60 years
polarized solid targets
for particle physics experiments

W. Meyer

Developments with the focus on
target materials and cryogenics
seen from my Bonn/Bochum view

A commemoration to the long-lasting Bonn PT-Laboratory

Dynamic Polarized Solid Target Method:

- Production of a high polarization degree in a suitable material with a high content on polarizable nucleons and 'free' electrons (radicals) by means of
- high magnetic field
- extreme low temperature
- microwave irradiation → (dynamic nuclear polarization (DNP))
- Polarization detection by Nuclear Magnetic Resonance (NMR) technique

In addition:

- Radiation hardness of the polarization
- Easy handling and fast target material exchange or polarization refreshment
- ESR- apparatus reinvented

The prehistory of the Dynamic Nuclear Polarization (DNP) - 1953 until 1962 -

- … can be traced back to the late **1940s**, especially to:
  - Discovery of the Nuclear Magnetic Resonance (NMR)
  - Simultaneous development of the Electronic Spin Resonance (ESR)
  - Elucidation of nuclear and electron spin lattice relaxation processes in solids

- **1953**
  OVERHAUSER EFFECT: For metals, radiofrequency (RF) saturation of the ESR-line of the conduction electrons would enhance the nuclear polarization by a large factor. Carver and Slichter: Experimental proof

- **1955**
  A. Abragam invented the OVERHAUSER EFFECT for nonmetal substances

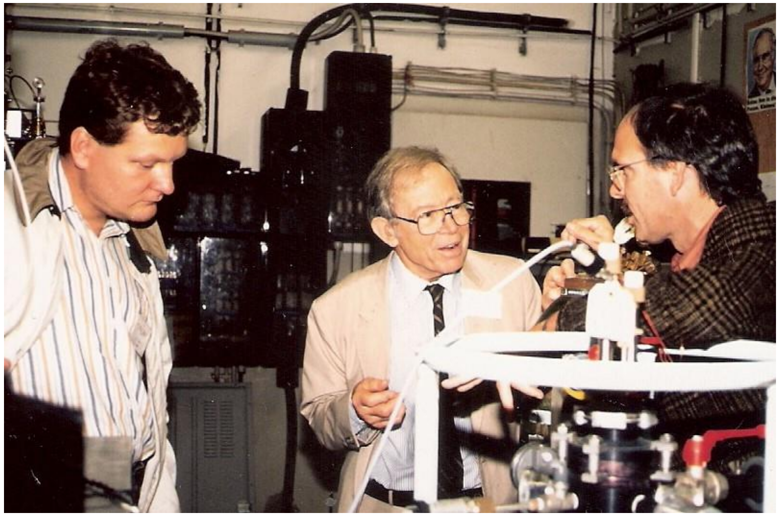
- **1957**
  A. Abragam (Saclay) and C.D. Jeffries (Berkeley): Polarization of nuclei by RF (microwave) saturation of so-called forbidden transitions I_+S_+ and I_+S_- in the dipolar case should give negative and positive polarization enhancement

- **1958**
  A. Abragam and W. Proctor (Saclay): Prototype experiment in a crystal of LiF at room temperature, in which the „S“ spins were ^{19}F nuclei and the „I“ spins the ^{6}Li nuclei in dipolar coupling → positive and negative Li NMR signals

→ l` effet solide – to distinguish this case from the OVERHAUSER EFFECT

For further reading: Opening Talk of C.D. Jeffries „History of the Development of polarized Targets“ at the 9th International Symposium for High Energy Spin Physics → held in Bonn 1990

C.D. Jeffries in the Bonn Laboratory during the spin symposium



First particle physics experiments
- 1962 until 1966 -

- **1962**
  Polarized target in SACLAY (A. Abragam, M. Borghini, P. Roubeau et al.)
  20 MeV polarized protons on a polarized crystal (0,12 mm thick):
  Lanthanum Magnesium Nitrate ($La_2Mg_3(NO_3)_{12}$ $24H_2O$ (LMN) with 0.2% of the La replaced with Ce: 20% pol.

- **1963**
  First polarized target for „High energy particle scattering" off 246 MeV pions in Berkeley (O. Chamberlain, G. Shapiro, C.D. Jeffries)

- **1966**
  Polarized target for high energy particle physics at CERN – Saclay (M. Borghini, P. Roubeau, C. Ryter)

- **1966**
  1st polarized target conference in Saclay:
  Reports on polarized Target in Saclay, Berkeley, CERN, Argonne, Dubna, Harward, Nagoya (INS-Tokyo), Rutherford.
  All using Nd:LMN (see Table I)

Problem 1: Only 3,1% free protons polarizable (f = dilution factor)

Problem 2: Poor radiation damage resistance of the polarization

Reminder: Characteristic of the target („Figure Of Merit“)

$$FOM_{target} = P^2 \cdot f^2 \cdot \kappa \cdot \rho \sim 1/t$$

P = Polarization value
κ = filling factor (heat removal from the material)
ρ = Density
t = measuring time for asymmetry A a given $\Delta A/A$

1966

TABLE I: Typical polarized target parameters (1966)

Material:	1% Nd:LMN 3.1% free protons
Thickness:	25 to 75 mm
Volume:	1.5 to 40 cm^3
Magnetic field:	17 to 20 kG
Temperature:	0.95 to 1.3K
Microwave frequency:	64 to 75 GHz; 1 watt
He consumption:	50 to 150 ℓ/day
Proton polarization:	40 to 75%
Radiation damage after:	10^{10} to 10^{11} particles/cm^2

1970

TABLE II: Materials in which significant nuclear polarizations by DNP had been achieved by August 1970

Sample	Nuc.	Pol. (%)	H (kG)	T (K)	% Nuc.	Refs.
Ce:LMN	^1H	20	13.3	1.2	3.1	1
Nd:LMN	^1H	72	19.5	1.5	3.1	2,3
Ethanol & prophyrexide	^1H	35	25	1.05	13.5	4
Butanol & prophyrexide	^1H	40	25	1.0	13.5	5
Butanol & prophyrexide	^1H	67	25	0.5	13.5	6,7
Ethylene glycol & Cr complex	^1H	50	25	1.1	9.7	8,9
Ethylene glycol & Cr complex	^1H	80	25	0.5	9.7	10
Glycerol + H_2O + porphyrexide	^1H	55	25	0.6	9.9	7

1. A. Abragam, M. Borghini, and M. Chapellier, Compt. Rend. 255, 1343 (1962).
2. T. J. Schmugge and C. D. Jeffries, Phys. Rev. 122, 1781 (1962); and Phys. Rev. 138, A1785 (1965).
3. O. Chamberlain *et al.*, Phys. Lett. 7, 293 (1963).
4. M. Borghini, S. Mango, O. Runolfsson, and J. Vermeulen, Proc. Int. Conf. on Polarized Targets and Ion Sources (Saclay, 1966), p. 387.
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8. V. N. Fedotov, Sov. Phys. JETP 26, 1123 (1968).
9. H. Glattli *et al.*, Phys. Lett. 29A, 250 (1969).
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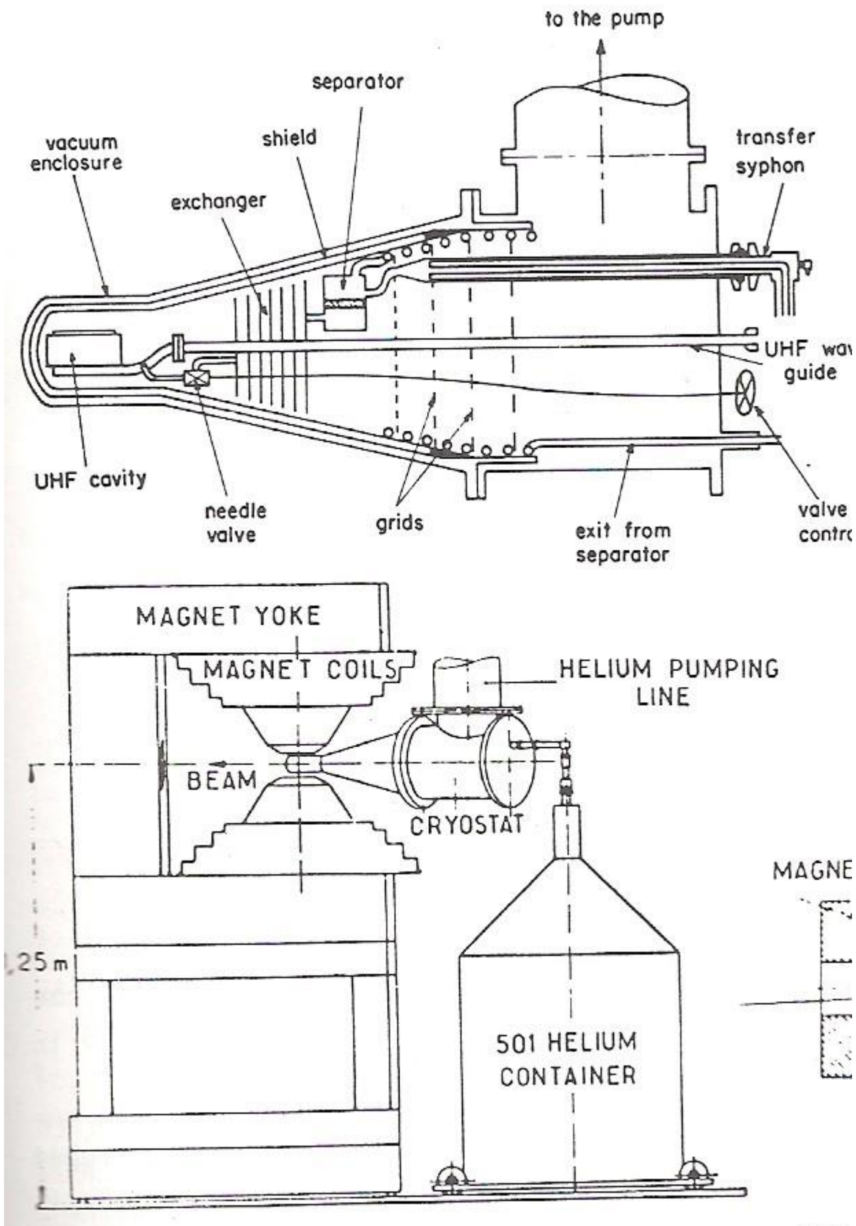
Polarized target start at CERN

- **1965**
  Michel Borghini moved from Saclay to CERN; „Better“ polarized target materials was a priority of research (Borghini - CERN Yellow Report 66-3, 1966, „Choice of substances for polarized proton targets“)

- **1968**
  - Breakthrough with a Butanol +5% H_2O + Prophyrexide mixture (f = 13.5%; Target material for better cooling in form of beads (1 - 2mm) (S. Mango ,O. Runolfsson, M. Borghini, NIM72, 1969, 456)
  - M. Borghini developed a new phenomenological model of DNP: “Spin temperature model” → Borghini Model. He also found a way to experimentally test the predictions in the low temperature regime.

- **Up to that time:**
  Provotorov theory correctly describes the behavior of dipolar coupled spin systems under saturation. With the exception of the solid state rate equations so far the considerations were restricted to the ‘so called' High Temperature Approximation. This assumption is far from being valid under usual conditions of a DNP experiment ($T < 1$ K). Within this framework an expression for the spin temperature can be derived (degree of saturation; width of the resonance line; time constants).

Cern-Saclay arrangement

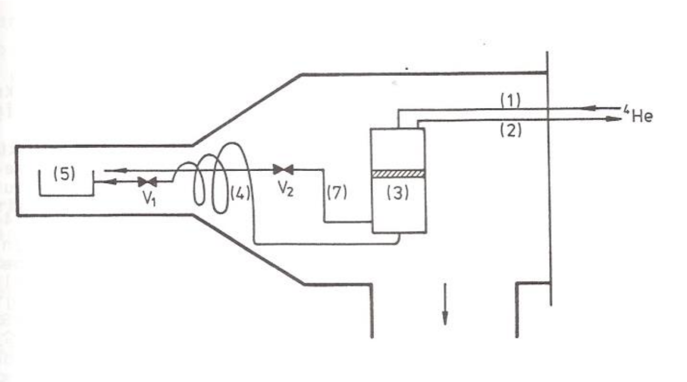


Continuous flow
^{4}He-cryostat
(Roubeau type)

Magnet design
for better particle
detection

1970s: Polarized Solid Targets came into fashion in every particle physics laboratory … also in Bonn

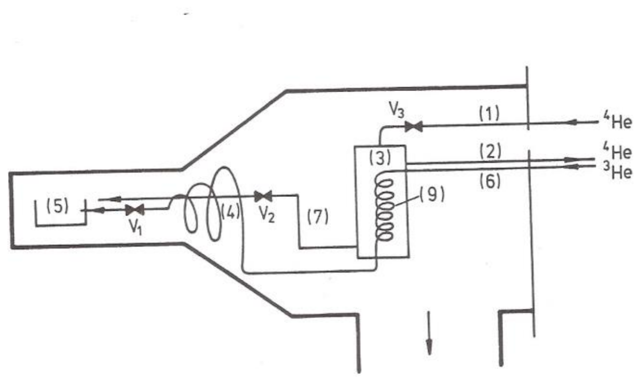
- **1970**
  First experiment at the 2.5 GeV Bonn synchrotron (CERN-copy of the Roubeau type ^{4}He-cryostat and 2.5 T iron-magnet



Schematisches Flußdiagramm des ^{4}He-Verdampfer-Kryostaten

- **1972**
  Start of a 50 year long cooperation between BONN/BOCHUM and NAGOYA/YAMAGATA (see later)

- **1973**
  ^{3}He- cryostat : New scheme (H. Herr et al. Nucl. Instr. & Meth. 204, 1974, 59)
  No separated ^{3}He and ^{4}He vacuum systems



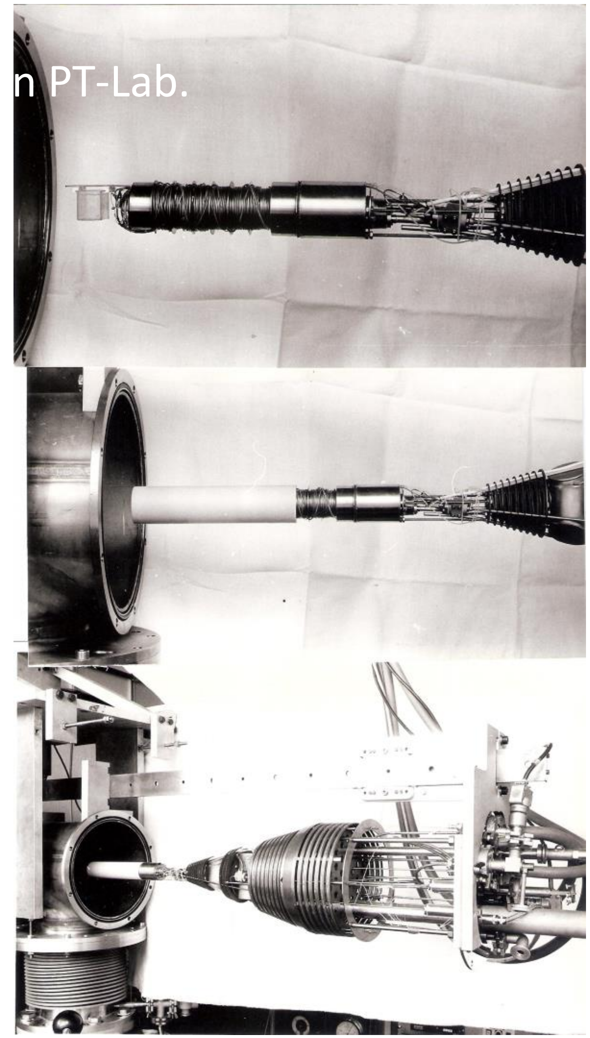
DESY-PT people comment: "Quick and dirty" version

- **> 1970**
  CERN: high power ^{3}He/^{4}He dilution refrigerators ($T < 100$ mK) developed by Tapio Niinikoski (new PT group leader)
  Modern ^{3}He/^{4}He dilution refrigerators based on the "Quick and dirty scheme" (i.e. One pumping system for the ^{4}He and ^{3}He/^{4}He mixture)
  Bonn (> 1982 W. Meyer et al.); Nagoya (> 1985 N. Horikawa et al.)
  Saclay/ Argonne (> 1988 – Chaumette et al.); CERN (> 1990 – T. Niinikoski, P. Berglund et al.); Bonn (> 1995 – H.Dutz et al.)
  TJNAF (> 2005 – Ch. Keith et al.) Dubna/Mainz (> 2008 – Usov et al.)

Bonn $^3He/^4He$ dilution refrigerator (1982)

From SACLAY ^4He refrigerator (Roubeau type) to ^3He refrigerator and finally to the $^3He/^4He$ refrigerator (copy of the front part from CERN) with lowest temperature of 150mK (see picture).

Main advantages: (1) Easy loading system
(2) Ready for measurements after about 3 hours



INTERMEZZO

“All roads lead to Rome”, but some in the context of dilution refrigeration to Bonn!

Heinz London – a Jewish German/British physicist

07.11.1907 born in Bonn

1926/27 Physics study at the Bonn University

1933 PhD in Breslau (Superconductivity) – left to England

1934 Clarendon Laboratory in Oxford
(London’sche equations – Ohm-law for superconductors)

1950/51 Invention of $^{3}He/^{4}He$-Mischungskühlung = dilution refrigeration

1964 1. construction of a $^{3}He/^{4}He$ dilution refrigerator at the Kamerlingh Ommes Laboratory in Leiden (Netherland)

Deuterated Target Materials

Proton target materials (alcohols) can be polarized almost completely at experimental conditions of 2.5 T and 5.0 T / 100 mK to 1K.

So far this was not the case for deuterated materials :

- lower' magnetic moment of the deuteron (Brillouin function)
- use of radicals sufficient to cool protons (Equal Spin Temperature concept)

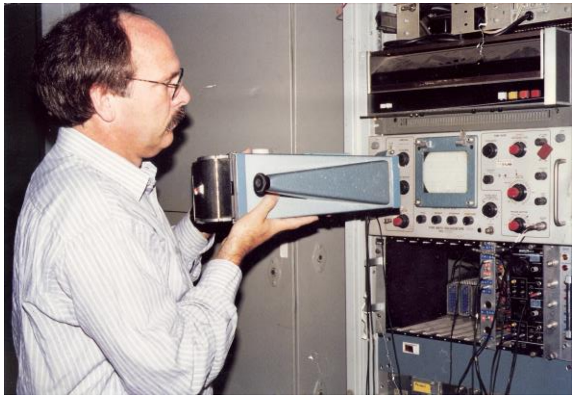
The effect of narrowing the ESR-line results in an increase of the inverse nuclear temperature, obtained by DNP. Borghini (1966): “What one loses due to the Brillouin function, one recuperates in the spin temperature” (see talk St. Goertz).

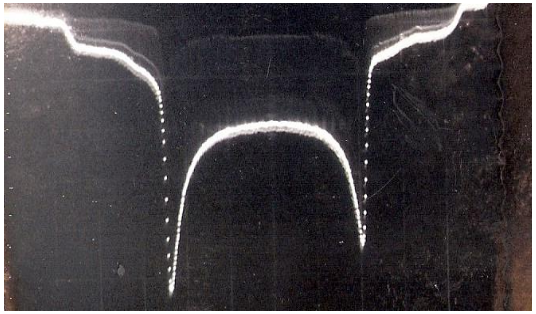
e.g. Bonn started 1974 with deuterated butanol + porphyrexide: P = 20%

Intermezzo

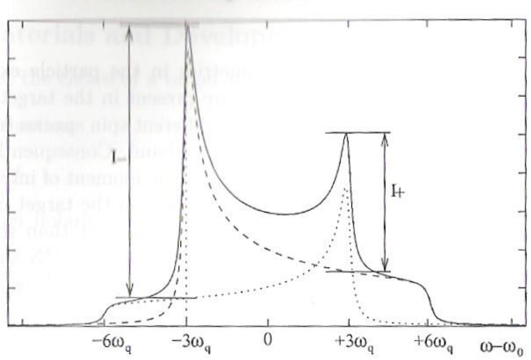
World first T.E. signal of deuterons in D-Butanol (24.07.1974) by O. Kaul in Bonn

P = 0,05%





P = -11%



Asymmetry Method

Ammonia

Next step in the PT-community: Material with higher f wanted

AMMONIA NH_3 with f = 17.3: Already prepared for polarization with different chemical radicals in the 1970s at CERN (Scheffler, Nucl. Instr.& Meth., 82 (1970), 205) → without reproducible results

What about radical production by irradiation?

First attempts already in the 1960s with X-rays (see talk C.D. Jeffries 1990)

1968 Solid H_2 and HD irradiated by bremsstrahlung (C. Solem and G.A. Rebka)

2nd Workshop on Polarized Target Materials in Abingdon (1979)

- Reports from D. Crabb (Univ. Michigan) and G. Court (Univ. Liverpool) about Butanol, irradiated with protons or photons or reactor neutrons.

- Proton irradiated Ammonia under liquid nitrogen at CERN (T.Niinikoski et al.) P = 92%; but explosions stopped activities

- Electron (20 GeV) irradiation of NH_3 at SLAC (V.Hughes, M.Seely). Fragile NH_3 beads turned over to powder. Activities stopped and V. Hughes moved to CERN and participates in the ongoing spin measurements.

- Electron (3 MeV) irradiation of ^{7}LiF, ^{7}LiH and ^{6}LiD (Y. Roinel); irradiation under liquid argon. P = 70% for Li^{6}D at 6.5 T and 0.2 K.

Combined all informations: NH_3 irradiation at the 20 MeV electron injection Linac at BONN, and NH_3 under liquid argon (87K) started 1980

New chemical dopant for alcohols and diols: Stable CrV-complexes in form of powder HMBA, BHHA and EHBA M. Krumpolc (Univ. Illinois, Chicago)
→ SMC- experiments at CERN with D-Butanol in the late 1990s

PT Activities from Bonn > 1980 (1)

- **1980**
  Spin Symposium in Lausanne → First report of a higher polarization resistance against radiation damage in $^{14}NH_3$ (U. Härtel, W.M.et al.)

- **1980**
  ND_3: P_D = 3% at 1 K , 12% at 0.5 K, ??? in a $^3He/^4He$ dilution refrigerator

- **1981**
  - W.M at CERN. Work in Scheffler`s Chemistry Lab. at CERN.
  - Production of glassy NH_3-beads by dropping in liquid isopentane.
  - Studied internal paper about an easy loading $^3He/^4He$ dilution refrigerator
  - First contact at CERN with EMC-collaborator G. Court (Liverpool)
  - First contact with K. Kilian (FZ-Jülich)

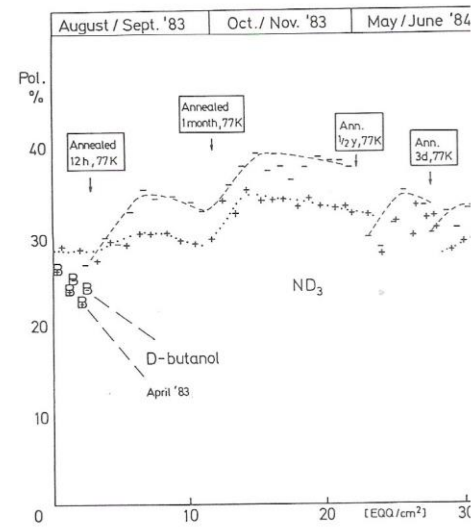
- **1983**
  Irradiation of 4 litres of NH_3 at the Bonn 20 MeV Linac for EMC-experiment at CERN → Important for a continuation of deep inelastic scattering experiments at CERN

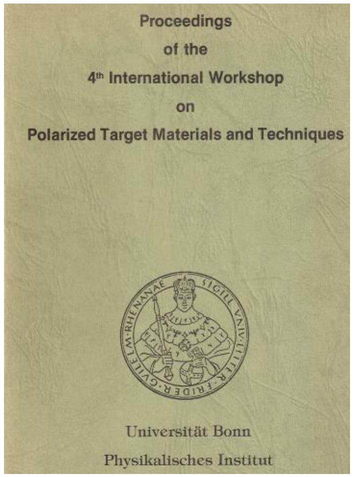
PT Activities from Bonn > 1980 (2)

- **1983**
  Photodisintegration experiment
  at the 2.5 GeV Bonn synchrotron
  using ND_3 with the easy loading
  $^3He/^4He$ dilution refrigerator

- **1984**
  4th International Workshop on Polarized
  Target Materials and Techniques
  Bonn ("Green Booklet" Editor: W. M.)
  Topic : Polarized Ammonia

- **1985**
  B. Mecking went to CEBAF (PT for CLAS)





PT Activities from Bonn > 1980 (3)

- **1985**
  Experiment with a tensor polarized ND_3 target at Bonn to disentangle the electrical and quadrupole form factors of the deuteron (B. Boden, V. Burkert, W. M. et al.,Z. Physik C 49 (1991) 175)

  The only one particle physics experiment (elastic electron deuteron scattering), using a tensor polarized ND_3 target with $P_{zz} \sim + 0,2$ (see talk E. Long)

- **1989**
  Frozen Spin Target activities started at Bonn with a copy of the SACLAY magnet system (see H. Dutz)
  The goal: To replace the huge superconducting magnets with their large fringe fields by “thin internal holding coils”.

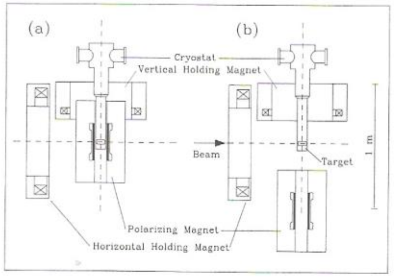


Figure 50: Set-up of a frozen spin target in the polarization mode (a) and in the holding mode (b), in which a vertical as well as a horizontal spin direction can be realized (SACLAY version). From Ref. [237]

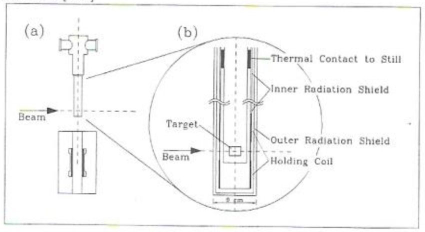


Figure 51: Holding mode (a) of a frozen spin target with an internal superconducting holding coil; (b) shows a close-up of the target area. This concept with a split-pair configuration of the holding coil was used for the measurement of the eta photoproduction on polarized protons at the Bonn tagged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

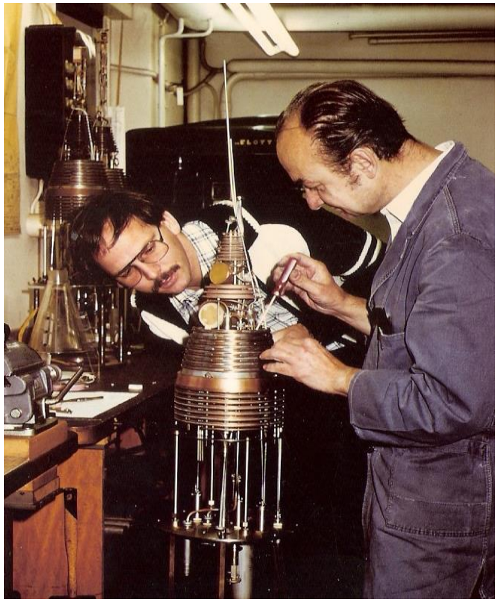
PT Activities from Bonn > 1980 (4)

- **1990**
  Lithium deuteride (^{6}LiD) activities with its high dilution factor f ~ 0.5 started (see talk St. Goertz)
- **1993**
  First eta-photoproduction experiment with the PHOENICS detector at ELSA and frozen spin target with internal holding coil for transverse polarization; A. Thomas went to the University Mainz (see talk A. Thomas)
- **1995**
  GHD activities started, triggered by a new horizontal ^{4}He/^{3}He refrigerator with internal holding coil (H. Dutz et al.) and gallium-arsenide superlattice crystals for polarized electron sources (T. Nakanishi et al.)
- **>1998**
  GDH measurements started at Mainz (MAMI) and Bonn (ELSA)
- **>2005**
  Photoproduction experiments at ELSA with Crystal-Barrel-detector, to measure double polarization observables (R. Beck, U. Thoma et al.)

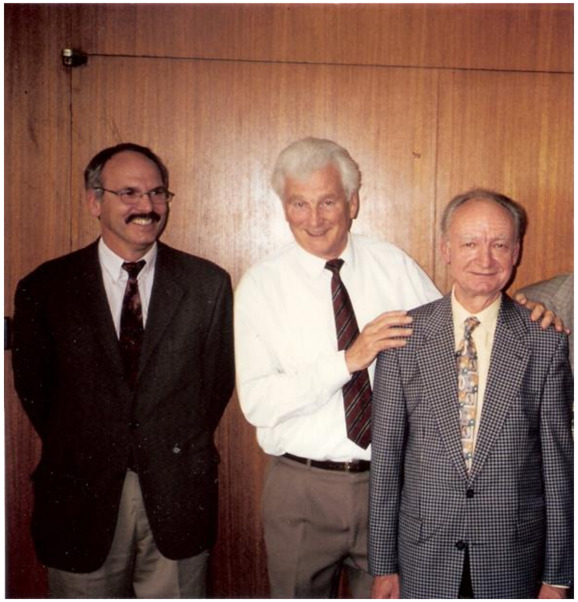
Herbert Peschel

„Quick and highly effective“ and „conditia sine qua non“:

The PT-Bonn precision-mechanic H. Peschel



1982



1998 - retirement of H. Peschel

He deserves thanks from all group members for his excellent work during 29 years.

PT Bochum Activities started 1995

- **1995**
  W. M., G. Reicherz, St. Goertz went to the University Bochum
  (see talks G. Reicherz and St. Goertz)

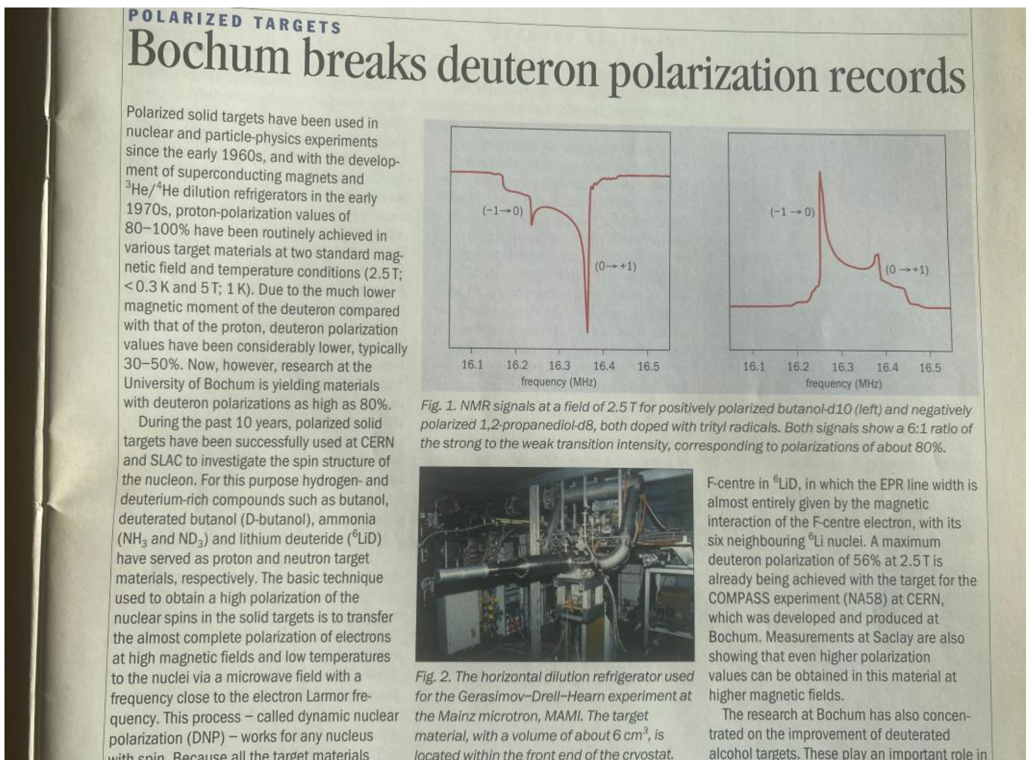
- **1996**
  Experiment with antiprotons and TEMPO doped Butanol frozen spin target at LEAR
  (H. Dutz/Bonn and G. Reicherz/Bochum)

- **1997**
  Ruhr University Bochum joined the COMPASS collaboration at CERN.
  Bochum and Nagoya (N. Horikawa, T. Iwata) responsible for the huge PT with 2 litres for target materials.

- **2000**
  Trityl radicals appeared for deuterated target materials. Cooperation with Malmö Group (K. Golman and J. H. Arkendjaer-Larsen)
  (see talk St. Goertz).

- **2002**
  COMPASS started with ^6LiD.
  Until 2022 13 experimental runs with $^{14}NH_3$ and ^6LiD
  (see talk N. Doshita).

- **2003**
  Bochum breaks deuteron polarization record
  (CERN Courier Sept. 2003)


POLARIZED TARGETS

Bochum breaks deuteron polarization records

Polarized solid targets have been used in nuclear and particle-physics experiments since the early 1960s, and with the development of superconducting magnets and $^3He/^4He$ dilution refrigerators in the early 1970s, proton-polarization values of 80–100% have been routinely achieved in various target materials at two standard magnetic field and temperature conditions (2.5 T; < 0.3 K and 5 T; 1 K). Due to the much lower magnetic moment of the deuteron compared with that of the proton, deuteron polarization values have been considerably lower, typically 30–50%. Now, however, research at the University of Bochum is yielding materials with deuteron polarizations as high as 80%.

During the past 10 years, polarized solid targets have been successfully used at CERN and SLAC to investigate the spin structure of the nucleon. For this purpose hydrogen- and deuterium-rich compounds such as butanol, deuterated butanol (D-butanol), ammonia (NH_3 and ND_3) and lithium deuteride (^6LiD) have served as proton and neutron target materials, respectively. The basic technique used to obtain a high polarization of the nuclear spins in the solid targets is to transfer the almost complete polarization of electrons at high magnetic fields and low temperatures to the nuclei via a microwave field with a frequency close to the electron Larmor frequency. This process – called dynamic nuclear polarization (DNP) – works for any nucleus with spin. Because all the target materials

Fig. 1. NMR signals at a field of 2.5 T for positively polarized butanol-d10 (left) and negatively polarized 1,2-propanediol-d8, both doped with trityl radicals. Both signals show a 6:1 ratio of the strong to the weak transition intensity, corresponding to polarizations of about 80%.

Fig. 2. The horizontal dilution refrigerator used for the Gerasimov–Drell–Hearn experiment at the Mainz microtron, MAMI. The target material, with a volume of about 6 cm³, is located within the front end of the cryostat.

F-centre in ^6LiD, in which the EPR line width is almost entirely given by the magnetic interaction of the F-centre electron, with its six neighbouring ^6Li nuclei. A maximum deuteron polarization of 56% at 2.5 T is already being achieved with the target for the COMPASS experiment (NA58) at CERN, which was developed and produced at Bochum. Measurements at Saclay are also showing that even higher polarization values can be obtained in this material at higher magnetic fields.

The research at Bochum has also concentrated on the improvement of deuterated alcohol targets. These play an important role in

Networking

- **1990**
  Organisation of the Spin Symposium in Bonn, preceeded by 4 workshops (Proc. High Energy Spin Physics, K. H. Althoff, W.M. Vol.1: Conf. Report; W. M., E. Steffens, W. Thiel, Vol.2: Workshops, Springer Verlag 1991)

- **1991**
  Visit to SLAC (15 months) – work with polarized ^{3}He gas target
  (Polarized H, D and ^{3}He Targets for Particle Physics Experiments, St. Goertz, W. M., G. Reicherz, Progress in Particle and Nuclear Physics 49 2002 (403-489)

- **1994**
  Proc. of the 7th Int. Workshop on pol. Targets Materials and Techniques (H. Dutz , W. M., NIMRD9 356, No.1 1-152 (1995)

- **2004**
  Proc. of the 9th Int. Workshop on pol. Targets Materials and Techniques (St. Goertz, W. M., G. Reicherz , NIM Vol. 526, 1-196 (2004)

50 years
Bonn/Bochum – Nagoya/Yamagata Network

- **Basic pillars:**
  - Common passionate interest in the research of elementary particle physics using polarized targets.
  - Mutual trust and friendship
  - Sufficient financial support by AvH, DFG, BMBF, JSPS and Monkasho

- **1st phase (1972 – 1983)**
  BONN (K.H. Althoff, H. Herr, W. Schwille, P. Feller, P. Nölle)
  NAGOYA (K. Kajikawa, S. Fukui, N. Horikawa) and KYOTO (A. Masaike)

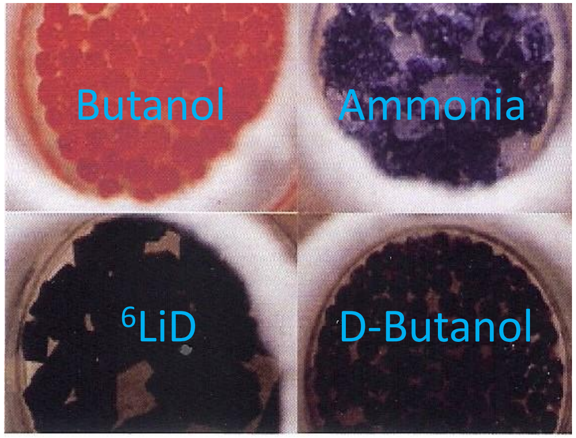
- **2nd phase (1984 – 1995)**
  BONN (K. H. Althoff, W. M., D. Menze, D. Husmann, W. von Drachenfels)
  NAGOYA (N. Horikawa, T. Iwata, T. Nakanishi, S. Nakamura)

- **3rd phase (1996 – 2014)**
  Bochum (W.M., G. Reicherz)
  Nagoya/Yamagata (N. Horikawa, T. Iwata)

- **4th phase (2015 – ...)**
  Bochum (G. Reicherz)
  Yamagata (T. Iwata)

  Intermezzo

  16th Int. Workshop on Polarized Sources, Targets and Polarimetry,
  Bochum Sept. 15 -19, PoS PSTP (2015)
  embedded:
  Celebration of my “Scientific” retirement



“Domo arigato” to my best friend N. Horikawa

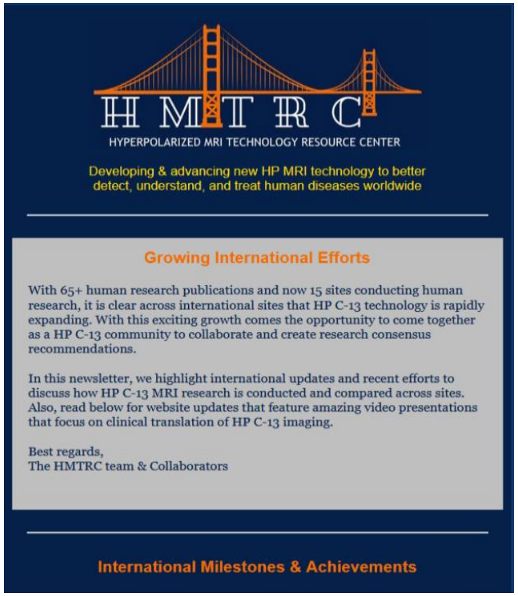
Bochum laboratory
for target material research

Future of Polarized Solid Targets?

(1) Dynamic Nuclear Polarization for medical applications

Bochum/Malmö-Cambridge cooperation: In the Bochum laboratory, focus on trityl radicals studies (deuteron polarization for particle physics experiments) and polarization measurement with pyruvate acid ($^{13}C_3H_4O_3$) at different fields in a $^3He/^4He$ dilution refrigerator (medical applications in human beings)

European patent: "WO2009150211A1-
DNP polarizer and method of producing
A hyperpolarized material"
(J. H. Arkendjaer-Larsen, J. Wolber,
H. Johannesson, W. M., G. Reicherz,
J. Heckmann)



(2) CRYPTA- Workshop devoted to the
future of Polarized Solid Targets for particle physics!


A.OTT
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EKAHA
Ti+ 65° CM
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