Polarized H, D and $^3\text{He}$ Targets for Particle Physics Experiments

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Abstract

The development, in the early 1960s, of the dynamic nuclear polarization scheme in solid diamagnetic materials, doped with paramagnetic radicals, led to the use of solid polarized H and D targets in numerous nuclear and particle physics experiments. In the 1980s this well established technology was supplemented by the developments of polarized H, D and $^3\text{He}$ gas targets, fed by atomic beams or by optically pumped gas. Since then steady progress has been made in all contributing sub-systems so that proton polarization values around 90% and deuteron as well as $^3\text{He}$ polarization values between 40% and 85% have been routinely achieved in various set-ups. These polarization values have been measured with a relative accuracy of ±5% or better using nuclear magnetic resonance techniques and other improved polarimetry methods. Many experiments with reasonably high luminosities have taken advantage of these developments and many more are being planned, especially with electromagnetic probes.

1 Introduction

A detailed understanding of the structure and interaction of hadrons has been the main motivation for strong interaction physics for many decades. The advent of solid polarized targets in the early 1960s opened up a new era in the study of particle interactions. The first experiment with a polarized target was done at Saclay [1], at low energy, and it was soon followed by an experiment at the Lawrence Radiation Laboratory Berkeley [2] and a rapid expansion in the use of such targets at most laboratories in the world. The availability of polarized proton and, then, deuteron (neutron) targets, whose spin could be oriented in any direction, allowed the detailed study of hadron-hadron-interactions. In interactions with spinless particles, such as $\pi$- and $K$-mesons, the spin-sensitive part could be extracted and used to discriminate among various theories. Together with polarized nucleon beams it became possible to study extensively the nucleon-nucleon interaction. The motivation of this research program was to probe and extend our knowledge of specific details of spin dependencies of various interactions and to check the theoretical models invoked to explain them.

At the same time, polarized proton and deuteron (neutron) targets were used to measure (double) polarization observables with real (polarized) photon beams to improve the knowledge about the electromagnetic properties of baryon resonances.

Meanwhile quantum chromodynamics (QCD) has led to a theoretical description of the strong interaction in terms of the fundamental constituents, quarks and gluons. In the high-energy limit the internal structure of the nucleon can be well described by perturbative quantum chromodynamics (pQCD) due to the weak coupling in this energy regime. Deep inelastic lepton-nucleon scattering, together with the $e^+e^-$ collision experiments, is well established as a powerful tool for the investigation of nucleon structure [3]. The measured structure functions have been successfully interpreted in terms

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of parton distributions showing that quarks and QCD are on firm footing. The Standard Model tells that the valence quarks and the sea quarks are spin-\(\frac{1}{2}\) objects bound by gluons which are spin-1 objects. The presence of constituents with spin inside the hadrons has moved the question of spin phenomena to the forefront of high energy physics experiments.

At the Stanford Linear Accelerator Center (SLAC) the development of a polarized electron beam meant that, in the mid 1970s, an attempt could be made to use deep inelastic scattering (DIS) to measure the spin content of the proton carried by quarks [4, 5]. After many considerable technical achievements in polarizable solid target materials, refrigerators, magnets and associated equipment, this type of experiment was continued at CERN in the mid 1980s with polarized muons. In 1988, this European Muon Collaboration (EMC) experiment told the world of physics that little of the spin of the proton is carried by the quark spins [6].

In the meantime electron beam polarization technology had also made rapid progress with optical pumping of strained GaAs photocathodes [7, 8] and with the Sokolov-Ternov effect [9] which yielded electron (positron) polarization in DESY’s hadron electron ring accelerator HERA [10]. The EMC result, together with new polarized target technologies led to a succession of polarized lepton-polarized nucleon DIS experiments taking a much closer look at what happens inside nucleons. Up to now three types of experiments were performed:

- At CERN with polarized muons and polarized solid H and D targets (SMC).
- At SLAC with polarized electrons and polarized solid H and D targets and polarized gaseous external \(^3\)He targets (E142, E143, E154, E155, E155x).
- At DESY with polarized positrons and polarized gaseous internal H, D and \(^3\)He targets (HERMES).

The CERN, SLAC and DESY experiments — each with its own specific experimental power — have confirmed, with greatly improved precision, the original EMC result that quarks alone could not be responsible for the spin of the nucleon. In addition, the SMC and in particular the HERMES collaboration measured precisely the contributions to the nucleon spin of each valence quark flavour. Recently the HERMES experiment at DESY with its polarized internal nuclei targets and the COMPASS experiment at CERN, which uses like the original EMC experiment polarized muons and polarized solid targets, have started to investigate the gluon spin contribution to the nucleon spin. In addition the SLAC experiment E161 with a polarized solid target is proposed to look at the gluon spin. Together with the polarized beam experiments at the RHIC collider at the Brookhaven laboratory a full understanding of how spin \(\frac{1}{2}\) quarks and spin 1 gluons play one’s part to give nucleons a spin of \(\frac{3}{2}\) cannot be far off.

However, up to now the interaction of hadrons at lower energies cannot be described by perturbative calculations and here the understanding of the saturation of the color forces is still incomplete. The fundamental quark-gluon degrees of freedom in QCD are replaced by so-called effective degrees of freedom of nucleons and mesons. These underlying effective field theories, which include the symmetry of QCD and describe to some extent in phenomenological models the internal structure of nucleons and mesons.

In the energy range up to several GeV, a rich program is in progress at the electron beam facilities at the University of Bonn, the University of Mainz, MIT-Bates and Thomas Jefferson National Accelerator Facility (TJNAF). In addition polarization experiments are being prepared at LEGS (Brookhaven), GRAAL (Grenoble) and SPRING8 (Osaka). From the list of proposed experiments at these facilities, it is clear that measurements involving spin degrees of freedom are becoming increasingly common, perhaps even prevalent. The common factor of all these experiments is that several ingredients contribute to the measured quantities and it is well known that spin observables often provide better discrimination separating one piece from another. Experiments are now underway that address outstanding fundamental questions in nucleon and nuclear structure. With polarized D and \(^3\)He targets the neutron polarization can be exploited to determine the elastic nucleon from factors of the neutron by spin dependent electron scattering. Measurements of the charge distribution or electric form factor \(G_E\) of the neutron have begun. This fundamental quantity, which is known with limited precision, is central...
to an understanding both nucleon and nuclear structure.

With a tensor polarized deuterium target the complete set of deuteron form factors crucial to the study of the nucleon-nucleon force is accessible by measuring the polarization dependence of elastic electron scattering from polarized deuterium. In this case the asymmetry depends sensitively on the ratio of the quadrupole to charge form factors of the deuteron. When combined with unpolarized measurements, the three form factors of the deuteron (charge, quadrupole, magnetic) can be separated.

Furthermore, electro- and photonuclear reactions are particularly clean experimental instruments to investigate the resonance region and to analyze the multipole content of the individual resonance contributions. With the advent of the new electron facilities, each with high duty-factors, new classes of experiments including polarization degrees of freedom have become possible. Especially, the recent developments in polarized solid targets have initiated the first experiment with both a circularly polarized photon beam and a polarized target to prove a sum rule derived by Gerasimov [11] and Drell and Hearn [12] in the mid 1960s.

To address a special problem by scattering experiments, first of all the given (polarized) beam characteristics e.g. stored beam, external beam, beam intensity etc. are decisive for the choice of the target type. Triggered by the many technical developments in different fields over more than three decades a variety of highly functional polarized target systems has been developed. The present day polarized targets can be divided into three types:

- Solid H and D targets for external beam experiments.
- Internal gaseous H, D and \(^{3}\text{He}\) targets for storage beam experiments.
- Gaseous \(^{3}\text{He}\) targets for external beam experiments.

Each of these target types has its own combination of highly sophisticated techniques, which result in specific advantages for the polarization experiments such as high nuclear polarization, high density, high purity of polarizable target material and reasonable polarization resistance during the experiments. All the manifold developments have made it possible to access progressively lower cross section interactions with better precision.

Recent review articles on optically pumped polarized gas targets have been written by T.E. Chupp, R.J. Holt and R.G. Milner in 1994 [13] and on polarized solid targets by D.G. Crabb and W. Meyer in 1997 [14]. In this review we limit the discussion to the light polarized nuclei H, D and \(^{3}\text{He}\) which are of greatest interest for particle physics experiments at intermediate and high energies. Furthermore we focus the description on those target technologies which are routinely used in the various experiments i.e. on the dynamically polarized solid H and D targets, on the atomic beam source H and D polarized gas target and on the polarized \(^{3}\text{He}\) gas targets. In particular we discuss such targets in the context of experiments performed with electromagnetic probes. In section 2 we start with general considerations concerning luminosities and the so-called 'figure of merit' and mention briefly the very general polarization principles. For details about the polarization mechanism of gaseous H, D and \(^{3}\text{He}\) atoms the reader is referred to the textbooks in atomic physics. The technology of the internal polarized H and D gas targets and the polarized \(^{3}\text{He}\) gas targets are summarized in section 3 and section 4, respectively. In these sections examples of presently working polarized H, D and \(^{3}\text{He}\) targets are given. The main emphasis is put on polarized solid H and D targets. In section 5 the principle of the dynamic nuclear polarization, which is the guidance for all polarized solid target material developments, is reviewed. Furthermore the equipment and techniques of producing polarized solid targets as well as their polarization measurement method are described. Examples of state-of-the-art polarized solid targets in the context of the experimental programs at various laboratories are given. For even more details we refer to the proceedings of several conferences and workshops during the last decade [15, 16, 17, 18, 19, 20].
2 General Considerations

2.1 Luminosities and Asymmetries

An important question in any experiment is the optimization of the reaction counting rate

\[ N = \mathcal{L} \frac{d\sigma}{d\Omega} \Delta\Omega \]  

(1)

where \( \frac{d\sigma}{d\Omega} \) is the cross section of interest and \( \Delta\Omega \) the detector solid angle. The luminosity

\[ \mathcal{L} = I \cdot n_t \]  

(2)

is defined as the product of beam current \( I \) (number of beam particles per second) and the areal target thickness \( n_t \) (number of target particles per cm\(^2\)).

Thus, luminosity is one of the main quality factors in determining how fast an experiment can be done. In general, the luminosities possible with polarized targets are smaller compared with typical luminosities of \( 10^{36} - 10^{37} \) cm\(^{-2}\) s\(^{-1}\) in present experiments using unpolarized targets. Limits on \( n_t \) may be imposed by the target technology, but also, in the case of storage rings, by requirements of accelerator operation. A target thickness vs. beam intensity diagram showing the regions for polarization experiments with external beams and storage ring experiments is plotted in Figure 1. The lines of equal luminosity are indicated. The limitations in the luminosities for the different target types are discussed later. Targets operated in storage rings have to be very thin (\( \leq 10^{15} \) atoms per cm\(^2\)) to guarantee a sufficiently long lifetime of the stored beam. Internal polarized H targets typically have densities \( n_t \).
on the order of $10^{13} - 10^{14}$ cm$^{-2}$ and are used with circulating electron beams in storage rings whose currents $I$ are in the range of $50 mA = 3 \cdot 10^{17} e^- s^{-1}$ giving luminosities of $3 \cdot 10^{30} - 3 \cdot 10^{31}$ cm$^{-2}$ s$^{-1}$.

The external targets fall into two classes. Solid polarized H targets using ammonia as the hydrogen carrier operate with a density of about $10^{22}$ cm$^{-3}$ but at a beam current of 100 nA or less yielding a luminosity of about $6 \cdot 10^{24}$ cm$^{-2}$ s$^{-1}$. A slightly higher luminosity can be obtained with an optically pumped, polarized $^3$He gas target, compressed to some atmospheres. In this case the target thickness is smaller, about few times $10^{21}$ cm$^{-2}$, but the tolerable beam current is much higher ($\leq 30 \mu A$). The quality of a polarization experiment is, however, not only given by the reaction counting rate, but is very much dependent on other factors like background from other sources and the polarization degree of the target. In designing an experiment, one has to consider seriously these factors in addition to the luminosity.

The counting rate in experiments with a pure transverse (up↑) to the production plane polarized target is $N \approx \sigma(1 + P_t A)$ where $\sigma$ is the differential cross section, and $P_t$ is the target polarization. $A$, the asymmetry of interest, can be obtained in the case of a spin $\frac{1}{2}$ target particle from the counting rate asymmetry

$$\varepsilon = \frac{N \uparrow - N \downarrow}{N \uparrow + N \downarrow}$$

by

$$A = \frac{1}{P_t} \cdot \varepsilon$$

If the target (as seen by the beam) contains, in addition to the fraction $f_A$ of polarized nuclei, a fraction ($1-f_A$) of unpolarized material, the asymmetry $A$ has to be corrected by the 'dilution factor' $f$ i.e.

$$f = \frac{f_A \sigma}{(1-f_A) \sigma_0 + f_A \sigma}$$

where $\sigma_0$ is the average cross section for the unpolarized atoms. Then we get

$$A = \frac{1}{f} \cdot \frac{1}{P_t} \cdot \varepsilon$$

Assuming the measured asymmetry $A$ to be small, the absolute error in $A$ is obtained from

$$(\Delta A)^2 = \left( \frac{1}{f} \cdot \frac{1}{P_t} \cdot \Delta \varepsilon \right)^2 + \left( \frac{A \Delta P_t}{P_t} \right)^2 + \left( \frac{A \Delta f}{f} \right)^2$$

where

$$\Delta \varepsilon = (N \uparrow + N \downarrow)^{-1/2}$$

From this expression it is evident that $P_t$ and $f$ should be determined as well as possible. However, as experience shows the first term of equation (7) is the most dominant one.

Since the number of counts $N = N \uparrow + N \downarrow$ is proportional to both the areal target thickness $n_t$ and the (unpolarized) beam intensity $I$ as well as the running time $T$ the expression for the uncertainty in the asymmetry $A$ is

$$\Delta A \approx \frac{1}{f} \cdot \frac{1}{P_t} \cdot \sqrt{\frac{1}{n_t \cdot I \cdot T}}$$

or in terms of running time

$$T \approx \frac{1}{f^2 \cdot P_t^2 \cdot n_t \cdot I \cdot \Delta A^2} \approx \frac{1}{f^2 \cdot P_t^2 \cdot \mathcal{L} \cdot \Delta A^2}$$

A 'figure of merit' for external targets $FOM_{ext}$, can then be defined

$$FOM_{ext} = n_t \cdot P_t^2 \cdot f^2$$

i.e. the larger $n_t \cdot P_t^2 \cdot f^2$ the shorter the running time required to achieve a chosen accuracy in the asymmetry. For internal window-less gas targets (see section 3.2) an appropriate definition of the 'figure
The 'dilution factor' depends on the cross sections of the respective experimental reaction as can be seen from equation (5). For comparison of 'bare' target materials the dilution factor $f$ can be simply defined as

$$f = \frac{\text{number of polarizable nuclei}}{\text{total number of nuclei in the target material}}$$

(12)

This simple minded dilution factor is 1 for H and D gas targets, 0.33 for $^3$He and varies between 0.13 and 0.5 for solid H and D target materials.

2.2 General Principles of Nuclear Polarization

Since 1962 a variety of polarized target systems have been developed to reach high values of nuclear polarization by various methods. However there are two major steps, which are common to all of them. The first step always consists of producing an atomic polarization in the sample. Because the electron magnetic moment exceeds that of the nucleus by a huge factor the polarization of an electron system is much easier to achieve than polarizing the nuclear spin system directly. In the second step the hyperfine interaction between the atomic spin $J$ and the nuclear spin $I$ is exploited to transfer the atomic polarization to the nucleus.

The different polarization techniques are schematically listed in Figure 2 resulting in the polarized nucleus species of interest H, D and $^3$He. They are used in (polarized) particle beams as gas targets for internal experiments in storage rings or as solid H and D targets and gaseous $^3$He targets for experiments with external beams, respectively.

Polarized solid targets have to rely on low temperatures $T (\leq 1 \text{ K})$ and high magnetic fields $B (\geq 2.5 T)$. The Boltzmann factor, $e^{-\mu_J B/kT}$, strongly favors the population of the atomic substate $m_J$ with the lowest magnetic energy, resulting in almost 100% polarization of paramagnetic radicals, which are embedded in diamagnetic solid samples. Dynamic nuclear polarization (DNP) by microwave irradiation is then used to select the wanted nuclear substates $m_I$.

Polarized H and D gas targets are routinely obtained by atomic beam source (ABS) technique. In a Stern-Gerlach setup e.g. the atomic beam traverses a sextupole magnet with a field gradient $dB/dz$ in such a way that the magnetic force perpendicular to the beam separates spatially the different magnetic substates of the atomic spin $J$. Rf frequencies in weak fields can be tuned to transitions between two specific hyperfine states characterized by the total spin $F$ and substate $m_F$. The strong field gradient of a sextupole can then select those substates with a specific $m_J$ and the wanted nuclear polarization $m_I$.

For polarized $^3$He targets the first step is optical pumping using circularly polarized photons from a laser system. The repeated resonant absorption of $|\Delta m_J| = 1$ photons can result in a nearly complete atomic polarization of a mediating species like rubidium. The atomic polarization can be transferred to $^3$He during $^3$He-Rb collisions or metastability-exchange collisions with excited-state $^3$He atoms leading to a nuclear polarization of the ground state $^3$He atoms.
2.3 Maximum Polarization and Polarization Measurement

The orientation of a system of spin $I$ along an axis $z$ can be described by so-called orientation parameters, which are defined as expectation values of irreducible spin tensors [21]. Throughout this review we consider only the vector polarization $P_z$, which is defined as

$$P_z = \langle I_z \rangle / I$$

and the tensor polarization $P_{zz}$, defined as

$$P_{zz} = \langle 3I_z^2 - I(I + 1) \rangle / I^2$$

For spin $I = \frac{1}{2}$ the tensor polarization is always zero and the orientation coincides with the (vector) polarization.

The degree of the polarization of an assembly of nuclei with $I = \frac{1}{2}$ is given by

$$P_z = \frac{N_{+\frac{1}{2}} - N_{-\frac{1}{2}}}{N_{+\frac{1}{2}} + N_{-\frac{1}{2}}}$$

and the vector polarization and tensor polarization of an $I = 1$ spin assembly by

$$P_z = \frac{(N_{+1} - N_0) + (N_0 - N_{-1})}{N_{+1} + N_0 + N_{-1}} = \frac{N_{+1} - N_{-1}}{N_{+1} + N_0 + N_{-1}}$$

If an assignment of the polarization to the spin species is needed the subscript of $P_z$ is substituted for $P_z$ = electron polarization, $P_t$ = nuclear polarization, $P_p$ = proton polarization, $P_n$ = neutron polarization, $P_D$ = deuteron polarization, $P_{\alpha\text{He}} = ^{3}\text{He}$ polarization or more generally $P_i$ = target polarization, $P_{\frac{1}{2}}$ = polarization of spin $\frac{1}{2}$ particles and $P_1$ = polarization of spin 1 particles.
and

\[ P_{zz} = \frac{(N+1 - N_0) - (N_0 - N_{-1})}{N_{+1} + N_0 + N_{-1}} \]

Using the normalization \( N_{+1} + N_0 + N_{-1} = 1 \) the tensor polarization can be written as \( P_{zz} = 1 - 3N_0 \).

\( N_{+\frac{1}{2}}, N_{-\frac{1}{2}} \) and \( N_{+1}, N_0, N_{-1} \) are the relative populations of the various spin projections on the quantization axis \( z \), normally chosen along the magnetic field direction.

The maximum value of the nuclear polarization which can be obtained is a competition of the polarization rate and the nuclear spin relaxation rate \( \Gamma_{rel} = T_1^{-1} \). In the case of polarized solid targets the polarization rate i.e. the polarization build-up time \( \tau \) combined with a high polarization, depends strongly on the electron (paramagnetic radical) density. It is well known that the nuclear spin relaxation proceeds mainly via the interaction with the electron spin system [22]. It was shown that the main cause for the nuclear spin relaxation in solids with paramagnetic impurities is the fluctuating magnetic field at the nucleus created by the transitions with the electron spin system. The electron spin relaxation as well as the nuclear spin relaxation rate is strongly dependent on the magnetic field strength and the target temperature, which imposes limits for the heat input by the microwave induced dynamic polarization process (DNP) and by the beam current. For \(^3\)He gas targets, where the polarization build-up is achieved by spin-exchange or metastability-exchange, the total rate at which polarized \(^3\)He is obtained is determined by the available laser power and by the efficiency for the transfer of the photon angular momentum to the \(^3\)He nuclei.

For all target types the maximum polarization critically depends on achieving long nuclear spin relaxation times \( T_1 \) in the target samples, which are kept in metallic or glass containers during the interaction with the beam particles. Careful sample preparation to eliminate harmful impurities is generally crucial. To avoid rapid depolarization in gaseous targets due to collisions of the atoms with the cell walls it is necessary to coat the metallic walls of the storage cell with a suitable substance, e.g. drifilm or Cs in the case of the glass walls [23].

A special problem for internal gas targets arises in electron storage rings where the beam bunches cause an intense time-varying magnetic field which results in a depolarization of the target nuclei. This problem can be overcome by applying a sufficiently strong magnetic holding field in the direction of the nuclear polarization. In external electron beam experiments with polarized \(^3\)He, a potentially serious source of relaxation is the formation of \(^3\)He\(^2\) molecules, which can be quenched by admixing a little amount of nitrogen to the compressed \(^3\)He. In solid targets the creation of additional paramagnetic radicals in the sample by the beam leads to depolarization and limits the lifetime of the samples. This problem is significantly reduced by the use of solid target materials, which are prepared for the DNP-process by irradiation (see section 5.6).

Finally, a measurement of the absolute value of the nuclear polarization in the target with high precision is necessary. In solid targets the measurement is typically done by the nuclear magnetic resonance (NMR) method. Prior to the polarization experiments the polarization is calibrated by so-called thermal equilibrium (TE) signals at a given temperature and magnetic field. The corresponding rf-frequency is than also used to monitor online the dynamically enhanced polarization signals during the experiments. For details see section 5.4.

In high density \(^3\)He targets the polarization is measured and reversed by Adiabatic Fast Passage (AFP) NMR. Here the polarization can be calibrated by measuring, at the same rf-frequency, signals from protons in a \( \text{H}_2\text{O} \) sample at room temperature in a cell of identical dimensions (see section 4.5.2). For less dense \(^3\)He targets or in case of metal target cells optical polarization measurement techniques are used (see section 4.3). In atomic beam set-ups the hyperfine states of a sample of the target gas can be analyzed by a combination of deflections in the inhomogeneous field of a sextupole magnet and by inducing rf-transitions (Breit-Rabi polarimeter). Here the method of the polarization analysis is essentially the same as that of producing the polarization.

The polarization value entering in the asymmetry calculation should be obtained from measurements which ideally sample nuclei from the entire beam interaction region. Normally this can only be achieved in special cases, where a reaction asymmetry, determined previously using an unpolarized target, can be used to measure the polarization of nuclei as seen by the beam.
3 Polarized H and D Gas Targets

3.1 Internal Gas Targets

Polarized internal gas targets to be used in storage ring experiments have to fulfill a number of requirements. They should be 'thick' enough to give sufficient counting rate, and at the same time 'thin' enough not to degrade the circulating beam too rapidly. The disturbance of the ring vacuum should be kept at a minimum and for the detector setups, a large solid angle should be available around the target.

They have several unique features:
- The polarized nuclei are present as pure atomic species.
- The open storage cells with no windows brings the dilution factor very close to 1.
- The sign of the target polarization can be reversed rapidly.
- Only a relatively low magnetic field at the target place is needed.
- The storage cell wall may be thin allowing the detection of heavily ionizing recoil particles from the reactions under study.

The use of polarized atomic beams as internal targets in storage rings (so-called polarized jet targets) has been considered for a long time. But the density achieved in polarized jets is too low for most applications. The idea to increase the target density compared to that obtained with polarized jet targets by injecting the atomic beam into a cell was already reported in 1980 [24, 25]. The polarized internal target technique using a storage cell for deuterium gas was first demonstrated at the VEPP-3 electron storage ring at Novosibirsk [26]. First experiments with a polarized hydrogen gas storage cell were performed at the TSR low energy ion storage ring at Heidelberg [27].

3.2 Storage Cell Target

The development of the storage cell technique was decisive for the success of new polarization experiments. Internal polarized gas targets for present day experiments consist of a source that generates a flow of polarized atoms directed into a windowless, conductance-limiting storage cell through which the circulating particle beam passes. A schematic drawing is shown in Figure 3.

![Figure 3: Generalized storage cell.](image)

The cell has an opening for the injection of the atomic beam, and apertures for entry and exit of the circulating particle beam. Rather than passing through the circulating particle beam only once, as the atoms do in a jet target, they undergo several hundred collisions with the wall of the cell before leaking out. Thus a much improved economy of polarized atoms, i.e. a much increased target thickness is obtained.

The density of the atoms in the storage cell is given by the flow $F$ (number of atoms injected per second) divided by the combined gas conductance $C$ (volume per second) of the openings of the cell:

$$\rho = \frac{F}{C}$$

(18)
The target thickness or target area density \( n_t \) is the product of the density \( \rho \) times the length \( l \) of the target cell along the circulating particle beam:

\[
 n_t = \rho \cdot l \quad (19)
\]

The openings of the cell, and thus the conductance, depend on the circulating particle beam size and, in the case of atomic beam source H and D gas targets, on the atomic beam size. The only other parameter available to affect the gas conductance is the wall temperature \( T_c \) of the storage cell. Cooling the cell increases the target thickness since the gas conductance decreases in proportion to the velocity of the atoms in the cell. For a given flow the target thickness is proportional to \( l/\sqrt{T_c \cdot d^3} \), where \( d \) is the diameter of the target cell.

Meanwhile a gain in the target thickness by a factor of approximately one hundred compared with the free atomic beam targets has been reached \[24\] by tailoring target cells to the beam dimensions. This gain has to be paid for by the narrow constrictions which may cause background problems. To reduce this problem, a so-called active storage cell was tested \[28\]. It is open while injecting electrons into the storage ring and is closed when the beam is cooled, maximizing the target thickness during data acquisition.

Further requirements include thin cell walls to minimize multiple scattering of the scattered particles, resistance to the radiation environment of the storage ring and compatibility with the high vacuum requirements of the ring. Special attention has also to be put on the cell wall coating and cooling to keep recombination and depolarization at low values. Some details about depolarizing effects, polarimetry and actually obtained polarization values are given in Sections 3.3.1, 3.3.2, 4.2, 4.4.1, 4.5.1 and 4.5.2.

### 3.2.1 Atomic Beam Source H and D Gas Targets

The Stern-Gerlach type Atomic Beam Source (ABS) for producing polarized protons or deuterons consist of a dissociator, a sextupole magnet for the selection of one electronic \( m_I \) state, a rf-transition unit in a weak field, followed by another sextupole magnet which selects only the wanted hyperfine component. The hyperfine structure (hfs) diagrams of hydrogen and deuterium are shown in Figure 4.

\[
\begin{align*}
\text{HYDROGEN} & : m_I = \pm \frac{1}{2} \\
\text{DEUTERIUM} & : m_I = \pm \frac{1}{2}
\end{align*}
\]

The energies are given in units of \( \Delta W = 1420.4 \, MHz \) and \( 327.4 \, MHz \), respectively, and the magnetic fields in units of \( B_c = 50.7 \, mT \) and \( 11.7 \, mT \). From Ref. \[29\]

In the case of hydrogen we have a simple atomic system with both nuclear and electronic spin equal to \( \frac{1}{2} \). The resulting hyperfine levels with \( F = 1 \) and 0 are split up into magnetic substates by an external magnetic field. In the strong-field limit there are two groups characterized by the electron spin with \( m_I = \pm \frac{1}{2} \). The proton spins which are the ones of interest in the experiments, are here \( m_I = \pm \frac{1}{2} \) in
the two groups. The states 1 and 3 are pure in \( m_I = \pm \frac{1}{2} \) and \(-\frac{1}{2}\), with a polarization \( P_z = +1 \) and \(-1\), respectively, independent of the external magnetic field. In the mixed states 2 and 4, the polarization depends on the strength of the field, \( P_z = -1 \) and \(+1\), respectively, in strong fields, going down to 0 in weak fields. In the case where several states are occupied, the polarization is given by the weighted mean value.

Figure 5: Schematic diagram of a polarized atomic beam target system with multipole magnets and rf-transitions to produce single substates. The trajectories of hydrogen atoms in different magnetic substates, see Figure 4, are indicated. (From Ref. [29])

A polarized beam of atomic hydrogen may be obtained by state selection in inhomogeneous magnetic fields and rf-transitions between the hyperfine states (see Figure 6). Molecular hydrogen is dissociated by an rf-discharge to give a beam of atomic hydrogen. The first sextupole focuses atoms in the states 1 and 2 with \( m_J = +\frac{1}{2} \), while atoms in 3 and 4 are defocused and lost. In the central field, an rf-transition is induced between the states 2 and 4, so, at the entrance of the second sextupole we have atoms in the states 1 and 4. In this magnet, atoms in the state 4 are again defocused, and we have a remaining hydrogen beam of state 1 atoms focused to the intersection with the circulating particle beam. The proton polarization is here \( P_z = +\frac{1}{2} \) since all atoms are in the state 1 with \( m_I = +\frac{1}{2} \). An rf-transition between the states 1 and 3 directly after the second magnet provides a rapid switching between the polarizations \( P_z = +1 \) and \(-1\).

Deuterium, with a nuclear spin \( I = 1 \), has a hfs diagram similar to hydrogen (see Figure 4). The \( m_I \) components are here 1, 0 and \(-1\), and the polarization of the deuteron may be described by a vector polarization \( P_z \) and a tensor polarization \( P_{zz} \). The only pure states are 1 and 4 with \( P_z = \pm 1 \) and \( P_{zz} = 1 \). For the other mixed states, the vector and tensor polarizations depend on the strength of the external field. By state selection and rf-transitions one may obtain deuterium beams with different combinations of the deuteron vector and tensor polarizations.

The resulting polarization, measured with an atomic beam sampling polarimeter is high, but the intensity from atomic beam sources is limited to typically \( 10^{16} - 10^{17} \) atoms/s. As seen from equation (18) the flow of polarized atoms \( F \) into the storage cell entrance tube has to be optimized. Great efforts over many years have been made seeking an optimum design by studying the operating properties of the individual parts of the ABS which is shown schematically in Figure 6.

The flow \( F \) of polarized atoms out of a source and injected into a target storage cell can be generally expressed as follows [31]:

\[
F \approx F^\prime \cdot \frac{\Delta \Omega}{\Omega} \cdot A \cdot T_{\text{trans}}
\]

\( F^\prime = 2an_{\text{H}_2(D_2)} \) is the total atomic flow which is given by the product of the degree of dissociation \( \alpha \) in the nozzle beam and the total \( \text{H}_2 \) (\( D_2 \)) - flowrate \( F_{\text{H}_2(D_2)} \). \( \frac{\Delta \Omega}{\Omega} \) is the relative geometric solid angle accepted by the first magnet element, \( A \) describes the attenuation of the beam e.g. due to residual gas scattering, and \( T_{\text{trans}} \) is a transport factor which expresses how many atoms in the focussing state can be transported through the magnet system and injected into the entrance tube of a storage cell.
This description demonstrates the necessity of a highly dissociated beam emerging from the dissociator nozzle with a high phase space density and a velocity profile adapted to the transmission of the separation magnets. Detailed studies have been made in order to understand quantitatively the mechanism of the rest gas attenuation, one of the major effects limiting the output intensity of the ABSs. The measured single scattering loss cross sections allow to explain the lower output intensity obtained with sources operating at low nozzle temperatures and the general lower output obtained for deuterium operation [30]. The ABS intensity limit and new ideas for further improvements are discussed in [32].

3.2.2 Laser-Driven H and D Gas Targets

The laser-driven source (LDS) operates on the principle of optical-pumping spin-exchange where photon angular momentum is transferred to the target nuclei through spin exchange collisions with laser-pumped polarized alkali-metal atoms, typically potassium. The optical pumping spin-exchange has long
been known [33] and is reviewed in Refs. [34, 13]. A circularly polarized beam of 770.1 nm photons ($\sigma^\pm, \Delta m_J = \mp 1$), e.g. from an Ar-ion pumped Ti-sapphire laser, is used to optically pump the $4^2S_{1/2} \rightarrow 4^2P_{1/2} D1$ line of potassium in the presence of a relatively high magnetic field (see below). A schematic level diagram of a potassium atom in a high magnetic field is shown in Figure 7. The rate equations [35] for optical pumping spin-exchange in a high magnetic field (where only the electron spin polarization is important) lead to a simple relation for the H(D) atomic polarization $P_{H(D)}$ in terms of the average potassium polarization ($P_K$).

\[
P_{H(D)} = \frac{\gamma_{SE}}{\gamma_{SE} + \Gamma_R} \langle P_K \rangle
\]

where $\gamma_{SE}$ is the K-H(D) spin-exchange rate, and $\Gamma_R$ is the H(D) spin relaxation rate. The K-H(D) spin-exchange rate is given by $\gamma_{SE} = (\sigma_{SE} \nu)(K)$ where $K$ is the potassium density within the spin-exchange cell, $\sigma_{SE}$ is the K-H(D) spin-exchange cross section ($7.4 \times 10^{-15} \text{ cm}^2$) [37], and $\nu$ is the thermally averaged relative K-H(D) velocity. The spin relaxation rate depends on all polarization losses, e.g. collisions with the cell walls and loss of polarized atoms leaving the cell.

![Figure 8: Schematic picture of the various spin-transfer and spin-exchange processes in a LDS operating in spin temperature equilibrium. From Ref. [41]](image)

The major problem for producing high intensities consists in the fact that, at low magnetic field, $P_K$ is only large at very low potassium densities ($< < 10^{12} \text{ atoms/cm}^3$), thus limiting the spin-exchange rate to values $\gamma_{SE} << \Gamma_R$. At high potassium concentrations, necessary to increase $\gamma_{SE}$ sufficiently, $P_K$ is substantially reduced by radiation trapping [38] i.e. the re-absorption of fluorescent photons ($\pi, \Delta m_J = 0$) de-exciting the excited state of potassium.

At high magnetic fields radiation trapping is known to occur at higher alkali densities ($< < 10^{13} \text{ atoms/cm}^3$) [39] because of the increased energy spacing between the $\sigma$ and $\pi$ light absorption due to Zeeman splitting. Thus, by applying a high field, high alkali polarization can be maintained at higher
alkali density corresponding to a higher spin-exchange rate. This has been demonstrated using magnetic fields up to 440 mT [40].

A more sophisticated model has been proposed that considers effects of the nuclear spin and the applied magnetic field on polarized H(D) production in an optical pumping spin-exchange apparatus [42]. The model suggests that, although high magnetic fields generally weaken the influence of the hyperfine coupling between the nucleus and the electron, frequent H-H(D-D) collisions increase the total probability for a hyperfine interaction to occur. Under such a condition, atoms within the optical pumping spin-exchange apparatus evolve to an equilibrium ratio of the electron and nuclear polarization (so-called spin temperature equilibrium) which results in a direct polarization of the H(D) nuclei without actively performing rf-ground state transitions. It has been shown that at sufficient high flow rates (> 3 \times 10^{17} \text{ atoms/s}) the Argonne LDS [40] and the Erlangen LDS [43] fulfill the condition of a spin temperature equilibrium for H(D). The LDS operation scheme running in spin temperature equilibrium is pictorially shown in Figure 8.

3.3 Realization of H and D Gas Targets

Scattering experiments in which pure and highly polarized gas targets are used in particle storage rings have many advantages (see Section 3.1). Nevertheless this target technique provides a challenge for attaining sufficient luminosities, especially when polarized hydrogen or deuterium is used. Furthermore, there exist many mechanisms that can depolarize the target atoms. Polarization losses can originate from the interaction of the electron spin with external fields associated with the stored particle beam and/or the presence of container walls. In the last decade significant effort has been made to produce intense atomic beams of hydrogen and deuterium [44, 45, 46] and to develop suitable wall coatings [47] to preserve the nuclear polarization of these atoms. Additionally several polarimetry schemes have been developed for internal target applications [48, 49, 50, 51] that can precisely measure the nuclear polarization of the target species. For polarized hydrogen and deuterium, mainly ABSs are used to inject atoms into the feed tube of a storage cell and several internal target experiments have been carried out at BINP [26], IUCF [52], NIKHEF [53] and DESY [54].

3.3.1 Atomic Beam Source Targets

The most recent internal gas target for polarized hydrogen and deuterium injected from an ABS into a tubular open-ended storage cell is the HERMES target operated in the HERA positron storage ring at DESY. The HERMES experiment is designed to study the spin structure of the nucleon in inclusive and semi-inclusive deep inelastic scattering [55]. The storage cell is a 29 x 9.8 mm² elliptical tube 400 mm long made from 75 μm thick aluminum. It confines the target gas near the high energy positron beam thus increasing the target thickness. By cooling the target cell to 100 K, an areal density of 7 \times 10^{13} hydrogen atoms/cm² can be achieved. The cell walls are coated with drifilm [56] to minimize the wall collision effects. Recombination is further suppressed by water absorbed on the cell surface [57, 58] as demonstrated during the target cell operation with deuterium [59]. The magnetic field throughout the cell is directed parallel to the positron beam to define the quantization axis and its value of 335 mT is chosen to avoid resonant depolarization of the protons by the pulsed magnetic field caused by the bunch structure of the HERA beam. The spin direction in the target can be reversed in less than one second by selecting different spin states in the ABS. A schematic layout of the HERMES target is shown in Figure 9. The atomic beam is injected into the storage cell via a side tube connected to the centre. A sample of the gas from the middle of the storage cell diffuses into a Breit-Rabi polarimeter (BRP) [51] which measures the nuclear and electron polarizations of the atoms. The atomic fraction is measured with a target gas analyzer (TGA), consisting of a beam chopper and a quadrupole mass spectrometer (QMS). The target polarization \( P_t \) as seen by the positron beam is given by

\[
P_t = \alpha_0 [\alpha_r + (1 - \alpha_r) \beta] P_{\text{atom}}
\]

\( \alpha_0 \) is the fraction of atoms with proton polarization \( P_{\text{atom}} \) injected into the cell. The atomic fraction accounts for the presence of the small number of molecules originating from the ABS and the residual
gas in the vacuum system. $\alpha_\ell$ is the atomic fraction surviving recombination. The values of $P_{\text{atom}}$ and $\alpha_\ell$ were corrected for the sampling efficiencies of the BRP and TGA. The quantity $\beta$ is defined as the ratio of the polarization of protons in molecules from recombination to the polarization of protons in the atoms. It has been demonstrated that about half of the proton polarization is retained by molecules after recombination [60]. A plot of the proton polarization $P_t$ versus time, measured with the polarimeter during the 1997 run period of HERMES is shown in Figure 10. After the corrections mentioned above the resulting target polarization $P_t$ was $(88 \pm 4)\%$.

The very good HERMES internal target performance for polarized protons could even be improved for polarized deuterium. The target thickness was almost doubled compared to the proton target operation and a thickness of $2.1 \cdot 10^{14}$ deuterium atoms/cm$^2$ was obtained. This could mainly be achieved by a reduction of the internal storage cell size and by lowering the cell temperature from $100 \, K$ to about $70 \, K$.

Using the general equation (22) for the determination of the target polarization, values for the deuteron vector polarization of $P_1 = (+85.1 \pm 3.1)\%$ and $P_1 = (-84.0 \pm 2.8)\%$ have been measured after negligible corrections for recombination and depolarization [59]. Detailed information about the HERMES polarized internal target and its polarimetry can be found in Refs. [58, 51].

Elastic electron scattering from tensor polarized internal D targets were performed at Novosibirsk (BINP) [26] and later at the NIKHEF electron storage ring [53]. The measured observable $T_{20}$, when combined with cross section measurements, allows the determination of the charge, quadrupole and magnetic form factor of the deuteron. A challenging task for all internal targets is to measure and monitor the average target polarization in the interaction region. At NIKHEF this was done by combining the results of several polarimeters [61]. In particular, an electrostatic ion extraction system and Wien filter were used to measure online the atomic fraction of the target gas in the storage cell. This device was supplemented by a tensor polarization analyzer using the neutron anisotropy of the $^3\text{He}(d,n)^4\text{He}$ reaction at $60 \, keV$ [48]. This method allows the determination of the averaged tensor polarization of the target gas directly, independent of its spatial and temporal variations [50]. Tensor polarizations of $P_{zz} \simeq +0.5$ and $P_{zz} \simeq 1.0$ have been reported [62]. The deviation from the maximum polarization

---

**Figure 9:** The set-up of the Breit-Rabi type polarimeter (BRP) in the HERMES environment. Both the ABS and BRP axis are tilted by $30^\circ$ with respect to the horizontal plane. The chamber between BRP and storage cell contains the gas analysis system (TGA), consisting of a beam chopper and a quadrupole mass spectrometer (QMS).
values can be explained by only 80% rejection efficiency of the second sextupole [63] and losses in the cell due to wall and spin-exchange collisions.

Measurements indicate, that spin-exchange reduces the tensor polarization in a storage cell by about 10% for densities of $\approx 10^{12}$ atoms/cm$^3$, even for an external magnetic field three times larger than the critical field [61]. This behavior was also measured at IUCF, where tensor polarization was obtained without transition units (without medium field transition (MFT) $P_T^{\text{max}} = \frac{1}{3}$ and $P_T^{\text{max}} = -\frac{1}{3}$), to exclude effects from inefficient transitions [64]. It should also be mentioned, that measurements at NIKHEF have shown, that molecules produced by recombination of atoms retain most of the atomic nuclear tensor polarization in a copper cell [62].

3.3.2 Laser-Driven Targets

The fundamental component of a LDS is a one-piece glass apparatus that is composed of the three regions (see Figure 11):

![Diagram of LDS component](image)

Figure 11: The fundamental component of a LDS is a one-piece glass apparatus that is composed of a dissociator region, a spin-exchange cell and a transport tube to an internal target storage cell.
A dissociator region.

An optical pumping spin-exchange cell.

A transport tube, e.g. to an internal target storage cell.

In the dissociator region, molecular H(D) is dissociated in an rf-inductive discharge. Then the H(D) atoms pass into the optical pumping spin-exchange cell which contains potassium vapor. The ratio of H(D) atoms to potassium atoms in the spin-exchange cell is typically 100 : 1. The spin-exchange cell, the transport tube and the target cell are coated with drifilm [66] to reduce spin-relaxation and recombination of the atomic H(D) during cell wall collisions.

Electron polarization of 60% for H atoms at flowrates above $10^{18}$ atoms/s have been measured [40]. For internal target experiments it is desirable to minimize any contamination from the potassium atoms, which dilutes the target with unpolarized potassium nuclei and necessitates the heating of the target cell to prevent potassium condensation and subsequent depolarization and molecular recombination on the target cell walls.

The effective nuclear polarization of the target, $P_t$, depends on several quantities: the electron polarization of the atoms, the atomic fraction, whether the system has achieved spin temperature equilibrium and on depolarization effects i.e. from collisions with surfaces and gas and from recombination effects. Tests of the LDS for H(D) targets (see Figure 12) were made in the proton storage ring at IUCF [65, 67, 68].

![Figure 12: Tests of the LDS for H(D) targets were made in the proton storage ring at IUCF. From Ref. [65]](image-url)

The storage cell was 40 cm long and was fed with a flow rate of $\approx 1 \cdot 10^{18}$ atoms/s of H(D) nuclei, resulting in a total target thickness of $4 \cdot 10^{14}$ atoms/cm$^2$. The magnetic field in the storage cell was 70 mT at the center and 40 mT near the ends of the storage cell. The magnetic field at the bottom of the spin-exchange cell was $\approx 70$ mT and the frequency of the laser light was tuned so that atoms in this region were optically pumped. The field near the top of the spin-exchange cell was significantly smaller, $\approx 20$ mT, so that the efficiency of H(D) - H(D) collisions for transferring polarization to the nucleus was higher in that region.

The nuclear polarization in the storage cell, diluted by the atomic fraction, was determined from the measured spin observables. Nuclear polarization values of $\approx 15\%$ for H targets and $\approx 10\%$ for D targets (vector polarization) have been reported [65]. More details about the recent status of the LDS technique, which needs further studies and optimization for their use in particle physics experiments can be found in Refs. [40, 41, 69].
3.4 Summary of Polarized H and D Gas Targets

With the well established ABS technology polarized atoms in their pure spin states can be routinely fed into windowless storage cells operated in storage rings. However, at present the flow of the atoms into the target cell seems to have reached a limit of \(7 \cdot 10^{16}\) atoms/s, so that target thicknesses slight higher than \(10^{15}\) atoms/cm\(^2\) have been obtained by cooling and fitting the storage cell dimension to the particle beam size. Correcting for all depolarizing effects nuclear (vector) polarization values of close to \(\pm 90\%\) in proton and deuteron targets have been achieved and a tensor polarization of half of its maximum values. These polarization values have been determined to a relative accuracy of \(\pm 5\%\) and better with so-called Breit-Rabi polarimeters and/or using nuclear reactions with known analyzing power. The sign of the polarization can be reversed rapidly e.g. every 45 seconds. Gas targets have been operated in high intensity beams giving luminosities between \(10^{31} - 10^{32}\) cm\(^{-2}\) s\(^{-1}\).

The LDSs deliver flow rates of about \(10^{18}\) atoms/s. In principle the flow rate is only limited by the available laser power. However, nuclear polarization values of \(\approx 10\%\) demonstrate that this relatively new technology needs further improvement before reliable particle physics experiments can be started. Internal polarized H and D targets were operated in the past in Novosibirsk (BINP), Heidelberg (TSR) and Amsterdam (NIKHEF). Presently there are only two in operation for particle physics experiments: The HERMES target at DESY and the PINTEX target at IUCF and a third target will be installed at MIT-Bates.

4 Polarized \(^3\text{He}\) Gas Targets

4.1 Polarizing Methods of \(^3\text{He}\)

Polarized \(^3\text{He}\) has long been recognized as an important nuclear target [70] for studying spin dependent neutron interactions [71, 72]. The Pauli exclusion principle provides a conceptual understanding of why polarized \(^3\text{He}\) may be considered as a polarized neutron, as far as the spin is concerned. The \(^3\text{He}\) ground state wave function is predominantly \(S\) state, with small admixtures of approximately \(8\%\) \(D\) state and \(2\%\) \(S'\) state. The \(S\) state is a spatially symmetric, spin-isospin antisymmetric state in which the spins of the protons are coupled to spin 0 and the neutron carries the \(^3\text{He}\) nuclear spin. The \(S'\) state is an \(L = 0\) mixed symmetry state that arises because of the spin dependence of the nucleon-nucleon interaction. The \(D\) state is an \(L = 2\) state in which the nucleon spins are oriented predominantly opposite the orbital angular momentum. Both the \(S'\) and \(D\) state components dilute the fraction of the nuclear spin carried by the neutron. Detailed calculations of the \(^3\text{He}\) nuclear wavefunction were made [72] and found that the unpaired neutron carried about 90% of the \(^3\text{He}\) spin.

Nuclear polarization methods using low temperatures and high magnetic fields have achieved high polarizations in solid \(^3\text{He}\) [73]. But especially the low heat capacity of the solid \(^3\text{He}\) make these targets not useful for applications in particle physics experiments. The only practical methods employ laser optical pumping of gaseous \(^3\text{He}\). There are two primary methods of polarizing \(^3\text{He}\) nuclei for target applications, the metastability-exchange procedure, first demonstrated by Walters, Colegrove and Scheerar [74], and the method of collisional spin-exchange with optically pumped alkali-metal vapor introduced by Bouchiat, Carver and Varnum for the case of \(^3\text{He}\) [75], and developed more generally by Happer and coworkers [76, 77].

Both methods have been developed for the use as high density \(^3\text{He}\) targets in external beam experiments (see section 4.5) whereas the metastability-exchange pumping method is also used to feed polarized \(^3\text{He}\) in (cryogenically cooled) storage cells [78]. Common to all polarized \(^3\text{He}\) targets is that only a small guiding magnetic field of \(1 - 3\) mT is required, normally produced by a so-called Helmholtz coil configuration.

4.1.1 Metastability-Exchange Optical Pumping of \(^3\text{He}\).

In the metastability-exchange scheme a sample of \(^3\text{He}\) in a cell is excited by an electric discharge so that a fraction of the atoms (\(\approx 10^{-6}\)) is in the metastable \(2^3S_1\) state. The cell is placed in a
homogeneous magnetic field with the field direction defining the spin quantization axis. Circularly polarized light at the wavelength of the $2^3S_1 \rightarrow 2^3P_0$ atomic transition (so-called $C_8$- or $C_9$-line at 1.083\,\mu m), incident upon the $^3$He atoms, induces $\Delta m = +(-1)$ transitions for right-handed (left-handed) circularly polarized light, where $m$ is the magnetic quantum number. The $^3$He atomic energy levels relevant to the optical pumping process are shown in Figure 13.

![Figure 13: Level diagram of the $2^3S_1$ and $2^3P_{0,1,2}$ hfs multiplets of $^3$He indicating four of the nine transitions.](image)

The electronic polarization is transferred to the nucleus through hyperfine interaction. The $2^3P_0$ state decays through photon emission with equal probability to any accessible state, and repeated absorption and emission polarize the $2^3S_1$ metastable atoms.

The polarization is transferred to the ground state nuclei through metastability-exchange collisions, which exchange the excitation of the atomic electrons without altering the nuclear spins of the atoms involved. The collision process can be written schematically as

$$3^3He(2^3S_1) + ^3He(1^1S_0) \rightarrow 3^3He(1^1S_0) + ^3He(2^3S_1)$$  \hspace{1cm} (23)

so that the relevant energy levels of the $^3$He atom are those shown in Figure 14.

![Figure 14: The important energy levels in the $^3$He atom for the optical pumping process.](image)

The metastability-exchange collisions occur with a very large cross section of the order of $10^{-15}$\,cm$^2$. The metastability-exchange cross section is a strong function of the temperature, with the exchange rate decreasing approximately by two orders of magnitude between room temperature and liquid helium temperature.

The density at which the pumping is efficient is set by the requirement of a homogeneous weak discharge in the gas and a long lifetime of the metastable atoms. This corresponds to a $^3$He pressure of about 1\,mbar [79]. The short spin-exchange time constant per $^3$He ground state atom in the order of $T_{ex} \approx 0.3$\,s [80] in combination with the availability of intense pump lasers (LNA laser, $P \approx 5\,\text{Watt}$ at $\lambda = 1.083\,\mu m$ [81]) make this pumping scheme very attractive. Under optimum conditions, samples of
$^3$He gas at a density of few times $10^{16}$ atoms/cm$^3$ can be pumped to high polarizations of more than 80\% at rates of $10^{16}$ atoms/s. The circular polarization of the 668 nm $3^1 D_2 \rightarrow 2^1 P_2$ light emitted from the discharge can be used as a relative polarization monitor [82].

4.1.2 Spin-Exchange Optical Pumping of $^3$He

In contrast to the metastability-exchange method, collisional spin-exchange with optically pumped alkali-metal vapors can take place at high pressures (some atmospheres) without sacrificing polarization, but with much slower spin-exchange rates. Spin-exchange optical pumping is a two stage process which begins with optical depopulation pumping of an alkali-metal vapor, in most of the cases rubidium. The optical pumping is accomplished by illuminating Rb vapor with circularly polarized laser light tuned to the Rb $D_1$ line, which is the transition from the $5S_{1/2}$ ground state to the $5P_{1/2}$ first excited state as shown in Figure 15. The result is a spin polarization of the valence electron. Under typical optical pumping conditions with optically thick Rb vapors (a few times $10^{14}$ atoms/cm$^3$), the Rb is nearly 100\% polarized since the photon-rubidium spin-exchange time constant is $\approx 10^{-8}$s compared to a depolarizing spin-destruction decay time of $\approx 10^{-3}$s [83, 35]. To avoid radiation trapping at these high alkali densities a small amount of nitrogen (50 - 100 mbar) is present to ensure radiationless de-excitation of the $5P_{1/2}$ excited state of Rb [35]. In principle any alkali-metal vapor can be polarized in this manner, but Rb is particularly convenient due to the commercial availability of Ti-sapphire lasers, which provide several watts of cw light and are easily tunable to the 795 nm Rb $D_1$ resonance.

![Figure 15: Illustration of Rb optical pumping for incident circularly polarized ($\sigma_+$) light. Only the ground state with $m_s = -\frac{1}{2}$ is depopulated by $\sigma_+$ light. Due to collisions with the $^3$He and N$_2$ gas in the pumping cell, the $P_{1/2}$ states are mixed and the probability for repopulating each ground state is $\frac{1}{2}$. From Ref. [84]](image)

Once the Rb vapor is polarized, that polarization is transferred to the $^3$He through spin-exchange collisions. During any Rb-$^3$He binary collision there is a small probability that the wavefunction of the Rb valence electron will penetrate through the $^3$He atom's electron cloud to the $^3$He nucleus. The hyperfine interaction between the $^3$He nucleus and the Rb valence electron can then induce both species to flip their spins, thereby transferring angular momentum to the $^3$He nucleus from the electron. The cross section for this interaction is very small, $\approx 10^{-24}$ cm$^2$ [85]. Consequently, the spin-exchange process is very slow.

The expected $^3$He polarization, calculated from a simple analysis of spin-exchange and $^3$He nuclear relaxation rates, starting from $P_{He} = 0$ at $T = 0$, is

$$P_{He}(T) = \left( \frac{\gamma_{SE}}{\gamma_{SE} + \Gamma_R} \right) \langle P_{Rb} \rangle \left( 1 - e^{-(\gamma_{SE} + \Gamma_R)T} \right)$$

(24)

where $\gamma_{SE}$ is the spin-exchange rate per $^3$He atom between the Rb and $^3$He, $\Gamma_R$ is the total relaxation rate of the $^3$He nuclear spin and $\langle P_{Rb} \rangle$ is the average polarization of the (Rb). Maximizing the $^3$He
polarization therefore requires making $T$ very long, maximizing $\langle P_{Rb} \rangle$ and $\gamma_{SE}$, and minimizing $I_R$. Since $F_{Rb}$ is $\approx 100\%$ wherever the laser light penetrates, $\langle P_{Rb} \rangle$ is maximized by carefully matching the spatial profile of the laser beam to the geometry of the pumping chamber and by adjusting the $(Rb)$ in the chamber to that the absorption length is nearly equal to the length of the chamber. The spin-exchange rate $\gamma_{SE}$ is also sensitive to $(Rb)$, and is defined by

$$\gamma_{SE} \equiv \langle \sigma_{SE} \nu \rangle \langle Rb \rangle$$

(25)

where, $\langle \sigma_{SE} \nu \rangle = 1.2 \cdot 10^{-19} \, \text{cm}^3/\text{s}$ is the spin-exchange cross section for $Rb-^3He$ collisions times the average relative $Rb-^3He$ velocity $\nu$ [85] and $(Rb)$ should be averaged over the cell. In contrast, the volume of rubidium vapor which can be fully polarized with a given laser intensity will eventually drop off as $(Rb)$ is increased, so $\gamma_{SE}$ cannot be arbitrarily increased without eventually sacrificing $\langle P_{Rb} \rangle$. The optimum Rb density is most easily found by experimentally tuning the pumping chamber temperature to find the highest $^3He$ polarization.

In targets typical time constants for the build-up of $^3He$ nuclear polarization are up to 40 hours, even though the $^3He$ is in constant contact with the $\approx 100\%$ polarized Rb vapor. Polarized $^3He$ targets of high density (up to 12 atm at 273 K) high polarization up to 60% [86] and large volumes up to 170 cm$^3$ have been produced (see section 4.5.2).

### 4.2 $^3He$ Polarization Relaxation

Generally the $^3He$ polarization in a target cell is obtained by transfer of polarized metastable or alkali atoms from the optical pumping cell with polarization $P_0$. For the spin-exchange targets, the polarization rate in the pumping cell is much less than the transfer rate and the asymptotic $^3He$ polarization is given by $P_{3He} = P_0 \left( \frac{\gamma_{SE} \Gamma_R}{\gamma_{SE} \Gamma_R + \Gamma_{wall} + \Gamma_{gas} + \Gamma_{field} + \Gamma_{beam}} \right)$ where $P_0$ is the average rubidium polarization $\langle P_{Rb} \rangle$. For the mechanically or cryogenically compressed metastability-exchange pumped targets the equilibrium $^3He$ polarization can be written as [13] $P_{3He} = P_0 \left( \frac{G}{\Gamma_R} \right)$ where, for the mechanically compressed targets, $G$ is the rate at which doses of $^3He$ atoms are added to the target cell, taking into account any polarization losses in the compression process. For the cryogenically compressed targets $G$ is the diffusion rate through the transfer tube.

In all cases, the total rate at which polarized atoms are delivered to the target cell is determined by the laser power and by the efficiency for the transfer of photon angular momentum to the $^3He$ nuclei. Characteristically, $N \cdot G$ is $\approx 10^{17} - 10^{18}$ atoms/s [13], where $N$ is the total number of $^3He$ atoms in the target. The spin-exchange pumped and the mechanically compressed metastability-exchange pumped targets have time constants ranging from a few hours to many tens of hours, and therefore $^3He$ relaxation mechanisms are very crucial. In contrast, the cryogenically compressed metastability-exchange pumped targets can achieve high $^3He$ polarization with pumping time constants of tens of minutes.

The total $^3He$ relaxation rate $\Gamma_R$ is a result of several effects, approximately $\Gamma_R = \Gamma_{bulk} + \Gamma_{wall} + \Gamma_{gas} + \Gamma_{field} + \Gamma_{beam}$. Bulk relaxation results from $^3He - ^3He$ collisions, where a dipolar interaction couples the nuclear spin to the orbital angular momentum of the colliding atoms [87, 88]. The relaxation $\Gamma_{bulk}$ is proportional to the $^3He$ density and varies slightly with the temperature. This limits the relaxation time $\tau = 1/\Gamma_{bulk}$ to approximately 80 h for a 10 bar target and to $\approx 850$ h for a 1 bar target [89]. This limiting lifetime will be further reduced by relaxation due to collisions with paramagnetic gaseous impurities such as $O_2$. At a $^3He$ density of 10 bar, 10 parts per million contamination of $O_2$ is sufficient to reduce $\Gamma_R^{-1}$ to $\approx 10$ h [90]. Wall relaxation rates can vary appreciably, depending most strongly on the glass material used to build the cells. Cell glasses can be characterized by magnetic impurities and an effective adsorption energy [91]. Borosilicate glasses such as Pyrex have high helium permeation, and it is believed that the $^3He$ is subject to long effective sticking times as it diffuses into the glass. Glasses with very low helium permeation such as alumino-silicate glasses (Corning 1720 and 1723; Schott 8290) have much longer wall relaxation times, reaching from 30 h to 100 h.

These three effects yield an inherent target lifetime. In addition, there are interactions not inherent to the target cell which further increase the $^3He$ relaxation rate. Both static and time-dependent magnetic fields lead to $^3He$ polarization relaxation. Inhomogeneities in the magnetic alignment field
induce relaxation in proportion to the diffusion constant for the cell and the square of the gradients transverse to the magnetic alignment field [92, 93]. This contribution to the $^3$He relaxation rate can be made negligibly small by controlling the field gradients through a proper magnet design (mostly large Helmholtz coils), by magnetic shielding (important in experiments where spectrometer magnets are close to the target) and by increasing the static field. The relaxation due to the time-dependent magnetic fields e.g. produced by pulsed electron beams depends on the time structure, peak currents and beam size. Three effects must be considered:

- Possible coincidence of the $^3$He Larmor frequency with harmonics of the electron beam pulse frequency.
- The sudden periodic appearance of transverse magnetic field components due to the beam microstructure.
- The magnetic field gradients produced by the beam.

The solution to the possible coincidence of the $^3$He Larmor frequency and pulse frequency harmonics lies in the detuning and shifting the Larmor frequency so that this effect becomes negligible [94, 95].

Further $^3$He depolarization occurs due to ionization. When a $^3$He atom is ionized, the hyperfine interaction couples the nuclear spin to the unpaired electron spin allowing a transfer of angular momentum if the two initially have opposite spin orientations. Furthermore, electrons from other $^3$He atoms can be transferred to the original ion, creating the potential for depolarizing another atom. This depolarization process continues until the ions are finally neutralized. It is also possible that molecular ions ($^3$He$^+$) form and depolarize $^3$He nuclei by resonant atom exchange. Admixing of $^2$He to the $^3$He mitigates radiation trapping effects and strongly suppresses the formation of molecular ions [95, 96, 35].

Details of the polarization transfer and loss mechanisms can be found for the spin-exchange pumped targets in Ref. [84], for the metastability-exchange pumped cryogenic target techniques in Ref. [97] and for the mechanical compression targets in Ref. [98, 91].

Sophisticated designs of these targets have been developed, where in all spin-exchange pumped $^3$He targets all-glass targets were used. For the cryogenically compressed metastability-exchange pumped targets, thermal conductivity and temperature control requirements have led to the use of copper or ultra-pure aluminum target cells [78].

### 4.3 $^3$He Polarization Measurement

It is clearly vital for the use of the various $^3$He targets for particle physics experiments that an accurate measurement of the $^3$He polarization at the beam target interaction point, i.e. in the target cell, has to be performed. Two methods of polarization measurement are used. A calibrated nuclear magnetic resonance (NMR) probe has been used for all targets [99]. It measures the total magnetization of the $^3$He from which the polarization is extracted using the $^3$He density. The calibration is done with the known thermal equilibrium Boltzmann polarization of the protons in a water sample of the same cell dimensions at room temperature. Although the water sample has a high density compared with the $^3$He gas its proton polarization is only $\approx 10^{-8}$. The small signals and the linearity of amplifiers over the several orders of magnetic dynamic range encompassing proton and $^3$He signals make this method challenging. The NMR calibration is reliable at the $3\% \rightarrow 7\%$ level. Some details about the method of the $^3$He polarization measurement are given in section 4.5.2.

For the cryogenically compressed metastability-exchange targets, the measurement of the polarization in the target cell cannot be done directly via NMR technique because the target cells are constructed from metal. However, the polarization of the $^3$He in the target cell can be determined indirectly from the polarization of the atoms in the pumping cell. It is monitored continuously using an optical technique, i.e. measuring the circular polarization of the $^3D_2 \rightarrow 2P_1$ fluorescence ($\lambda = 667$ nm) excited by the discharge [100, 101, 82]. Furthermore the number of atoms in each cell, the transfer time between the two cells, and the relaxation time of the coupled system have to be known. The relaxation time and the transfer time are measured frequently during the course of an experiment. The optical polarization measurement (fluorescence method) has to be calibrated against a direct measurement of the nuclear polarization using NMR and is also reliable to a few percent [102, 103].
4.4 Realization of $^3\text{He}$ Internal Gas Targets

The fast polarization build-up of $^3\text{He}$ makes the metastability-exchange optically pumped $^3\text{He}$ targets an excellent system for internal target operation in storage rings. The one drawback of this method is the need to produce the metastable $^3\text{He}$ atoms in a rf-discharge at pressures of about 0.7 mbar. Thus an increase of the density of $^3\text{He}$, to achieve sufficiently high luminosities in the experiment, is evident which also implements a spatial separation of the optical pumping and target region. In order to create an internal target, $^3\text{He}$ gas flows through a glass pumping cell of volume $V$ - where the $^3\text{He}$ gas is polarized by metastability-exchange pumping - into the target cell at a flow rate of $F$ atoms/s. Internal target experiments with polarized $^3\text{He}$ are technically easier than experiments with polarized H or D internal targets. Optical pumping permits higher production and flow rates than it is possible for H or D delivered by atomic beam sources. Consequently $^3\text{He}$ internal targets of sufficient target density can be obtained with storage cells of larger aperture than is needed for H or D internal targets.

A schematic diagram of a polarized $^3\text{He}$ internal gas target is shown in Figure 16. As indicated the $^3\text{He}$ gas has an input density of $\rho_1$ and traverses an input conductance $C_1$. The gas in the pumping cell has an average density of $\rho_p$ and exits through a conductance $C_2$ to the target cell. The average residence time $T_{res}$ of an atom in the pumping cell is given by $T_{res} = \rho_p \cdot V/F$. For an equilibrium flow with a target cell density $\rho_0 \approx 0$ and $T_{res} = V/C_2$. Typically $\rho_1$ is $\approx 5\, \text{mbar}$, $\rho_p \approx 0.6\, \text{mbar}$, and $\rho_0 \approx 10^{-3}\, \text{mbar}$. High polarization in the flowing system is obtained only if the pump-up time is much shorter than $T_{res}$. A pumping cell of volume $500\, \text{cm}^3$ at a pressure of $0.66\, \text{mbar}$ and a $^3\text{He}$ flow rate of $10^{17}$ atoms/s will have a residence time $T_{res} = 75\, \text{s}$. For a pump-up time of $\approx 10\, \text{s}$ it is expected that the $^3\text{He}$ atoms are polarized to an equilibrium polarization of greater than 50% [104].

The feasibility of this technique was demonstrated in a series of elastic scattering measurements with polarized protons at the Indiana University Cyclotron Facility Cooler Ring [105]. The $^3\text{He}$ atoms were polarized through metastability-exchange optical pumping in a Pyrex cell which was connected by a Pyrex capillary to an aluminum storage cell. The storage cell was an open-ended tube of $400\, \text{mm}$ length and of rectangular cross section $(13 \times 17\, \text{mm}^2)$ with sides of Mylar foil coated with $1.7\, \mu\text{m}$ aluminum. The $^3\text{He}$ atoms were directed to the pumping cell through a precision capillary so that the equilibrium pressure in the pumping cell was about $0.7\, \text{mbar}$ at a flow rate $F = 1.2 \cdot 10^{17}\, \text{atoms/s}$ resulting in a target areal density of $1.5 \cdot 10^{14}\, \text{atoms/cm}^2$. The average $^3\text{He}$ polarization in the pumping cell was $(45 \pm 2)\%$ and was measured using the detection of the $667\, \text{nm}$ line in the $^3\text{He}$ discharge.

![Figure 16: Schematic diagram of a polarized $^3\text{He}$ internal gas target. Figure notation is defined in the text. From Ref. [104]](image-url)
4.4.1 Cryogenically Compressed $^3$He Storage Cell Target

The $^3$He target density can be further increased with cooled $^3$He storage cells as demonstrated by the HERMES experiment at DESY. In this experiment polarized $^3$He was used as a polarized neutron target to study the neutron spin structure function $g_1(x)$ [106]. The HERMES $^3$He target (Figure 17) was a second generation of the target described in section 4.4. The storage cell was a long elliptical cylinder with 9.8x29.0 mm$^2$ cross section and an effective target length of 400 mm, formed by two sheets of 125 μm thick, ultra-pure (99.999 %) aluminum.

![Diagram of the HERMES polarized $^3$He internal gas target system.](image)

Figure 17: Cross sectional schematic of the HERMES polarized $^3$He internal gas target system. $^3$He gas flows into a glass pumping cell where it is polarized by interaction with laser light. The $^3$He atoms then flow into the aluminum target cell through which the beam is directed. From Ref. [104]

The cooling was accomplished by a contact of the storage cell to flowing cryogenic helium gas. By operating at $\approx 25\,\text{K}$, the target density could be increased by a factor of 3.5 compared to a room temperature storage cell. Below this temperature, surface spin relaxation effects, which determine the minimum operating temperature, become non-negligible [107, 108]. The $^3$He target area density was $1.0 \cdot 10^{15}$ atoms/cm$^2$ and was chosen by balancing the density versus the reduction in the beam lifetime of the HERA storage ring. As the target system was designed to be able to deliver either longitudinal or transverse target polarization, the coil configuration and the laser optics were rather complicated. The target was operated at a field of 3.4 mT. In addition to the pumping cell polarimeter (see section 4.3) a so-called target optical monitor (TOM) was used to measure the $^3$He polarization in the storage cell directly [109]. The TOM is based on the principle that the positron beam excites atomic levels in the storage cell via Coulomb interaction. For a suitably chosen atomic level the circular polarization of the emitted atomic photons can be related to the $^3$He polarization. The main use of the TOM is not in the absolute measurement of the polarization, but as a monitor of the relative polarization to study beam depolarization as well as temperature dependent wall depolarization effects. An example of simultaneous polarization measurements made with TOM and the pumping cell polarimeter is shown in Figure 18. The polarization was reversed every 10 minutes and the polarization averaged over the data taking period was 47% with a relative error of about ±9%. More details about the HERMES polarized $^3$He internal target can be found in Ref. [104].
target polarization measured with TOM

Figure 18: Polarization vs. time measured with the TOM (target optical monitor) and PCP (pumping cell polarimeter). Each point represents the average of 100 s of data taking. Larger errors for the TOM measurements are due to a small photon flow. From Ref. [104]

4.5 Realization of $^3$He External Gas Targets

The density of polarized $^3$He targets for the operation in external beams has to exceed that of polarized internal $^3$He targets by many orders of magnitude. To achieve a luminosity of about $2 \times 10^{34} \text{ cm}^{-2} \text{ s}^{-1}$ with a typical accelerator beam current of $10 \mu\text{A}$, e.g., a 10 cm long target cell filled with 1 bar of $^3$He at room temperature, corresponding to a target thickness of $\approx 3 \times 10^{20}$ atoms/cm$^2$ has to be produced. Sufficient density is attained by cryogenic [110, 78, 86] or mechanical compression methods [98]. Sufficiently long relaxation times of the $^3$He polarization in high density target cells fed by metastability-exchange pumped $^3$He were achieved by the advances in cryogenic coatings of cell surfaces with H$_2$ [111, 112] and N$_2$ [113], and with Cs coating in room temperature cells [114]. As mentioned before, spatial separation of the optical pumping and target regions for $^3$He targets polarized by the metastability-exchange method is mandatory. This separation is not necessarily required for the $^3$He targets which are polarized by the spin-exchange scheme.

In addition to the $^3$He polarizing methods, the possibility of a transportation over large distances of highly polarized sealed $^3$He target cells - put in a small magnetic field - opened new applications in the field of medicine [115, 116] and also some options for the use of high density $^3$He target cells in particle beams.

Density compression of the spin-exchange pumped $^3$He targets by cryogenic methods is always done prior to the particle physics experiment. Sealed single glass cells are first polarized in the laboratory and then transported to the experimental area, whereas sealed high density double glass cells are placed at the target position in the beam and finally polarized (see section 4.5.2). Mechanical compression of metastability-exchange pumped $^3$He targets can be either done in the laboratory, followed by the transportation of the polarized target cell to the experimental area, or directly in a system of circulating
\( ^3 \text{He} \) gas with the target cell in the beam position. In the second case the entire polarization apparatus has to be built up in the experimental area like in the case of cryogenic compression of metastability-exchange pumped \(^3\text{He} \), where the target cell needs a direct connection to the optical pumping cell.

In the following examples mechanically and cryogenically compressed metastability-exchange pumped \(^3\text{He} \) targets and a high density double cell spin-exchange pumped \(^3\text{He} \) target are briefly described.

### 4.5.1 Mechanically Compressed \(^3\text{He} \) Targets

The main functions of a polarized \(^3\text{He} \) target operated by mechanical compression are shown in Figure 19.

Figure 19: Flow diagram of a mechanically compressed \(^3\text{He} \) target.

In addition to the high polarization in the optical pumping cell and a long relaxation time in the target cell it has to fulfill the following two demands:
- Preservation of polarization during compression.
- Removal of any impurity in the gas before repolarization.

Figure 20: Schematic set-up of a mechanically compressed \(^3\text{He} \) target (OPC = optical pumping cell, PCP = pumping cell polarimeter). From Ref. [23]
The complete set-up of a mechanically compressed (Toepler compressor) polarized $^3$He target is schematically shown in Figure 20.

This $^3$He polarized target has been specially developed for an experiment at the MAINZ Microtron MAMI, to measure the electric form factor of the neutron $G_e^2$ in double polarized exclusive $^3$He$(e,e'n)$ scattering [23]. The polarized gas was compressed into the target cell ($p = 1\ \text{bar, } V = 100\ \text{cm}^3$) by means of a nonmagnetic Toepler compressor, which uses a liquid mercury piston. In order to compensate for $^3$He polarization relaxation in the target cell a continuous flow of about $10^{18}\ \text{atoms/s}$ was circulated from the target cell, via capillary tubes and getter purifier, back into the optical pumping cells for repolarization.

$^3$He polarization within the OPCs was determined from the degree of circular polarization of fluorescence light, as measured by the PCP unit (see section 4.3). A steady state polarization in the optical pumping cell of 70% was achieved. The $^3$He polarization within the target cell was monitored by NMR. The NMR signal was calibrated periodically by returning a certain fraction of the gas directly into the previously evacuated OPCs and then performing the polarization measurement with the PCP. An overall accuracy $\Delta P_t/P_t = \pm 10\%$ is estimated for this measurement.

The $^3$He polarization in the target cell is reduced with respect to that in the optical pumping cells due to a loss during compression and to the gas recycling time with respect to the spin relaxation time. The relaxation rate $\Gamma_R$ is composed of several components (see section 4.2). A serious source of relaxation is the formation of $^3\text{He}_2^+$ molecules by the electron beam [118, 95]. The average beam current was 7 $\mu\text{A}$ at the target. The formation rate of $^3\text{He}_2^+$ was quenched by admixing 0.1 $\text{mbar}$ of nitrogen to the compressed $^3$He. The dominant component is due to collisions with the walls of the container. A decisive improvement was achieved by coating the walls with Cs [114]. The $^3$He polarization history in the target cell and optical pumping cells during the scattering experiment is shown in Figure 21.

Instead of decompression the $^3$He target cell can be decoupled from the compression circuit and transported in a small magnetic holding field to the target position in the particle beam (see Figure 22). This operation scheme was used to measure the neutron electric form factor $G_e^2$ via $^3\text{He}(e,e'n)pp$ using a 3-spectrometer facility at the Mainz microtron MAMI [119]. The $^3$He gas was compressed by a two-stage titanium piston compressor to 6 $\text{bar}$ in a special three-cell design, which results in a small overall thickness of the beam entrance and exit glass windows of the target cell. The target polarization achieved was approximately 50%. During the beam time the polarization and relaxation of the $^3$He in the target was controlled by NMR and AFP. A single AFP measurement leads to a polarization loss of less than 0.2% and the absolute polarization was measured with a relative error of $\pm 3.5\%$. The influence of the beam ($2 - 10\ \mu\text{A}$) and impurities in the cell reduced the relaxation time to 20 - 30 h, which made a cell change necessary twice a day. The time-averaged polarization was about 30%. More
4.5.2 Cryogenically Compressed $^3$He Targets

Cryogenic compression of metastability-exchange pumped $^3$He targets requires a double cell technique with the target cell located in the particle beam and kept at low temperature. The apparatus is similar to that for the polarized internal $^3$He targets (see section 4.4.1) with additional thin foil windows for the entrance and exit of the external particle beam. A schematic layout of a cryogenically compressed $^3$He target for external particle beam experiments is shown in Figure 23.

This double cell system was used in different experiments performed with longitudinally polarized electrons at the MIT-Bates Linear Accelerator Center [120, 107, 113]. The metastable $^3$He atoms were optically pumped in a Pyrex pumping cell, where the $^3$He density is $7 \times 10^{16}$ atoms/cm$^3$ ($\approx 2.6 \text{ mbar}$). The optical pumping light was supplied by a Nd-doped lanthanum magnesium hexaluminate crystal (LNA) pumped by a krypton arc lamp in a Laser Application 9560 cavity. The $^3$He polarization was transferred with negligible polarization loss to a cooled copper target cell, where the $^3$He density is $x \times 10^{18}$ atoms/cm$^3$, i.e. with a high thermal gradient between the room-temperature pumping cell and the target cell at $\approx 15 \text{ K}$ a considerable compression ratio can be achieved. The beam entrance and exit windows were $5 \mu m$ thick copper foil. The target cell had an effective target length seen by the spectrometers of $10 cm$, corresponding to a target areal density of $1.2 \times 10^{19}$ atoms/cm$^2$. In target cells, which are operated at low temperatures, depolarization by field gradients becomes less important because the atoms diffuse more slowly throughout the volume, and the surface effects become more important at lower temperatures because the sticking time of the $^4$He atoms on the walls is longer. Therefore the target walls were coated with a thin layer of N$_2$ to reduce surface-induced spin relaxation. The spins were aligned in a holding field of approximately $3 mT$ provided by Helmholtz coils. The polarization of the $^3$He in the pumping cell was monitored continuously by the detection of the circular polarization of...
the 667 nm line excited by the discharge. The $^3$He polarization in the target cell was inferred from the polarization of the pumping cell and the time constants of the coupled system. With a 25 $\mu$A electron beam, a target polarization of up to about 40% could be achieved. With no depolarization by the beam, the target polarization was typically higher by a factor of 1.15.

Using the spin-exchange technique with optically pumped rubidium vapor, such targets have been shown to be capable of operating at even higher $^3$He pressures (more than 10 bar [86]) and in intense electron beams [84]. The target cells are prepared by baking out under vacuum for up to 7 days at 450 °C. During the $^3$He filling procedure they are cooled to about liquid $^4$He temperature to reach a pressure below atmospheric. After sealing the small entrance tube for $^3$He the glass cells finally reach high pressure at room temperature. All such filling procedures were used for experiments at TRIUMF [121, 86], at LAMPF [87, 88], and at SLAC [122].

In principle, a single cell chamber could be laser pumped to maximum polarization in a location outside the experimental area, then installed in the beam line to replace another cell when the polarization of the latter has dropped too low. However, there are several reasons not to proceed in this way, e.g. the glass cell walls could darken from radiation damage and prevent repumping of the cell to the optimum polarization. Also serious is the ionization effect on Rb which must be eliminated [123]. In addition any change of the target cell requires a long access time to the accelerator and operating with different target cells during the experiment is certainly not advantageous for the optimization of the systematic polarization error. Here a two-cell target (see Figure 24(b)) solves or minimizes these problems, and one good cell ($\Gamma = \Gamma_{\text{bulk}} + \Gamma_{\text{wall}} + \Gamma_{\text{gas small}}$) could operate for the whole particle experiment. $^3$He is polarized in the upper (pumping) cell and diffuses through the transfer tube to the lower (target) cell with a time constant of about 10 min, which is small compared with the characteristic spin-exchange and relaxation times. Due to the low thermal conductivity of glass, the target cell remains at a low enough temperature (about 60 °C) so that the rubidium vapor density there is negligible. Then the two-cell target can be considered as a single total volume equal to the sum of the pumping cell volume and target cell volume with the Rb confined to the heated pumping cell. The disadvantage of this design is the increase of the effective spin-exchange time. A description of the dynamics of the two-cell target can be found in [84].

This $^3$He target type (see Figure 24(a)) was successfully used in experiments at SLAC to study the spin structure of the neutron [124, 125, 126]. The high pressure target operated at a $^3$He density of
Figure 24: (a) Schematic overview of the SLAC $^3$He polarized target. Not shown are four of five identical Ti-sapphire lasers, the argon-ion lasers used to pump the Ti-sapphire, and the optics used to combine the five laser beams. (b) A schematic illustration of the SLAC E142 experiment two-cell target. The upper cell is the pumping volume and the lower cell is the target volume. The Rb is confined to the upper cell (surrounded by an oven at $\approx 160^\circ$C as shown in (a)) by a thermal gradient along the transfer tube.

2.3 $\times$ $10^{20}$ atoms/cm$^3$ in a 30 cm long scattering region, giving an areal density of about $7 \times 10^{21}$ atoms/cm$^2$. The average $^3$He polarization was about 36%, as measured with the NMR technique of AFP. The AFP system uses an rf-drive field orthogonal to the main field at a fixed frequency of 92 KHz, while the main holding field strength is varied from the normal value of 2 mT to a maximum of 3.8 mT, passing through resonance for $^3$He at this frequency when the field is about 2.9 mT. If the strength of the drive field and the rate of change of the main field strength are chosen correctly, essentially all of the $^3$He nuclear spins flip over during this process and the target polarization ends up reversed. The precessing nuclear spins induce a signal in the small pickup coils located around the center of the target cell. In Figure 25 a typical $^3$He signal resulting from this procedure is shown. The main field is then swept back to the normal value, reversing the process and restoring the polarization to the direction being pumped, while providing another measurement of the $^3$He signal. Repeated measurements show that very little polarization ($< 0.1\%$) is lost in this procedure. The average of 25 proton signals from the calibration measurements (see section 4.3) is also shown in Figure 25 and the $^3$He polarization history during the experiment is shown in Figure 26.

The glass windows of the target cell, where the SLAC polarized electron beam enters and exits, are within the acceptance of the spectrometers, and must be therefore made as thin as possible in order to minimize background events from unpolarized materials. Developments of very thin concave spherical windows were made to eliminate their dominance of the event rate [127]. A further source of background events results from the scattering of the beam halo from the target cell walls and from $N_2$ gas. It is essential (especially for inclusive measurements) to know the dilution of the observed asymmetries resulting from target material other than $^3$He, which can be determined by measurements with empty or partially empty target cells. The dilution factor $f$ (see equation (5)) for the SLAC experiment varies between 0.3 and 0.35, which is further reduced when the polarized neutron results from polarized $^3$He are extracted [125]. A second generation of this $^3$He target type is operated at TJNAF for measurements at $Q^2 < 1.2 \text{ GeV}^2/c^2$ [128]. Using a 10 - 15 $\mu A$ polarized electron beam a luminosity of about $7 \times 10^{36} \text{ cm}^{-2} \cdot \text{s}^{-1}$ could be handled, which is the highest value so far reached in a double polarization experiment.
4.6 Summary of Polarized $^3$He Gas Targets

Model calculations of the $^3$He wavefunction show that the unpaired neutron is carrying about 90% of the $^3$He spin. Thus additionally to polarized deuterons, polarized $^3$He is used as a polarized neutron target. There are two methods of polarizing $^3$He nuclei for target applications: The metastability-exchange method and the method of collisional spin-exchange with laser pumped Rb vapor. Cryogenic and mechanical compression methods have been developed to achieve high target thicknesses between $5 \times 10^{20}$ atoms/cm$^2$ and about $10^{22}$ atoms/cm$^2$ giving luminosities of up to $5 \times 10^{35}$ cm$^{-2}$ s$^{-1}$ in combination with electron beam currents of several microamperes. Target thicknesses of up to $1 \times 10^{15}$ atoms/cm$^2$ have been obtained in cooled storage cells, which are placed in electron beams with up to 80 mA. This gas thickness fits to the acceptable one of $10^{15}$ atoms/cm$^2$ for storage ring applications. In that case luminosities in the order of $10^{32}$ cm$^{-2}$ s$^{-1}$ are obtained. $^3$He polarizations of 40% - 50% are routinely achieved during the experiments and the polarization degree is measured with NMR and/or optical methods with a relative accuracy of about ±5% or better. $^3$He polarized targets for past, present and future particle physics experiments have been or are operated at DESY, IUCF, JLAB, LAMPF, NIKHEF, MAMI, MIT-Bates, TJNAF, SLAC and TRIUMF.

5 Polarized Solid Targets

A polarized solid target can be assumed to be an ensemble of particles with spin, placed in a high magnetic field and cooled to very low temperature. The basic idea - to obtain a high polarization of nuclear spins - consists in using a microwave field, to transfer the polarization of electron spins to these nuclei. This process is called dynamic nuclear polarization (DNP) and is described in detail in section 5.2. The main practical problem with DNP is finding a suitable combination of hydrogen - or deuterium - rich material and a paramagnetic dopant, i.e. material with an unpaired or quasi-free electron. Suitable means that the relaxation time of the electron spins is short ($ms$) and that of the nucleons (nuclei) is long ($min$), resulting in a high nucleon (nuclei) polarization.

Molecular hydrogen would be the ideal proton target material, except that at low temperature it devolves to the magnetically inert para-state, with spin zero and, hence, is unpolarizable. Various
practical polarized solid target materials have been developed in which a diluted amount of paramagnetic centers has been introduced by chemical or radiation methods (see section 5.5).

Polarized solid targets have some special advantages, such as:

- High density, which makes them unique for particle experiments with secondary beams e.g. (tagged) photon beams or muon beams.
- High construction flexibility, which enables an integration into almost any detector set-up.

The most evident disadvantage is the small dilution factor \( f < 0.5 \) of the polarizable target materials, which is less serious in the case of exclusive particle reactions.

There are two operation modes depending on the intensity of the particle beams:

- Continuous mode, i.e. permanent microwave irradiation for DNP.
- Frozen spin mode, i.e. after DNP microwave irradiation is stopped.

A very important feature of polarized solid targets is the fact, that the DNP scheme works for any nucleus with spin. However, the efficiency of the DNP in the different target materials, i.e. to get highest nuclear polarization values, depends strongly on the nature and the interaction of the added paramagnetic centers in the respective solids.

### 5.1 Thermal Equilibrium Polarization

The starting-point of any general discussion of polarized solid targets is the magnetic moment of each of the particles of interest, i.e. the electron and proton or deuteron. In the so-called high temperature approximation, being valid, if the Zeeman energy \( \mu \cdot B \) of a system of spin \( J \) is much lower than its thermal energy \( kT \), the entropy at a magnetic field \( B_0 \) is given by

\[
S = kN \ln(2J + 1) - \frac{C}{2} B_0^2 + \frac{B_l^2}{T^2},
\]

where \( N \) is the number of spins and \( C \) is the Curie constant

\[
C = N \frac{\hbar^2 \gamma^2 J (J + 1)}{3k}.
\]

As long as the external field \( B_0 \) is much larger than the local field \( B_l \), which is the additional field seen by one spin due to the presence of all the others, the magnetic order \( S_0 - S \) is proportional to \( (B_0/T)^2 \).
but also proportional to the square of the gyromagnetic ratio \( \gamma = gJ\mu_M/\hbar \). In the same approximation the polarization is calculated from

\[
P_J = \frac{1}{3} \frac{g_J\mu_M}{kT} B_0 \quad \text{or in general from} \quad P_J = B_J(x) = B_J\left(\frac{g_J\mu_M J B_0}{kT}\right)
\]

with no restriction for the value of \( B_0/T \). Herein \( B_J(x) \) is the Brillouin function, which reduces to the hyperbolic tangent in the case of spin-1/2. The polarization also depends on both quantities, \( B_0/T \) and \( \gamma \), but in a linear manner. Whereas the value of \( B_0/T \) may be chosen at will within the limits given by the respective technologies, the gyromagnetic ratio \( \gamma \), i.e. the product of the particle's g-factor \( g_J \) and the magneton \( \mu_M \), is an invariable feature of the particular particle. As a consequence of its low value for nuclei the ratio \( B_0/T \) must be at least as large as \( 10^3 T/K \), in order to create a sizeable nuclear thermal equilibrium (TE) polarization. Although these values are technically achievable, they are not useful for the majority of the polarized targets for particle physics experiments (see section 5.8.2).

5.2 Dynamic Nuclear Polarization

The technique, which is able to provide the necessary high nuclear polarization at reasonable temperatures and magnetic fields, is the DNP method mentioned above. Here one makes use of the high gyromagnetic ratio \( \gamma_S \) of unpaired electrons, which leads to an electron polarization near to unity under magnetic fields of some Tesla even at temperatures as high as \( 1 K \). With the help of a strong (i.e. saturating) microwave field, whose frequency is chosen slightly off the electron Larmor frequency \( \omega_s = \gamma_S B_0 \), the high magnetic order of the electron spin system can be transferred to the nuclear spins. The most simple model, which is able to describe the physical process behind that, is the solid state effect in its resolved version.

5.2.1 The Resolved Solid State Effect

In 1953 Carver and Slichter [129] for the first time were able to observe an enhanced nuclear polarization in metals by rf-saturating the conduction electrons, an effect, which had been predicted by Overhauser [130] in the same year. The method of DNP described in this chapter is based on the so-called solid state effect discovered in 1958 [131, 132, 133]. It permits the build-up of an enhanced nuclear polarization in solid insulators by a similar mechanism. In order to bring out the basic ideas as clearly as possible, the following presentation is restricted to a nuclear spin system with \( I = 1/2 \). A situation is considered, in which a solid of \( N_I \) nuclear spins with Larmor frequency \( \omega_I \) and \( N_S \) electron spins with Larmor frequency \( \omega_S \) is exposed to a magnetic field at a certain temperature.

Figure 27 shows the term scheme of a dipolarly coupled electron-nucleus pair. The respective state of lower Zeeman energy for the electron is that with \( S \) antiparallel and for the nucleus that with \( I \) parallel to the external magnetic field. As long as these spins are isolated from each other, the only possible transitions under a microwave field with frequency near \( \omega_S \) are those with \( \Delta S_z = \pm 1 \) and \( \Delta I_z = 0 \). The corresponding transition probability is denoted by \( W \). However, due to the magnetic field \( B_S = h\gamma_S/r^3 \), produced by the electron at a site of a nucleus in the distance \( r \), the four 'pure' states \( |++>, |+->, |-+>, |--> \) must be replaced by the states \( |a>, |b>, |c>, |d> \), each containing a small admixture of size \( \xi = h/sin \theta \cos \phi \)

\[
|q| = \frac{3}{2} \frac{h\gamma_S}{\omega_I r^3} \sin \theta \cos \phi \simeq \frac{B_S}{B_0} \simeq 10^{-4} \ll 1
\]

from the nuclear spin state with opposite magnetic quantum number. The smallness of this number implies that \( |p| = (1 - |q|^2)^{1/2} \simeq 1 \). The presence of the dipole-dipole interaction gives thus rise to a nonvanishing probability \( V \) for simultaneous spin flips of an electron and a nucleus. If one defines (see also section 2.3) the respective polarizations of the particles by

\[
P_S = \frac{n_--n_+}{n_- + n_+} \quad \text{and} \quad P_I = \frac{N_--N_+}{N_- + N_+} = \frac{N_--N_+}{N_I}
\]
with the occupation numbers $n_\pm$ for the electron spins and $N_\pm$ for the nuclear spins, one may write down the evolution equations

$$\frac{dP_s}{dt} = -V(P_s - P_t) + \frac{1}{T_s}(P_s^L - P_s)$$

$$\frac{dP_t}{dt} = \frac{N_s}{N_I}V(P_s - P_t) - \frac{1}{T_I}(P_t - P_t^L),$$

where the respective lattice (TE) values are indicated by the superscript $L$. The relaxation times are denoted by $T_s$ and $T_I$, respectively. The stationary polarizations are evaluated by the condition $dP_{s,t}/dt = 0$:

$$P_s = P_s^L \left(1 + T_s V\right) + P_s^T \left(1 + (N_s/N_I)T_I V\right) = \frac{P_s^T \left(N_s/N_s T_I V\right) + P_s^L \left(N_I/N_s T_I V\right)}{1 + V(T_s + (N_s/N_I)T_I)}$$

$$P_t = P_t^L \left(1 + T_s V\right) + P_t^S \left(N_s/N_I)T_I V\right) = \frac{P_t^T \left(N_s/N_s T_I V\right) + N_T T_s}{N_s T_I V + N_s T_s T_I}$$

From equation (32) it is readily seen that the nuclear polarization grows the faster the higher the concentration $N_s/N_I$ of the electrons and the longer the nuclear relaxation time $T_I$. Furthermore, its growth rate is also proportional to the difference of the nuclear and the electron polarization, so that $T_s$ should be as short as possible. Thus the condition for achieving a high nuclear polarization can be summarized by:

$$\frac{N_s T_s}{N_s T_I} \ll 1$$

In other words, the total rate of spontaneous electron spin flips $N_s/T_s$ should considerably exceed the rate of spontaneous nuclear spin flips $N_I/T_I$. In this case equations (33) and (34) reduce to

$$P_s = \frac{P_s^L}{N_s T_s} \left(1 + \frac{N_s}{N_s T_s V}\right) \approx P_s^L$$

because of $P_t^L \ll P_s^L$

and to

$$P_t = \frac{P_t^L \left(N_s/N_s T_I V\right) + P_t^S}{N_s T_I V + N_s T_s T_I}.$$
Furthermore, if the condition

$$\frac{N_S}{N_I} T_I V \gg 1$$

is also valid, meaning that the induced transitions of the electron spins are taking place at a rate $N_S V$ much faster than the nuclear relaxation rate $N_I/T_I$, the nuclear polarization is limited only by the TE value of the electrons:

$$P_{I}^{\text{max}} = P_S^L$$

This is the prediction for the maximum nuclear polarization in the case of the 'ideal' solid state effect, i.e. of the 'resolved' solid state effect (see below) under the additional neglect of the following two circumstances, which may be present in a real material to some extent [134]:

1. Due to material impurities or crystalline imperfections, other relaxation channels than those via the electron spins may exist. To account for them, an additional relaxation term has to be introduced in equation (34), which then obviously reduces the maximum nuclear polarization. Typically the electron concentration is $10^{-4} - 10^{-3}$ electron spins per nucleus, so that one electron has to polarize not only the nuclear spins in its direct vicinity, but also those, which are several lattice constants away. The 'transport mechanism' – the so-called 'spin diffusion' – proceeds via resonant mutual flips of two neighboring nuclear spins. The longer the distance to be bridged by the spin diffusion the more important is the strength of the 'leakage' relaxation rate $1/T_1$, which is usually quantified by the leakage factor $\gamma = T_1/T_1$.

2. The electron relaxation rate $1/T_S$ may be considerably slowed down by the so-called phonon bottleneck: When an electron spin relaxes from its upper to its lower Zeeman level, a phonon is emitted carrying away the energy difference $\hbar \omega_S$. In order not to disturb the equilibrium distribution of the lattice phonon bath, the flow rate of the phonons emitted by the electron spins has to be small compared to the rate of their thermalization, i.e. the rate, with which scattering processes between these phonons and the phonons of the thermal bath are taking place. A possible violation of this condition has to be taken into account by a replacement of $T_S$ in equation (33) by $\sigma T_S$ with the phonon bottleneck coefficient $\sigma$.

One of the earliest polarized target materials developed in 1963 was lanthanum magnesium nitrate La$_2$Mg$_3$(NO$_3$)$_{12}$·24H$_2$O (LMN), in which the lanthanum was replaced by neodymium (Nd) to a small extent, in order to create the necessary amount of unpaired electrons. Its water protons could be polarized via the resolved solid state effect up to a value of 70% at a magnetic field of 2.0 T and a temperature of about 1.5 K, achieved by a liquid $^4$He evaporation refrigerator [135]. This is the highest dynamical enhancement of the nuclear polarization ever observed in a polarizable solid material under these conditions and even very close to the theoretical value of 83%. Nevertheless, from the particle physics point of view LMN suffers from two major drawbacks. One is its small dilution factor, being only 0.031 in LMN. The second problem of this material is its small polarization resistance against radiation damage produced by the particle beam. A maximum allowed integrated particle flux of only $10^{10} - 10^{11}$ part./cm$^2$ is much too low for investigating electromagnetic processes with their comparatively small cross sections.

The situation concerning both of these problems was greatly improved by the introduction of the alcohols and their chemical relatives, the diols, during the late 1960s. Dilution factors ranging from 0.1 to more than 0.13 were now available together with a radiation hardness a factor of 1000 higher than that of LMN. About one decade later, in the end of the 1970s and the beginning of the 1980s, it was the development of ammonia, which again led to an enormous improvement of the performance of the polarized target materials [137, 138, 139]. A (proton) dilution factor of more than 0.17 and a radiation hardness of $10^{15}$ part./cm$^2$ opened up the possibility to use particle beams of high intensity to look for rather faint spin dependent effects in polarized scattering experiments. From the discovery of the lithium hydrides in 1980 [140, 141] it took a further decade in order to find the optimum preparation method.
for these face-centered, cubic lattice (FCC) type crystals [142]. Nowadays the isotope combination $^6\text{LiD}$ is the polarized target material with the highest content of quasifree protons and neutrons available in combination with an unsurpassed radiation hardness of about $10^{16}$ part./cm$^2$. The commonly used doping method for all of these later substances, i.e. the technique of implanting the necessary amounts of unpaired electrons into them, consists either of admixing a certain percentage of a stable chemical radical to the liquid state or of exposing the solid or solidified materials to an ionizing radiation. As different as these methods are, they have one thing in common: They lead to paramagnetic species, whose electron spin resonance line widths are of approximately the same size as the nuclear Larmor frequency or even considerably larger.

On the other hand the solid effect in its resolved version relies on the condition of a narrow electron paramagnetic resonance (EPR) line width $D$ compared to the nuclear Larmor frequency $\omega_I$. A violation of this condition leads to the possibility, that both of the 'forbidden' transitions $|a>\rightarrow |d>$ and $|b>\rightarrow |c>$ (see Figure 27) are driven simultaneously by the microwave field, thus leading to a reduction of the achievable nuclear polarization. A model, which tries to take these counteracting processes into account, is the so-called 'differential' solid state effect [143]. The weakness of this model is that it assumes the 'spin packets', i.e. the parts of the EPR line which are separated in frequency due to the presence of inhomogeneous interactions, to be saturated independently. This assumption is valid only for spin systems, in which the concentrations are so small that a dipolar interaction between the spins need not to be considered. However, the usual electron concentrations in a polarizable target material are of the order of some $10^{19}$/cm$^3$. A concentration of that magnitude leads to a static component of the local field seen by the electrons, which is on the order of a tenth of the line width in typical inhomogeneously broadened systems. In these cases the dipolar interaction is able to 'connect' the individual spin packets via the operator $S_i^z S_j^z$ as a part of the dipolar Hamiltonian. Let $\omega_i$ be the Larmor frequency of a particular spin packet $i$, whose Zeeman transitions are driven by a saturating microwave field of frequency $\omega_i$. With no or only a weak dipolar interaction present it is only the spin packet $i$, whose polarization is diminished. However, if the dipolar coupling strength exceeds a certain limit, i.e. if the probability of mutual spin flips is high enough, the whole resonance line is affected by the action of the microwave field. The distribution of the occupation numbers of the non-Zeeman part of the interaction, i.e. the frequency distribution within the resonance line, reach a state of internal equilibrium, which is characterized by a temperature, different from the one of the lattice. This so-called spin temperature eventually determines the nuclear polarization, too.

5.2.2 The Spin Temperature Theory in the High Temperature Approximation

The theory which correctly describes the behaviour of a dipolarly coupled spin system under saturation is the Pровоторов theory. Within this framework an expression for the spin temperature can be derived, which depends not only on the degree of saturation but also on the width of the resonance line as well as on the time constants of the spin system. The present section attempts to give a brief overview over the major ideas and consequences of this theory as far as it concerns the DNP process. It starts with an introduction to the basic principle of adiabatic demagnetization. The authors would greatly emphasize that the following three paragraphs were included only in order to make the concept of spin temperature as clear as possible. They are much too oversimplified to be taken as valid theoretical descriptions of this technique.

**Adiabatic Demagnetization in the Laboratory Frame:**

In the high temperature approximation the magnetization $\vec{M}$ of a spin system exposed to a certain magnetic field $\vec{B}$ is given by Curie’s law

$$\vec{M} = C \frac{\vec{B}}{T_2},$$

wherein $T_2$ denotes the Zeeman temperature of the system, i.e. the temperature, which together with the magnetic field, determines the Boltzmann distribution of the spin states. In the case of thermal
equilibrium the Zeeman temperature is equal to the temperature of the lattice $T_L$. However, a situation, in which $T_Z$ is different from $T_L$ can be easily created by changing the external field strength as long as this process is performed on a time scale much shorter than the spin-lattice relaxation time $T_1$ but considerably longer than the spin-spin relaxation time $T_2$. Both conditions together ensure that the process takes place adiabatically, i.e. fast enough to avoid any energy exchange between the spin system and the lattice but slow enough for the system to maintain a common temperature at all times. If for instance the magnetic field is lowered from a certain initial value $B_i$ to a final value $B_f$ (still much larger than the local field $B_l$ of the system), the magnetization is a constant of motion. Therefore, the temperature $T_Z$ of the spin system must decrease from the initial (lattice) value $T_L$ to some final value $T_f$, so that with $B_i / B_f = T_L / T_f$ Curie's law (40) is still valid.

If on the other hand the magnetic field is lowered down to zero or at least to a value comparable to the local field, another temperature has to be considered, namely the spin-spin temperature $T_{SS}$. It determines the distribution of the occupation numbers corresponding to the non-Zeeman interaction of the system, which, in the most simple case, only consists of the dipolar interaction between the individual spins. In complete analogy with the Zeeman energy in the high temperature approximation

$$E = -\hat{M} \cdot \vec{B} = -C \frac{B_0^2}{T_Z}$$

the non-Zeeman energy is given by the expectation value of the dipolar Hamiltonian

$$\langle H_D^0 \rangle = -C \frac{B^2}{T_{SS}}$$

where the superscript '0' indicates that only the secular part, i.e. the part which commutes with the Zeeman Hamiltonian has to be considered. During the adiabatic demagnetization the spin-spin temperature stays at its initial value $T_L$ until the magnetic field reaches a value so low that the Zeeman and the dipolar energy are of comparable size. In this situation an exchange of energy between the two systems starts to take place, which eventually leads to an equalization of the Zeeman and the spin-spin temperature. In contrast to the first example of a demagnetization at high fields in which both the entropy and the magnetization is a constant of motion, in the low field case it is only the entropy, which stays constant. By comparing the corresponding expressions (26) in the beginning and in the end of the demagnetization process it is easily seen that the final temperature common to both systems is given by:

$$T_{SS} = T_Z = T_L \left( \frac{B_i^4 + B_l^4}{B_i^4 + B_l^4} \right)^{1/2} \approx T_L \frac{B_i}{B_l} \ll T_L$$

Already two far reaching statements can be made from this very simple example of an adiabatic process without any rf-field present.

1. The spin-spin temperature as well as the Zeeman temperature can be made much lower than the lattice temperature.

2. The degree of 'cooling' of the spin system depends on the magnitude of both the local and the (initial) external magnetic field.

These dependencies stay valid also in the case of the 'dynamical' cooling process via the application of a saturating rf-field near the Larmor frequency of the spin system.

**ADIABATIC DEMAGNETIZATION IN THE ROTATING FRAME:**

The next step towards an intuitive picture of the DNP process via the dynamic cooling of the spin-spin interaction (non-Zeeman) reservoir is to allow for an additional strong rf-field perpendicular to the external magnetic field. As usual the linearly polarized field can be decomposed into two circularly polarized components, one of them rotating in the same sense and one of them rotating in the opposite sense with respect to the Larmor precession. Since the time average of the counter-rotating component
seen by the spins vanishes, it is only the co-rotating one which is able to induce transitions. The problem is greatly simplified if viewed from a coordinate system, in which the latter component is at rest. Seen from this frame of reference, which thus rotates around the axis of the external field with the frequency $\omega$ of the rf-field the electron spin experiences a time independent effective field $\vec{B}_{\text{eff}}$. It is given by the vector sum of the reduced external field $\vec{b}_0 = (B_0 - \omega/\gamma_S) \hat{e}_z$ and the rf-field $\vec{B}_1$ (see Figure 28). In this situation an adiabatic$^3$ demagnetization of the spin system (being initially in thermal equilibrium with the lattice) may be performed by switching on the rf-field far below the resonance condition followed by an adiabatic increase of the rf-frequency towards the resonance frequency $\omega_S = \gamma_S B_0$, while the external field is kept constant. During this process the spin temperature is again given by equation (43) with $B_i$ and $B_f$ replaced by $B$ and $B_{\text{eff}}$. A complete demagnetization corresponding to the minimum temperature of the non-Zeeman reservoir can be achieved by a subsequent reduction of the rf-field amplitude down to zero once the system is at exact resonance. Because of the invariance of the entropy the complete order initially stored in the magnetization has been put into the dipolar (non-Zeeman) system.$^4$ Again the ratio of the local to the external field determines the maximum cooling of the dipolar system.

**Figure 28: The effective magnetic field in the frame of reference rotating with the frequency of the rf-field.**

Steady State Demagnetization in the Rotating Frame:

From the very definition of the processes of adiabatic demagnetization it is clear that they do not permit a continuous cooling of the dipolar reservoir, since steady state conditions automatically lead to an exchange of energy between the spin system and the lattice. A continuous application of a strong rf-field near resonance – although a non adiabatic process – still has the effect of reducing the temperature of the dipolar reservoir. The most simple and intuitive treatment of such a situation goes back to the earliest ideas about spin temperature given by Redfield [144]. In order to include spin-lattice effects it is best to start up with the energy balance of the spin system. Combining equations (41) and (42) and replacing again $B_i$ by $B_{\text{eff}}$ a first expression for the change of its total energy

$$\frac{d}{dt} E = C (B_{\text{eff}}^2 + B_f^2) \frac{d}{dt} \left( \frac{1}{T} \right) = -C (\vec{b}_0^2 + B_f^2 + B_{\text{eff}}^2) \frac{d}{dt} \left( \frac{1}{T} \right)$$

$^3$Similarly to the process in the laboratory frame the demagnetization in the rotating frame is adiabatic, if the process takes place in a time much shorter than $T_1$ but much longer than $T_2$.

$^4$The technique of adiabatic fast passage (AFP) makes use of this very fact. By sweeping a strong rf-field from low frequencies ($\omega \ll \gamma_S B_0$) through the resonance up to high frequencies ($\omega \gg \gamma_S B_0$) or vice versa the effective field and thus the magnetization are reversed from a parallel to an antiparallel orientation with respect to the external field.
can be obtained, wherein it has been assumed that the rf-field strength is high enough for the Zeeman and
the dipolar (spin-spin) reservoir to share the same temperature in the rotating frame: \( T_Z = T_{SS} =: T \).\(^5\)
On the other hand the total energy is by definition
\[
E = -\vec{M} \cdot \vec{B}_{eff} + \langle \mathcal{H}_D^0 \rangle 
\]
With the components \( u \) and \( M_z \) of the magnetization in the rotating frame along \( x' \) and \( z \) a second
expression for the change of the energy reads
\[
\frac{d}{dt} E = \frac{d}{dt} \left( -M_z b_0 - u B_1 + \langle \mathcal{H}_D^0 \rangle \right) 
\]
One of Redfield's major ideas was that for a spin system in the solid state the Bloch equations have
to be modified. In particular he realized that in the case of a very strong rf-field (in excess of the
resonance line width) not only the relaxation of the component \( M_z \) of the magnetization along \( \vec{B}_0 \) or \( \vec{B}_0 \),
respectively, but also the relaxation of \( u \) along \( \vec{B}_1 \) leads to an exchange of energy with the lattice. Thus,
the corresponding time constants \( T_1 \) and \( T_1' \) possess the character of 'longitudinal' relaxation times and
are of comparable size. Finally – on the basis of the same argument – a time constant \( T_D \) has to be
assigned to the process of thermalization of the dipolar reservoir towards \( \langle \mathcal{H}_D^0 \rangle_L \), which again is of
the order of \( T_1 \). To summarize, the energy exchange with the lattice can be expressed by the following three
equations:
\[
\frac{dM_z}{dt} = \frac{M_0 - M_z}{T_Z}, \quad \frac{du}{dt} = \frac{u}{T_Z}, \quad \frac{d\langle \mathcal{H}_D^0 \rangle}{dt} = \frac{\langle \mathcal{H}_D^0 \rangle_L - \langle \mathcal{H}_D^0 \rangle}{T_D} 
\]
Herein \( T_1 \) and \( T_1' \) have been renamed by \( T_Z \) and \( T_Z' \) in order to stress their association to the Zeeman
reservoir. Bearing in mind the high temperature expressions for the components of the magnetization
and the dipolar energy
\[
M_0 = C \frac{b_0}{T_L}, \quad M_z = C \frac{b_0}{T}, \quad u = C \frac{B_1}{T}, \quad \langle \mathcal{H}_D^0 \rangle = -C \frac{B_1^2}{T} 
\]
a comparison of equations (45) and (47) together with equations (48) leads to an equation of evolution
for the spin temperature \( T \)
\[
\frac{d}{dt} \left( \frac{1}{T} \right) = \frac{1}{T} \left( \frac{1}{T_{eq}} - \frac{1}{T} \right) 
\]
which is equivalent to the following equation for the absolute value of the magnetization:
\[
\frac{dM}{dt} = \frac{1}{T} \left( M_{eq} - M \right) 
\]
The time constant \( T \) denotes the time needed by the spin temperature \( T \) to reach its (dynamic)
equilibrium value \( T_{eq} \) under the action of the rf-field:
\[
\frac{1}{T} = \frac{b_0^2/T_Z + B_1^2/T_Z + B_1^2/T_D}{b_0^2 + B_1^2 + B_1^2}, \quad \frac{1}{T_{eq}} = \frac{b_0 b_0}{T_L T_Z} \left( \frac{b_0^2/T_Z + B_1^2/T_Z + B_1^2/T_D}{b_0^2 + B_1^2 + B_1^2} \right)^{-1} 
\]
Both quantities are functions of the effective magnetic field \( b_0 \) in the direction of the external field.
With the relation \( \gamma_S b_0 = \gamma_S B_0 - \omega = \omega_S - \omega \) they are thus functions of the frequency difference of the
rf-field with respect to the Larmor frequency. In particular the equilibrium inverse spin temperature
\( 1/T_{eq} \) shows the following behavior:
\(^5\)This restriction is essential for the simplified theory to be valid and can be lifted only in the framework of the
Provotorov theory (see next paragraph).
- It is an odd function of $\omega_S - \omega$ meaning that (a) it is zero for an irradiation in the center of the resonance line and (b) its sign depends on whether the rf-frequency is lower or higher than the Larmor frequency.

- Independently of the particular rf-frequency its absolute value increases with (a) a decreasing local field $B_l$ and (b) a decreasing ratio of the relaxation times $T_z/T_D$.

The first item implies an interesting consequence for the equilibrium value of the magnetization which is still given by Curie’s law:

$$\tilde{M}_{eq} = C \frac{1}{T_{eq}} \tilde{B}_{eff}$$

In contrast to a spin system in a liquid, which can be saturated completely by a strong rf-field applied everywhere within the resonance line, the magnetization of a spin system in a solid only vanishes if the rf-frequency is tuned exactly to the center of the line. Figure 29 illustrates the three situations, in which the rf-frequency is smaller (a), equal (b) or larger (c) than the Larmor frequency. In all cases (unless $\omega = \omega_S$ where $M_{eq} = 0$) the magnetization is aligned along the direction of the effective field, but independently of the actual value of $\omega_S - \omega$ it is pointing into the upper hemisphere, i.e. into the direction of the external field.

![Figure 29: Orientation of the magnetization in the rotating frame of reference under the influence of a saturating rf-field: (a) $b_0 = B_0 - \omega/\gamma_S > 0$: $\tilde{M}_{eq}$ parallel to $\tilde{B}_{eff}$, (b) $b_0 = 0$: $\tilde{M}_{eq} = 0$, (c) $b_0 < 0$: $\tilde{M}_{eq}$ antiparallel to $\tilde{B}_{eff}$.](image)

This last feature is a consequence of the non-adiabaticity of the process, which proceeds under a continuous exchange of energy with the lattice. The spin-lattice relaxation tends to "bend" the magnetization into the direction of the external field, while the strong rf-field forces the magnetization to be aligned with $\tilde{B}_{eff}$. These counter-acting processes cause the spin temperature not only to be of much smaller size than the lattice temperature, but also permit an arbitrary choice of its sign: The spin temperature can be made positive for $\omega_S - \omega > 0$ and negative for $\omega_S - \omega < 0$.

To summarize, already these basic considerations covering only a special case of the spin temperature theory, namely the case of a very strong rf-field, are able to explain how dynamic nuclear polarization works: By means of a strong rf-field applied near resonance to the diluted electron system of a solid, its spin temperature can be tuned almost at will. Since in most polarizable target materials the non-Zeeman energy is of similar size as the nuclear Larmor frequency, a further process of equalization is possible, in which energy is exchanged between the electron and the nuclear spin system. As a consequence the nuclear Zeeman temperature comes into equilibrium with the combined Zeeman and dipolar spin temperature of the electrons. Thus the nuclei become dynamically polarized with a sign depending on the sign of the spin temperature, which is then common to all of the three thermal reservoirs.
THE PROVOTOROV THEORY OF SATURATION:

Complementary to Redfield's ansatz, the Provotorov theory is able to predict the behavior of a spin system in a solid for the whole range of saturation as long as the amplitude $B_1$ is kept considerably smaller than the local field $B_I$. Because of its non-trivial quantummechnical background as well as its complexity the derivation will not be outlined here. Preferentially the reader is referred to M. Goldman's monograph about spin temperature effects in nuclear magnetic resonance [145]. However, the expression (52) can be used as a starting point for the discussion of the Provotorov theory. The restriction $B_1 \ll B_I$ can be accounted for by neglecting the term containing $B_1$. Thus, expressing all magnetic fields by their corresponding frequencies

$$\omega_S = \gamma_S B_0, \quad \omega_1 = \gamma_S B_1, \quad D = \gamma_S B_I$$

and introducing the symbol $\Delta = \omega_s - \omega$ for the difference between the Larmor and the rf- field frequency, equation (52) reads:

$$\frac{T_L}{T_{eq}} = \frac{\omega_S \Delta}{\Delta^2 + (\frac{T_L}{T_D}) \frac{D^2}{2}}$$

This expression is identical with the asymptotic (strong rf-field) solution for the common spin temperature in the rotating frame deduced from the Provotorov theory. The assumed smallness of $\omega_1$ in comparison to the line width $D$ is actually not a serious restriction, since for all practical cases a rf-amplitude can be found which is - although much smaller than the usual EPR line width of several Gauss - high enough to completely saturate the spin transitions. Under these conditions the spin system is still governed by only one spin temperature in the rotating frame $T_{eq}$ which determines the occupation distribution of both the dipolar and the Zeeman levels. However, in the general case of lower rf-amplitudes in which the system is either unsaturated or only partially saturated, two different temperatures are needed for a complete description of the system. One of the them, the Zeeman temperature, is a measure for the magnetization, i.e. for the occupation distribution of the Zeeman levels. For a spin-1/2 system this distribution reduces to the difference

$$n_- - n_+ \sim \exp \left( -\frac{h \omega_S}{2kT_Z} \right)$$

wherein $T_Z^{lab}$ denotes the Zeeman temperature in the laboratory frame. The other temperature, the frame independent spin-spin temperature $T_{SS}$, describes the non-Zeeman occupation distribution, i.e. the occupation numbers within the Zeeman levels $n_\pm$ according to the non-Zeeman interactions

$$n_\pm (\omega' - \omega_S) \sim \exp \left( -\frac{h(\omega' - \omega_S)}{2kT_{SS}} \right)$$

where the symbol $\omega'$ has been used in order to distinguish the running variable from the rf-frequency $\omega$. The occupation difference between the Zeeman levels as a function of the position $(\omega' - \omega_S)$ within the levels is then proportional to the combined expressions (56) and (57):

$$(n_- - n_+) (\omega' - \omega_S) \sim \exp \left[ -\frac{h}{2k} \left( \frac{\omega_S}{T_Z^{lab}} - \frac{\omega_S - \omega'}{T_{SS}} \right) \right] T_L \exp \left( -\frac{h \omega'}{2kT_L} \right)$$

The last equality is valid, if the system is in thermal equilibrium (TE). This case, in which both the Zeeman temperature in the laboratory frame and the spin-spin temperature are equal to the lattice temperature, is shown in Figure 30 (a). Figures (b) and (c) correspond to situations in which a rf-field with frequencies $\omega < \omega_S$ and $\omega > \omega_S$ is applied, respectively. Transitions are induced with probabilities $W(\Delta)$ (dashed arrows) which are frequency dependent by their proportionality to the natural line shape of the resonance $g(\Delta)$:

$$W = \pi \omega_S^2 g(\Delta)$$

Due to fast flip-flop processes within the spin system caused by the dipolar interaction there is no local saturation of the resonance line at the particular frequency $\Delta$. Instead, the influence of the rf-field is
Figure 30: Occupation distribution of a spin-1/2 system in a solid with no rf-field (a): \( T_Z = T_{SS} = T_L \) and with a saturating rf-field of frequency \( \Delta > 0 \) (b): \( T_{SS} > 0 \) or \( \Delta < 0 \) (c): \( T_{SS} < 0 \).

spread over the whole non-Zeeman distribution, which is now given by a spin-spin temperature \( T_{SS} \) different from \( T_{SS}^{ab} \). Its absolute value may be much lower than the lattice temperature and its sign depends on the sign of the frequency offset \( \Delta \).

In the framework of the Provotorov theory the problem is again treated in the rotating coordinate system. Here the effective field is reduced by the factor \( \Delta / \omega_S \). Due to the fact that the Zeeman occupation distribution must not change by a simple transformation of the coordinates the Zeeman temperature \( T_Z^{rot} \) – as viewed from the rotating frame – has to be smaller by the same factor. Defining the inverse Zeeman temperature \( \alpha = \hbar / kT_Z^{rot} \) and the inverse spin-spin temperature \( \beta = \hbar / kT_{SS} \) equation (58) reads

\[
(n_- - n_+) (\Delta') = \exp\left(-\frac{1}{2} (\alpha \Delta - \beta \Delta') \right), \tag{60}
\]

wherein the notation \( \Delta' = \omega_S - \omega' \) has been used. With only a weak rf-field present both inverse temperatures are given by their respective lattice values:

\[
\alpha_L = \frac{\omega_S}{\Delta} \frac{\hbar}{kT_L}, \quad \beta_L = \frac{\hbar}{kT_L} \tag{61}
\]

The situation changes completely, if the rf field is strong enough to at least partially saturate the Zeeman transitions. In this case the inverse Zeeman temperature \( \alpha \) decreases, but the inverse spin-spin temperature \( \beta \) increases due to a thermal contact between the Zeeman and the non-Zeeman reservoir, the latter being a consequence of the similar transition energies of both reservoirs in the rotating frame of reference. The behavior of the inverse spin temperatures after switching on a rf-field near resonance is given by the so-called Provotorov equations which can be found in [145] together with their rigorous derivations. In the steady state the solutions of these equations are:

\[
\frac{\alpha}{\alpha_L} = \frac{1 + WT_D(\Delta^2/D^2)}{1 + W(T_Z + (\Delta^2/D^2)T_D)} \tag{62}
\]

\[
\frac{\beta}{\beta_L} = \frac{WT_D(\Delta^2/S^2)}{1 + W(T_Z + (\Delta^2/D^2)T_D)} \tag{63}
\]

\[
\frac{\alpha - \beta}{\alpha_L} = \frac{1}{1 + W(T_Z + (\Delta^2/D^2)T_D)} \tag{64}
\]
In the limit of complete saturation \((WT_z, WT_D \gg 1)\) it is immediately seen that both spin temperatures are equal and consistent with the modified result (55) of the simple theory. On the other hand, if the system is essentially unsaturated \((WT_z, WT_D \ll 1)\), \(\alpha\) and \(\beta\) correspond to their respective lattice values as mentioned above.\(^6\) Figure 31 shows the behavior of the inverse spin temperatures under partial as well as under complete saturation as functions of the frequency difference normalized to the strength of the non-Zeeman interaction \(D\).\(^7\) As already stated as a result of the strong field case, the magnetization only vanishes for \(\Delta = 0\). Under complete saturation the common inverse spin temperature displays extrema

\[
\left| \frac{\beta_{\text{max}}}{\beta_L} \right| = \frac{\omega_S}{2D} \sqrt{\frac{T_D}{T_z}} = \frac{\omega_S}{2|\Delta_{\text{max}}|} \quad \text{located at} \quad \left( \frac{\Delta_{\text{max}}}{D} \right)^2 = \frac{T_Z}{T_D} \quad . \tag{65}
\]

Their absolute values are again the larger the smaller both the line width \(D\) and the ratio of the Zeeman to the dipolar relaxation time are.

Figure 31: Magnetization \(\langle M_z \rangle / \langle M_0 \rangle = \alpha / \alpha_L\) and inverse spin-spin temperature \(\beta / \beta_L\) under partial saturation (dashed curves) and complete saturation (solid curves) as functions of the normalized frequency difference. In this example the Zeeman and the dipolar relaxation times \(T_Z\) and \(T_D\) are assumed to be equal.

**Coupling of the Nuclear Zeeman to the Electron Non-Zeeman Reservoir:**

As already mentioned in the previous section the very fact that the nuclear Larmor frequency is of similar size as the local frequency \(D\) of the electrons leads to an exchange of energy between the two systems. The corresponding polarization mechanism is usually called 'thermal mixing'. The consequences of the coupling of both reservoirs may be understood in analogy with two bodies of certain heat capacities being in thermal contact with each other.

The heat capacity of a spin system with respect to a certain interaction \(\mathcal{H}\) is defined by:

\[
C = \text{tr}\{\mathcal{H}^2\} \quad . \tag{66}
\]

In the high temperature approximation the heat capacities \(C_Z^S\) of the electron Zeeman reservoir and \(C_D^S\) of the electron dipolar reservoir can be calculated in a straightforward manner. In a coordinate system rotating with a frequency close to the electron Larmor frequency they are

\[
C_Z^S = \hbar N_S \frac{S(S+1)}{3} \Delta^2 \quad , \quad C_D^S = \hbar N_S \frac{S(S+1)}{3} D^2 \quad . \tag{67}
\]

\(^6\) In equation (63) the term containing \(\beta_L\) has been neglected.

\(^7\) To simplify matters, the dependency of \(\beta\) on the lineshape function \(g(\Delta)\) in the case of partial saturation is ignored, i.e. the transition probability \(W\) is assumed to be independent of \(\Delta\).
where $N_s$ denotes the electron concentration. Already the mechanism of the dynamical cooling of the electron dipolar reservoir can be thought of as a thermal contact of two bodies in the rotating frame, one of heat capacity $C^D_2$ and one of heat capacity $C^S_2$. Furthermore, the advantage of a small non-Zeeman interaction - parameterized by the local frequency $D$ - gets a simple interpretation: The common equilibrium temperature of two bodies in thermal contact is the lower the smaller the heat capacity of the body, which is to be cooled.

With the nuclear Zeeman reservoir having a heat capacity

$$ C'_2 = \hbar N_I \frac{I(I+1)}{3} \omega_I^2 $$

(68)

a third body enters the game. Due to its close thermal contact to the electron dipolar reservoir the body, which is now to be cooled, consists of the combined electron dipole and nuclear Zeeman system with heat capacity

$$ C = C'_2 + C^S_2 = \hbar N_I \frac{I(I+1)}{3} \omega_I^2 + \hbar N_S \frac{S(S+1)}{3} D^2 $$

(69)

It may be rewritten in the form

$$ C = \hbar N_s \frac{S(S+1)}{3} D^2 $$

(70)

by which a combined local frequency

$$ D^2 = D^2 + \frac{\omega_I N_I I(I+1)}{N_S S(S+1)} $$

(71)

is defined. Due to the much larger number $N_I$ of nuclear spins compared to the number $N_s$ of electron spins one may argue that the cooling efficiency of the combined body is worse by roughly a factor $N_S/N_I = 10^{-4} \ldots 10^{-3}$. But luckily the coupling of the nuclear spins forces not only the local frequency to be changed, but also the dipolar relaxation time. With no leakage relaxation channels present, $T_D$ is enhanced by the same factor as the local frequency

$$ T_D = T_D \frac{C'_2 + C^S_2}{C^S_2} = T_D \frac{D^2 + \omega_I (N_I/N_S) (I(I+1))/(S(S+1))}{D^2} $$

(72)

with the result that the equation (65), which now has to be interpreted as the maximum inverse spin temperature for the combined system, stays unchanged. The replacement of $T_D$ by $T'_D$ takes into account nothing else than the fact, that the time, which is needed by a body to heat up, is prolonged by the same factor as its heat capacity is enlarged. The only assumption here is that no further 'heat leaks' are introduced due to the coupling of the nuclear to the electron reservoir. In practice, however, such relaxation channels may exist. Although not yet experimentally proven the most obvious one is based on the spin-phonon coupling of a system with $I > 1/2$ due to the interaction of the particles quadrupole moment with the lattice electric field gradient at the site of the particle. In order to account for these effects, a leakage factor $f$ may be defined such that the actual nuclear relaxation rate is given by:

$$ \frac{1}{T'_I} = \frac{1}{T_D}(1 + f) $$

(73)

The maximum inverse spin temperature then reads:

$$ |\beta|_{\text{max}} = \frac{\omega_S}{2|\Delta_{\text{max}}|} \beta_L = \frac{\omega_S}{2\sqrt{T'_D/T_D(1 + f)}} \beta_L \quad \text{with} \quad \Delta_{\text{max}}^2 = \frac{T'_D}{T_D} (1 + f) D^2 $$

(74)

With these results an expression for the maximum nuclear polarization in the high temperature approximation can be given:

$$ |P_{\text{max}}| = \frac{|I|}{I} \left( \frac{I+1}{3} \right) \omega_I |\beta_{\text{max}}| = \left( \frac{I+1}{3} \right) \omega_S \beta_L \frac{\omega_I}{2|\Delta_{\text{max}}|} $$

(75)
For instance in the case of a proton system \((I = \frac{1}{2})\), whose Larmor frequency is equal to the electron line width at a given magnetic field, one gets for zero leakage

\[
|P_{\text{max}}^n| = \frac{1}{2} \left( \frac{\omega_S \beta_L}{2} \right),
\]

wherein the ratio \(T_Z/T_D\) is set to unity. It is half of the thermal equilibrium electron polarization under the given conditions and corresponds to a dynamical enhancement of \(\omega_S/2\omega_I \simeq 330\).

**THE RESOLVED SOLID STATE EFFECT FROM THE THERMODYNAMICAL POINT OF VIEW:**

The spin temperature theory is not only able to describe the situation, in which a thermal contact exists between the electron dipole and the nuclear Zeeman reservoir, i.e. when the condition \(\omega_I \approx D\) is fulfilled. It also includes the opposite case of a complete separation of the two systems. If at a given magnetic field the nuclear Larmor frequency is considerably larger than the electron line width \((\omega_I \gg D)\), it is not possible to realize a cooling of both of the reservoirs with the same frequency offset \(\Delta\). For instance with the choice \(\Delta \approx D\) the electron dipole reservoir is efficiently cooled but the nuclear Zeeman reservoir is not, because the energy difference \(\hbar \omega_I\) cannot be balanced by a change in the distribution of the electron non-Zeeman occupation numbers. Nevertheless, the application of a strong rf-field with frequency offset \(\Delta\) leads in any case to a cooling of the electron Zeeman reservoir. Without a thermal contact to another 'body' its inverse temperature, viewed from the rotating frame of reference, is:

\[
\alpha_S = \alpha_L = \beta_L \frac{\omega_S}{\Delta}
\]

If for the frequency offset \(|\Delta| = \omega_I\) is chosen, the nuclear Zeeman system is directly coupled to the electron Zeeman system via forbidden simultaneous spinflips (solid state transition) of an electron-nucleus pair and is thus be cooled without any participation of the electron dipolar reservoir. The equivalence of this picture of the resolved solid effect with the results from section 5.2.1 may be seen as follows.

In the strong rf-field limit the inverse Zeeman temperature \(\alpha_S\) of the electrons is given by

\[
\alpha_S = \alpha_L = \beta_L \frac{\omega_S}{\Delta} \frac{\omega_S \Delta}{\Delta^2 + (T/Z/T_D) D^2}
\]

where the quantities \(D\) and \(T_D\) should be understood as a measure of the heat capacity and the relaxation time of any 'body' being in thermal contact to the electron system. If this 'body' is identified with a nuclear spin-1/2 system, one has to make the following identifications:

\[
\Delta = \omega_I, \quad T_D = T_I \quad \text{and (see equation (71))} \quad D^2 = \omega_I^2 \frac{N_I}{N_S}
\]

With the assumption of a common equilibrium temperature of the nuclear and the electron Zeeman reservoir equation (78) results in

\[
\alpha_I = \alpha_S = \beta_L \frac{\omega_S}{\omega_I} \frac{1}{1 + \frac{N_I}{N_S} \frac{T_S}{T_I}}
\]

leading to

\[
\alpha_I = \alpha_L = \beta_L \frac{\omega_S}{\omega_I} \quad \text{for} \quad \frac{N_I}{N_S} \frac{T_S}{T_I} \ll 1
\]

Thermodynamically interpreted the last condition claims that the temperature of the electron Zeeman reservoir keeps its lattice value, even if the nuclear Zeeman reservoir is coupled to it. Again it is fulfilled, if there is no leakage relaxation of the nuclear spins. For the nuclear polarization one gets the same result as from the elementary treatment of section 5.2.1

\[
P_L = \frac{1}{2} \omega_I \alpha_I = \frac{1}{2} \omega_S \beta_L = P_{\text{S}}^L
\]
Compared to the expression (76) for the situation of an additional involvement of the electron dipole reservoir, the maximum nuclear polarization in the 'non-contact' case is twice as high. From the point of view of the spin temperature theory the higher efficiency of the resolved solid effect is caused by the absence of the nuclear relaxation channel via the coupling of the electron dipolar reservoir to the lattice. This explains the extremely high dynamical enhancement of the proton polarization in the early target material LMN, which up to now has never been achieved again in any of the later materials at such low $B_0/T_L$ values. The particular challenge is to create a paramagnetic system in the respective material, whose non-Zeeman interaction is low enough to avoid the coupling of the corresponding reservoir to the nuclear Zeeman reservoir. But luckily, although the resolved solid state effect is (at least theoretically) the most effective DNP process, it is not the only way to achieve high dynamic polarizations. A situation, in which the electron non-Zeeman interaction is not much stronger than the nuclear Zeeman interaction, also allows a high degree of nuclear cooling and thus a high degree of polarization. In proton target materials this demand may rather easily be met because of the comparatively high proton Larmor frequency. On the contrary, special care has to be taken in choosing an appropriate paramagnetic system for materials, in which nuclei with much smaller magnetic moments (e.g. deuterons) are to be polarized.

As one of the few existing examples for an almost pure solid state effect Figure 32 shows the polarization versus the applied microwave frequency in propanediol. This material was doped with a methyl type stable radical, which possesses only a very weak non-Zeeman interaction. The dominating polarization mechanism is given by the resolved solid effect, whereas the process of thermal mixing plays only a minor role.

5.2.3 Borghini's Equations

With the exception of the solid state rate equations outlined in section 5.2.1 so far the theoretical considerations were restricted to the case of a small Zeeman energy $\langle H^2_z \rangle$ of the electron spins compared...
to the thermal energy $kT_L$ of the lattice (high temperature approximation). Due to the fact that this assumption is far from being valid under the usual conditions of a polarization experiment it is necessary to find a way, which allows the predictions of the spin temperature theory to be tested also in the low temperature regime. Fortunately it is possible to do so under the following special conditions for the electron system:

1. The EPR line consists of a number of individual spin packets, whose respective Larmor frequencies $\omega_i^S$ differ from the free value $\omega_S$ due to the presence of an anisotropic g-factor or due to a hyperfine interaction with the surrounding nuclei. The frequency deviations of the individual spin packets are quantified by $\Delta_i = \omega_S - \omega_i^S$.

2. The dipolar interaction is weak enough for the EPR line width to be almost exclusively given by the inhomogeneous interactions mentioned above, but it is strong enough to ensure the existence of a common spin temperature for the occupation distribution of all spin packets.

3. The electron non-Zeeman and the nuclear Zeeman interaction reservoir share at all times the same temperature due to a strong coupling between them. Then the nuclear polarization is given by the Brilliouin function $B_i(x)$ as the generalization of Curie's law for arbitrary ratios of $B/I$, in which the lattice temperature is simply replaced by the common spin temperature:

$$B_i(x) = \frac{1}{2} \frac{\tanh \left( \frac{1}{2} \beta \omega_i \right)}{\tanh \left( \frac{1}{2} \beta \omega_i \right) + \tanh \left( \frac{1}{2} \beta \omega_i \right)}$$

Instead of going through the details, which can be found in [134], only the basic idea as well as some consequences of Borghini's equations will be discussed. Furthermore the following presentation is restricted to the simpler one of the two cases, in which the non-Zeeman interaction consists of a spread of the g-factors of the individual spin packets, a situation most common to solutions of stable chemical radicals.

The Hamiltonian of a combined nuclear and electron spin system, which consists of $n$ spin packets with Larmor frequencies $\omega_i^S$, is given by:

$$\mathcal{H} = \hbar \omega_S S_z - \hbar \sum_{i=1}^{n} \Delta_i S_z^i - \frac{\hbar \omega_i I_z}{\mathcal{H}_{I_z}}$$

wherein $S_z^i$ denotes the average magnetic spin quantum number of packet $i$. The EPR line shape is determined by the statistical weights $f_i$ of the packets, which are normalized to unity and centered around $\omega_S$:

$$\sum_i f_i = 1, \quad \sum_i \Delta_i f_i = 0$$

The respective polarizations of the packets under the action of a rf-field of frequency $\Delta$ are given according to equation (60) by:

$$P_i^f = \frac{1}{2} \beta (\Delta - \Delta_i) \tanh \left( \frac{1}{2} \beta \omega_i \right)$$

If the amplitude of the rf-field is of saturating strength, the Zeeman and the non-Zeeman reservoir share the same temperature in the rotating frame of reference. With $\alpha = \beta$ equation (87) simplifies to:

$$P_i^f = \frac{1}{2} \beta (\Delta - \Delta_i)$$
Using the polarizations instead of the magnetic spin quantum numbers the expectation values of the different parts of the Hamiltonian read:

\[
\langle \mathcal{H}^S_{\perp} \rangle = -\frac{1}{2} N_S \hbar \omega_S \sum_i f_i P^i_S \\
\langle \mathcal{H}^S_{nz} \rangle = \frac{1}{2} N_S \hbar \sum_i \Delta_i f_i P^i_S \\
\langle \mathcal{H}^I \rangle = -I N_I \hbar \omega_I P_I
\]

Taking into account the coupling of \( \langle \mathcal{H}^S_{nz} \rangle \) to \( \langle \mathcal{H}^I \rangle \) one has to evaluate the conditions

\[
\frac{d}{dt} \langle \mathcal{H}^S_{\perp} \rangle = 0 \quad \text{and} \quad \frac{d}{dt} \langle \mathcal{H}^S_{nz} + \mathcal{H}^I \rangle = 0
\]

in order to find an equation for the stationary polarizations of the spin packets

\[
\sum_i f_i P^i_S \left( \Delta - \frac{T_S}{T_D} \Delta_i \right) = \Delta P^I_S + 2 I \frac{N_I T_S}{N_S T_I} \omega_I P_I - 0
\]

Herein \( P^I_S \) denotes the common equilibrium (lattice) polarization of the spin packets, \( T_S \) and \( T_D \) are the Zeeman and the dipolar relaxation times of the electrons, respectively. With the help of equation (88) an equivalent expression for the inverse spin temperature \( \beta \) can be written down:

\[
\sum_i f_i (\Delta - \delta \Delta_i) \tanh \left( \frac{1}{2} \beta (\Delta - \Delta_i) \right) = \Delta P^I_S \quad \text{with} \quad \delta = \frac{T_S}{T_D}
\]

In going from equation (91) to equation (92) it was once again used that the number of electron relaxation processes per unit time \( N_S/T_S \) is much larger than those \( N_I/T_I \) of the nuclei. The replacement of the sum over the statistical weights \( \sum_i f_i \) by an integral over the corresponding distribution function \( \int d\Delta' f(\Delta') \) leads to Borghini’s equation

\[
\int_{-\infty}^{+\infty} f(\Delta') (\Delta - \delta \Delta') \tanh \left( \frac{1}{2} \beta (\Delta - \Delta') \right) d\Delta' = \Delta P^I_S
\]

for the case of a fixed occupation distribution for the spin packets. In contrast to this situation, which is usually caused by a g-factor anisotropy, the corresponding equation for the case of a hyperfine broadened EPR line is more complicated. Here the occupation distribution of the spin packets, i.e. the EPR line shape, depends on the actual nuclear polarization. Thus, the particular microwave frequency, which provides an optimum cooling of the nuclear Zeeman reservoir, varies during the polarization process. This prediction, which is a consequence of the extension of the spin temperature theory to high values of \( B_0/T \), was in fact observed in highly polarized \( ^6\text{LiD} \). In this material the diluted electron system consists of so-called F-centers, whose EPR is governed by a hyperfine interaction of the unpaired electron with six adjacent \( ^6\text{Li} \) nuclei. The derivation as well as a discussion of Borghini’s equation for the case of hyperfine broadening is given in [134].

In general, equation (93) is an integral equation for \( \beta \), which has to be solved numerically. Nevertheless, using the conditions in (86) it is easy to show that for high temperatures, i.e. for

\[
P^I_S \approx \frac{1}{2} \beta_L \omega_S \quad \text{and} \quad \tanh \left( \frac{1}{2} \beta (\Delta - \Delta') \right) \approx \frac{1}{2} \beta (\Delta - \Delta')
\]

it reduces to:

\[
\beta_L \omega_S \Delta = \beta \left( \Delta^2 + \delta \int_{-\infty}^{+\infty} \Delta'^2 f(\Delta') d\Delta' \right)
\]

The above integral is nothing else but the definition of the EPR line width. Its identification with the quantity \( D \), used so far for the strength of the dipolar energy, reproduces the high temperature expression for \( \beta \) under strong rf-field conditions

\[
\beta = \beta_L \frac{\omega_S \Delta}{\Delta^2 + \delta D^2}
\]
Figure 33 shows the results of a numerical calculation for the general case, in which the EPR line was assumed to be Gaussian with a width equal to \( D \). In diagram (a) the temperature coefficient \( \beta D \) as a line width independent measure of the inverse nuclear Zeeman temperature is plotted versus the normalized frequency offset \( \Delta/D \) for different electron polarizations. The most important features of the low temperature solutions are:

1. The maximum inverse spin temperature considerably increases with the electron polarization even when \( P_L^S \) is almost unity.

2. The frequency offset \( \Delta(\beta_{\text{max}})/D \), at which the maximum occurs, increases with an increasing electron polarization.

![Figure 33](image)

Figure 33: Numerical solution of equations (92) or (93), respectively, for a Gaussian EPR line of width \( D \) and for \( \delta = T_S/T_D = 1 \). (a) shows the positive side of the frequency curves for different electron polarizations. The straight lines (proportional to \( \Delta/D \)) as well as the asymptotic curves (proportional to \( (\Delta/D)^{-1} \)) correspond to approximative solutions of the above equations. In (b) the maximum nuclear Zeeman temperatures and their respective frequency positions are drawn versus the electron Boltzmann factor \( \beta_L \omega_S \), which determines the electron polarization \( P_L^S = \tanh(1/2 \beta_L \omega_S) \). The error bars represent the uncertainty due to the final numerical resolution in \( \Delta/D \).

The behaviors of both, the maximum value of the inverse nuclear Zeeman temperature and its frequency position as functions of the electron Boltzmann factor \( \beta_L \omega_S \) are shown in diagram (b). The dependence
Thus Borghini's equation delivers not only a natural extension of the Provotorov theory down to low temperatures, a comparison of equation (97) with equation (74) shows that the expressions for the maximum inverse spin temperatures are even identical in both cases within the uncertainties due to the numerical calculation. This result is most important in two respects: Firstly, it provides a thorough understanding of the DNP process in the most common situation, when the 'naive' model of the solid state effect is not applicable. Secondly, the parameters of the electron spin system, i.e. the EPR line width $D$ and the ratio of the relaxation times $T_S/T_D$, keep their relevance for the achievable nuclear polarization also under the realistic conditions of high magnetic fields and low temperatures. This statement is of particular importance with respect to future efforts in optimizing the performance of polarizable target materials.

5.2.4 Thermal Mixing and Equal Spin Temperatures

In the literature the term 'EST theory' (equal spin temperature theory) is frequently used as a synonym for the polarization mechanism via thermal mixing of the electron non-Zeeman and the nuclear Zeeman reservoir. It originates from the common observation that the polarizations of different nuclei in the same material can be calculated from their respective Brillouin functions using a single spin temperature. This behavior was explicitly demonstrated in a variety of different target materials. Examples are the partially and fully deuterated alcohols and diols (all of them chemically doped), in which the proton polarization was either compared to the deuterion or to the $^{13}$C polarization [146, 147, 148]. Recently a similar study was performed in $^6$LiD [149], which is used as the polarized target material for the COMPASS (NA58) experiment at CERN. In this case the polarizations of three different nuclei, $^6$Li, deuterion and $^7$Li (unsaturated residual of $\approx 5\%$) were measured during the build-up phase of the polarization process. In Figure 34 the polarizations of the two different lithium nuclei are plotted versus the deuteron polarization together with the respective predictions. They behave in perfect agreement with the assumption of a common Zeeman temperature for all nuclei with a lowest value of less than $1mK$. Unfortunately, the determination of the polarization of the unsubstituted protons was not possible because of their very low concentration (less than 0.2% of the deuterons). This measurement would have been of particular interest, because the protons are the only nuclear spin species in the material, whose Larmor frequency $\omega_p$ is considerably larger than the EPR line width. On the basis of the picture of the DNP outlined in this chapter it is most probable that an rf-irradiation near $\omega_S \pm \omega_p$ would change the proton Zeeman temperature independently of the common Zeeman temperature of the other nuclei. A related situation is known from the polarization behavior of ammonia, in which the Larmor frequency of the $^{14}$N nucleus is smaller than the line width of the radiation induced NH$_2$ radical, but the proton Larmor frequency is a factor of two larger. In this material a common Zeeman temperature of both nuclei is observed only under the action of the microwave field, which provides a 'forced' thermal contact between both species and the electron system [150]. With no rf-field present the 'spontaneous' contact between the proton Zeeman and the electron non-Zeeman reservoir is not strong enough to avoid a separation of the two temperatures caused by the respective relaxation processes. These examples show that also within the same material different DNP mechanisms may be active. Thus, a discrepancy of one of the nuclear Zeeman temperatures with respect to others not necessarily means that the process of thermal mixing is completely absent. It simply means that the Zeeman reservoir of this particular spin species does not share the electron non-Zeeman temperature, which is common to the other nuclear species. The only known reason for a behavior like that is the impossibility of a nuclear spin flip to be energetically balanced by simultaneous spin flips of two electrons. Such a situation occurs, if the transition energy for both processes are too different, i.e. if the nuclear Larmor frequency is considerably larger than the electron line width.
5.3 Equipment and Techniques

Figure 35 shows a schematic of a generic polarized target, showing all the necessary sub-systems. The polarizable material is cooled to a low temperature (≤ 1 K), placed in a homogeneous magnetic field of 2.5T or higher and irradiated with microwaves. A NMR system measures the degree of polarization. Not shown are the standard vacuum and pumping systems and instrumentation.

Parallel to the developments of new target materials (see Sections 5.5) great efforts have been made to improve the $\frac{P_0}{P}$ ratio and hence the achievable polarizations.

After many years of operation with $^4$He evaporation refrigerators, $^3$He evaporation refrigerators started to be used and provided the ability to cool to ≈ 0.5K rather than the 1 K possible with $^4$He. Since the polarization is related to $\exp \left( -\frac{\mu B}{kT} \right)$, the achievable polarization could, in fact, be doubled (see Figure 36). However, the thermal properties of $^3$He limit the amount of beam intensity that can be tolerated before the polarization is reduced to an unacceptable level.

$^3$He/$^4$He dilution refrigerators soon followed. As they became easier and more reliable to use, they supplanted the $^3$He refrigerator. For polarizing, temperatures in the range of 150 – 400 mK could be maintained and lowered to ≈ 50 mK for frozen spin operation when the microwaves were turned off (see section 5.8). This added flexibility and ease of operation, similar to a $^3$He refrigerator, has made the dilution refrigerator the current choice for a large range of polarized target experiments.

5.3.1 Cryogenics

$^4$He and $^3$He Evaporation Refrigerators:

This type of horizontally working refrigerator, designed by Roubeau [151] and shown in Figure 37, was used in the first polarized target experiment [1] and copied by many laboratories.

Here, liquid $^4$He is fed into a separator pot where the liquid phase is separated from the vapor phase by a sintered copper plate. The cold vapor is pumped away and used to cool the radiation shields and baffles that intercept the radiation heat load. Liquid helium flows through the separator plate into a heat exchanger and then is metered into the target holder (or evaporator) via a needle valve. The pool of liquid in the target holder is pumped on by large capacity Roots pumps to reduce the temperature to
Figure 35: A generic polarized target showing the major sub-systems and typical operational parameters. Not shown are the standard vacuum and Roots pumping systems.

Figure 36: Positive $\sigma$ and negative $\pi$ polarizations observed in propanediol as a function of the lattice temperature $T$. From Ref. [148]

As the cold vapor is pumped away, it exchanges heat with and cools the incoming warm liquid. Services such as microwaves and NMR are also brought into the target cavity. For the best polarization performance, it is necessary to operate the $^4\text{He}$ refrigerator with a high magnetic field of $5T$ as is the case with the University of Virginia target operating at SLAC and TJNAF (see section 5.8.1).

In the $^3\text{He}$ evaporation refrigerator, a $^4\text{He}$ refrigerator as described above is wrapped around and mechanically isolated from the $^3\text{He}$ section. In a more elegant version [152], the $^4\text{He}$-part is directly incorporated into the $^3\text{He}$ refrigerator. The incoming $^3\text{He}$ gas is cooled and eventually liquefied by the $^4\text{He}$ section. It proceeds via heat exchangers and a metering valve into the target holder in a similar way to that described for the $^4\text{He}$ refrigerator. The liquid $^3\text{He}$ is pumped on by sealed Roots pumps to obtain temperatures of $\approx 0.5\, K$. Because the gas is costly, it is recirculated through the refrigerator into a sealed system, which includes the pumps. $^3\text{He}$ refrigerators are now rarely used in polarized targets because of the versatility of $^3\text{He}/^4\text{He}$ dilution refrigerators.

$^3\text{He}/^4\text{He}$ Dilution Refrigerator:
The operation of a $^3\text{He}/^4\text{He}$ dilution refrigerator is based on special properties of liquid mixtures of $^4\text{He}$ and $^3\text{He}$. Above $0.88\, K$ $^4\text{He}$ and $^3\text{He}$ are mixable in any ratio, whereas below $0.88\, K$ the liquid consists of two separate phases: a diluted phase, which is $^3\text{He}$ poor and a $^3\text{He}$ rich 'concentrated' phase [153].
Important for the operation is the fact that in the diluted phase there is a reasonable amount of $^3$He (6.6%), down to the lowest temperatures, that can be pumped and used for cooling. The cooling takes place when a $^3$He atom is removed from the concentrated phase to the diluted phase. The dilution unit of a $^3$He/$^4$He refrigerator consists of a mixing chamber, containing the target material, a heat exchanger and a still. The still temperature is normally adjusted via a heater to 0.8 K. Because of its much higher vapor pressure, compared to $^4$He, the $^3$He is preferentially boiled off and recirculated. It is liquified inside the refrigerator by heat contact with a separate $^4$He circuit and then proceeds through different heat exchangers into the mixing chamber, where the target material can be cooled to about 50 mK. The flow diagram and the $^3$He concentration of a schematic dilution unit is shown in Figure 38. A remarkable cooling power can be attained even at the lowest temperature by the development of special heat exchangers for the dilution part [154, 155]. The cooling power of the world largest $^3$He/$^4$He dilution refrigerator is about 1 mW at 50 mK, 15 mW at 100 mK, 400 mW at 300 mK and reaches 1.3 W at 500 mK; a set of eight Roots pumps in series with a nominal pumping speed of 13500 m$^3$/h for $^3$He is used [156]. Nowadays target volumes between 0.2 cm$^3$ and 3000 cm$^3$ can be handled by fast loading the mixing chamber of various types of $^3$He/$^4$He dilution refrigerators [154, 157, 158]. These are the standard devices for future modern experiments with low intensity particle beams (see section 5.8.2).

HEAT DISSIPATION AND KAPITZA RESISTANCE:
A specific feature of $^3$He is its superfluidity below 2.17 K. The high thermal conductivity (even better than the best metallic conductors) and the good circulation around the target material due to super-
fluidity and film flow make $^4$He a particularly suitable cooling agent. High cooling powers of $\approx 2 \, \text{W}$ can be achieved with specially designed $^4$He refrigerators, which allow high $^4$He mass flows across the liquid-gaseous boundary to be pumped away at low pressures ($\approx 0.1 \, \text{mbar}=1 \, \text{K}$) with sufficiently high pumping speed. Thus $^4$He refrigerators operating at 1 $\text{K}$ are best suited to withstand relatively high heat input from particle beams. However, this additional heat input, which can be tolerated without affecting the polarization too much, is limited by the thermal boundary resistance between liquid helium and solid referred to as Kapitza resistance $\rho_{\text{kap}}$ [159]. It arises from the fact that the acoustic impedances, i.e. the product of the density and first sound velocity of most solids differ by more than two orders of magnitude from that of liquid helium. The acoustic mismatch interferes with the passage of thermal phonons across the interface so that their transmission is reduced and the target material (beads) temperature is higher than the measured bath temperature [160].

The temperature dependence of the Kapitza resistance is predicted as $\rho_{\text{kap}} \approx T^{-3}$, which has practical consequences for the construction of $^3$He/$^4$He dilution refrigerators. That means, that it is not necessary to achieve temperatures below 30 $\text{mK}$, as many dilution refrigerators do, since in this temperature range the Kapitza resistance of the target material prevents an efficient removal of the beam heating from the target beads. This fact and the smaller cooling power of $^3$He/$^4$He dilution refrigerators limit their use for experiments with low intensity particle beams up to $1 \times 10^6$ particles/s or to about $10^8$ particles/s in the so-called frozen spin target operation mode (see section 5.8).

### 5.3.2 Polarizing Magnets

The polarization that can be obtained with a given material is linked to the value of $\frac{B_p}{T}$. Early polarized targets used iron magnets operating at fields of $2 - 2.5 \, \text{T}$ and operated at temperatures of $1 - 1.5 \, \text{K}$ so that $\frac{B_p}{T}$ was $\approx 2$. The development of low temperature refrigerators and high field superconducting magnets meant that $\frac{B_p}{T}$ of 50 - 100 were easily achievable. But for various technical reasons DNP is applied mainly at $\frac{B_p}{T} \approx 5 - 10$.

The main limitation of the iron magnets is that, in order to boost the operational field to $2.5 \, \text{T}$ and maintain the required uniformity, a substantial mass of iron and copper coil restricts access and reduces the acceptance of the experiment. This is particularly true if the polarization direction is required to be in the plane of scattering. Superconducting magnets that could be built with large material-free apertures changed this. Magnets built in the Helmholtz configuration that have a 100° aperture for scattering particles in the forward direction and about 35° for scattering near 90° are now available [161]. Nearly a 4$\pi$ aperture for the outgoing particles can be achieved by operating the targets in the frozen spin mode. For such applications superconducting solenoid magnets are available up to 7 T, which must be used in particle experiments together with so-called holding magnets (see section 5.8.2).

### 5.3.3 Microwaves

The frequency needed for DNP is about $28 \, \text{GHz}/T$, i.e. $140 \, \text{GHz}$ at 5 $\, \text{T}$. As a rough number the required power at a temperature of 1 $\, \text{K}$ is $1 - 2 \, \text{mW/g}$ of target material at $2.5 \, \text{T}(70 \, \text{GHz})$ and $\approx 20 \, \text{mW/g}$ at $5 \, \text{T}(140 \, \text{GHz})$. The required power is also material dependent. The highest power devices available at these frequencies are extended interaction oscillators (EIO); for lower power applications, klystrons, IMPATT- and Gunn diodes are available. Cericinotrons, which used to be the tube of choice, are no longer manufactured. The power that can be extracted from a tube is inversely related to its operational frequency, and the power absorption in microwave components increases with frequency. Therefore, from the standpoint of microwave power, there is at present a practical limit for operation at about 210 $\, \text{GHz}$, corresponding to 7.5 $\, \text{T}$, unless very low mass targets are being considered.

The power supply for a given tube should be capable of frequency modulating the microwaves by up to about $\pm 40 \, \text{MHz}$ around the central frequency. In some materials substantial improvements in maximum polarizations and increased polarization rate have been observed with the frequency modulation method [162].
5.4 Nuclear Magnetic Resonance and Polarization Measurement

5.4.1 Principles of the Nuclear Magnetic Resonance

The nuclear magnetic resonance was discovered in the year 1945 by F. Bloch [163] and E.M. Purcell [164]. If a magnetic field is applied to an ensemble of nuclear spins I, it exhibits \((2I + 1)\) magnetic sublevels (Zeeman levels) with energies \(E_{\text{mag}}(m) = m\hbar\omega_0\), where \(m = -I, -I + 1, \ldots, I\) denote the magnetic quantum number and \(\omega_0\) the Larmor frequency. In thermal equilibrium the population among the sublevels is given by the Boltzmann distribution:

\[
N(m) \sim \exp \left( -\frac{E_{\text{mag}}(m)}{kT} \right)
\]

The polarization at a temperature \(T\) and at a magnetic field \(B_0\) can be calculated from the Brillouin-function

\[
P_I = \frac{2I + 1}{2I} \coth \left( \frac{2I + 1}{2I} x \right) - \frac{1}{2I} \coth \left( \frac{1}{2I} x \right)
\]

with \(x = g\mu_N B_0 (kT)^{-1}\). Herein \(g\) is the g-factor and \(\mu_N\) the nuclear magneton.

The macroscopic magnetization \(\vec{M}\) of the sample is connected to the polarization of the ensemble \(P_I\), consisting of \(N\) spins in a volume \(V\), by

\[
\vec{M} = N g\mu_N \vec{P}_I - \sum_i \frac{\vec{\mu}_i}{V},
\]

where \(\vec{\mu}_i\) denotes the magnetic moment of each individual spin. A rotating rf-field with amplitude \(\vec{B}_1\), which is generated by a rf-resonance circuit with frequency \(\omega_1\), is applied perpendicular to the axis of the external magnetic field \(B_0\). If one superimposes \(\vec{B}_1\) onto \(B_0\), transitions can take place among the nuclear Zeeman levels, whose probability depends on the difference between the rf-field frequency \(\omega_1\) and the Larmor frequency \(\omega_0\). Furthermore the number of transitions per unit time is proportional to the population difference of the levels and is thus a measure for the polarization of the spin system. Depending on its sign energy is absorbed from or emitted into the rf-coil, which surrounds the sample. Correspondingly one measures a net energy loss or gain induced into the rf-circuit. Speaking in terms of the macroscopic magnetization the principle of the CW (continuous wave) NMR experiment consists of measuring the voltage which is induced by the transverse magnetization \(\vec{M}_\perp\) as it sweeps through the rf-coil with frequency \(\omega\) of the rf-field.

To simplify matters the Bloch equations are used to give an explicit expression for the behavior of the transverse magnetization, even though they are valid only for liquids and gases where spin temperature effects are absent. In addition it is assumed that the two following conditions are fulfilled:

- The rf-frequency is varied slowly enough through the resonance in order to keep the spin system in an internal as well as in an external equilibrium at all times.
- The amplitude of the rf-field is chosen weak enough to ensure the spin system to be essentially unsaturated \((\omega_1^2 = \gamma^2 B_1^2 \ll 1/T_1 T_2)\).

Then the transverse magnetization can be expressed by

\[
\vec{M}_\perp = M_\perp \begin{pmatrix}
\cos(\alpha) \\
\sin(\alpha) \\
0
\end{pmatrix}
\]

with

\[
M_\perp = \sqrt{M_u^2 + M_v^2} = \frac{\omega_1 T_2 M_z}{\sqrt{1 + T_2^2 (\omega_0 - \omega)^2}}
\]

where the angle \(\alpha\) between the transverse magnetization and the rf-field is determined through the relation \(\tan(\alpha) = M_u/M_v = (T_2 (\omega_0 - \omega))^{-1}\). Figure 39 illustrates the situation as seen from a frame of reference \((x', y', z' = z)\), which rotates around the axis of the external (static) magnetic field with the frequency of the rf-field.
Figure 39: Transverse magnetization $\tilde{M}_1$ in the rotating frame. When $\omega \approx \omega_0$ within the width of the resonance line, $M_1$ is different from zero. Seen from this frame the magnetization is static, whereas the rf-coils rotates, which causes a voltage being induced into it.

In order to cover the whole resonance line, the rf-frequency is swept from a value of $\omega$ lower than the Larmor frequency $\omega_0$ through resonance up to a value $\omega > \omega_0$. At the same time the transverse magnetization describes a circle in the $(x', y')$ plane. Exactly at resonance the absorptive part of the magnetization $M_1$, i.e. the component along $y'$ being 90° out of phase with respect to $B_1$ is at maximum. The dispersive part $M_d$ (along $x'$ in phase with $B_1$) vanishes at that point. Although in principal both absorption and dispersion can be used in order to determine the polarization, it is usually the component $M_1$, which is registered in CW NMR experiments. In the unsaturated case, which is the common choice in order not to diminish the polarization due to its measurement, $M_1$ is proportional to the unsaturated line shape function, a statement which holds for liquids and gases as well as for solids.

5.4.2 Q-Meter-Technique

In order to calculate the power, which is gained or lost by the rf-circuit due to the induced spin transitions, usually the susceptibilities are used instead of the magnetizations. The susceptibility of a system is defined as the constant of proportionality between the magnetization and the applied magnetic field.

$$M_1 = \chi_0 H_0 = \frac{1}{\mu_0} B_0$$

$\mu_0$ is the permeability. The same way as the static susceptibility according to the external field $B_0$ one may define the complex dynamic susceptibility $\chi = \chi' - i\chi''$ according to the rf-field $B_1$

$$\chi' = \frac{M_u}{2B_1/\mu_0} \quad \text{and} \quad \chi'' = \frac{M_d}{2B_1/\mu_0} \quad (103)$$

With these quantities the impedance $Z(\omega)$ of the rf-coil, i.e. its complex resistance, under the influence of the resonating spins can be formulated as follows:

$$Z(\omega) = R_c + i\omega L_c (1 + 4\pi \eta \chi(\omega))$$

or with the dynamic susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$

$$Z(\omega) = \frac{R_c + \omega L_c \cdot 4\pi \eta \chi''(\omega) + i\omega L_c (1 + 4\pi \eta \chi'(\omega))}{\chi'}$$

The parameter $\eta$ represents the effective filling factor of the target material in the coil, $L_c$ the inductance when $\chi = 0$ (empty coil), $R_c$ the resistance of the coil and $\omega$ is given by the frequency of the rf-oscillator.
The real part of the complex susceptibility (dispersion) represents the resulting inductance $Z_L$, whereas the imaginary part (absorption) is determined by the energy exchange caused by the Zeeman transitions. The average power loss or gain $\bar{P}_c$ in the coil is given by $\bar{P}_c = \frac{1}{2} I_c^2 (Z_R - R_c) = \frac{1}{2} I_c^2 \omega L_c \cdot 4\pi \eta \chi''(\omega)$ or in terms of the NMR-circuit parameters by

$$P_c = \frac{1}{2} I_c^2 (Z_R - R_c) = \frac{1}{2} I_c^2 \omega L_c \cdot 4\pi \eta \chi''(\omega)$$

(104)

where $I_c$ is the current in the coil.

Since the energy exchange (and the number of transitions) depends on the population of the Zeeman levels, the absorptive part of the susceptibility is a direct measure of the polarization $P$ of the target material. The Zeeman levels are broadened, so the imaginary part of $\chi$ has to be integrated over the resonance region:

$$P = c \cdot \int_{\omega_0}^{\omega_1} \chi''(\omega)d(\omega)$$

(105)

with $c \propto (g^2 / N)^{-1}$.

The proportional factor $c$ for the polarization is obtained by a calibration measurement in thermal equilibrium (TE measurement) at a known temperature and magnetic field (see section 5.4.3). Knowing $c$, the polarization can also be calculated during the microwave pumping of the DNP.

In Figure 40 a schematic diagram of a series Q-meter circuit is shown. The coil inside the refrigerator is connected by a coaxial transmission cable to the room temperature electronics of the so-called Liverpool NMR module [165, 166, 167]. In order to reduce influences by an additional capacity and inductance, the length of the transmission cable is adjusted to an integer number of half-wavelengths at the Larmor frequency. The Liverpool NMR module contains a rf-amplifier, a phase sensitive detector (PSD) and a LF-amplifier. The use of wide-band rf-amplifiers ($10 - 250 MHz$) is necessary for polarization measurements of protons ($\approx 106 MHz$ at 2.5T) as well as deuterons ($\approx 16 MHz$ at 2.5T) although it is disadvantageous for the signal-to-noise-ratio.

As shown in Figure 40 the circuit is driven by a frequency synthesizer $V_o$, which sweeps the rf-frequency $\omega$ through the Larmor resonance. This causes a change of the inductance of the coil as the target material absorbs or emits energy. The inductance change in turn causes an impedance change in the circuit, which is proportional to the complex output voltage $V(\omega, \chi)$ as long as the current is kept constant. At the last stage the PSD allows the selection of the real part of the voltage by using the input rf-signal as a reference. The voltage is a superposition of both the signal, proportional to $\chi$, and the so-called Q-curve, which is the response of the Q-meter to $\omega$ in the absence of $\chi$. The Q-curve is
measured by setting the $B_0$-field so that the Larmor frequency $\omega_0$ of the spin species is well outside the range of the frequency scan of the Q-meter, where $\chi''$ vanishes and $\chi'$ is negligible. The two signals are subtracted and the result is the NMR-signal,

\[ S(\omega) = \text{Re}\{V(\omega, \chi) - V(\omega, 0)\} \approx \chi''(\omega) \]  

(106)

The design of a NMR-coil depends on the target material, the target size, the Larmor frequency of the nuclei (depending on the magnetic field) and the desired quality factor. The quality factor has to be large enough to obtain a clear NMR signal, but small enough to stay within the linear region of the NMR module. For these reasons also the wire of the NMR coil is coated by a Teflon tube. This tube keeps the target material away from the strong rf-field close to the wire and thus limits the size of the signal. However teflon tubing on the NMR coils will not work in intense beams, it radiation damages and falls off. Instead thin wall tubing is used.

5.4.3 Polarization Calibration by the TE Method

The polarization is calibrated using the calculable polarization $P_{TE}$ at thermal equilibrium of the nuclear spin by means of the Brillouin function given in equation (99). This is normally done at a given magnetic field and at three different temperatures $T$, so that the proportional factor $c$ (see equation (105)) can be determined by linear regression. These measurements are performed at temperatures in the range of $1 - 2 \, K$ (i.e. running a $^{3}$He/$^{4}$He dilution refrigerator in the $^{4}$He-mode), since in this range temperatures can easily be measured and the polarization build-up times are short (less than a few hours). The absolute polarization $P$ is finally obtained by comparing the TE signal area $A_{TE} \approx \int_{\omega_1}^{\omega_2} S_{TE}(\omega) d\omega$ with that of the enhanced signal area $A_{enh} \approx \int_{\omega_1}^{\omega_2} S_{enh}(\omega) d\omega$ under DNP conditions,

\[ P = \frac{\int_{\omega_1}^{\omega_2} S_{enh}(\omega) d\omega}{\int_{\omega_1}^{\omega_2} S_{TE}(\omega) d\omega} \cdot P_{TE} \]

The polarization can be monitored continuously or sampled. Typical highly polarized NMR signals of protons and deuterons are shown in Figure 41.

![Figure 41](image_url)

Figure 41: Dynamically enhanced NMR signals of protons in butanol (a) and deuterons in D-butanol (b), which see a nonvanishing electric field gradient at their sites. Polarizations correspond to 82% for protons and 33% for deuterons.

For spin-$\frac{1}{2}$ particles like protons the Brillouin function is reduced to

\[ P_{1/2} = \tanh \left( \frac{\hbar \omega}{2kT} \right) \]

(107)

while the vector polarization of spin-1 particles like deuterons reads

\[ P_z = P_1 = \frac{4\tanh \left( \frac{\hbar \omega}{2kT} \right)}{3 + \tanh^2 \left( \frac{\hbar \omega}{2kT} \right)} \]

(108)
Figure 42: TE signal obtained in butanol (a) and D-butanol (b) at 1 K and 2.5 T. Polarization correspond to 0.25% for protons and 0.05% for deuterons.

$k$ is the Boltzmann constant. At $T = 1$ K and $B_0 = 2.5$ T the TE polarization for protons is 0.25% and for deuterons 0.05%. Especially the detection of the TE deuteron signal (see Figure 42(b)) is complicated due to its smaller polarization and to its larger signal width compared to that of the proton signal.

The uncertainty in the determination of the polarization is mainly due to the calibration measurements. The main error sources of a single TE measurement are the determination of the temperature and the noise of the signal. Therefore a precise measurement of the temperature by calibrated resistors or $^3$He evaporation pressure thermometers and a careful tuning of the NMR circuit (to avoid gain ratio errors of the low frequency amplifiers and non-linearities of the high frequency amplifiers) are done. In addition, the TE signal errors are reduced by using the signal averaging technique to overcome the signal-to-noise ratio problem.

Especially in the case of the deuteron NMR detection the subtracted signal $S(\omega)$ may still contain a residual Q-curve or residual wings caused by changes in the circuit parameters between the times when the Q-curve and the NMR signal are taken. To get rid of the residuals an additional background subtraction by fitting the wings of $S(\omega)$ is made. In a less serious case a third degree polynomial baseline fit is used. If this is not sufficient, a baseline function with integrated rf-circuit properties for the background subtraction is more advisable [168].

Finally a polarization accuracy $\Delta P/P$ of $\pm 2\%$ for protons and $< \pm 5\%$ for deuterons is obtained [169, 170].

5.4.4 Deuteron Signal Shape and Asymmetry Method

The NMR signal line shape depends on the spin species, the lattice and the degree of the polarization. Solving the Bloch equations the natural line shape is given by a Lorentzian function. However, in a solid state material the resonance line shape is determined by homogeneous and inhomogeneous effects like dipole-dipole interactions with neighboring nuclei or quadrupol interactions (see below) for spin species with $I > 1/2$. A further broadening may occur due to instrumental shortcomings like field inhomogeneities and saturation effects by the applied rf-field as well as due to other electronic effects. In Figure 42(a) a proton TE signal is shown which line shape may be nicely fitted with a convolution of a Lorentzian and a Gaussian function.

The line-shape of deuteron NMR signals, e.g. in D-butanol or ND$_3$, is rather complicated (see Figure 41(b)), but on the other hand it offers another possibility to determine the absolute deuteron polarization directly. Under a high magnetic field the energies of the three sublevels can be written as

$$E_m = -h\omega_0 m + h\omega_q(3\cos^2\Theta - 1)(3m^2 - I(I + 1)),$$

where $\omega_0$ is the deuteron Larmor frequency and $\omega_q = (eq\psi_{zz}/h)/8$ denotes the so-called quadrupol frequency. The quantity $eq$ is the deuteron quadrupole moment and $\psi_{zz}$ is the value of the electric
The quadrupole interaction shifts the levels depending on the angle \( \Theta \) between the direction of the magnetic field and the principal axis of the electric field gradient tensor. \( \theta \) is the angle between this axis and the direction of the magnetic field. The quadrupole interaction shifts the levels depending on the angle \( \Theta \) as shown in Figure 43. This causes two transitions, which are different in frequency. The corresponding lines are smeared out, since we do not have a single crystal. The two lines partially overlap and the observed deuteron NMR signal is a superposition of them as indicated in Figure 44. The two peaks correspond to \( \Theta = 90^\circ \), the pedestals to \( \Theta = 0^\circ \). The higher frequency peak and the lower frequency pedestal correspond to the \( m = +1 \) to \( m = 0 \) transition with an intensity \( I_+ \) and the other peak and pedestal to the \( m = 0 \) to \( m = -1 \) transition with an intensity \( I_- \). At any polarization the intensities, which are proportional to the population differences of the corresponding levels, become different and the NMR signal shows an asymmetry. This asymmetry is the more pronounced the higher the polarization degree is. Defining \( I_+/I_- = r \) and taking the weak quadrupole interaction into account \( (\omega_q/\omega_B \approx 10^{-3}) \) the deuteron polarization is independent of \( \Theta \) \[171\]. Thus with the so-called asymmetry parameter \( r \) the deuteron vector polarization can be written by

\[
P_z = \frac{r^2 - 1}{r^2 + r + 1}
\]

and the tensor polarization is written by

\[
P_{zz} = \frac{r^2 - 2r + 1}{r^2 + r + 1}
\]

as follows from the definitions given in equations (16) and (17). This so-called asymmetry method offers a simple way to determine the deuteron polarization by measuring the NMR signal peak heights \( (\Theta = 90^\circ) \) with a ruler (see Figure 44). In practice the asymmetry parameter is determined by a lineshape function, which describes the entire complicated deuteron NMR signal \[172\]. The Boltzmann distribution is not valid during fast alterations of the polarization induced by the DNP process. The polarization build-up time must be long compared to the spin-spin interaction in the nucleon system. A comparison of the polarization determination in D-butanol obtained by the TE- and asymmetry methods is shown in Figure 45 and indicates very good agreement. The polarization value between -20% and +12% are excluded in the plot because of the above mentioned reason. It should be mentioned that in
Figure 44: Theoretical deuteron line shape, which is the sum of the two possible transitions $m = +1$ to $m = 0$ (dotted line) and $m = 0$ to $m = -1$ (dashed line). Some line broadening has been taken into account. Otherwise the peaks would tend to infinity.

Figure 45: Comparison of the polarization determination in D-butanol obtained by the TE- and asymmetry methods. The dashed line is a fit to the data and indicates a good agreement of both methods.

$^6$LiD there is no asymmetry splitting of the energy levels in these spin-1 systems due to the vanishing electric field gradient in the FCC lattice structure. Here the NMR line width (nearly gaussian shape) is about 5 kHz (FWHM) compared to about 150 kHz in D-butanol or ND$_3$. In that case the tensor polarization $P_{zz}$ cannot be determined by the asymmetry method. If the small quadrupole interaction is neglected, $P_{zz}$ is given by

$$P_{zz} = \frac{4 \tanh^2 \left( \frac{\hbar \omega}{2kT} \right)}{3 + \tanh^2 \left( \frac{\hbar \omega}{2kT} \right)}$$

(112)

Together with equation (108) it follows that at thermal equilibrium $P_{zz}$ and $P_z$ are related by

$$P_{zz} = 2 - \sqrt{4 - 3P_z^2} \approx \frac{3}{4} P_z^2$$

(113)

In practice, the tensor polarization is calculated from the measured vector polarization. Assuming the normal case of a Boltzmann distribution among the magnetic sublevels, it is evident that only positive values for $P_{zz}$ can be obtained with polarized solid deuteron targets. Up to now only one particle physics experiment (elastic electron deuteron scattering) was performed using a tensor polarized solid ND$_3$ target with $P_{zz} \approx +0.2$ [173].
5.4.5 Cross-Calibration

For a precise determination of the cross section asymmetries in the particle experiments an exact knowledge of the polarization degree of all nuclei, which are present in the target sample, is obvious. However a direct comparison of NMR signals obtained from different spin species is possible only, if the NMR circuit is not changed, i.e. is working at a fixed frequency band. Consequently the magnetic field strength has to be changed to get into resonance with the magnetic moment of interest. This technique has to be used in the case of the polarization determination of $^{14}$N in the target material $^{14}$NH$_3$. The quadrupole moment of the spin-1 particle $^{14}$N is about ten times bigger than that of the deuteron, resulting in a total $^{14}$N-line width of about 2.5 MHz. Thus only part of the $^{14}$N-signal can be detected (see Figure 46) in a frequency range where the proton polarization $P_p$ in $^{14}$NH$_3$ is calibrated by the TE method. The connection between the polarization, the properties of the nuclei and the area $A_i$ under the signal lineshape is given by equation (105):

$$P_i \approx (g^2 I N)^{-1} A_i$$

(114)

The total area under the $^{14}$N-signal can be calculated with a lineshape fit [172]. Taking the ratio

$$\frac{P_{^{14}N}}{P_p} = \frac{g_p^2 I_p N_p A_{^{14}N}}{g_{^{14}N} I_{^{14}N} N_{^{14}N} A_p}$$

and inserting the known ratios of $g_p^2/g_{^{14}N}^2 = 191.3, I_p/I_{^{14}N} = \frac{1}{2}$ and $N_p/N_{^{14}N} = 3$ it follows:

$$P_{^{14}N} = 287 \cdot \frac{A_{^{14}N}}{A_p} \cdot P_p$$

Figure 46: Comparison of a proton TE signal ($P_p = 0.016\%$ at $\approx 1.5 K$ and $\approx 0.24 T$) and a $^{14}$N enhanced signal (low frequency peak) in NH$_3$ corresponding to -10% $^{14}$N-polarization.

On the other hand this cross calibration method can be used to determine an unknown content of spin species in the target sample, e.g. the $^6$Li, $^7$Li or proton content in $^6$LiD compared to its deuteron content [174]. More details about the NMR techniques for the detection of the polarization degree in solid target materials can be found in Refs. [169, 175, 176, 177, 178, 179, 180]
5.5 Target Materials and Developments

Important criteria for the choice of a target material suitable for particle physics experiments are:

- Degree of polarization.
- Dilution factor $f$.
- Presence of other polarizable nuclei.
- Short polarization build-up time.
- Good resistance of the polarization against radiation damage with the possibility to repair the radiation damage.

5.5.1 Chemically Doped Materials

The very nature of the DNP process as described in section 5.2 demands that a paramagnetic dopant be introduced into the host target material. A priori, it cannot be predicted for sure which host and dopants are best matched (see below), although theory predicts that a dopant having a narrow EPR line also at high magnetic fields should be advantageous, independently of the host material. The influence of the width of the EPR line, i.e. of the strength of the spin system's internal interaction, was demonstrated already in the early days of target material research. The polarization of LMN could be increased from 20% [181] to more than 70% [182, 2] under comparable values of $B_0/T$ simply by switching from cerium to neodymium as the paramagnetic dopant. Nevertheless, the materials used in current polarized targets were only established after many measurements of combinations of host and dopant [183, 184]. For example, butanol is combined with porphyrexide, which is easily mixed in. On the other hand, the diols, including the deuterated forms, worked best with a chromium radical Cr(V) which was introduced via a chemical reaction. Then EHBA, a synthesized form of Cr(V), was introduced [185], followed later by its deuterated counterpart EDBA [186]. This form was mixed in solution with the host, which allowed better control of the doping.

The solid target materials have to be in the form of small pieces, preferably of equal sizes, to provide good and uniform cooling of the material with the liquid helium. For example 1-butanol in its normal or perdeuterated version containing 5 wt% of water and 0.5 – 2 wt% of porphyrexide (not available anymore) as the paramagnetic dopant was an often used material. Butanol is preferred, because, compared to ammonia it lacks a polarized background and, compared to propandiol, its dilution factor is higher. The material is frozen into spherical beads of about 1.5 mm – 2 mm in diameter by dripping the liquid mixture into liquid nitrogen. Rapid freezing ensures that the material transforms into a glassy state, which is required for the homogeneous distribution of the paramagnetic radicals. Details about the preparation method are given in [187, 188].

Some materials [189] must be handled at temperatures below that of liquid nitrogen to maintain their amorphous character, while others are spontaneously combustible [190] when exposed to air; neither characteristic lends itself to confidence in being able to produce an operational target. On the other hand pentanol-2 stays amorphous up to room temperatures and, when doped with EHBA, it has produced polarizations of $\approx 85\%$ in a $^3$He/$^4$He dilution refrigerator [191].

Results of DNP were obtained in thin polymer foils and tubes [192, 193, 194]. Protons, deuterons, and $^{19}$F were polarized at 2.5T and below 0.3 K using the stable nitroxyl radical TEMPO [195, 196]. This radical can be easily introduced via a diffusion process. In addition, good results were obtained by doping an organic scintillator material with TEMPO. At the Paul Scherrer Institut (PSI) it was possible to extract scintillator light from a 10x10x1 mm$^3$ sample that had been polarized to 55% at 0.3 K and 2.5T [197]. Such polarizable materials, which are solid and can be handled at room temperature for at least several hours, could provide new experimental possibilities in nuclear and particle physics along with their obvious practical advantages in the loading of refrigerators. Table 1 lists the properties of the most common target materials used in polarization experiments.
Table 1: Polarized target materials commonly used in particle physics experiments

<table>
<thead>
<tr>
<th>Materials and chemical composition</th>
<th>Dopant and method(^a)</th>
<th>nucelons % by weight</th>
<th>(B/T) Tesla/K</th>
<th>Polarization %</th>
<th>Radiation Characteristic flow(^b) (10^{14}) particles/cm(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LMN (\text{La}_2(\text{Co, Mg})_3(\text{NO})_6 \cdot 24\text{H}_2\text{O})</td>
<td>Neodymium Ch</td>
<td>3.1</td>
<td>2.0/1.5</td>
<td>±70</td>
<td>(\approx 0.01)</td>
</tr>
<tr>
<td>1,2 Propanediol (\text{C}_3\text{H}_6(\text{OH})_2)</td>
<td>Cr (V) Ch</td>
<td>10.8</td>
<td>2.5/0.37</td>
<td>+ 98</td>
<td>(\approx 1)</td>
</tr>
<tr>
<td>1,2 Ethanediol (\text{C}_2\text{H}_4(\text{OH})_2)</td>
<td>Cr (V) Ch</td>
<td>9.7</td>
<td>2.5/0.5</td>
<td>± 80</td>
<td>(\approx 2)</td>
</tr>
<tr>
<td>Butanol (\text{C}_4\text{H}_9\text{OH})</td>
<td>EHBA Cr (V) Ch</td>
<td>13.5</td>
<td>2.5/0.3</td>
<td>± 93</td>
<td>3 - 4</td>
</tr>
<tr>
<td>EABA (\text{C}_2\text{NH}_2\text{BH}_3\text{NH}_3)</td>
<td>EHBA Cr (V) Ch</td>
<td>16.5</td>
<td>2.5/0.5</td>
<td>+75</td>
<td>7 (+), 3.5(-)(^c)</td>
</tr>
<tr>
<td>Ammonia (\text{14NH}_3, \text{15NH}_3)</td>
<td>(\text{NH}_2) Ir</td>
<td>17.5, 16.6</td>
<td>5/1.0</td>
<td>+97</td>
<td>70, 175(^d)</td>
</tr>
<tr>
<td>D-Butanol (\text{C}_4\text{D}_9\text{OD})</td>
<td>EDBA Ch</td>
<td>23.8</td>
<td>2.5/0.3</td>
<td>± 50</td>
<td>Not measured</td>
</tr>
<tr>
<td>D-Butanol (\text{C}_4\text{D}_9\text{OD})</td>
<td>(\text{C}_3\text{D}_7\text{CD}-\text{OD}) Ir</td>
<td>23.8</td>
<td>5.0/0.2</td>
<td>-70</td>
<td>Not yet measured</td>
</tr>
<tr>
<td>D-Ammonia (\text{14ND}_3, \text{15ND}_3)</td>
<td>(\text{ND}_2) Ir</td>
<td>30.0/28.6</td>
<td>3.5/0.3</td>
<td>+49</td>
<td>130(+), 260(-)</td>
</tr>
<tr>
<td>Lithium Deuteride (6\text{LiD})</td>
<td>F-center Ir</td>
<td>50</td>
<td>6.5/0.2</td>
<td>± 70</td>
<td>400</td>
</tr>
</tbody>
</table>

\(^a\) Ch: chemically doped, Ir: doped by irradiation.
\(^b\) Accumulated particle flow which reduces the polarization by \(e^{-1}\) of its initial value.
\(^c\) For positive and negative polarizations, respectively.
\(^d\) In \(\text{NH}_3\) there are two distinct regions of decay.
5.5.2 Radiation Doped Materials

AMMONIA:

Another method to introduce the paramagnetic radicals into solidified target materials (frozen beads or chips) is to irradiate the pure samples. This preparation technique for the DNP was first successfully used with NH3 [137] and ND3 [198, 199]. Two different irradiation techniques can be employed and both make use of high intensity ionizing particle beams to produce an adequate number of radicals (some $10^{19}$ spins/ml) in a reasonable amount of time: (a) high-temperature irradiation at 80 – 90 K and (b) low-temperature irradiation at 1 K.

Low-temperature irradiation means that the radicals necessary for DNP are produced during the experiment. As 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 or protons.

Detailed studies have shown that it is more advantageous to operate with high temperature pre-irradiated material [200]. The accumulated flux on ammonia, obtained by high intensity electron beams from injection linacs e.g. the 20 MeV injection linac of the BONN electron accelerator is about $10^{17}$ electrons/cm$^2$. Detailed information about the ammonia preparation for DNP can be found in [201].

Electron paramagnetic resonance (EPR) measurements performed with high-temperature irradiated NH3 and ND3 have indicated that the radicals NI$I^2$ and ND$I^2$, respectively, are responsible for DNP in ammonia [138, 139]. If the samples are kept under liquid nitrogen, the lifetimes of the radicals are on the order of years.

As experience shows, the polarization behavior of ND3 is more complicated than that of NH3. Whereas it is sufficient to dope NH3 via high-temperature irradiation only, in order to create high proton polarizations under all conditions, the preparation of ND3 demands an additional low temperature irradiation at experimental conditions ($\approx 1$ K) [202, 161]. The typical beam flux to achieve best deuteron polarization values is about $4 \cdot 10^{14}$ e$^-$/cm$^2$ [202] to $10^{16}$ e$^-$/cm$^2$ [161].

A disadvantage of ammonia compared to the alcohol and diol targets is the existence of background nuclei $^{14}$N (spin-$1$) or $^{15}$N (spin-$\frac{1}{2}$) in $^{14}$NH$3$, $^{15}$NH$3$, $^{14}$ND$3$ and $^{15}$ND$3$. The $^{15}$N polarization is relatively easy to measure [161] but the $^{14}$N nucleus, whose quadrupole moment is about 10 times bigger than that of the deuteron, challenges the NMR measurement. With a combination of techniques, this problem was solved (see section 5.4.5).

LITHIUM HYDRIDES AND DEUTERIDES:

$^6$LiD is of special interest as a polarized neutron target with its high dilution factor $f \approx 0.5$ for neutrons (see section 5.7). Compared to other deuterated target materials the deuteron and $^6$Li TE polarization signal is easy to detect, because there is no quadrupole splitting of the energy levels in these spin-$1$ systems due to the vanishing electric field gradient in the face-centered, cubic lattice structure. This circumstance allows an accurate measurement of both the deuteron and the $^6$Li polarization via the TE method. Investigations demonstrate a high polarization resistance against radiation damage [203].

Pioneering work to prepare $^7$LiH, $^6$LiH, and $^6$LiD for the DNP process has been done by different groups at Saclay [204, 205, 206, 207]. Systematic studies of the optimum preparation method including the behavior of the materials under an intense electron beam have been also performed in Bonn [203].

At room temperature the lithium compounds are in their solid state phase, and the paramagnetic impurities (so-called F-centers) are introduced by irradiation. In contrast to the ammonia targets, the temperature during irradiation has to be between 180 and 190 K. EPR studies of the F-center concentration showed that it is not the production efficiency which defines the rather narrow temperature window, although it seems to be maximum around these temperatures. In the framework of the most recent study of $^6$LiD for the CERN COMPASS experiment [208] the EPR of these paramagnetic centers was investigated under the condition of partial and complete saturation [209, 210], i.e. if $WT_1 > 1$ with the induced transition probability $W$ and the longitudinal relaxation time $T_1$ of the electrons. Figure 47(a) and (b) show the saturation curves of the F-centers in $^6$LiD, irradiated at different temperatures, in which the peak-peak amplitudes $\Delta V_{pp}$ as well as the magnetic field distances $\Delta B_{pp}$ of the extrema of the EPR signals are plotted versus the relative microwave amplitude $\omega_1/\omega_{1,\text{max}}$. The most striking features of the 'cold irradiated' (140 K, 170 K) samples are the appearance of a local maximum
in the width of the signals as well as the existence of two stages of a linear increase of the signal amplitudes; neither is seen in the case of the 'warm irradiated' (190 K, 200 K) samples. This behavior can be understood on the basis of the Provotorov theory (see section 5.2.2). Moreover, theoretical expressions for the saturation curves can be found, which depend on the undistorted (low power) width $D$ of the resonance and on the relaxation times of the Zeeman and the dipolar occupation distribution, $T_Z$ and $T_D$, respectively. These expressions, which are valid up to a certain strength of the transition probability $W$, can be fitted to the experimental data. It follows from this analysis that the Zeeman relaxation time $T_Z$ decreases by almost two orders of magnitude when the irradiation temperature is increased from 140 to 200 K, while the dipolar relaxation time is much less affected. From the point of view of the spin temperature theory the large differences in the maximum polarization are simple consequences of the strongly varying ratio $T_Z/T_D$ in samples of different irradiation temperatures. It should be noted that the hyperfine structure of the undistorted EPR signal is practically unchanged by a variation of the irradiation temperature. Thus, the identity of the paramagnetic center (as an electron captured in a vacancy of the anion sublattice) seems not to be altered. It is still unclear which physical process is responsible for the change of the time constants of the electron system. It is interesting to notice that alternatively to an irradiation in the high temperature region one may use much lower temperatures followed by an exposition of the sample to room temperature for some minutes. The corresponding saturation curves are shown in Figure 47(c) and (d). Not only the saturation behavior of the EPR but also the polarization build-up and maximum values are essentially the same in material obtained from both preparation methods. Taking into account all of the observations listed above it would appear that it is not the electron system itself but rather its environment, which undergoes certain temperature dependent transformations with the consequence of a highly improved spin-lattice coupling of the F-center system.

Figure 47: (a) and (b): Saturation curves of $^6$LiD irradiated at temperatures between 140 and 200 K taken at liquid nitrogen temperature and at a microwave frequency of 9.35 GHz. (c) and (d): Same for a $^6$LiD sample irradiated at 140 K with (open symbols) and without (closed symbols) a subsequent annealing at room temperature.
CARBOHYDRATE COMPOUNDS:
Although \(^{6}\text{LiD}\) delivers the highest overall polarization of all the established deuterated target materials at given magnetic field and temperature conditions (e.g. more than 50% at 2.5\(T\) in a \(^{3}\text{He}/^{4}\text{He}\) dilution refrigerator) it is best suited only for those experiments, in which the highly polarized proton and neutron within the \(^{6}\text{Li}\) nucleus can be regarded as quasi free. A large step forward towards a highly polarized deuteron (neutron) target for physics experiments at intermediate energies was recently done by the extension of the radiation doping method to the carbohydrate compounds, which are free of any polarized background. Using an electron beam in the \(MeV\) range the application of a total dose of some \(10^{15}\) e\(-/cm^{2}\) at liquid argon temperature is sufficient to create paramagnetic defects in the respective CH\(_{2}\)-chains with a concentration high enough for the DNP process to function. Systematic EPR studies in a variety of different carbohydrate compounds have shown that these defects, independent of the particular material, consist of a proton or deuteron vacancy along the chain, i.e. the paramagnetic center can be identified with the alkyl radical (\(-\text{CH}-\)). In the special case of the normal (straight chained) alcohols like 1-butanol, frozen in glassy beads, the vacancy is most likely located at the hydroxyl carbon of the molecule having the supposed structure \(-\text{CH-OH}\).

![EPR spectra at liquid nitrogen temperature of perdeuterated ethanol (a) and perdeuterated 1-butanol (b).](image)

Figure 48: EPR spectra at liquid nitrogen temperature of perdeuterated ethanol (a) and perdeuterated 1-butanol (b). In both cases the hydroxyalkyl radical is responsible for the observed hyperfine structures. Numbers 1 - 9 indicate the positions of the expected nine hyperfine lines.

Figure 48(a) shows the EPR line of irradiated ethanol in its perdeuterated form. After an exposition of the sample to visible light for some minutes a central single line (see inlet in Figure 48(a) showing the difference of the two signals), which may be attributed to electrons captured at interstitial lattice sites, has vanished. The remaining structure corresponds to the unpaired hydroxyalkyl electron, which possesses a certain probability to be found at the sites of the deuterons bound to the hydroxyl or to the methyl group carbon atoms, respectively. Due to the approximative equivalence of these \(n = 4\) deuterons, the EPR spectrum consists of \(2n + 1 = 9\) hyperfine lines. The coupling to the hydroxyl (\(-\text{OD}\)) deuteron is much weaker, only contributing to the width of the nine lines produced by the carbon bound deuterons. In Figure 48(b) the corresponding spectrum of perdeuterated butanol is shown, from which the individual hyperfine lines are much harder to identify. Nevertheless, by means of a comparison of the spectra of partially deuterated 1-butanol, in which the protons are substituted by deuterons at different positions in the molecule, it is possible at least to strongly motivate the hydroxyalkyl radical to be the most prominent radical type [209].

In contrast to the commonly used chemical dopants the radiation induced radicals possess a much smaller g-factor anisotropy leading to a considerably narrower EPR line at the high magnetic fields of a
polarization experiment. This circumstance is advantageous in two respects. Firstly it is immediately clear from the spin temperature theory that a narrower EPR line enables the creation of higher inverse spin temperatures and thus of higher nuclear polarizations. Secondly the magnetic field may be increased to values higher than in the case of chemically doped butanol, in which the linearly increasing EPR line width eventually prevents the existence of a unique spin temperature throughout the electron system. Experimentally the maximum deuteron polarization in chemically doped alcohols and diols at the temperatures of a $^3\text{He}/^4\text{He}$ dilution refrigerator was measured to be maximum around $2.5\,T$ followed by a rapid decrease with increasing magnetic field [211, 212]. In contrast, the maximum deuteron polarization in irradiated butanol increases with increasing magnetic field at least up to $5\,T$. Although systematic investigations concerning the optimum irradiation dose have not yet been done, deuteron polarizations of more than 50% at $2.5\,T$ and more than 70% at $5\,T$ have already been achieved [212]. Besides the alcohols first studies of radiation doped polyethylene are under way, which show that in these substances not only radicals of the alkyl type can be created but also other types, some of which being stable at room temperature for weeks [209, 210]. Together with first successful attempts to polarize radiation doped perdeuterated polyethylene [213] the techniques of radiation doping appears to open new horizons in preparing polarized target materials with much higher performance for particle physics experiments at intermediate particle energies.

5.6 Radiation Damage

The reduction of the polarization by radiation damage is a serious problem in the experiments performed with the alcohol and diol materials, where the radicals for the DNP process are provided by chemical dopants. This phenomenon may be understood qualitatively as follows.

Additional radicals in the target materials are created during the irradiation, for which most probable candidates are atomic hydrogen or deuterium. In contrast to the temperatures, under which the radiation doping procedure is normally performed, these atoms may be stable at the much lower temperatures of a polarization experiment. The respective g-factors of these low temperature stable radicals may be considerably different from 2. In the case of atomic hydrogen (deuterium) it is the strong hyperfine coupling of the S-wave electron to the nucleus magnetic moment, which shifts the effective g-factor away from the free resonance value. Due to the Larmor frequency of these radicals, which is then far away from the frequency of the applied microwave field, they do not contribute to the DNP process, but they do contribute to the nuclear relaxation processes via dipolar coupling to the nuclear spins on the one hand and via spin-phonon coupling to the lattice on the other hand. As the radical density increases, this effect becomes more and more important eventually leading to a reduction of the nuclear polarization.

In most cases, however, the original degree of polarization can almost be recovered by heating (annealing) the target material up to, but not higher than, the devitrification temperature [184]. The observation that with each subsequent annealing process the initial polarization (the one obtained directly after the annealing) is more and more reduced, clearly shows a second mechanism of radiation damage to be present. It may consist of a slowly proceeding destruction of larger and larger amounts of the paramagnetic molecules. As a consequence the material has to be changed after several annealing procedures. The polarization decrease combined with the time-consuming annealing process limited the use of these alcohol and diol materials in experiments with intense proton, electron, or photon beams.

At this point the development of ammonia, with its high polarization resistance to radiation damage, made it the choice for improved experiments of a new generation (see section 5.8). It turned out that the depolarizing dose in NH$_3$ is more than one order of magnitude higher compared with that of butanol [200]. In addition, the polarization loss in NH$_3$ can be completely recovered by annealing the sample at a lower temperature ($\approx 77\,K$) than in the case of a butanol sample ($\approx 120\,K$) and can be repeated many times without loss.

On the other hand, the presence of an intense particle beam may also have positive influence on the polarization behavior of some materials. For instance, a steep increase of the deuteron polarization is observed in ND$_3$ during the initial phase of an irradiation at low temperatures, an observation, which 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 ND₃ 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 [200]. A second example is ⁶LiD, whose radiation hardness is again almost one order of magnitude higher than that of ammonia. Here the nuclear polarization also benefits from an additional low temperature irradiation during the first 5 × 10¹⁵e⁻/cm² [214]. From the fact that in contrast to ND₃ no dynamic polarization effect was observed from ⁶LiD samples without any preirradiation it can be concluded that the observed increase of the maximum polarization as well as the reduction of the polarization build-up time is not caused by the creation of additional DNP active radicals at low temperatures. Taking into account the strong dependence of the achievable maximum polarization on the temperature during pre-irradiation it appears that the reasons for both phenomena may be found in a changing environment of the F-center and thus in different spin-lattice time constants.

Even though the phenomenon of radiation damage has been studied extensively in most of today’s polarized target materials, so far no consistent picture exists about the actual physics processes, which are responsible for this effect. Nevertheless, taking all results together, it seems most probable that the high polarization resistance to radiation damage in ammonia and in lithium deuteride is a consequence of (a) the initial radiation doping of both materials and (b) the much lower efficiency in producing paramagnetic vacancies compared to that in organic molecules like butanol. The latter presumption is reflected by the much lower radiation dose needed for the carbohydrates (compared to the inorganic substances) to produce a paramagnetic system of a concentration high enough to be useful for DNP.

5.6.1 EPR Spectroscopy

Although in the light of the spin temperature theory the physics of dynamic nuclear polarization can be looked at as a special field of magnetic resonance in solid state materials, the technique of electron spin resonance has become only recently a standard tool for the purpose of target material research [209, 210, 215]. Already 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, a wealth of informations can be obtained from the electron system, which plays the role of a ‘generator’ for the nuclear polarization. The most simple but nonetheless very important application is the measurement of the concentration of the unpaired electrons. In particular, when the material is doped by irradiation, the comparison of the EPR signal strength of a given sample to that of a standard sample with known radical density is the only way to pin down the optimum irradiation dose. In the same way, i.e. through the analysis of the unsaturated EPR signal line shape, it is possible in many cases to reveal the physical nature of a radiation induced paramagnetic species. Examples are the ammonia radical, the F-center in lithium deuteride as well as the hydroxyalkyl radical in the alcohols.

In going from unsaturated (low power) measurements to a region of the microwave amplitude, in which the occupation distributions start to be affected by the Zeeman transitions, it is possible to obtain informations about the dynamical behavior of the electron spin system. The saturation curves taken from irradiated ⁶LiD and their comparison to the spin temperature theory may be seen as a first step towards a much better understanding of the role of the electron spin system for the DNP process. This application of EPR, which demands a much more sophisticated analysis compared to low power measurements, is to be extended towards temperatures and magnetic fields, which are representative for the usual conditions of polarization experiments.

For this purpose, but also in order to determine the fundamental parameters of the non-Zeeman interaction of the electron spin system more accurately, a EPR spectrometer is currently under construction, which will allow measurements at a temperature of 1 K and at magnetic fields as high as 2.5 T [216]. The hyperfine splitting of a given paramagnetic center is studied best in the X-band region, whereas for the determination of the g-tensor elements higher magnetic fields are more advantageous, because the splitting due to g-anisotropy increases with increasing field values. Furthermore, an EPR spectrometer, which operates in situ with a polarization experiment, can be regarded as the most promising tool, in order to understand the physical processes of radiation damage. A high field EPR spectrometer, which itself is part of a target apparatus, provides the unique possibility of studying online what happens to the initial paramagnetic system during a continuous irradiation at low temperatures. In addition a
possible growth of other paramagnetic species can be observed.

Although the above list of applications for electron spectroscopy in the field of polarized targets is far from being complete, the reader may get an impression about the importance of systematic EPR studies for further improving the performance of polarized solid state target materials.

5.7 Spin Structure of Nuclei

For spin structure studies of the neutron polarized deuterons in D-butanol, ND₃ or ⁶LiD are used in the case of solid targets. As the NMR system is only sensitive to the total magnetization of the nuclei, the measured nuclei polarization has to be corrected in order to obtain the neutron spin asymmetries. Furthermore the possible kinematic dependence of the nuclei polarization has to be understood. Both together requires that a proper allowance is made for the nuclear properties of the spin I = 1 particles e.g. deuterium and lithium.

Two approaches are commonly considered. In the case of isospin singlets, like D, ⁶Li, and ¹⁴N, model independent analyses using only isospin conservation can be applied, based on the decomposition of the total nuclear angular momentum and magnetic moment in terms of spin and orbital components [217]. In the case of mirror nuclei, like ³H and ³He, or ¹⁵N and ¹⁵O, this analysis can be supplemented by information from the beta decays of one of the pair members [218, 219]. The cluster model description of light nuclei, such as lithium [220], allows the extension of this method to the cluster components of ⁷Li.

The second approach involves models of the nuclear structure of the target nuclei. This approach has been used to describe the nucleon polarization in D [221], ³He [222], ⁶Li [221, 223], ¹⁴N [6], and ¹⁵N [224]. At the simplest level, the relevant information to be extracted from the models consists of the probabilities of the different nuclear states and their corresponding angular momentum decompositions. This information results in a proportionality factor between the nucleon and the nucleus polarizations.

Using the first approach, the values for the nucleon polarization in D and ⁶Li are estimated to be 0.94 and 0.85 of the nuclear polarization, respectively. These values are in excellent agreement with the corresponding results of the nuclear structure models.

In these models the deuteron ground state is described by about 95% S-state and 5% D-state occupation probability [225]. While the nucleons in the S-state are always aligned along the nuclear spin, the nucleons in the D-state have their spins aligned opposite to the nuclear spin a net 50% of the time. Consequently the neutron polarization is lower than the measured deuteron polarization as calculated in the following:

In an external magnetic field the deuteron states split into the triplet

\[ |I, m\rangle = |1\ 1\rangle, \ |1\ 0\rangle, \ |1\ -1\rangle \]

For the D-state these deuteron states are composed of the orbital angular momentum \( L = 2 \) and the sum of the nucleon spins via Clebsch-Gordon coefficients \( (|I, M_L| |I m\rangle) \):

\[
\begin{align*}
|1\ 1\rangle &= \frac{\sqrt{3}}{2} |2\ 2\rangle |1\ -1\rangle_I - \frac{\sqrt{3}}{10} |2\ 1\rangle |1\ 0\rangle_I + \frac{\sqrt{10}}{10} |2\ 0\rangle |1\ 1\rangle_I \\
|1\ 0\rangle &= \frac{\sqrt{3}}{10} |2\ 1\rangle |1\ -1\rangle_I - \frac{\sqrt{5}}{5} |2\ 0\rangle |1\ 0\rangle_I + \frac{\sqrt{5}}{5} |2\ -1\rangle |1\ 1\rangle_I \\
|1\ -1\rangle &= \frac{\sqrt{10}}{10} |2\ 0\rangle |1\ -1\rangle_I - \frac{3}{10} |2\ -1\rangle |1\ 0\rangle_I + \frac{3}{5} |2\ -2\rangle |1\ 1\rangle_I
\end{align*}
\]

The states \( |I m\rangle_I = |1\ 1\rangle_I, |1\ 0\rangle_I, |1\ -1\rangle_I \), which are given by the sum of the nucleon spins in the magnetic field, have to be decomposed in order to obtain an information about the alignment of the nucleon spin:

\[
\begin{align*}
|1\ 1\rangle_I &= \frac{1}{\sqrt{2}} \left( |\frac{1}{2}\ \frac{1}{2}\rangle |\frac{1}{2}\ \frac{1}{2}\rangle + |\frac{1}{2}\ -\frac{1}{2}\rangle |\frac{1}{2}\ -\frac{1}{2}\rangle \right) \\
|1\ 0\rangle_I &= \frac{1}{\sqrt{2}} \left( |\frac{1}{2}\ \frac{1}{2}\rangle |\frac{1}{2}\ -\frac{1}{2}\rangle + |\frac{1}{2}\ -\frac{1}{2}\rangle |\frac{1}{2}\ \frac{1}{2}\rangle \right) \\
|1\ -1\rangle_I &= \frac{1}{\sqrt{2}} \left( |\frac{1}{2}\ -\frac{1}{2}\rangle |\frac{1}{2}\ \frac{1}{2}\rangle + |\frac{1}{2}\ \frac{1}{2}\rangle |\frac{1}{2}\ -\frac{1}{2}\rangle \right)
\end{align*}
\]
The probability of a nucleon state in a given deuteron state can be calculated with the square of the Clebsch-Gordon coefficients. Thus for the deuteron states with \( L = 2 \) the probability of the nucleon spin to be aligned opposite to the spin of the deuteron is

\[
\begin{align*}
\text{for } |1 1\rangle &= \left(\sqrt{\frac{3}{5}}\right)^2 + \left(\sqrt{\frac{3}{10}} \cdot \sqrt{\frac{1}{2}}\right)^2 + \left(\sqrt{\frac{1}{10}} \cdot 0\right)^2 = 0.75 \\
\text{for } |1 0\rangle &= \left(\sqrt{\frac{3}{10}}\right)^2 + \left(\sqrt{\frac{3}{5}} \cdot \sqrt{\frac{1}{2}}\right)^2 + \left(\sqrt{\frac{1}{10}} \cdot 0\right)^2 = 0.50 \\
\text{and for } |1 -1\rangle &= \left(\sqrt{\frac{3}{5}}\right)^2 + \left(\sqrt{\frac{3}{10}} \cdot \sqrt{\frac{1}{2}}\right)^2 + \left(\sqrt{\frac{1}{10}} \cdot 0\right)^2 = 0.75.
\end{align*}
\]

Using these results the population numbers \( n_{1/2} \) and \( n_{-1/2} \) of the two neutron spin levels can be calculated from the population number \( N_1, N_0 \) and \( N_{-1} \) of the three deuteron levels:

\[
\begin{align*}
n_{1/2} &= 95\%N_1 + 5\% \cdot 75\%N_{-1} + 5\% \cdot 25\%N_1 + 50\%N_0 \\
n_{-1/2} &= 95\%N_{-1} + 5\% \cdot 75\%N_1 + 5\% \cdot 25\%N_{-1} + 50\%N_0
\end{align*}
\]

Using the definition of the polarizations given in equation (15) and (16) the neutron polarization is

\[
P_n = 0.925 \cdot \frac{N_1 - N_{-1}}{N_1 + N_0 + N_{-1}} = 0.925 \cdot P_D
\]

Including the error for the \( D \)-state probability the neutron polarization is \((92.5 \pm 1.5)\%\) of the deuteron polarization \( P_D \).

With \(^6\text{Li}\), the situation is somewhat more complex. A three body cluster model of \(^6\text{Li}\) \((\alpha + p + n)\) indicates that \(P\)- and \(P'\)-states are also present, in agreement with the predictions of the model independent approach \([217]\), if small isospin breaking effects are neglected. Taking from Ref. \([223]\) the average of seven models with adequate predictions for properties of \(^6\text{Li}\), such as the magnetic moment, charge radius, and charge form factor, one obtains for the neutron polarization \(P_n = 0.866 \pm 0.012\%\) of the nuclear polarization, which is in good agreement with the model independent prediction. More details can be found in Ref. \([226]\).

The presence of \(^7\text{Li}\) in the \(^6\text{Li}\text{D}\) used in the experiments requires that the contributions of this nucleus to the cross section asymmetry is understood. While the model independent analysis is valid for \(^7\text{Li}\), there is no measurement of the magnetic moment of its very short lived mirror pair \(^7\text{Be}\) that could be used for this purpose. \(^7\text{Li}\) is very well described as an \(\alpha + \text{triton}\) pair of clusters \([220]\). This feature allows to approximate the nucleon polarization in \(^7\text{Li}\) in terms of the corresponding nucleon polarization in tritium. Following the model independent approach, the polarization of the proton in \(^3\text{He}\) is 0.94 of the tritium polarization. Since the tritium and alpha clusters form an \(I = \frac{3}{2}\) system, the spin of the tritium cluster is aligned parallel to the lithium spin 66\% of the time. Thus the net proton spin polarization in \(^7\text{Li}\) is 0.627 \(\pm 0.014\) of the nuclear polarization.

For the kinematic range of Bjorken \(x < 0.75\) in the deep inelastic scattering experiments it was shown recently \([227]\), that for the deuteron the use of a constant factor to correct for the presence of \(D\)-state nucleons in the deuteron results in a deviation of less than 1.5\% from the more accurate convolution or covariant descriptions. Since the uncertainty in the \(D\)-state probability itself is on the order of \(\approx 1\%\) (20\% relative), the use of a constant correction factor, at least in the measured kinematic range \((0.015 < x < 0.75)\), is valid in the case of the deuteron. For the extraction of the neutron spin structure, more refined descriptions of the nucleon polarization may be necessary, given the small size of the deuteron asymmetry at low \(x\). The level of required refinement becomes more important with increasing nuclear complexity, as in the cases of \(^3\text{He}\) and \(^6\text{Li}\).

5.8 Realization of Polarized Solid Targets

As already mentioned in the Introduction, spin effects using polarized solid state targets have been studied in innumerable experiments. The results obtained at laboratories that have operated polarized proton and deuteron targets over many years, i.e. Bonn, Brookhaven, CERN, Dubna, Gatchina, KEK,
Kharkov, LAMPF, IHEP Scrupkho, PSI, Saclay, SLAC, TRIUMF, are given in the proceedings of several symposia [228, 229, 230, 231, 232, 233, 234, 235]. Depending on the beam intensities there are two modes of operation for a polarized solid state target system. (Polarized) electron beams have to be operated at intensities of less than 100 nA in order to handle cooling requirements and radiation damage of the target materials. In this case polarized targets with $^4$He evaporative cooling are used, which have to work in a continuous mode, i.e. with permanent microwave irradiation to maintain the DNP process at $1 \, K$. At this temperature the nucleus (nucleon) polarization relaxation time $T_1$ is relatively short. This continuous mode operation puts strong constraints on the design of polarizing magnets. Due to the field inhomogeneity requirements over the entire target volume, the large dimensions of the magnet coils limit the angles for the outgoing particles (see Figure 49). Other electromagnetic probes such as tagged photon beams are intensity limited by their production techniques to about $10^8$ particles/s. To obtain a reasonable counting rate, a wide opening angle with the ability to simultaneously measure a large kinematical range is needed. This can be achieved with the concept of the frozen spin target [236].

![Figure 49: The E143, E155 and E155x target set-up at SLAC with a vertically operated $^4$He evaporation refrigerator and a 5 T polarizing superconducting magnet.](image)

The operation of the frozen spin target is based on the experimental fact that the nucleon polarization relaxation time $T_1$ is a very strong function of the temperature and magnetic field. $T_1$ characterizes the polarization decay after switching off the DNP mechanism (microwaves). Typical values for $T_1$ are minutes at a temperature of $1 \, K$ and days below $100 \, mK$. The principle of the frozen spin target operation is to polarize the target material at a high field (e.g. 2.5 T) and in the temperature range 0.15 – 0.3 K. Once the target material is optimally polarized, the microwaves are switched off and the polarization is frozen at temperatures around 50 mK (frozen spin mode). Due to the very long $T_1$ at these temperatures, it is possible to reduce the field to a value of 0.3 – 0.4 T where the polarization decay is acceptable (holding field).

An appropriate setting of the holding magnets allows the target polarization to be oriented in different directions [237, 238, 239] and provides good experimental access (see Figure 50). A new concept
of a small superconducting holding magnet placed inside a $^3$He/$^4$He dilution refrigerator (see Figure 51) has been developed [240, 241]. Thus the fringe field and its influence on nearby detector components is minimized and that makes it possible to operate polarized solid targets in a $4\pi$-detector. Since the cooling power of the $^3$He/$^4$He dilution refrigerators, operating at such temperatures, is relatively low and the Kapitza resistance [153] large, the beam intensity for experiments with a frozen spin target is limited. The maximum flow hitting a target area of 1 $cm^2$ is $10^7 - 10^8$ particles/s [237, 242].

![Figure 50](image1.png)

Figure 50: Set-up of a frozen spin target in the polarization mode (a) and in the holding mode (b), in which a vertical as well as a horizontal spin direction can be realized (SACLAY version). From Ref. [237]

![Figure 51](image2.png)

Figure 51: Holding mode (a) of a frozen spin target with an internal superconducting holding coil; (b) shows a close-up of the target area. This concept with a split-pair configuration of the holding coil was used for the measurement of the eta photoproduction on polarized protons at the Bonn tagged photon facility [243].

In the following we discuss some modern polarized solid target equipment which is used to address some physics goals at various laboratories.

5.8.1 Target for High Intensity Beam Currents

**E143, E155 and E155x Experiments at SLAC:**

With the benefit of improvements in target design and radiation resistant materials, the experiments E143, E155 and E155x were able to provide high statistics data on the spin structure function $g_1$ for the proton, deuteron (neutron) as well as good data for the spin structure function $g_2$. 
Figure 52: The polarization history of (a) protons in $^{15}$NH$_3$ and (b) deuterons in $^{15}$ND$_3$ as a function of the charge on the target. One unit of charge is equivalent to $10^{14}$ electrons. Bumps in the curve at one polarization sign indicate the annealing of the target sample. From Ref. [161]

Figure 53: Deuteron polarization of a $^6$LiD sample as a function of the accumulated dose in the SLAC 48 GeV electron beam. Deuterated ammonia shows a similar behavior, but with a maximum polarization at about $10^{15}e^-/cm^2$, only.

The target set-up used in these experiments is shown in Figure 49 and described in Ref. [161]. In the E143 experiment a beam of about $5\cdot10^{11}$ electrons/s at 29 GeV was scattered from a 3 cm long polarized ammonia target ($^{15}$NH$_3$ and $^{15}$ND$_3$), which resulted in a luminosity of higher than $10^{35}cm^{-2} \cdot s^{-1}$. The targets were cooled by a $^4$He evaporation refrigerator operating at around 1K with a cooling power of about $1.5W$ in a 5T field. The proton polarization in $^{14}$NH$_3$ had previously been shown [244] to have a rapid rise to values over 90%, after pre-irradiation at 90 K. Tests before E143 showed that this was equally true for $^{15}$NH$_3$. On the other hand, the deuteron polarization in $^{15}$ND$_3$ rose to 13% only after the high-temperature irradiation, but a modest additional low-temperature irradiation (see section 5.5.2) boosted the achievable polarization finally to $\approx 42\%$, with frequency modulation applied.

The beam was rastered over the face of the target to prevent local depolarization. Typically, after turning the beam on the target, polarization dropped by a few percent over the course of about 10 minutes due to beam heating of the target volume and, then, declined at a lower rate due to radiation damage. After reducing to a given level, the polarization was refreshed by a target annealing, the
polarization reversed, and the cycle repeated. A short history of a $^{15}$NH$_3$ and $^{15}$ND$_3$ target is shown in Figure 52(a) and (b). In the case of the deuteron, the effect of the low-temperature irradiation pushing the polarization to > 40% is apparent. The proton polarization, on the other hand, stays relatively flat. NMR measurements of the $^{15}$N polarization in both targets and of the residual proton polarization in the deuteron target were made to correct the asymmetry data. More target details can be found in [161].

Because the electron beam spin direction was flipped randomly on a pulse-by-pulse basis, target polarization did not need to be reversed. However, because of the rapid polarization build-up, it was done once or twice a day for consistency checks and a study of systematic effects. For the same reason a set of data was taken with the magnetic field of the target reversed. Data on these spin structure measurements have been published in Refs. [224, 225, 245, 246].

In the E155 experiment $^6$LiD was used as target material [214]. During the experiment performed with a 48 GeV electron beam a maximum deuteron polarization of - 32% and $^6$Li polarization of - 31% were achieved. The $^6$Li polarization was regularly monitored and followed the deuteron polarization as expected from EST (see Figure 34). A full polarization history is shown in Figure 53 for target material, which was prepared for DNP at 185K with a pre-irradiation dose of $2.0 \times 10^{17}$e$^-$/cm$^2$. When a sample reached about a dose of about $10^{16}$e$^-$/cm$^2$ it was annealed to 185K, the initial optimum pre-irradiation temperature. After the annealing the same maximum polarization was attained. However, the rate of polarization decay with increasing charge seems to be faster than before the annealing. An increase of the polarization after further irradiation at low temperature was already seen at earlier measurements [203]. For a detailed understanding of this behavior, which is similar to that observed with ND$_3$, in situ EPR studies at low temperatures are needed (see section 5.6.1).

In comparison with other target materials like ND$_3$, $^6$LiD with its excellent dilution factor when taking the $^6$Li polarization into account, shows good polarization values at 1K and 5T and a superior polarization resistance against radiation damage. The major disadvantage of $^6$LiD can be seen in its unfavorably long polarization build-up time, requiring extensive calibration time for the TE polarization.

5.8.2 Targets for Low Intensity Beam Currents

CONTINUOUS MODE OPERATION:

Figure 54: The SMC dilution $^3$He/$^4$He refrigerator with the two cells target and the superconducting 2.5T magnet.

The SMC experiment at CERN was the first one to access the neutron spin structure and allow
a test of the Bjorken sum rule. 190 GeV polarized $\mu^+$ mesons were scattered from polarized protons or deuterons in a very large target; the target material was butanol, D-butanol or $^{14}$NH$_3$. With a beam intensity of $\approx 10^7 \mu^+/s$, which was more than four orders of magnitude less than at the SLAC experiments, the luminosity was increased by having a target two orders of magnitude longer than at SLAC.

The $^3$He/$^4$He dilution refrigerator [156], connected to a 2.5 T solenoid, is shown in Figure 54. The mixing chamber was divided into three parts, with two containing the target material, each 60 cm long and a 30 cm divider the third part. The target sections were polarized in opposite directions and the divider filled with microwave absorber to prevent cross coupling between the sections. A volume of 2.7 l of butanol or $^{14}$NH$_3$ was required to fill both sections of the mixing chamber. The dilution refrigerator was capable of a cooling power of 1.3 $W$ at 500 mK, yet could cool to 50 mK, where the cooling power was 1 $mW$ for frozen spin operation. The build-up time for the polarization was several hours, but without any worries of beam heating or radiation damage, polarizing could go on during data taking for several days before saturation occurred. Maximum proton polarizations of $+93\%$ and $-94\%$ were obtained in butanol, while an average polarization of more than $\pm 90\%$ was reached in $^{14}$NH$_3$. Typical deuteron polarizations in D-butanol were $\pm 50\%$.

In this experiment it was not possible to flip the beam polarization (unless one changed to $\mu^-\!$) and with a build-up time of hours, it was not very easy to change the target polarization frequently. So a strategy of rotating the solenoid field to the opposite direction was adopted. With 190 GeV muons and a longitudinal field, the effect on the scattered particles was minimal. The field was rotated about every five hours while the target was in frozen spin mode. A dipole was turned on at a strategic time so that the total magnetic field never went below 0.5 T while the solenoid was crossing zero. Each rotation lost only a fraction of 1% of polarization.

During the course of the experiment it was found that frequency modulating the microwaves [247] improved the deuteron polarization by almost a factor of 2 and by about 20% for the proton. Up to now there is no satisfactory explanation for this huge modulation effect seen in the SMC target. The data from the latest SMC publications are found in Refs. [248, 249, 250].

Meanwhile a new experiment at CERN, COMPASS, has started to measure the gluon spin contribution to the nucleon spin [251]. $^6$LiD with its highest content of polarizable nucleons in solid state
materials is polarized in the SMC target set-up. The target material was prepared for DNP by irradiation at a temperature of \((190 \pm 2)\) K with a total dose of \(10^{17}\) electrons/cm\(^2\). The irradiation was performed at the 20 MeV Bonn injection linac [208]. So far maximum polarization values of \((+54 \pm 2)\) % and \((-47 \pm 2)\) % have been reached. The polarization build-up curves as measured with NMR coils in each target cell are shown in Figure 55. Similar polarization results in a small sample of \(^6\)LiD are reported in Ref. [252].

**Frozen Spin Mode Operation:**
Modern polarized solid targets as discussed above have mainly been used to obtain information about the spin structure of nucleons at high \(Q^2\) (perturbative regime). However there also is a strong call for spin structure studies at low \(Q^2\). As can be seen from table 2, the measurement of the Gerasimov-Drell-Hearn (GDH) sum rule is one major topic at various laboratories. The GDH sum rule relates the helicity asymmetry of the total cross section for polarized photons \((Q^2 = 0)\) on polarized nucleons to the anomalous magnetic moment of the nucleon [11, 12]. Being based on general principles of physics, the GDH sum rule is an important consistency check for the understanding of the hadronic structure.

![Figure 56: Schematic drawing of the Bonn \(^3\)He/\(^4\)He dilution refrigerator and the target area (a) in the polarizing mode and (b) during the data taking in the frozen spin mode. (c) Sideview of the front part of the \(^3\)He/\(^4\)He dilution refrigerator containing the internal holding coil. From Ref. [253]](image)

More than thirty years after the derivation of the sum rule in 1965 it is now possible to perform a direct experimental test up to energies of several GeV. Decisive was the development of the internal holding coil concept for frozen spin operation in a 4\(\pi\) detector set-up. To reach the center of the DAPHNE detector (see Figure 56(b)), a special 2.4 m long \(^3\)He/\(^4\)He dilution refrigerator has been
Figure 57: Proton polarization measured in butanol during three typical frozen spin cycles.
From Ref. [253]

constructed, which incorporates the beam pipe in the backward region of the detector. The DAPHNE
detector and a 6.5 T polarizing magnet (see Figure 56(a)) are mounted on a precise rail system and can
be placed around the $^3\text{He}/^4\text{He}$ dilution refrigerator corresponding to the polarizing or frozen spin (data
taking) mode. The spin direction in the frozen spin mode is maintained by a small superconducting
magnet. The internal superconducting holding coil is wound on a radiation shield and surrounds the
target completely. Its total thickness of only 780 $\mu$m including the 300 $\mu$m thick copper carrier enables
the majority of the outgoing particles to pass through. With a coil current of 10.8 A a holding field
of 0.42 T is reached. The homogeneous field of the internal holding coil allows, for the first time in a
scattering experiment with a frozen spin target, the determination of the target polarization (proton)
during data taking. In this way the systematic error of the target polarization is considerably lowered
in comparison to former experiments where the target polarization had to be interpolated between the
starting and the end values of the frozen spin period. The measurement of the target polarization is
done by the NMR technique with an error of 1.3 % for protons and 4.5 % for deuterons at the high field
(2.5 T) of the polarizing magnet. In the low field (0.42 T) of the holding magnet an additional statistical
uncertainty of about 0.2 % has to be considered [158, 253].

In the GDH experiment a maximum polarization of 87.7 % for protons and of 35.3 % for deuterons was
achieved at build-up times of about 2 hours using butanol and D-butanol as target material, respectively.
Temperatures below 60 mK in the frozen spin mode led to long relaxation times exceeding 200 hours
for protons and 150 hours for deuterons, such that the nucleon polarization had to be refreshed only
everone two days (see Figure 57), resulting in more efficient data taking.

GDH measurements with a polarized proton target were performed at MAMI (Mainz) in the domain
of low photon energies (140 - 800 MeV) and at ELSA (Bonn) for higher energies up to 3 GeV. First
physics results are given in Refs. [254, 255].

**Brute Force Polarized Operation:**
The method of brute force (or equilibrium) polarization in principle works for every material containing
nuclei with spin. From equation (28) it follows that, to get high polarization values, very high magnetic
fields and very low temperatures are required. The brute force polarization method requires a very
long time for the polarization to build-up, unless the material contains a certain amount of unpaired
electrons or other centers, which mediate a coupling between the nuclear spins and the lattice. In this
case the build-up time may be short enough to be acceptable, but then the high magnetic field must be
maintained also during the data taking period of the experiment, in order to prevent the polarization
from a fast decay. On the other hand, magnets, which are capable of producing fields of more than 5 T,
do generally not allow the particle beam to access the target material or the reaction products to be
detected over a large solid angle. In addition, the ultra low temperatures, which are necessary for the
brute force polarization method can only be achieved, if the respective refrigerator is working almost at its base temperature. Consequently, the useful current of the particle beam is strongly limited by the maximum heat load, which is still acceptable, in order to maintain those temperatures.

The only way out would be a method, which allows the coupling of the nuclear spins to the lattice to be switched off, and thus to slow down the relaxation rate once the material is polarized. In fact a procedure like that was already proposed in 1967 to polarize HD gas, the only polarizable isotopical combination of hydrogen gas [256].

After the development of superconducting magnets placed inside a $^3\text{He}/^4\text{He}$ dilution refrigerator (see before) this idea was picked up again. Since the early 1990s brute force polarized HD targets are prepared by the SPICE collaboration [257] for measurements at the LEGS facility at Brookhaven and by an Orsay group for measurements at GRAAL (Grenoble) [258]. In these cases the material is exposed to a very high magnetic field ($17\,T$) at an extremely low temperature ($15\,mK$) for a period of some weeks. During this time the protons and deuterons are polarizing towards their thermal equilibrium values. Meanwhile the ortho-$^3\text{H}_2$, which had been added ($\approx 10^{-4}$) to the HD in order to deliver the needed nuclear relaxation channel, converts into its non-magnetic ground state, the para-$^3\text{H}_2$. After the conversion is completed, the target material is extracted from the production refrigerator, using a special retrieval device, and stored and/or transported in a holding dewar. Finally, the target material can be installed in the in-beam refrigerator in combination with a magnet for particle experiments. The holding temperature and field will depend on the experimental configurations, but 0.5 K and 1 T will allow a sufficiently long lifetime to perform some experiments.

Following this production path a proton polarization of $\approx 60\%$ has been obtained up to now [259]. With about 20% proton polarization an experiment was recently performed at the LEGS facility [260]. These proton polarization values are achieved after waiting for several decay constants ($6.25$ days) of the ortho-$^3\text{H}_2$. For para-$^3\text{D}_2$, with a decay constant of 18.2 days, holding the HD material at high field and low temperature for months is not practical. Therefore the solution for the production of reasonable high deuteron polarization [261] is to use ortho-$^3\text{H}_2$ to build-up the proton polarization and then, by the technique of adiabatic fast passage, to transfer the high proton polarization to the deuteron. By this procedure deuteron polarization values up to 14% were obtained [259].

The HD target material is attractive because it has a high dilution factor. However, the target itself is not completely pure HD since there are about 20% by weight of aluminium wires imbedded to remove the heat of conversion of ortho-$^3\text{H}_2$ to para-$^3\text{H}_2$. From the FOM point of view a drawback is its low density ($0.13\,g/cm^3$), which however can be advantageous for experiments, where the interaction of the reaction products with large target volumes is critical. It is obvious that additional heat input into the target will accelerate the polarization decay. Independent of the acceptable heat load also the ability of the particle beam to produce unwanted paramagnetic defects in the solid (leading to an extra relaxation channel for the nuclear spins) restricts the use of HD to experiments performed with low intensity beams. In addition, if for any reason the polarization is lost, there is no way of replenishing it, and the target material must be changed, unlike in the case of DNP operated targets.

### 5.9 Summary of Polarized Solid State Target

The first polarized solid state target and the subsequent developments of all aspects of a polarized solid state target system have opened up new experimental possibilities and allowed a choice of approaches to a particular physics problem. There are a variety of polarizable solid state target materials available with dilution factors varying between 0.13 and 0.5. However, in exclusive reactions kinematical overdetermination allows a clear event separation from background nuclei, which results in a dilution factor of nearly one.

The highest polarization resistance against radiation damage is shown in materials which are prepared for DNP by irradiation. Polarizing them in $^4\text{He}$ evaporation refrigerators with their high cooling power at 1K beam intensities upon the targets up to $6 \cdot 10^{11}$ particles/s can be tolerated. Thus luminosities of higher than $10^{35}\,cm^{-2}\cdot s^{-1}$ can be achieved.

Polarized target experiments performed with low intensity beams such as polarized muons or (polarized) tagged photons have to rely on solid target materials. At intensities up to $10^8$ particles/s the
Table 2: Experimental sites with polarized solid state targets performing experiments with electromagnetic probes.

<table>
<thead>
<tr>
<th>Laboratories</th>
<th>Physics goals</th>
</tr>
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<tbody>
<tr>
<td>CERN (Geneva)(^a)</td>
<td>Nucleon spin structure</td>
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<td>ELSA (Bonn)(^a)</td>
<td>Nucleon resonances, GDH</td>
</tr>
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<td>Nucleon resonances, GDH, formfactors, nucleon spin structure</td>
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<tr>
<td>SLAC (Stanford)(^a)</td>
<td>Nucleon spin structure</td>
</tr>
<tr>
<td>SPRING8 (Osaka)</td>
<td>GDH</td>
</tr>
</tbody>
</table>

\(^a\) Currently running dynamically polarized solid state targets.

\(^b\) Brute force polarized solid state targets.

Frozen spin mode operation of the target system is favorable in order to allow a particle detection over a large (4π) angular acceptance.

Maximum polarization values for protons > 90% and for deuterons between 50% and 70% are achieved. Polarization measurements are done by the NMR technique to a relative accuracy of < ±3% for protons and < ±5% for deuterons. Table 2 lists the various experimental sites where experiments with polarized solid targets using electromagnetic probes will be performed.

6 Conclusion

Since the discovery of the parity violation in the weak interaction, spin has become an important handle in nuclear and particle physics experiments to study the structure and interaction of hadrons. Early polarized solid targets allowed studies of spin effects in various reactions performed with (polarized) hadron or meson beams. Subsequent developments of all aspects of a polarized target system and the parallel developments of polarized lepton beams have opened up new experimental possibilities to study some of the outstanding and fundamental problems of particle spin physics. These experimental possibilities were further enhanced by the developments and uses of the polarized gaseous H, D and \(^3\)He targets in storage ring experiments or in external beams, respectively. The Bjorken sum rule in polarized deep inelastic lepton nucleon scattering was formulated in 1966, but it has only been confirmed through experiments in the 1990s, using state-of-the-art polarized targets. There is now a wealth of polarization data which have shed light on the question how spin \(\frac{1}{2}\) quarks and spin 1 gluons conspire to give nucleons a spin of \(\frac{1}{2}\). Using multipronged polarization techniques at several laboratories, however, a full understanding cannot be far off.

At the same time the new electron machines at medium energies, with their battery of polarized targets, will ensure that the questions what is the relation of particle degrees of freedom and the low energy structure of the hadrons can be answered. First double polarization measurements towards the test of the validity of the Gerasimov-Drell-Hearn sum rule and its generalized description are actual examples to study the relation between hadronic degrees of freedom and quark gluon degrees of freedom. New developments in the technically challenging field of polarized targets are still on the horizon which will further improve the quality of the data in particle physics experiments as well as will enable new types of experiments.

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