SOLID POLARIZED TARGETS FOR PARTICLE PHYSICS EXPERIMENTS

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Abstract

This talk will cover developments in techniques and the operation of solid polarized targets for scattering experiments. These range from the traditional dynamically polarized targets to the emerging HD targets as well as the possibility of room temperature targets. In addition a recent workshop on NMR in polarized targets is summarized.

1 Introduction

This talk will cover some of the developments in solid polarized targets of the last few years and will be based on a recent review article by myself and Werner Meyer[1].

Solid polarized targets were first used in experiments in the early 1960s [2,3] and quickly developed so that they were soon in use at most particle physics laboratories. Figure 1 shows the subsystems required for a standard solid polarized target. These include a polarizing magnet, usually in the range from 2.5T to about 7T, a refrigerator to cool the target material to a temperature of 1K or less and associated instrumentation, a microwave system to generate the polarization via the Dynamic Polarization method (DNP) and a nuclear magnetic resonance (NMR) system to measure the polarization. This talk will briefly review the DNP method, and discuss the various subsystems and their development, in the context of recent targets used in particle experiments. Also included is the current status of HD targets [4] and some information on the development of warm targets [5] .

The Workshop on NMR held at the University of Virginia last April will be summarized at the end of this talk.

2 Polarization Mechanisms

The DNP method is used to obtain a high polarization of nuclear spins through irradiation with microwaves in which the high polarization of unpaired electrons is transferred to those nuclei.

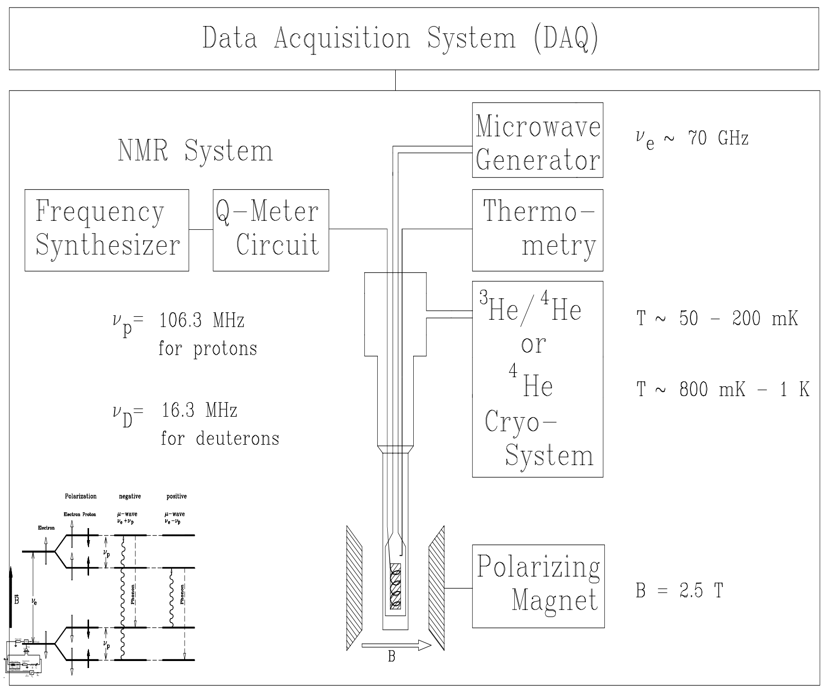

Figure 1: A generic polarized target showing the major subsystems and typical operational parameters. Not shown are the standard vacuum and Roots pumping systems.

In a discussion of polarized targets, the key is the magnetic moment, μ, of the particle of interest i.e., electron, proton, deuteron (neutron). A polarized target is an ensemble of such particles placed in a high magnetic field and cooled to low temperatures. The energy level diagram for spin - 1/2 protons, shown in Fig. 2, results because of the Zeeman interaction of μ with the magnetic field, B. The μ_e of quasi-free electrons, introduced into the parent material through doping by paramagnetic radicals, splits into two levels, each with two sub-levels due to μ_p. The population of each level is described by the Boltzmann Law and from it the polarization P(1/2) and P(1) for protons and deuterons are obtained. The TE polarizations are

$$P(\tfrac{1}{2}) = \tanh\left(\frac{g_i \mu_i B}{2 k_B T}\right) \tag{1}$$

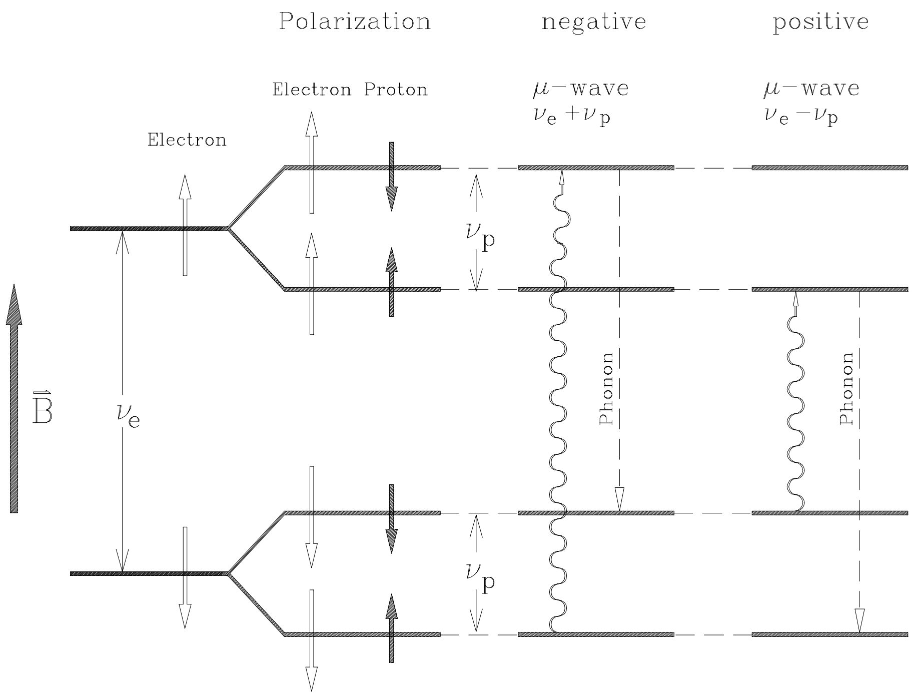

Figure 2: Schematic of the energy levels of for the solid state effect.

$$P(1) = \frac{4\tanh\left(\frac{g_i \mu_i B}{2k_B T}\right)}{3+\tanh^2\left(\frac{g_i \mu_i B}{2k_B T}\right)} \tag{2}$$

where μ_i is the magnetic moment of the spin species, g_i is its g-factor and T the temperature.

Unfortunately the magnetic moment of the proton is small and gives only small polarizations for a normal range of temperatures and magnetic fields, eg., P(1/2) = 0.25% at 2.5T and 1K, and P(1) = .05%. These are not very useful for particle physics experiments. However the magnetic moment of the electron is 600 times that of the proton leading to polarizations of greater than 90% under the same conditions. The key to DNP is being able to transfer the electron polarization to the proton. Figure 2 shows the process in the solid-state effect.

¿From Fig. 2, a microwave field of suitable frequency is applied, inducing a spin flip of both electron and proton. DNP works because the electron relaxation (msecs) is much faster than that of the proton (hours). Thus the polarization is transferred from the electron to proton and the electron is available to flip another proton spin. Finally spin diffusion allows the polarization to be transferred to protons far away from the electron site. Then a spin temperature, T_S, can be defined relating the degree of alignment to the polarization such that

$$P(1/2) = \tanh\left(\frac{\mu_p B}{k_B T_S}\right) \tag{3}$$

However, many materials in use today are non-crystalline and have energy bands rather than discreet energy levels. So the simple solid-state scheme does not apply and the theory of equal spin temperature applies. In this case the microwaves induce fast cross relaxation of the electrons i.e. electron spin flips take place, and so long as this is fast compared to the spin-lattice relaxation of the electrons, proton flips will occur and thus polarization. Other spin species in the material will also be flipped, but it is assumed that they are all at the same spin temperature and that their polarizations depend only on their magnetic moments.

3 Equipment and Techniques

Figure 1 shows the major components of a polarized target: The polarizable material is cooled to a temperature of 1K or less, in a homogenous field of 2T or greater and irradiated with microwaves. A nuclear magnetic resonance system measures the degree of polarization. Not shown are the necessary pumping and vacuum systems and instrumentation.

Cryogenics

The first experiments [2,3] used ^{4}He evaporation refrigerators operating at ~1K in conjunction with the LMN material which could be polarized to ~ 70%. As proton rich materials such as butanol came to be used, polarizations of only ~40% could be obtained with such refrigerators. However the development of ^{3}He evaporation refrigerators operating at 0.5K meant that the achievable polarization could be doubled, because of the lowering of the spin temperature [6].

Dilution refrigerators soon followed and as their reliability and ease of operation improved, have mostly replaced the ^{3}He evaporation refrigerators as the choice for polarized targets. The ability to polarize at 200mK - 500mK and then reduce the temperature to ≤100mK introduced the possibility of frozen spin targets. The ultimate dilution refrigerator, used in the SMC experiment at CERN[7], is shown in Fig. 3. With a mixing chamber of 1.5 meters in length the approximately 1.5kg of target material could be polarized at 300mK with a cooling power of 2W and reduced below 100mK for frozen spin operation.

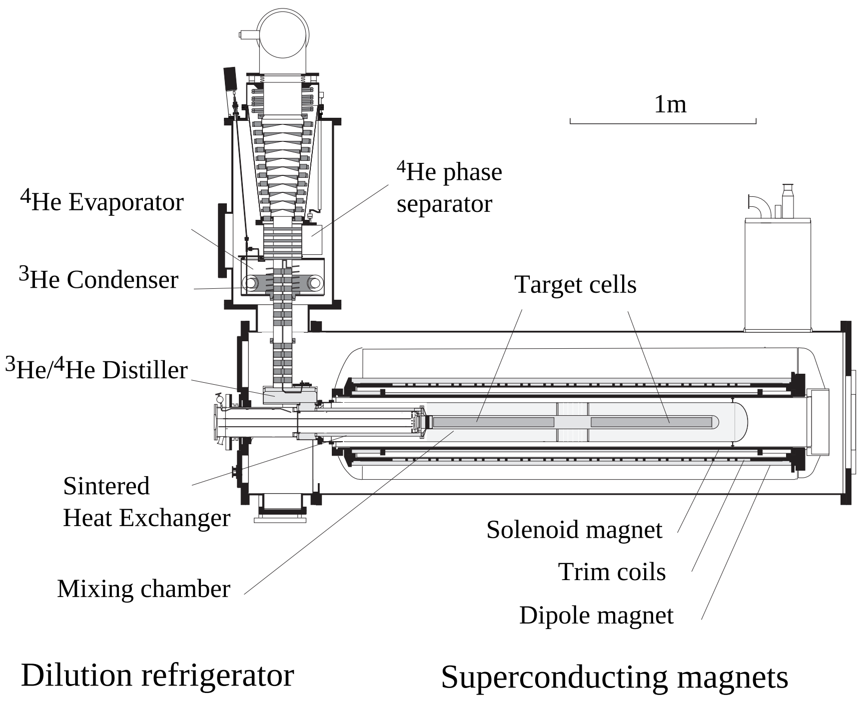

Figure 3: The target and magnet used in the SMC experiment at CERN.

The development of high field open geometry magnets opened up a new opportunity for ^{4}He evaporation refrigerators, particularly for use in high intensity beams. Making use of the highly superior heat transfer properties (compared to ^{3}He), high polarizations could be maintained with beam intensities of $6{\cdot}10^{11}$ particles/sec or better. Such a target system is shown in Fig. 4 and was used in two experiments at SLAC. Details of the performance are given in ref.[8].

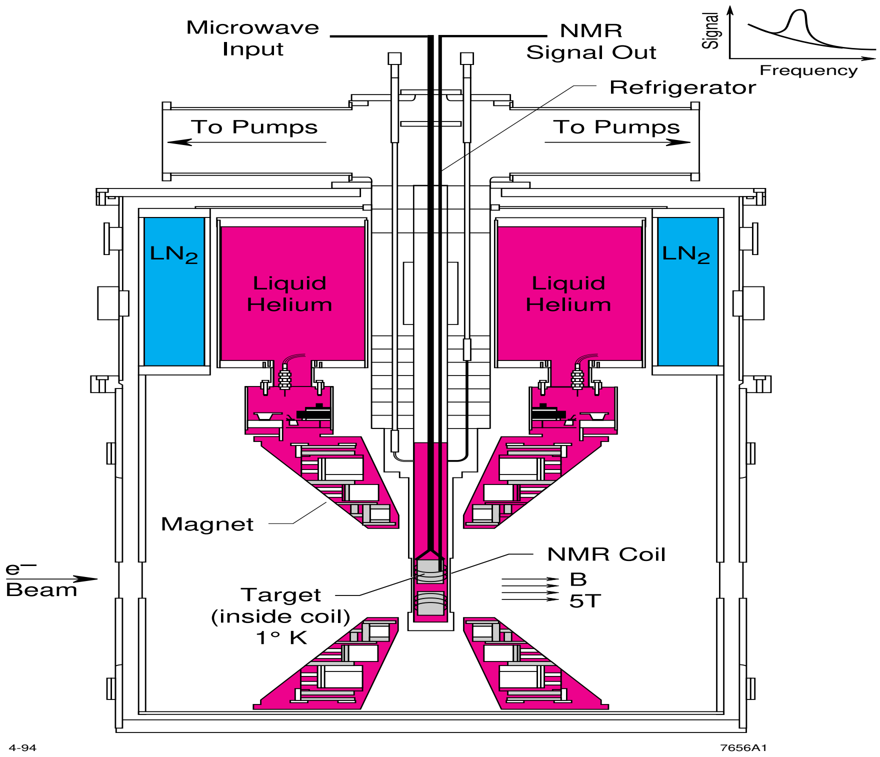

Figure 4: The target and magnet used in experiments E143 and E155 at SLAC

Recently a similar target has been in use at the Jefferson Lab. Here though the constraints of fitting into the CLAS spectrometer has led to a different refrigerator design with it entering the magnet at a 30 degree angle and having the target moved vertically and separately from the refrigerator.

Magnets

The polarizations that can be achieved are linked to B/T and the lower T is, the lower T_S can be and thus the higher the polarization that can be achieved. Early polarized targets operated with ^{4}He at T = 1 - 1.5K and iron magnets of B = 2 - 2.5T, giving B/T $\approx$ 2. The development of low temperature refrigerators and high field superconducting magnets meant that values of B/T of 50 - 100 were available. But for various technical reasons DNP works best when B/T $\approx$ 5 - 10.

The main limitation of iron magnets is that in order to boost the operational field to 2.5T, and maintain the required uniformity, the substantial mass of iron and copper present restricts access and reduces the experimental acceptance. On the other hand superconducting magnets can be built with high fields and a very open geometry [8].

The use of superconducting magnets has divided into two main routes: For reasons of high cooling power and heat extraction in intense particle beams, high field magnets, 5T and above, are used in conjunction with ^{4}He evaporation refrigerators [8]; a combination of a dilution refrigerators and moveable or large acceptance superconducting magnets allow frozen spin polarized Targets to be used in particle physics experiments.

Microwaves

The microwave frequency needed for DNP is approximately 28GHz/T, i.e. 140GHz at 5T. The power required varies from 1 - 2 mW/g of target material at 2.5T/0.5K to 20 - 40mW/g at 5T/1K. The high power tubes in use are EIOs while for low power applications klystrons, IMPATT diodes and Gunn diodes are available. The power that can be extracted from these tubes is inversely related to its operating frequency as is the power loss in microwave components. This leads to a practical limit of for operation of about 210GHz or 7.5T.

Power supplies for these tubes should have the capability for frequency modulating the tube as considerable improvements in polarization can occur in this mode.

NMR and Polarization Measurement

A spin system I placed in a magnetic field shows a Zeeman splitting into 2I + 1 levels. Figure 2 shows the case for I=1/2. When the spin system is irradiated by rf at the Larmour frequency, the spin system either absorbs or emits energy. The NMR circuit as shown in Fig. 5 detects the loss or gain of energy by measuring the voltage across the inductive coil in or around the target material. A phase sensitive detector extracts the absorptive part of the signal. Signals for the proton and deuteron are shown in Fig. 6 and the area under the curve is proportional to the polarization. The circuit is calibrated by measuring the area of the TE signal at a known temperature and using the Boltzmann relation to obtain the polarization at that temperature. The enhanced polarization is then calculated by scaling the measured areas.

I will discuss NMR questions some more in the workshop summary.

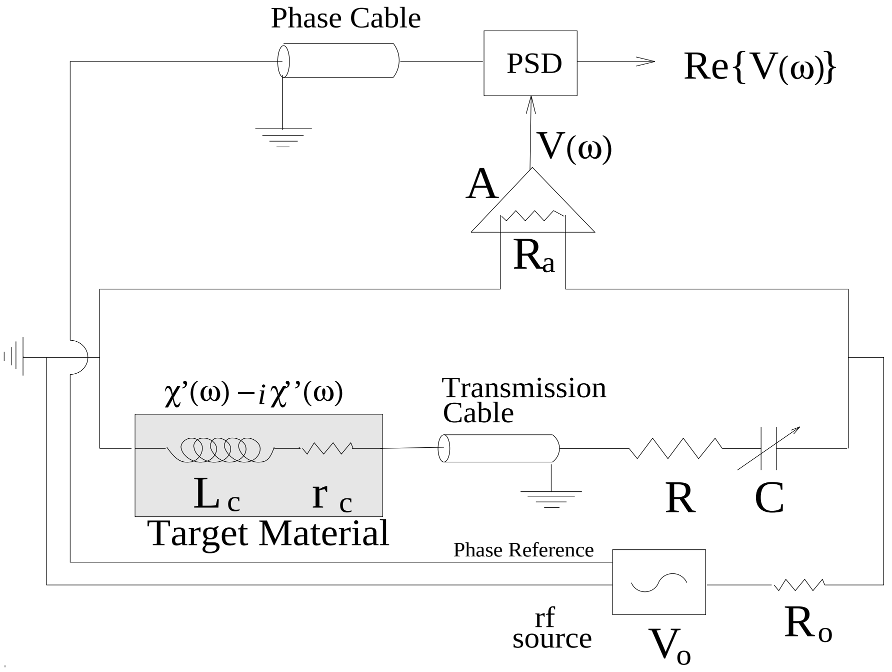

Figure 5: Schematic of the Q-meter circuit for NMR measurements.

Materials

Table 1 shows the properties of the more commonly used materials in polarized targets.

The DNP process requires that a paramagnetic radical be introduced into the host material. This is done chemically, or by irradiation. These days the most widely used chemical dopant is a synthesized form of CrV, EHBA [9] or in its deuterated form EDBA [10]. These forms of CrV are powders, which can easily be mixed in with the host and thus with better control of the doping. Recently, [11] the nitroxyl radical TEMPO has been used to dope scintillator, thus providing an active polarized targe.

In general, chemically doped materials are not very radiation resistant and for experiments with intense beams, ammonia has been found to have a radiation resistance of at least an order of magnitude better than the best chemically doped materials. It also has the advantage that repeated anneals retrieve the original polarization and reduce the polarizing times - the opposite

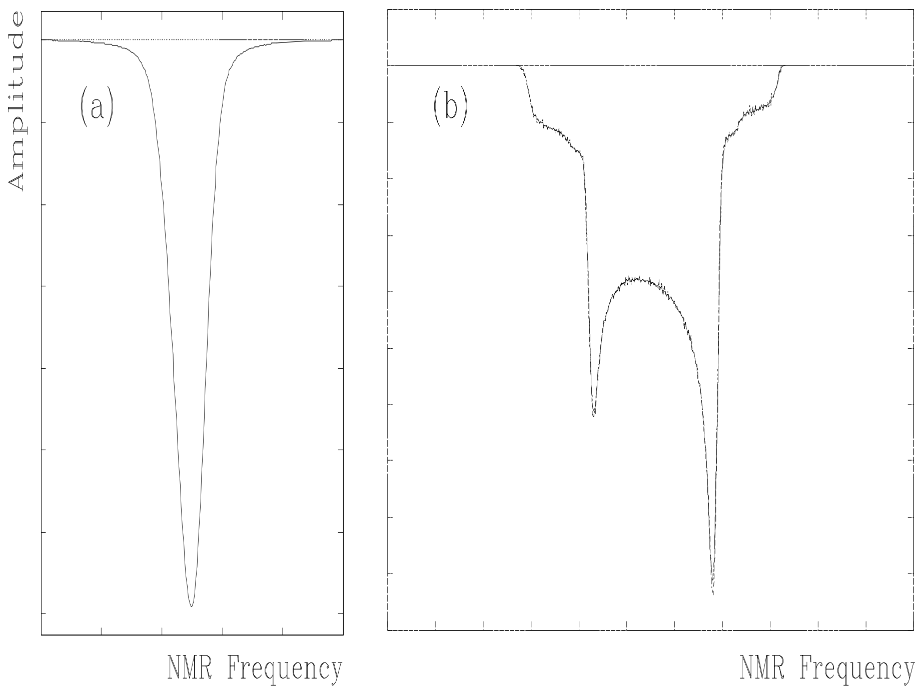

Figure 6: Dynamically enhanced NMR signals for (a) proton with polarization = 92% and (b) deuteron with polarization = 37%.

of the chemically doped materials. However it has recently been confirmed that after approximately 10^{17} charged particles/cm^2 have passed through ammonia it ceases to anneal back. No such limit has yet been found in deuterated ammonia.

Recently ^{6}LiD has been used in a scattering experiment [12] and found to have a radiation resistance a factor of five better than ammonia. One anneal restored the polarization, but subsequent anneals at the same temperature (182K) had little effect. Since, to a very good approximation, the ^{6}Li nucleus can be considered as a combination of a deuteron and ^{4}He nucleus and that ^{6}Li polarizes to the same level as the deuteron, the ^{6}LiD can be considered to contain two deuterons. For those experiments where the nuclear effects are not too intrusive this is an excellent material.

Table 1 Polarized target materials commonly used in particle scattering experiments

Materials & Chem. Comp.	Dopant[a] & Method	Polarizeable Nucleons % by weight	B/T Tesla/K	Polarization %	Radiation Characteristic Flux[b] 10^{14} particles/cm^2
LMN $La_2(Co, Mg)_3$ $(NO)_3 \cdot 24H_2O$	Neodymium Ch	3.1	2.0/1.5	± 70	~ 0.01
!,2 Propanediol $C_3H_6(OH)_2$	Cr (V) Ch	10.8	2.5/0.37	+98 −100	~ 1
1,2 Ethanediol $C_2H_4(OH)_2$	Cr (V) Ch	9.7	2.5/0.5	± 80	~ 2
Butanol C_4H_9OH	EHBA Cr (V) Ch	13.5	2.5/0.3	± 93	3 - 4
EABA $C_2NH_7BH_3NH_3$	EHBA Cr (V) Ch	16.5	2.5/0.5	+75 −73	7(+), 3.5(−)[c]
Ammonia $^{14}NH_3$, $^{15}NH_3$	$NH_2\bullet$ Ir	17.5, 16.6	5.0/1.0	+97 −100	70, 175[d]
d-Butanol C_4D_9OD	EDBA Ch	23.8	2.5/0.3	± 50	not measured
d-Ammonia $^{14}ND_3$, $^{15}ND_3$	$ND_2\bullet$ Ir	30.0, 28.6	3.5/0.3	+49 −53	130(+), 260(−)
Lithium deuteride ^6LiD	f-center Ir	50	6.5/0.2	± 70	400

[a] Ch: chemically doped, Ir: doped through irradiation.

[b] The radiation dose which reduces the polarization by e^{-1} of its value.

[c] For positive and negative polarizations, respectively.

[d] In NH_3 there are two distinct regions of decay.

4 Frozen Spin Targets

Frozen spin targets are those targets which have been polarized and then had their cryogenic state changed such that the relaxation time of the polarization is very long. The two types; dynamically polarized and brute force polarized approach this state from opposite temperature extremes.

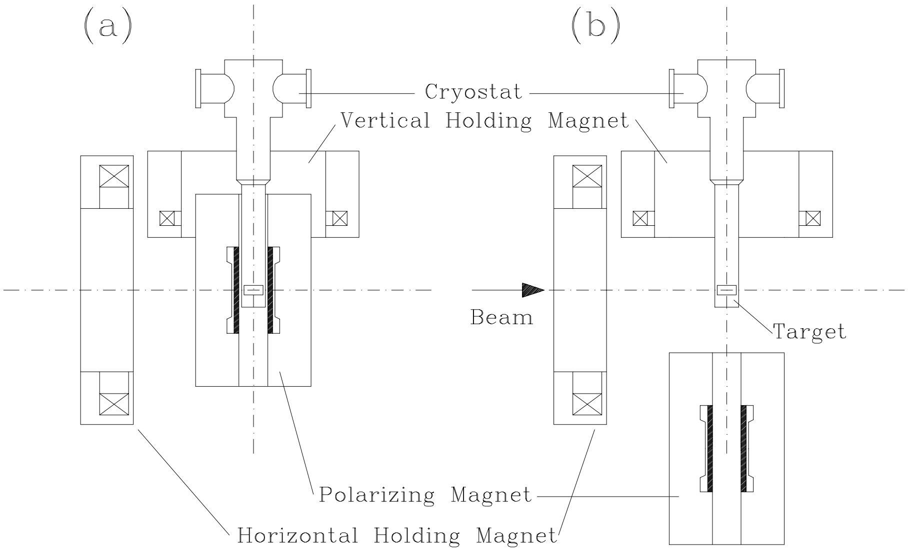

Figure 7: (a) Frozen spin target in 'polarizing mode', (b) in 'holding' mode in which either a vertical or horizontal spin direction can be maintained

4.1 Dynamically Polarized

Using a dilution refrigerator and magnetic fields of, typically, 2.5T, the material is dynamically polarized to as high a value as possible. The temperature is then reduced from $\sim$300mK to $<$100mK where the polarization decays very slowly because the relaxation times are very long. Reduction of the magnetic field, usually by physically moving the magnet, compromises the polarization decay but opens up the acceptance. Fig. 7 shows how this can be done. Such refrigerators are installed for experiments at a number of laboratories around the world, including Bonn, Dubna, PSI, Saclay, TRIUMF and KEK. Figure 7 shows a novel way of achieving the holding field of $\sim$0.5T with a thin solenoid wound into the dilution refrigerator. This has already been used in experiments at Bonn, on LEAR at CERN, and as reported at this conference, at Mainz.

4.2 Brute Force Polarized

The method of brute force (or equilibrium polarization) works for every polarized target, but as can be seen from Eqn. 1, in order to get high polarizations very low temperatures and very high magnetic fields are necessary. Molecular hydrogen cannot be polarized because of ortho to para conversion i.e., conversion to a magnetically inert state, at low temperatures. Honig [4] in 1967 proposed polarizing frozen HD by the brute force method. An HD target is attractive because a) the molecule is polarizable at low temperatures, b) it has a high dilution factor, c) it contains one neutron which can be polarized and d) the degree of polarization of H and D can be manipulated independently.

As in the case of DNP type targets, the HD must be doped, otherwise the relaxation times are too long for practical purposes. In the case of HD the dopant is ortho-H_2 and/or para-D_2, which allows spin-lattice coupling and a faster polarization build-up. As the polarization increases the density of ortho-H_2, as it converts to its magnetically inert state and the relaxation becomes very slow again. Proton polarizations achievable with this method are $sim80\%$, but deuteron polarizations are only about 20%. Moreover these polarizations are only reached after waiting for several decay constants of the impurities. Holding at fields of 17T and temperatures of 15mK for 3-4 weeks for ortho-H_2 (decay constant of 6.25 days) is practical but holding for months for para-D_2 (decay constant of 18.2 days) is not.

In order to enhance the deuteron polarization, a technique of adiabatic fast passage can be used to transfer proton polarization to the deuteron. This can be repeated several times and values of 50% are expected to be reached.

The targets are produced in a dilution refrigerator operating at 10 - 15 mK in a field of 15 - 20T and for the case of deuteron polarization must be maintained in this environment for 40 - 50 days and then extracted and stored while maintaining the polarization. Use in a scattering experiment requires that the target be inserted into the in-beam cryostat and maintained under less stringent conditions than production eg., 1T and 0.5K. The lifetime in the experiment will also depend on beam effects and so is limited to low intensity photon or neutron beams, for once the polarization has decayed in a target it has to be replaced.

At the time of this talk, proton targets of polarization ~60% are stored waiting to be used for the first time in experiments at LEGS at Brookhaven National Lab. The efficiency of the adiabatic fast passge method for the deuteron polarization is low, due to heating effects in the cables transmitting the RF power. Research is continuing on this.

5 Warm Targets

All the targets discussed so far require a considerable amount of sophisticated cryogenic equipment. Investigations at Kyoto University [5] have focussed on the possibilty of targets which can be polarized from liquid nitrogen to room temperatures. At room temperature and low magnetic field, the TE polarization of the electron is too small to use the standard DNP technique. However molecules such as napthalene or p-terphenyl can be doped with pentacene and grown as single crystals. The population differences in the triplet state of pentacene can be manipulated and the protons polarized in a DNP process. This process relies on optical pumping of the singlet ground state of the pentacene to an excited singlet state. A very fast transition to the lowest excited singlet state, S_1 occurs by internal conversion. Then due to a spin-orbit interaction, there can be a transition to the lowest of the triplet states T_0 and because of the selection rules there is alignment. During the time that the pentacene is in the T_0 state microwaves are applied in a DNP process and if the various relaxation times cooperate, protons are polarized. As the sequence of laser excitation and microwave sweep are repeated the proton polarization grows. A rather complicated process but at liquid nitrogen temperatures napthalene has been polarized to 30% and p-terphenyl to 19%. Room temperature polarizations are .12% and 1.3% respectively.

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SUMMARY of NMR WORKSHOP

The International Workshop on NMR in Polarized Targets was held in Charlottesville, Virginia on April 15th and 16th, 1998 and hosted by the University of Virginia. In association with the Workshop, a NMR clinic was held on April 20th in Newport News, Virginia and hosted by the Thomas Jefferson National Accelerator Facility.

Although specifically aimed at NMR questions in polarized targets, this workshop was considered to be 9th in the series of workshops on Polarized Target Materials and Techniques started at Argonne National Laboratory in 1978 and continued through the 7th at Bad Honnef (1994) and Vancouver (1996).

This Workshop had 39 participants from 16 institutions representing most of the places at which there is strong activity in polarized targets and was supported in part by the International Committee on High Energy Spin Physics.

The last workshop in which substantial time was devoted to the question of target polarization measurement by NMR techniques was the 1979 Coseners House Workshop at the Rutherford Laboratory. At this time two major steps forward were taken: the idea of using a series LCR circuit for tuning, instead of the more customary parallel tune, was introduced from CERN; and circuitry establishing phase-sensitive detection as a way of isolating the absorptive part of the signal was presented from the University of Liverpool. The proceedings can be found in Rutherford Laboratory Report RL-80-080.

After this of course, there have been a number of conferences and workshops at which advances in NMR measurement techniques have been presented, but none have focussed on NMR as a separate and complete topic. Hence holding a NMR workshop after 20 years was considered timely.

Since its introduction around the time of the Coseners House workshop, the Liverpool Q-meter has become the workhorse of the solid polarized target community. Originally designed for the measurement of proton polarization, in the intervening years it has been used in many different applications. However, with improvements in polarized target design, and new materials, a new generation of experiments have placed increasing demands on the versatility of the Q-meter. Again it was appropriate to hold the workshop at this time to see how Q-meter measurements and design could be improved to fit the demands of experiments.

The underlying themes of the workshop, in my opinion, were driven by requirements from modern experiments to improve the precision of the polarization measurement itself, and help improve the accuracy of that measurement by making the appropriate corrections. Into this category falls the correction of contributions from polarizable nuclei in any material. In some sense we are in a period of consolidating the gains of the past few years. In the case of protons, polarizations of close to 100% are routinely obtained, with ammonia having the highest fraction of protons. In my introduction to the workshop I showed a graph in which I plotted the increase in signal size, as represented by the product of polarization and proton content by weight, over time. An increase in signal size of almost an order of magnitude has led to the present day concerns about signal distortion and non-linearities and the corrections that have to be made. It is clear that further progress will be made only by developing new materials which have higher proton content and can be highly polarized. The situation with deuterons is similar, though recent measurements with lithium deuteride and the use of frequency modulation have given this area a boost. Of course with deuterons the concern is not with large signals, but with very small ones. This workshop has made clear that there is room for improvement in detecting and measuring small signals and that in measuring large signals we must pay careful attention to the details if we are to have credible measurements.

We have already seen evidence of measurement problems, which bear directly on our knowledge of target polarizations. There has been a trend to higher magnetic fields and, therefore higher NMR frequencies. This means that the effect of stray capacitance becomes more severe. Large signals have to be corrected and can lead to a false asymmetry between the positive and negative enhancements. There is the 'E155 problem' in which the coil design and its environment, exacerbated by the use of ammonia with its large dielectric constant, allowed the constant current assumption in the Q-meter design to be seriously violated in such a way that a parallel LCR resonance interfered with the normal tuned series one.

Signals from very small targets and the inherently small signals from some spin species compromise the precision of the polarization measurement because of poor signal to noise and techniques are being explored to enhance the signal. Also experiments carried out in high intensity beams can have problems with the NMR measurement, especially if there are differential heating and radiation damage effects.

These problems were discussed at this workshop and are being attacked in different ways at the many polarized target centers around the world.

One of the pleasures of this workshop for me was to see how the computer simulations of NMR circuits have evolved. It appears that we can now trouble shoot by using the simulations, provided we know all the rf parameters. In a series of talks, Geoff Court and Mike Houlden of the University of Liverpool showed that their simulations described how the circuitry of the Liverpool Q-meter dealt with large signals and indeed reproduce the odd effects observed in the E155 set-up. These simulations will be very helpful in solving the problems raised in several talks about odd effects in NMR measurement.

At the other end of the signal scale, Mike Houlden used his simulation package to show how one could modify the Liverpool Q-meter to subtract the Q-curve dynamically and then be able to boost the gain on very small signals. In this way the limitation on the gain, due to saturation of the Q-curve is removed. The simulation indicates that it is better to do the subtraction on the phase signal, rather than on the initial capacitive resonant tune. However Patrick Hautle presented evidence from PSI that subtracting the Q-curve before the phase signal does work and was being used in measuring the polarization of thin scintillator targets. Raffaella DeVita presented data from Genoa and Jefferson Lab. on small targets with small coil filling factors in which the polarization was obtained in the more traditional way of using a very large number (50,000) of sweeps to obtain an acceptable signal. This is an example of a target which would benefit from a subtraction of the Q-curve.

The choice of a target material for an experiment is dictated by several factors and quite often the choice of material containing polarizeable nuclei other than hydrogenous is made because of other compelling factors. The polarization of these nuclei must be measured and corrected for in the scattering experiment. It is useful if the Equal Spin Temperature (EST) theory holds, but generally the polarization has to be measured. Gerhard Reicherz presented work done at Bonn and Bochum on measurements with ^{15}N and ^{14}N ammonia. It is impossible to detect the TE signal for ^{14}N so another method had to be employed. On the other hand ^{15}N is much easier to measure, but ^{15}N ammonia is expensive and so is used in only small amounts. Nevertheless, such measurements are necessary for improving the accuracy of experiments such as performed by SMC and E143.

A way around the detection problem of the deuteron in most materials is to use ^{6}LiD, which has a narrow, easily detectable signal. Of course there are problems with using this material as a target, as Stephen Bültmann showed in presenting results from E155 at SLAC, but the advantage accruing from the quality of the NMR signal is a better measurement of the polarization. Another advantage is that, to a good approximation, the ^{6}Li nucleus can be considered a combination of an alpha particle and a deuteron, thus giving a second 'deuteron' for scattering. How good this approximation is has been considered elsewhere and was not really a topic for this workshop. What was considered here was the polarization of the ^{6}Li and of the small amount of ^{7}Li in the compound compared to the deuteron alone. Chris Harris of the U. of Virginia presented data that showed that each nucleus obeyed EST.

One presentation showed how corrections for the effects of heating and radiation damage could be made, while another looked at the question of whether the NMR technique gives a true measure of the polarization, i.e. is it that which a beam of incident particles sees?

One topic that must not be forgotten is that of control and data taking associated with the analogue measurement. Issues such as speed, linearity and other effects must be taken into account. Speaking as a user rather than provider, ease of use and choice of various options are also important considerations! Several presentations showed that there is a move to VXI/VME based control systems using LabView.

Finally, Art Brill of the U. of Virginia, bravely suggested, at a NMR workshop, that we should consider alternatives and suggested a non-destructive EPR technique. It would be desirable if the time and funding could be found to investigate such possibilities.

One topic that was not discussed at this workshop, and Patrick Hautle tells me that as far as he knows there is nothing new here, was adiabatic fast passage. Since some experiments will be using lithium deuterides and hydrides, which can have very long polarizing times, perhaps we should revisit this topic.

Another lesson of this workshop was that sanitary rf practices are extremely important: This was emphasized by Geoff Court and Mike Houlden in the clinic. Early design and testing is crucial and the use of rf instruments such as TDRs to characterize cables and connectors and vector impedance meters to aid in tuning the system, will in the long run save much time. Ideally, using a network analyzer to characterize a working target and inputting the measured rf parameters into a simulation program is the approach to take. However, because many of us are also responsible for other systems in a polarized target the demands on our time mostly do not allow this programmed approach and a 'seat of the pants ' culture has grown up. I hope that we can make the case that a careful design and testing period, followed by sufficient calibration time during an experiment, is the way to better polarization measurements.

In this summary I have tried to at least mention the topics presented and apologize for not mentioning everyone by name. The Workshop was valuable in pulling together many issues and perhaps indicating more coherent approaches to solutions. I would also suggest that the next workshop of this type be held before another 20 years have passed, preferably before the retirement of the veterans in this field.