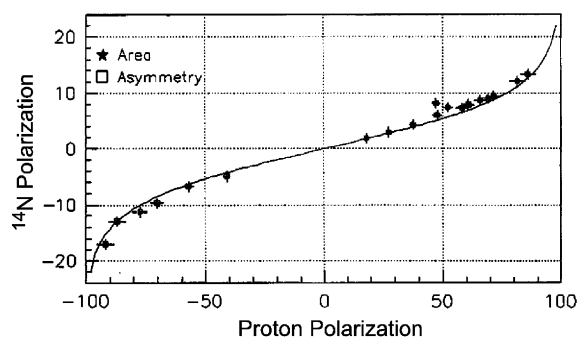


Fig. 10. The figure shows the relation between the proton and the ^{14}N polarization in $^{14}\text{NH}_3$. The solid line corresponds to the EST prediction and the data points were measured with two different methods. They agree within the errorbars.

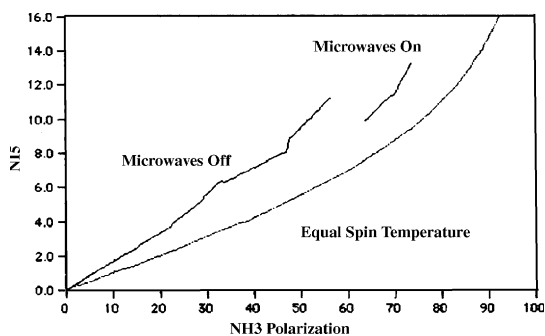


Fig. 11. Measured ^{15}N polarization versus proton polarization for microwaves on and off and the EST relation (from Ref. [23]).

Work in Bonn [9,17] has been done to understand the DNP mechanisms in high-temperature-irradiated $^{14}\text{ND}_3$. These results indicate that the differential solid-state effect can be considered as a possible DNP mechanism. However, the situation will change completely in the course of the particle physics experiments, where new radicals are created, which certainly influence the DNP conditions (see Fig. 8). Detailed studies to understand the DNP mechanisms in irradiated ammonia have still to be done.

7. DNP in hydrogen-rich materials other than pure ammonia

Motivated by the very good polarization results in irradiated ammonia DNP was studied in a variety of other materials with a high content of polarizable protons [15]. These materials were doped either by irradiation (see Table 2) or by the chemical doping agent EHBA (2-ethyl-2-hydroxybutyric acid) [32].

Concentration was put on glasses based on amines and ammonia with a high hydrogen content of 16.0–18.6 wt%. Their successful doping by chemical agents is possible by the developments of Krumpolc and Rozek in the late 1970s [33]. EHBA, as well as BHHA (2-butyl-hydroxyhexanoic acid), and HMBA (2-hydroxy-2-methylbutyric acid) are stable powders at room temperature, soluble in many polar solvents (water, alcohols, glycol) and their solutions undergo negligible decomposition during the preparation time of the

Table 2

Materials studied at SLAC with low-temperature irradiation (from Ref. [15]).

Material	Formula	H or D fraction	Studied at doses up to	Maximum polarization
Ammonia	NH_3	0.175	$4 \times 10^{15} \text{e}^-/\text{cm}^2$	75%
Deuterated ammonia	ND_3	0.175	5×10^{15}	25
Borane ammonia	BH_3NH_3	0.194	1×10^{15}	30
Ammonium hydroxide	NH_4OH	0.143	5×10^{13}	Not measured
Butanol/5% water	$\text{C}_4\text{H}_9\text{OH}$	0.135	8×10^{14}	21
Ethane	C_2H_6	0.200	6×10^{15}	10
Lithium borohydride	LiBH_4	0.183	1×10^{15}	10
Amino methane	CH_3NH_2	0.161	3×10^{14}	4

target materials. They replace previously used glycol-Cr(V) solutions formed by a chemical reduction of potassium dichromate dissolved in glycol.

However, these substances were no longer considered as target materials for the following reasons:

1. the radiation resistance as measured with ethylamine (85 wt%) + borane-ammonia (15 wt%) + EHBA-Cr(V) = $C_2NH_7BH_3NH_3$ ($f = 0.165$) is only comparable to that of butanol + porphyrexide samples i.e. more than a factor of 10 worse compared to that of irradiated ammonia [34];
2. radiation doping of borane-ammonia (BH_3NH_3) with $f = 0.196$ and of ammonium borohydride (NH_4BH_4) with $f = 0.246$ results in low polarization values [15,35]; and
3. there are more problems with the spins of the spectator nuclei such as ^{10}B (spin 3) and ^{11}B (spin $\frac{3}{2}$) in addition to ^{14}N (spin 1) (see Section 6).

8. First particle physics experiments with ammonia

In 1982 the first particle physics experiment, which profited from the good polarization resistance against ionizing radiation and from the high proton polarizations obtained with high-temperature-irradiated $^{14}NH_3$, was performed at the Brookhaven AGS in proton–proton elastic scattering [36,37]. In 1983 high-temperature-irradiated ND_3 was used in a photon beam of the Bonn electron synchrotron for the first time, to study the photodisintegration reaction $\gamma d \rightarrow pn$ by a target asymmetry measurement [17]. At that time ND_3 was also used in Bonn as a tensor polarized deuteron target in an elastic electron deuteron scattering experiment [38].

In 1988, the European Muon Collaboration (EMC) experiment at CERN told the world of physics that little of the spin of the proton is carried by the spin of the quarks [39]. These deep inelastic polarized muon polarized proton scattering results were obtained with 3 of $^{14}NH_3$, high-temperature-irradiated in Bonn by the Bonn–CERN–Liverpool activities [40]. A series of deep inelastic polarized proton/deuteron scattering ex-

periments at SLAC followed, for which the target materials $^{15}NH_3$ and $^{15}ND_3$ were essential [23]. Parallel to the SLAC measurements, the deep inelastic scattering data at CERN were improved by the Spin Muon Collaboration (SMC) using $^{14}NH_3$ as target material, too [41].

In these experiments proton polarization values above 90% at 5 T/1 K or 2.5 T/0.3 K have been routinely achieved, whereas the deuteron polarization has reached values around 40% after an additional in situ irradiation at 1 K during the course of the experiments. The latest results are given in Ref. [42].

Finally it should be mentioned, that some of these polarization results were achieved with frequency modulation of the microwaves [23,41].

9. Conclusion

After 5 years (1979–1984) of intense studies the high-temperature-irradiation method to prepare ammonia for the DNP was found to be the most effective and most practical one. $^{14}NH_3$, $^{15}NH_3$, $^{14}ND_3$ and $^{15}ND_3$ are used in new particle physics experiments, which up to that time suffered either from the lower quality factor or from the modest polarization resistance against radiation damage of the existing alcohol- and diol target materials. In the meantime, the development of irradiated 6LiD with its extremely good polarization hardness against ionizing radiation (almost one order of magnitude higher than that of ammonia) and its superior quality factor ($f \approx 0.5$) has replaced ND_3 as a polarized deuteron or neutron target, respectively [43,44]. However, in particle physics experiments with low intensity beams, where in addition the polarized background nuclei are problematical, the ‘old-fashioned’ alcohol targets like D-butanol or D-propanediol doped with trityl radicals or by irradiation are an excellent alternative for a polarized deuteron or neutron target [45].

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