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RECENT DEVELOPMENTS IN POLARIZED TARGETS AT CERN

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ABSTRACT

This report describes some of the development work done at CERN on polarized targets during past two years. The main emphasis is given to target material research, but some technical developments are also described.

INTRODUCTION

The major handicaps in today's polarized targets are their low hydrogen content and poor resistance to radiation, and sometimes the infrequent reversal due to slow polarization build-up. Although in the long term dramatic improvements in these parameters can be expected¹⁾, present users of polarized targets might profit from some more classical improvements which could possibly be implemented straight-away. In view of this we have tried to reintroduce the old butanol doped with the new Cr(V) complexes²⁾, and give some preliminary results here. Some tests have been done with deuterated butanol and propanediol as well.

Instrumental developments may also improve the figure of merit and availability of a given polarized target. In particular, optimization of the microwave parameters and automation of reversal are discussed with a view to shortening the reversal period. Cooling of the target by evaporation or dilution of ³He are compared, and a novel "easy loading" dilution refrigerator is briefly described. This refrigerator is now being used in our target material research.

DEVELOPMENT OF POLARIZABLE MATERIALS

The results of our recent work on 1-butanol alcohol doped with porphyraxide or a new stable Cr(V) complex BHHA-Cr(V)³⁾ are summarized in Table 1. We have also performed one measurement with deuterated propanediol PD-D₆ doped with another new stable complex EHBA-Cr(V)³⁾; the results are also given in Table 1. The advantages of the above materials are i) the direct chemical doping by dissolving a stable paramagnetic impurity allows large quantities of uniform material to be prepared, and ii) the effective chemical composition of BuOH is CH_{1.88} in comparison with propanediol which is equivalent to CH_{1.41}, i.e. BuOH has 1.33 times more hydrogen per carbon atom than PD. The second advantage might be compensated by the somewhat lower polarization so far obtained in BuOH, but we hope to find better ways of preparing solid solutions of the new complexes. In the following we shall briefly discuss the results of Table 1.

BuOH-PR: The two samples were prepared in the usual way by adding 5% water to BuOH and dissolving the complex in it after having deoxygenated the solution by bubbling dry N₂ through the liquid. Porphyraxide decays with a time constant of some tens of minutes in a

Table I

Material	Dopant and concentration a) (spin/ml) or (%)	f^+/f^- (GHz)	$\frac{\max P_H^+}{\max P_H^-}$ (%)	$\frac{\max P_D^+}{\max P_D^-}$ (%)	$\tau_{0.7}$ b) (min)	τ c) T (K)	Refr. c)
BuOH +5% H ₂ O	PR "1/2%"	70.235 70.580	67 -65	-	25	20 min @ 0.91 700 h @ 0.04	DR
BuOH +5% H ₂ O	PR "2%"	70.235 70.580	80 -78	-	10	11 min @ 0.98	DR
BuOD(D ₁₀) no water	BHHA-Cr(V) 10 ²⁰	69.180 69.685	(±94) d)	+30 -30	5	59 sec @ 0.98	DR
PD(D ₆) no additive	EHBA-Cr(V) 10 ²⁰	69.230 69.708	(±97) d)	+40 -40	10	3.5 min @ 0.98	DR
BuOH no water	EHBA-Cr(V) 3 × 10 ¹⁹		+75 -77		15	21 min @ 0.78	³ He

a) Abbreviations: PR = porphyrine; BHHA-Cr(V) = chromium (V) complex with 2-butyl-2-hydroxyhexanoic acid (see Refs. 2); EHBA-Cr(V) = chromium (V) complex with 2-ethyl-2-hydroxybutyric acid (see Refs. 2).

b) $\tau_{0.7}$ means the time necessary for reaching $0.7 \times (\max P)$ starting from opposite maximum polarization.

c) DR = dilution refrigerator; ³He = helium three evaporation refrigerator.

d) These numbers are in parentheses because they refer to free proton polarization of a deuterated substance, which is normally not used in a proton target.

solution thus prepared, which gives time to make beads from quantities of the order of some tens of millilitres. The first of our samples was taken out of the last 25% of beads prepared from a 100 ml solution; the estimated effective concentration is indicated as $\frac{1}{2}\%$, referring to the concentration equivalent to that of a freshly prepared solution. The second sample was prepared rapidly into beads after having dissolved the nominal amount of 2% of porphyrexide. The polarization results show a marked difference between these two samples, both of which were cooled in the mixing chamber of a large dilution refrigerator³⁾ at 25 kG field.

The optimum microwave frequencies observed for the two signs of polarization correspond rather well to those observed earlier. Also, the relaxation and polarization times roughly agree with earlier work. Contrary to the case of PD-Cr(V), the lower temperature available in the dilution refrigerator does not seem to greatly improve the polarizations which have already been reached in ³He evaporation refrigerators. This is in agreement with the predictions of Borghini⁴⁾. Another point of comparison is that the optimum frequency in BuOH-PR is much flatter than that in PD-Cr(V); in the above samples, little polarization change was observed in the interval of ± 10 MHz around the optimum frequencies.

BuOD(D₁₀)-BHHA-Cr(V): The butanol sample was 98% deuterated and no water was added to it, because the BHHA-Cr(V) complex was soluble in it at room temperature. Upon solidification, however, the complex may have crystallized to form clusters. This is supported by the line shape of the residual hydrogen nuclei in the sample, which is shown in Fig. 1. The narrow line corresponds to the usual hydrogen NMR frequency in butanol; the broad line is supposed to belong to the hydrogen in the BHHA-Cr(V) molecules, which have formed clusters and therefore see an effective field smaller than the weakly magnetized BuOD matrix. Also, the maximum polarization $P_D = \pm 30\%$ and the large distance (about 500 MHz) between the optimum positive and negative frequencies suggest that the material does not have a uniform distribution of paramagnetic spins. However, the spin temperature equilibrium was attained rapidly under microwave irradiation. The spin-lattice relaxation was unusually rapid in this sample.

PD(D₆)-EHBA-Cr(V): The sample was prepared by dissolving the complex directly into the partly deuterated propanediol CD₂OHCD₂OHCD₃. The stability of the complex in the deuterated solution at room temperature and exposed to usual atmosphere was far better than the stability in an undeuterated solution. The somewhat higher polarization obtained in this sample compared with the BuOD(D₁₀) might be explained by a better distribution of the complex in the material; no second hydrogen line could be distinguished. However, the maximum polarizations of +40% and -40% are worse than those in PD(D₆)-Cr(V) mixed with PD(D₈), where +44% and -47% maximum deuteron polarizations have been measured in a large target⁵⁾.

BuOH-EHBA-Cr(V): This test was done with a large sample (40 cm³) cooled in the copper cavity of a ³He evaporation refrigerator. No water was added to the butanol. The polarization results suggest that this material might be a potentially good replacement for the standard PD-Cr(V). The advantages of propanediol -- routinely about 90% polarization and a fast polarization speed (60% in 10 min) --

Fig. 1. The proton NMR line shape in 98% deuterated butanol alcohol. The narrow line occurs at the usual frequency in pure butanol and is interpreted as belonging to the 2% unsubstituted protons. The broad line is supposed to be due to clusters of the complex molecules which have a larger magnetization and therefore a shifted resonance frequency.

might well be compensated in some experiments by the higher hydrogen content of butanol.

The work with the new complexes is in progress, and we expect that improvements can be made quite soon in the preparation of these materials.

INSTRUMENTAL DEVELOPMENTS

Microwave optimization: It has been noted on several occasions that the optimum microwave frequency in PD-Cr(V) changes with the accumulating dose of radiation in the target. If a computer is used for deducing the polarization from a digitized NMR signal, then it is possible to control the microwave frequency in such a way that one always stays at the optimum. This was implemented at CERN by F. Udo; the microwave frequency was "modulated" synchronously with the target polarization measurement cycle, and the time derivative of the polarization was then "demodulated" at the same frequency in the computer. This demodulated signal was fed back to the frequency control of the microwave supply. In this way the system searches out the frequency at which the polarization time derivative is maximum, ending in a steady state with maximum polarization. The microwave power was stabilized at the cavity by feedback from the ^3He vapour pressure to a motorized attenuator.

The same scheme could be applied to maximizing the polarization growth speed in conditions where the target polarization reversal should take place as rapidly as possible.

Rapid polarization reversal: Several schemes could be considered, of which the adiabatic fast passage and rotating polarization are not discussed here. We focus our attention on two very simple and easily realizable cases: rotating field with frozen spin technique, and combined field zeroing and strong microwave irradiation. The first case is relevant to experiments where one does not have high beam intensity, say below 10^7 charged particles/(sec cm 2); the latter is compatible with high beam intensity.

Rotating field technique: The target polarization always follows the direction of a slowly rotating field, which can be obtained for example with a rotating magnet or with orthogonal fields. The latter case has been implemented at CERN by winding a thin superconducting solenoid axially around the vacuum jacket of the CERN frozen spin target³). The additional wall thickness around the target increased by 0.4 mm, including the wire, isolation, and bonding epoxy. This solenoid can carry 20 A current, giving a field of 6.5 kG. The solenoid can be used either for holding the target polarization axially to the beam, or for helping to rotate a transverse field to the opposite direction without passing through zero. In the former case, the main (polarizing) field can be used for rotating the polarization to the opposite axial position without passing through zero.

The asymmetry measured in an experiment where the target polarization is rotated together with field will be at least partly masked by the varying field. This can be taken into account by reversing the target polarization with respect to the magnetic field with regular intervals. Statistically, two experiments instead of one have to be done, and the somewhat increased counting time has to be compared

with the suppression of drifts in the apparatus and beam gained by frequent reversals of the polarization. It is clear that the false asymmetry created by the rotating field is relatively smaller in the case of polarization axial to the beam.

Field zeroing technique: Some magnets can be swept rather rapidly to zero field and back up to a stable 25 kG field. This offers a rapid means of zeroing the target polarization, and therefore a significant speeding up of the time necessary for reaching the polarization value, say 60%, at which the data-taking will be started. The gain in the reversal time would be rather insignificant if the polarization build-up would be exponential. This is rarely the case, and therefore the time spent from 0 to 60% (for example) depends strongly on the initial polarization at the moment when one switches on the microwaves at the frequency of the opposite polarization. Some physical insight into the situation can be gained by first noting that some parts of the target polarize slower than others, depending on the effective value of the magnetic field. Another cause of non-uniform polarization growth might be material inhomogeneity, in particular in large targets. Furthermore, microscopic polarization non-uniformities will definitely arise when the spin diffusion is needed for spreading the polarization around paramagnetic centres or clusters, a typical case being ammonia doped with PD-Cr(V) ⁶⁾.

Combined with microwave optimization, the field zeroing technique brings the reversal time from 25 min to 10 min in a large (40 cm³) propanediol target cooled in a ³He refrigerator. With microwave optimization, some further gain in reaching 60% could be expected, and if the experimenter would be happy to start counting at 40% polarization, only a few minutes are spent in the reversal.

Dilution refrigerators: It is a rather common belief that dilution refrigeration does not bring about any significant improvement in the dynamic polarization in comparison with the ³He evaporation refrigeration, because at the power levels needed, the helium temperatures are almost equal in both cases. This is true: as an example we might compare our ³He refrigerators and dilution refrigerators, which use the same size of pumps and cool 50 mW load at 485 and 305 mK temperatures, correspondingly. The ratio of these two temperatures is 0.63. However, these are cooling fluid temperatures, which are not directly relevant in dynamic polarization. The important parameter is the target material lattice temperature T_L.

The essential difference between these two cooling systems lies in the fact that the heat transfer mechanism in them is fundamentally different: in evaporating ³He the dominating heat resistance is the so-called nucleate boiling resistance⁷⁾, whereas in the dilution refrigerator it is the Kapitza resistance. In the former case the rise of the lattice temperature T_L can be determined from the boiling correlations, which have never been studied systematically for ³He and very little for ⁴He at low pressure. Generally, in nucleate boiling the temperature rise is proportional to the heat flux, giving

$$T_L = T_3 + \Delta T = T_3 + \frac{\dot{Q}_m}{\sigma_t \cdot c} , \quad (1)$$

where T_3 is the ^3He temperature ($T_3 = L_3^0 / (k \log \dot{Q}_m / A)$), $\dot{Q}_m$ is the microwave power dissipated in the target material, and c is a coefficient which relates the heat flux to the temperature rise ΔT in nucleate boiling; c may depend weakly on the vapour pressure, which we omit in the following. In a dilution refrigerator the lattice temperature can be expressed by⁸⁾

$$T_L = \left(\frac{1}{\sigma_t S_t} + \frac{1}{12.5 \sigma S} \right)^{1/4} \dot{Q}_m^{1/4}, \quad (2)$$

where σ is the effective surface area of the heat exchanger⁸⁾ and S the Kapitza surface conductivity constant. Taking the heat input 50 mW of the above example and applying it to a 40 cm³ target made in the form of 1.5 mm beads, we find lattice temperatures of 800 mK in the ^3He refrigerator and 365 mK in the dilution refrigerator, by taking $c = 0.15 \text{ mW cm}^{-2} \text{ K}^{-1}$ and $S_t = 2.5 \times 10^{-3} \text{ W K}^{-4} \text{ cm}^{-2}$. The ratio of these temperatures is now 0.46, illustrating well the large spin temperature differences that can be found particularly in the dynamic polarization of deuterium nuclear spins.

Easy-loading dilution refrigerator: As can be deduced from Eq. (2), with a limited target surface area σ_t it is not necessary that the dilution refrigerator runs at a temperature much below, say, 0.3 K. Such a refrigerator can theoretically run without isolating vacuum separating the mixing chamber and the still, allowing very easy loading of the target material into the mixing chamber. We have constructed a dilution refrigerator insert⁹⁾ replacing an old ^3He refrigerator; in this dilution refrigerator the heat exchanger and mixing chamber are covered with a simple teflon jacket, which just holds the helium mixture inside. The creeping superfluid film on the outside surface of this jacket, together with other residual heat leaks, limits the ultimate low temperature of this refrigerator to about 140 mK, probably preventing its use in a frozen spin target, but allowing, for example, high deuteron polarizations to be reached in the continuous mode. This refrigerator can be loaded in about 30 min, and material tests can be performed in it on a basis of one sample per two days, calibration included. The maximum cooling power is 20 mW, allowing rather sizeable targets to be polarized. For details we refer to the original publication⁹⁾.

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REFERENCES

- 1) G.R. Court et al., Report of the workshop on polarized target materials, these proceedings.

- 2) M. Krumpolc and J. Roček, Synthesis of stable chromium (V) complexes of tertiary hydroxy acids, to be published in J. Amer. Chem. Soc.
M. Krumpolc and J. Roček, J. Amer. Chem. Soc. 100, 145 (1978).
J. Amer. Chem. Soc. 99, 137 (1977).
J. Amer. Chem. Soc. 98, 872 (1976).
- 3) T.O. Niinikoski and F. Udo, Nucl. Instrum. Methods 134, 219 (1976).
- 4) M. Borghini, Proc. 2nd Internat. Conf. on Polarized Targets, Berkeley, 1971 (Ed. G. Shapiro) (National Technical Information Service, Springfield, Virginia, 1972), LBL 500, UC 34 Physics, p. 1.
- 5) T.O. Niinikoski, Proc. AIP Conference on High-Energy Physics with Polarized Beams and Targets, Argonne, 1976 (AIP Conf. Proceedings No. 35), p. 458 (1976).
- 6) K. Scheffler, Proc. 2nd Internat. Conf. on Polarized Targets, Berkeley, 1971 (Ed. G. Shapiro) (National Technical Information Service, Springfield, Virginia, 1972), LBL 500, UC 34 Physics, p. 271.
- 7) See, for example, I.R. McDougall, Cryogenics 11, 260 (1971) and references therein.
- 8) T.O. Niinikoski, Proc. ICEC 6, Grenoble, 1976, p. 102.
- 9) T.O. Niinikoski and J.-M. Rieubland, Proc. IIR Commission A 1-2, Zurich, 1978 (International Institute of Refrigeration, Paris, 1978), p. 181.