Early polarization studies with AMMONIA targets

W. Meyer
Ruhr-University Bochum
27 October, 2003
Bad Honnef

Why review of earlier polarization results with AMMONIA ?

Early attempts to polarize AMMONIA is also a story of failures !

- Failures are normally not published
- But in our case, if correctly interpreted, these results might also help to understand the physics of polarizable target materials
- Avoid needless duplication of enormous work

Aims and objectives of all previous WORKSHOPS:

- to improve the quality of the data in particle physics experiments
- to enable new types of experiments
- save money, as particle physics experiments are very expensive

This Workshop:

Look intensively also for other applications $\rightarrow$ (talk J. Ardenkjaer-Larsen)

Data Quality Improvement

Critical value of any experiment:

Optimization of the reation counting rate : $N = L \frac{d\sigma}{d\Omega} \Delta\Omega$

Luminosity : $L = I \cdot n_{target}$

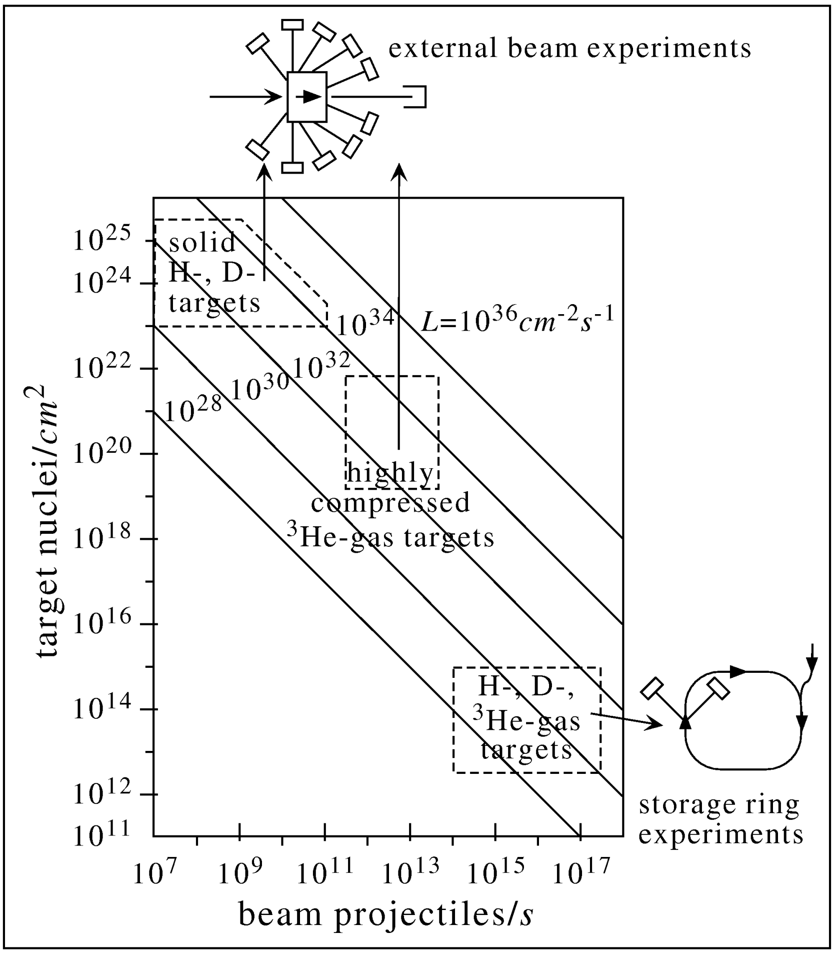
I = beam current

n_{target} = areal target thickness

Limits

- Avogadro constant
- Target technology

ighest Luminosity: $\approx 10^{35} cm^{-2} \cdot s^{-1} \rightarrow$ AMMONIA $\rightarrow {}^{6}$LiD



Highest Luminosity connected to

1. polarizable nucleon content
2. radiation damage resistance

ad 1: two distinct connotations

- a. the hydrogen content, by weight, of the material or
- b. the fraction f of polarizable protons or neutrons, whether or not they are found in larger nuclei

ad 2: two distinct things

- a. the resistance of the target polarization P to degradation by ionizing radiation and
- b: the ease of reserval of such degradation

‘Figure of Merit’ FOM $\approx \frac{1}{\text{measuring time}}$ at a given data error $\frac{\Delta A}{A}$

$$\text{FOM} = n_{target} \cdot P^2 \cdot f^2$$

↑ to optimize

New Types of Experiments
e.g. SPIN-PHYSICS in photo-absorption

Test of Bjorken Sum-rule (1966)

- Fundamental to QCD
- $$\int_0^1 \left[g_1^p(x) - g_1^n(x)\right] dx = \frac{1}{6}\left|\frac{g_A}{g_V}\right| K$$

⇒ **Target Material Research !** ⇒ **AMMONIA decisive**

Test of Gerasimov-Drell-Hearn (GDH Sum-rule (1965/66)

- $Q^2 = 0$ (real photons)
- Connects well known static properties of the nucleon with dynamical ones
- $$\int_0^\infty (\sigma_{3/2} - \sigma_{1/2}) \frac{d\omega}{\omega} = 2\pi^2 \alpha \frac{\kappa^2}{m^2}$$

ω = Photon energy
α = fine structure constant
κ = anomalous magnetic momnet
m = nucleon mass

⇒ **Development in Magnet Technology !** (→ talks Ch. Rohlof, H. Dutz)

⇒ **Deuteron Target polarization** (→ talk St. Goertz)

AMMONIA PROPERTIES

- ammonia (NH_3) – ammoniac : ammoniakon (greek)

  ↓

  applied to a salt and a gum resin said to come from near the Shrine of Ammon in Libya

- ammonia gas discovered 1773 by J. Priestley
- NH_3 usually produced by the direct combination of nitrogen and hydrogen gases → Haber-Bosch-procedure
- NH_3 : colourless gas; pungent smell → danger for eyes and muscous membrane
  : suffocating → max. concentration 35 mg/m^3

Physical properties:

Density	: 0.817 g/cm^3 at 194 K
Melting point	: 195.5 K
Boiling point	: 239.8 K

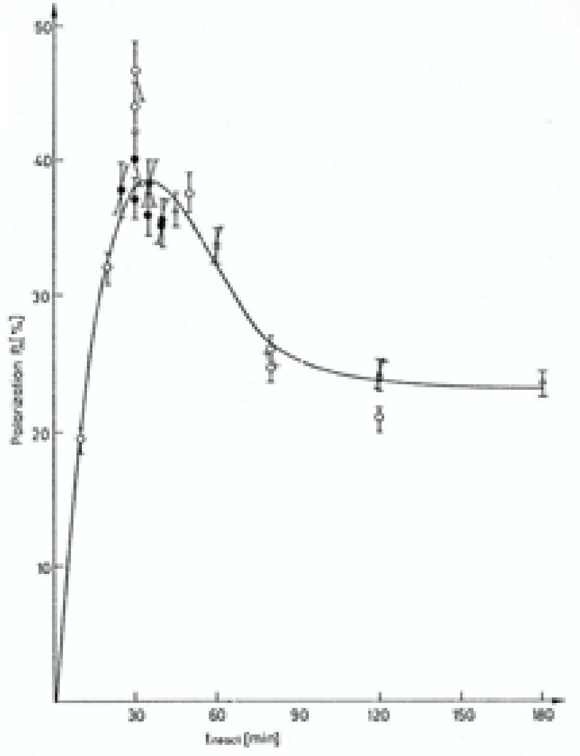
$$f = 0.176 \quad \Rightarrow \quad FOM \approx f^2 = \frac{f^2_{NH_3}}{f^2_{But}} = 1.7$$

1970 : First dynamic proton polarization in AMMONIA

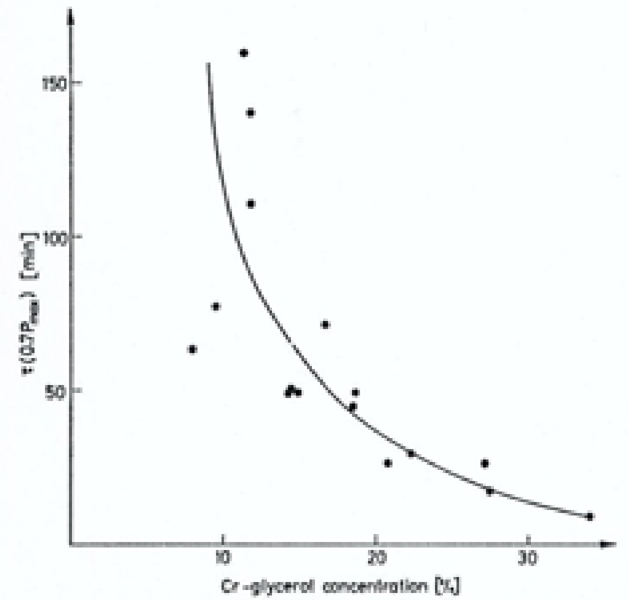
K. Scheffler (CERN); NIM 82 (1970) 205

"After trying a number of paramagnetic centers such as sodium, porphyrexide, porphyrindine, diphenylpicryl-hydrazil, Ziegler's radical, di-tert.-butyl-nitroxide, per-chlorotriphenylmethyl and violanthrone, all of which gave low polarization or none at all, we doped the liquefied ammonia with Cr^{V} glycerol complexes."

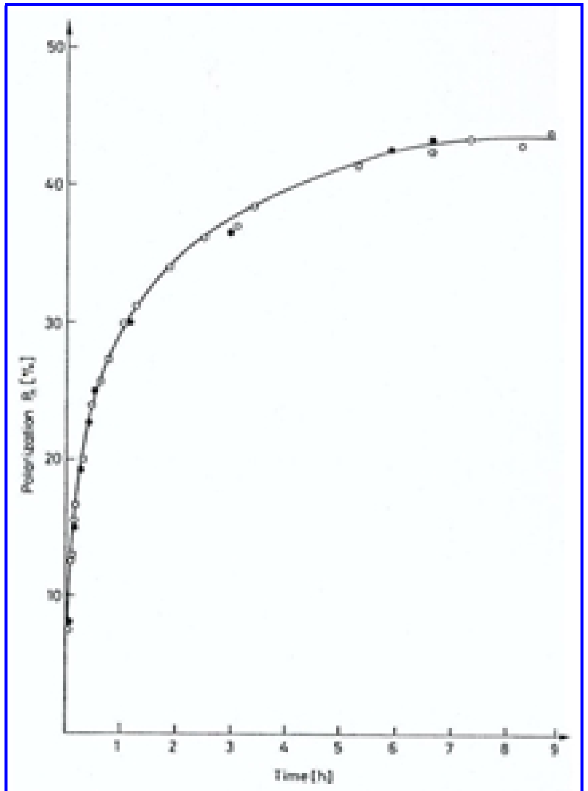
NH_3 with Cr(V) glycerol complexes $P \approx 40$ % at 1 K and 2.5 T



Polarization of ammonia versus reaction time of $K_2Cr_2O_7$-glycerol. Different signs correspond to different series of complex preparation.



Time needed to achieve 0.7 of the maximum polarization versus Cr-glycerol concentration (weight-%) of the ammonia target.



Polarization of ammonia versus time of polarization.

glycerol or glycerine : $C_3H_5(OH)_3 \rightarrow f = 0.087$

Further polarization results by *Scheffler (CERN) 1971*

NH_3 with ethanediol-Cr(V) : $P \approx 70$ % at 1 K and 2.5 T

Not confirmed by later work at CERN

→ *W. de Boer (unpublished 1971)*

→ *K. Guckelsberger (unpublished 1974)*

Slow groth of polarization:

Model based on the formation of clusters of the complex, surrounded by a relatively pure ammonia matrix?

ethylene glycol or ethanediol ar glycol: $C_2H_4(OH)_2 \rightarrow f = 0.097$

1979 : Dynamic Nuclear Polarization in Irradiated Ammonia below 0.5 K

T.O. Ninikoski and J.-M. Rieubland (CERN)
Phys. Lett. 72A (1979) 141

– just before 2nd Workshop in Abingdon
– triggered 2nd Workshop in Abingdon ?

- Irradiation with 580 MeV proton beam of the CERN synchrocyclotron
- NH_3 in LN_2 during irradiation
- Accumulated flux: $0.95 \cdot 10^{15}$ protons/cm^2
  $\rightarrow P_+ = 90.5$ %; $P_- = -93.6$ % at ≈ 0.2 K and 2.5 T
- Polarization build-up time : 7 - 8 hours due to low concentration of radicals $\approx 5 \cdot 10^{18}$ spins/cm^3

Authors: „These results may indicate a breakthrough in the development of better polarized target materials for high energy physics experiments"

Irradiation activities started see Proc. of the 2nd Workshop – Abingdon

- Reactor irradiated Ammonia at LN_2: *G. Court* – small P, long τ
- Electron irradiated NH_3 at SLAC at 1 K: *V. Hughes; M. Seely*
  NMR-signal after $1.5 \cdot 10^{15}$ e^- disappeared due to disintegration of beads (frozen in LN_2 → a mechanically week solid)
- Electron irradiated butanol with 5 % water at 1 K (why recepy of Mango et al.?)
  $2.3 \cdot 10^{15}$ $e^-/cm^2 \rightarrow P \approx -11$ % at 1 K and 2.5 T
- Proton irradiated (ZGS at Argonne)
  Butanol at LH_2 *(D. Crabb et al.)*
  $\approx 2.5 \cdot 10^{14}$ p $\rightarrow P \approx 12$ % at 0.5 K and 2.5 T
- Further proton irradiated NH_3 at CERN
  → **Explosions !** Stop of activity, because there was no firm conclusion for the cause of explosions
- Electron irradiated ^7LiF; ^7LiH; ^6LiD under LN_2 at Saclay
  3 Explosions ! Nitrogen ?
- Successful irradiations under LAr for ^7LiF; ^7LiH; ^6LiD at Saclay
  ⇒ $P_{6LiD} = 70$ % at ≈ 0.2 K and 6.5 T
  ⇒ today still highest deuteron pol. value in ^6LiD (see talk J. Ball)

1980 – 82 : 2 methods for the preparation of AMMONIA for the DNP on the market

a) High-temperature irradiation in liq. Ar (87 K)
→ BONN accelerator – 20 MeV electrons
U. Härtel et al., Birkhäuser Verlag, Basel, 1981, p. 447; p. 451

Bead type: Fast-frozen in liq. N_2
Irradiation dose: ≈ $10^{17} e^-/cm^2$

NH_3 : P = 66 % at 0.5 K and 2.5 T
Build-up time: 9min
Annealing temp.: liq. N_2
Original radical reasonable stabled 77 K

ND_3 : P = 3.5 % at 1.0 K and 2.5 T
= 11 % at 0.5 K and 2.5 T
= 31 % at < 0.3 K and 2.5 T

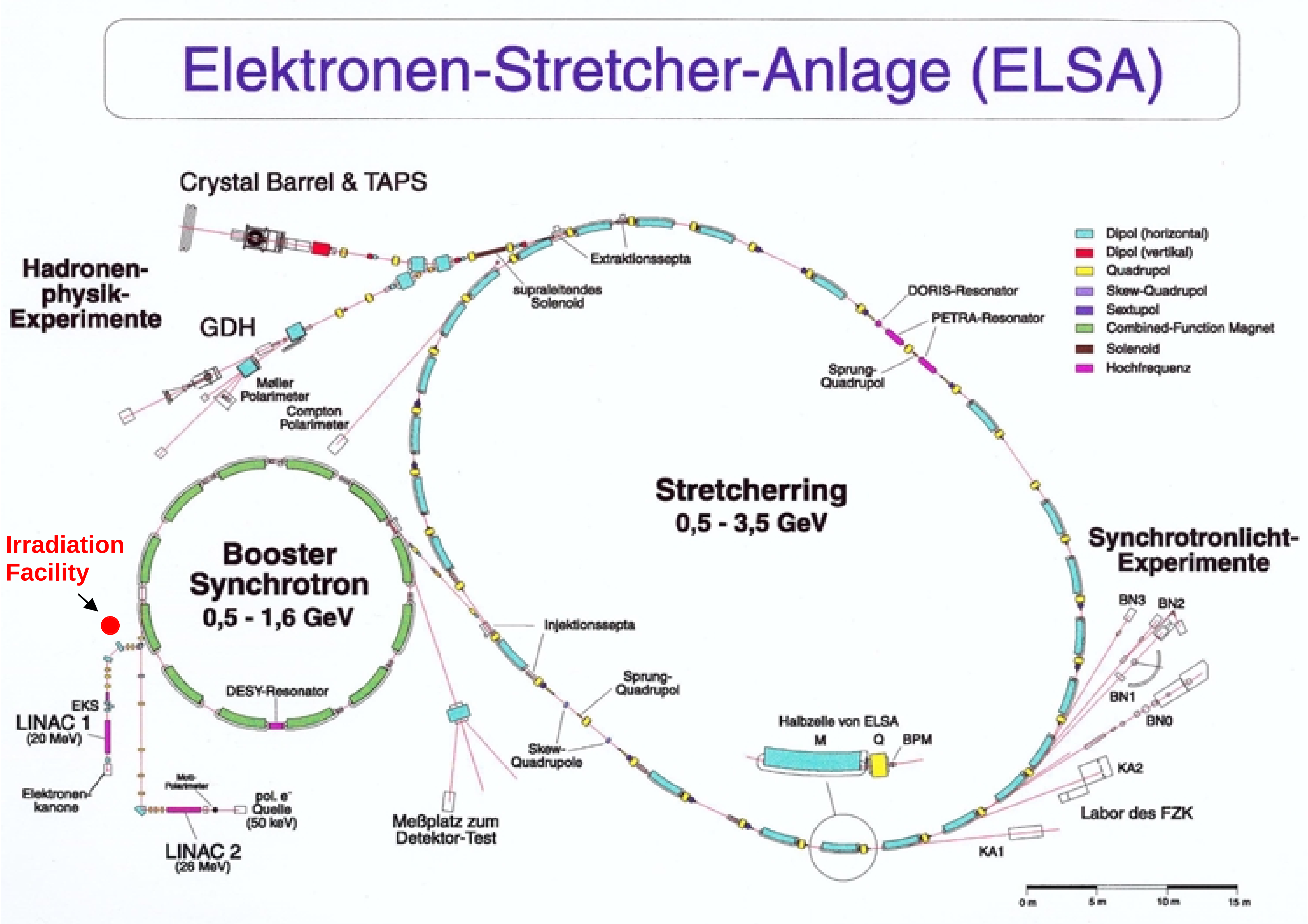
b) Low-temperature irradiation at 1 K with permanent DNP monitoring
→ SLAC – 20 GeV electrons

M. Seely et al., Birkhäuser Verlag, Basel, 1981, p. 453

Bead type: Slowly frozen
Irradiation dose: $\approx 4 \cdot 10^{15} e^{-}/cm^{2}$

NH_3 : P = 75 % at 1 K and 2.5 T
ND_3 : P = 25 % at 1 K and 2.5 T

Important : Radiation resistance order of magnitude better than that of butanol/porphyrexide



Elektronen-Stretcher-Anlage (ELSA)
Crystal Barrel & TAPS
Hadronen-physik-Experimente
GDH
Møller Polarimeter
Compton Polarimeter
Extraktionssepta
supraleitendes Solenoid
DORIS-Resonator
PETRA-Resonator
Sprung-Quadrupol
Dipol (horizontal)
Dipol (vertikal)
Quadrupol
Skew-Quadrupol
Sextupol
Combined-Function Magnet
Solenoid
Hochfrequenz
Stretcherring
0,5 - 3,5 GeV
Booster Synchrotron
0,5 - 1,6 GeV
Irradiation Facility
Synchrotronlicht-Experimente
BN3
BN2
BN1
BN0
KA2
KA1
Labor des FZK
Injektionssepta
Sprung-Quadrupol
Skew-Quadrupole
Halbzelle von ELSA
M
Q
BPM
DESY-Resonator
EKS
LINAC 1
(20 MeV)
Elektronen-kanone
pol. e⁻ Quelle
(50 keV)
LINAC 2
(26 MeV)
Meßplatz zum Detektor-Test
0 m
5 m
10 m
15 m

First particle physics experiments with Ammonia

1982 : NH_3: Brookhaven AGS

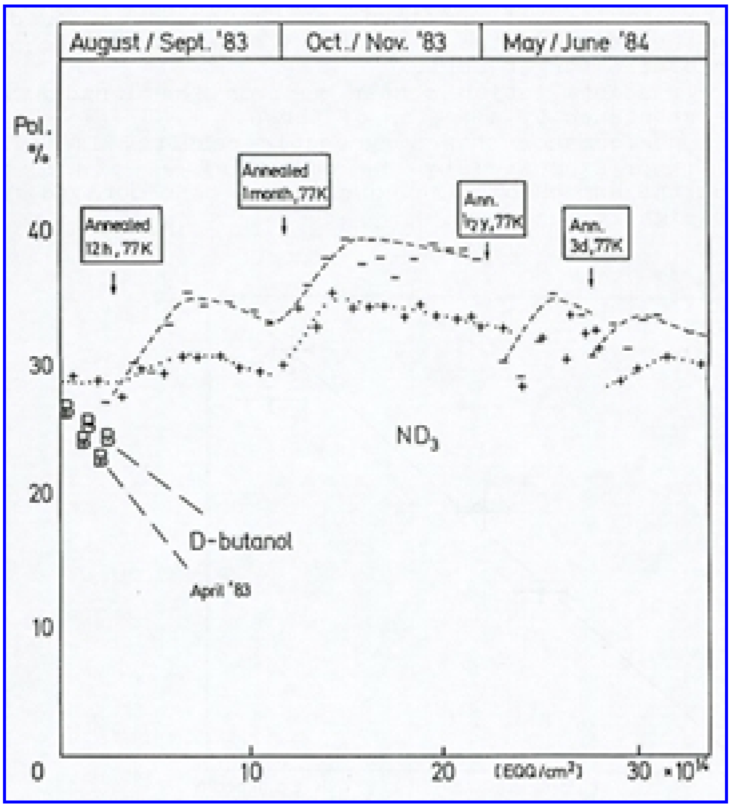
$\vec{p}\vec{p}$ elastic scattering

D. Crabb et al.
Proc. 3rd workshop (Brookhaven) p. 488

1983 : Bonn-Electron-Synchrotron

ND_3 $\quad \gamma \vec{d} \rightarrow pn$

R. Dostert et al., Proc. 4th Int. Workshop (Bad Honnef) p. 13



1985 : Bonn-Electron-Synchrotron

ND_3 $\quad e\vec{d} \rightarrow ed$

1. Measurement with a tensor polarized deuteron target
→formfactor of the deuteron
separation of the electric and the quadrupole formfactor

W. Meyer et al., NIM A244 (1986) 574

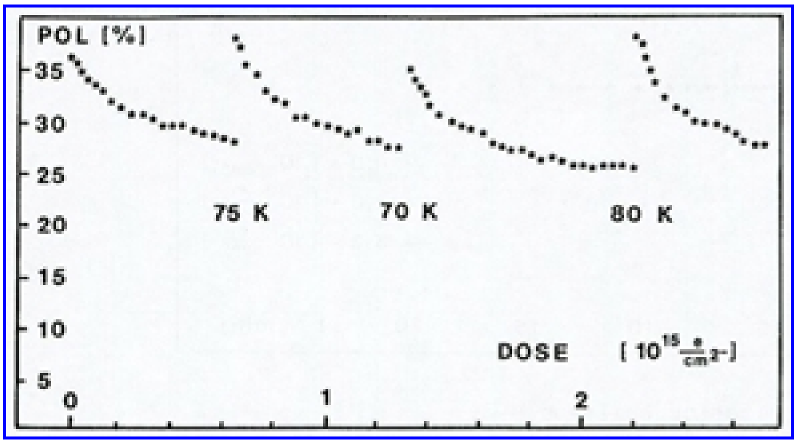
Conclusions on the 3th Workshop (Brookhaven)

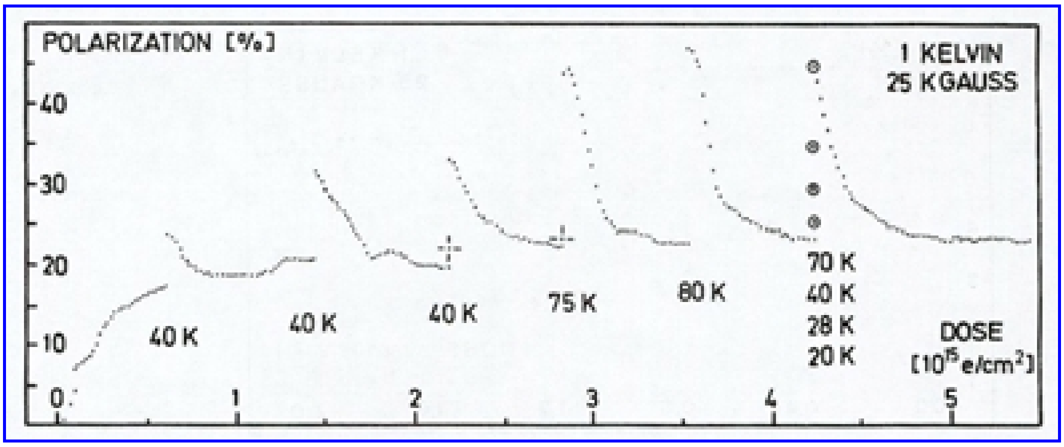
- Try low- and high temperature irradiated material in the same apparatus

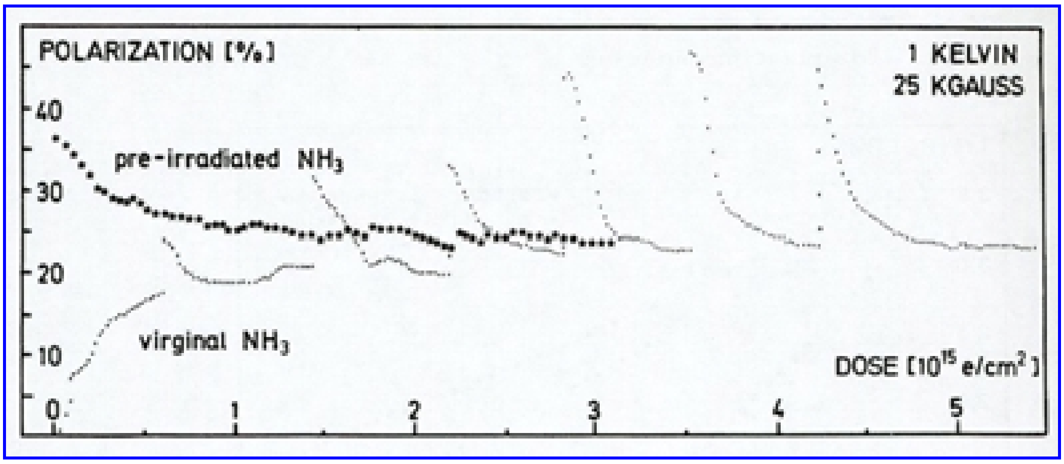
1983 : **Bonn – dedicated runs at 1 K and 2.5 T**

high temperature irradiation :   20 MeV electrons
low temperature irradiation :   1.5 GeV electrons
K.H. Althoff et al., Proc. 4th Workshop (Bad Honnef) p. 23

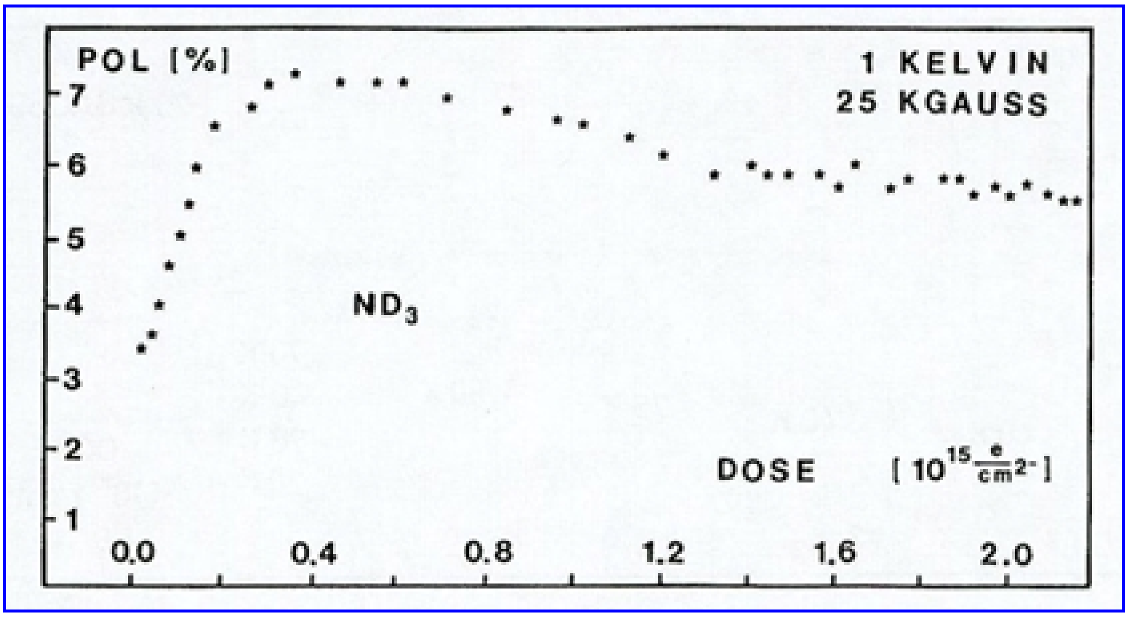
NH_3

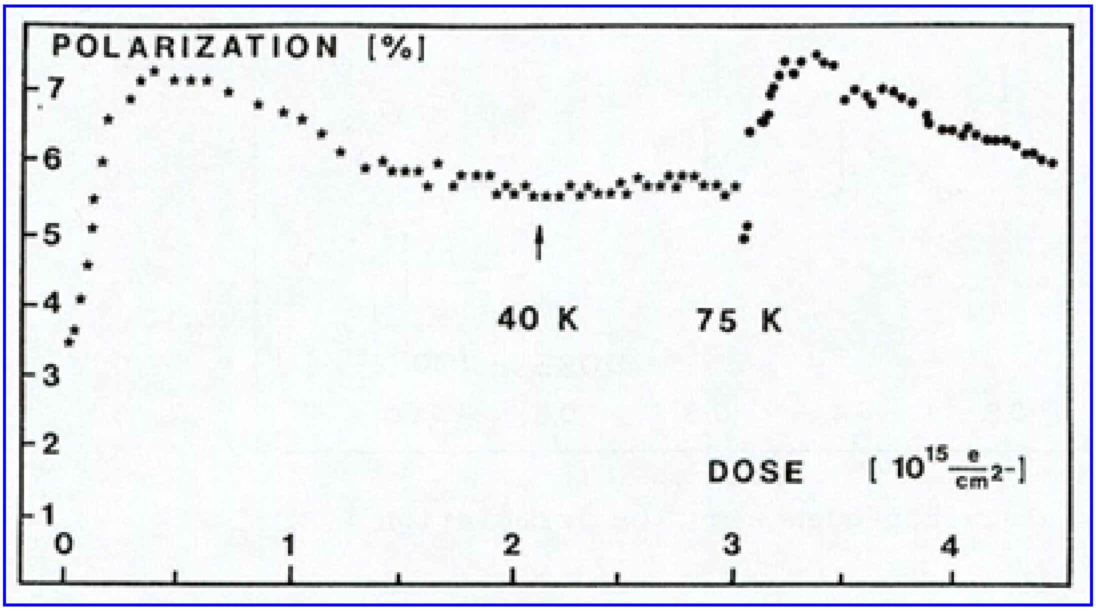


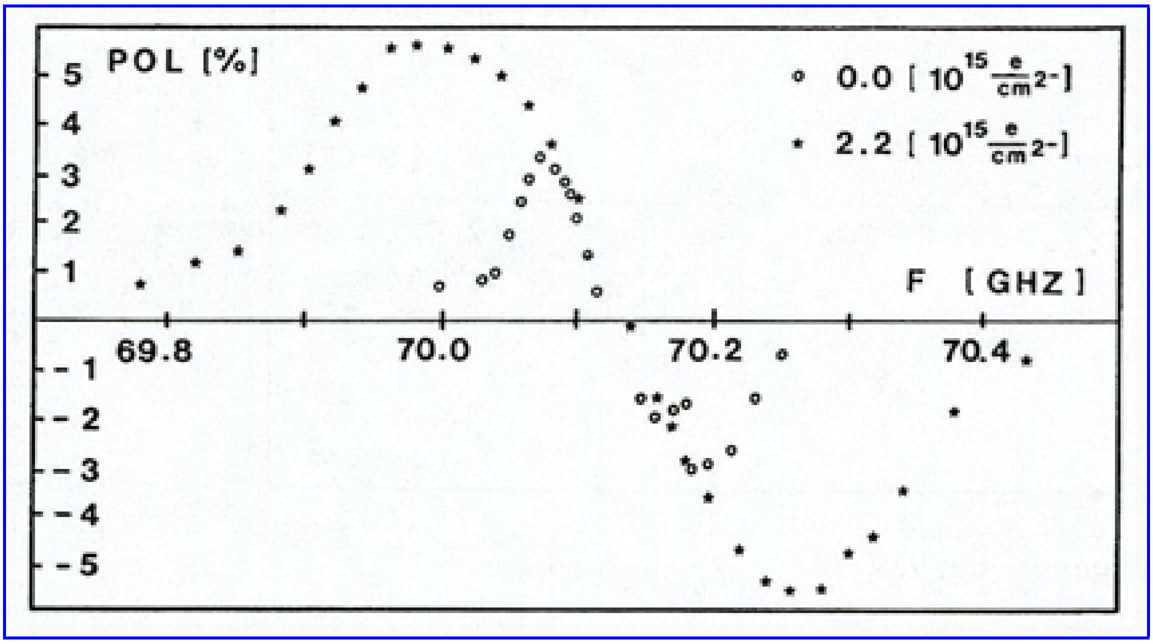




ND_3


POL [%]
1 KELVIN
25 KGAUSS
ND3
DOSE [10^15 e/cm2]
7
6
5
4
3
2
1
0.0
0.4
0.8
1.2
1.6
2.0


POLARIZATION [%]
40 K
75 K
DOSE [10^15 e/cm2]
7
6
5
4
3
2
1
0
1
2
3
4

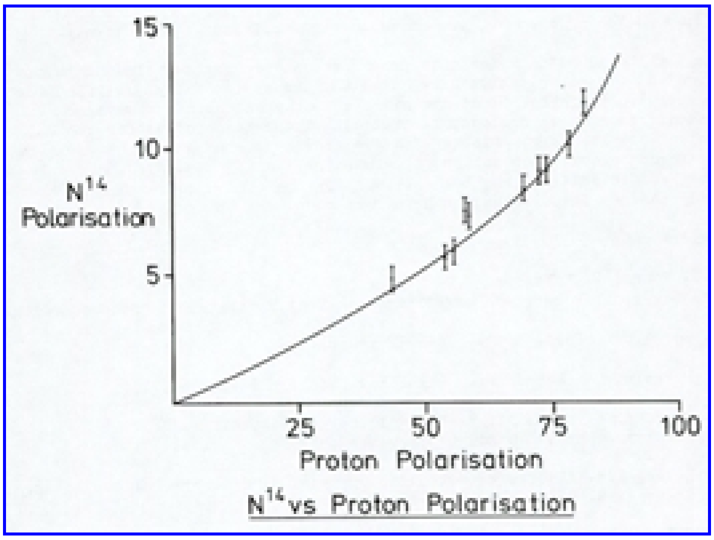

POL [%]
0.0 [10^15 e/cm2]
2.2 [10^15 e/cm2]
F [GHZ]
5
4
3
2
1
-1
-2
-3
-4
-5
69.8
70.0
70.2
70.4

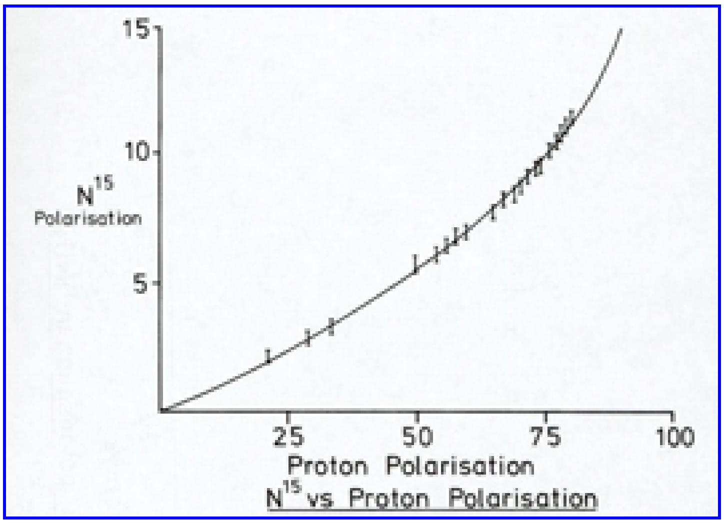
Polarization mechanisms in AMMONIA

- Polarization of background nuclei
- ^{14}N: Spin 1 → quadrupole moment
  → wide NMR signals
  → problems for NMR detection
  solved: *Ch. Dulya, PhD thesis*
  St. Goertz, W. Meyer, G. Reicherz
  Prog. Part. Nucl. Phys. 49 (2002) p. 464
- ^{15}N: Spin $^{1}/_{2}$ → easier NMR detection

$^{14}NH_3$; $^{15}NH_3$: EST established by Bonn-Liverpool activities

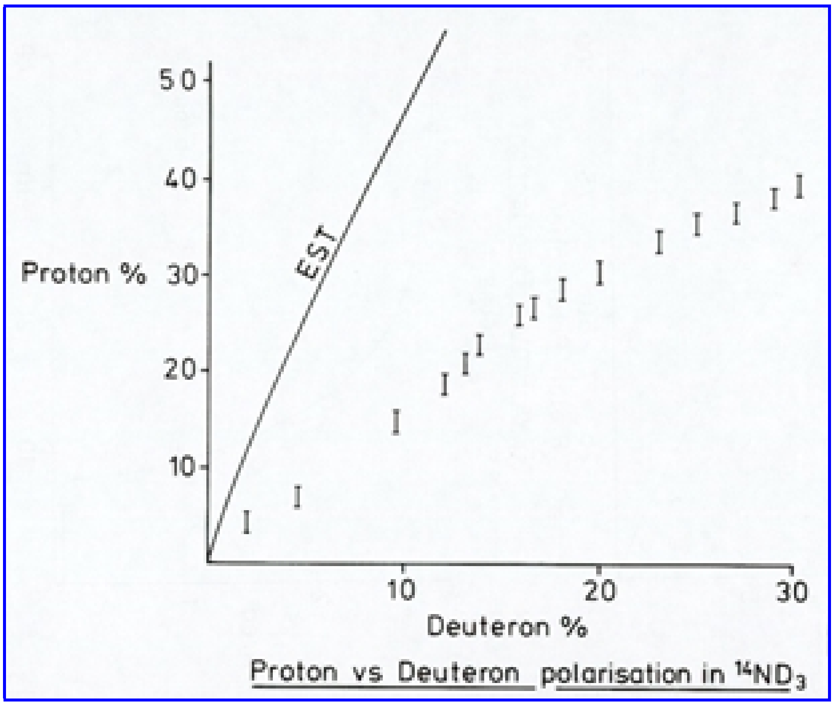
R. Dostert et al., Proc 4th workshop (Bad Honnef) p. 33
G.R. Court et al., Proc 4th workshop (Bad Honnef) p. 53

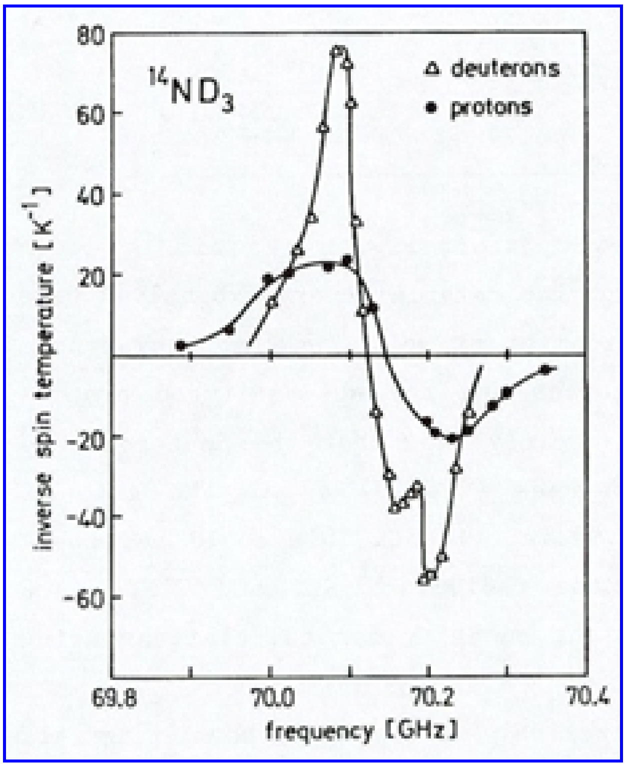


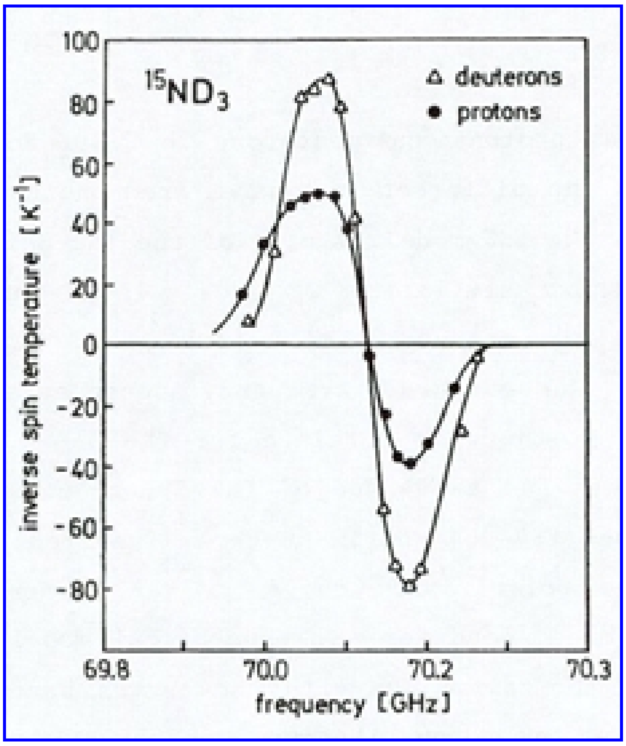


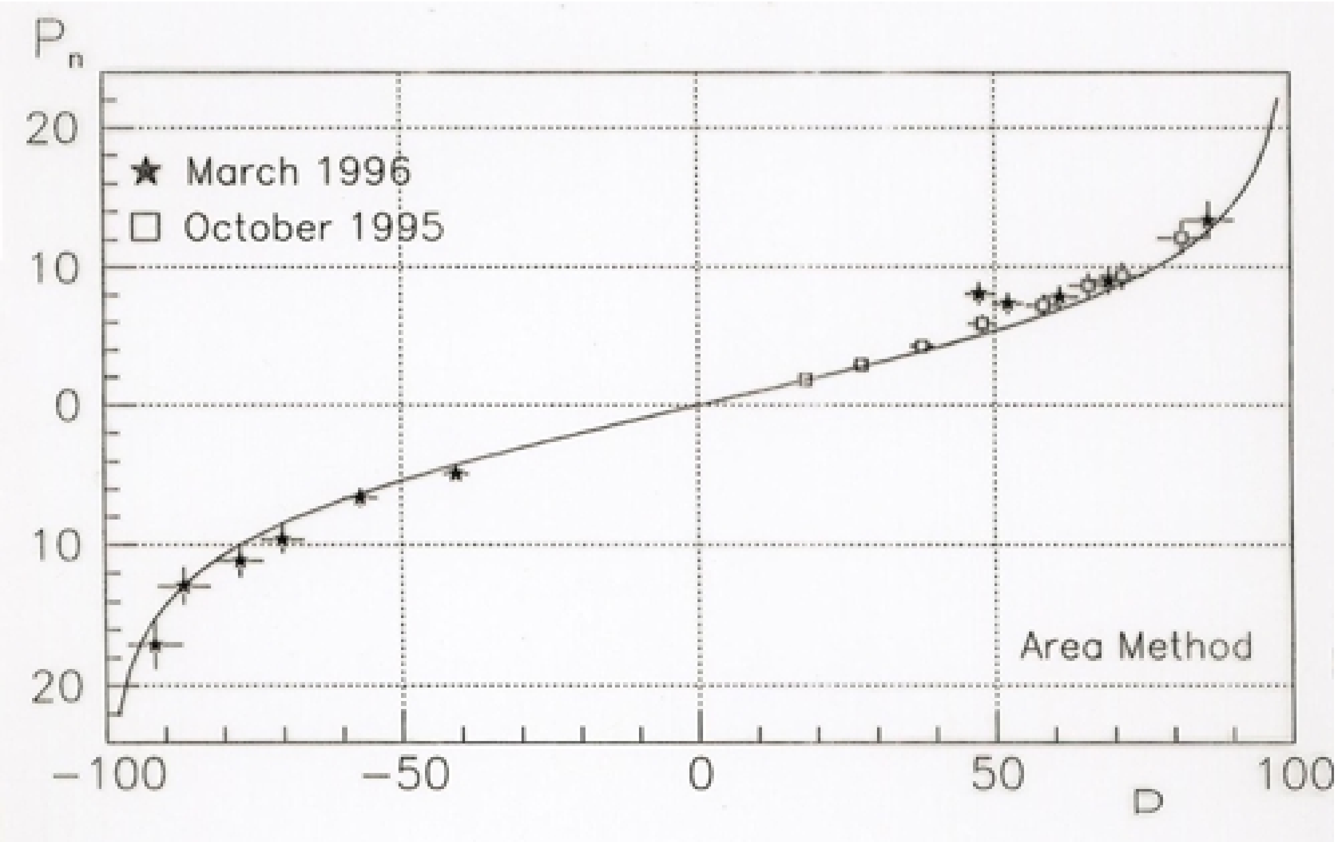
$^{14}ND_3$; $^{15}ND_3$: EST between D and ^{14}N

~~EST~~ between D and P (1 % P unsubstituted)

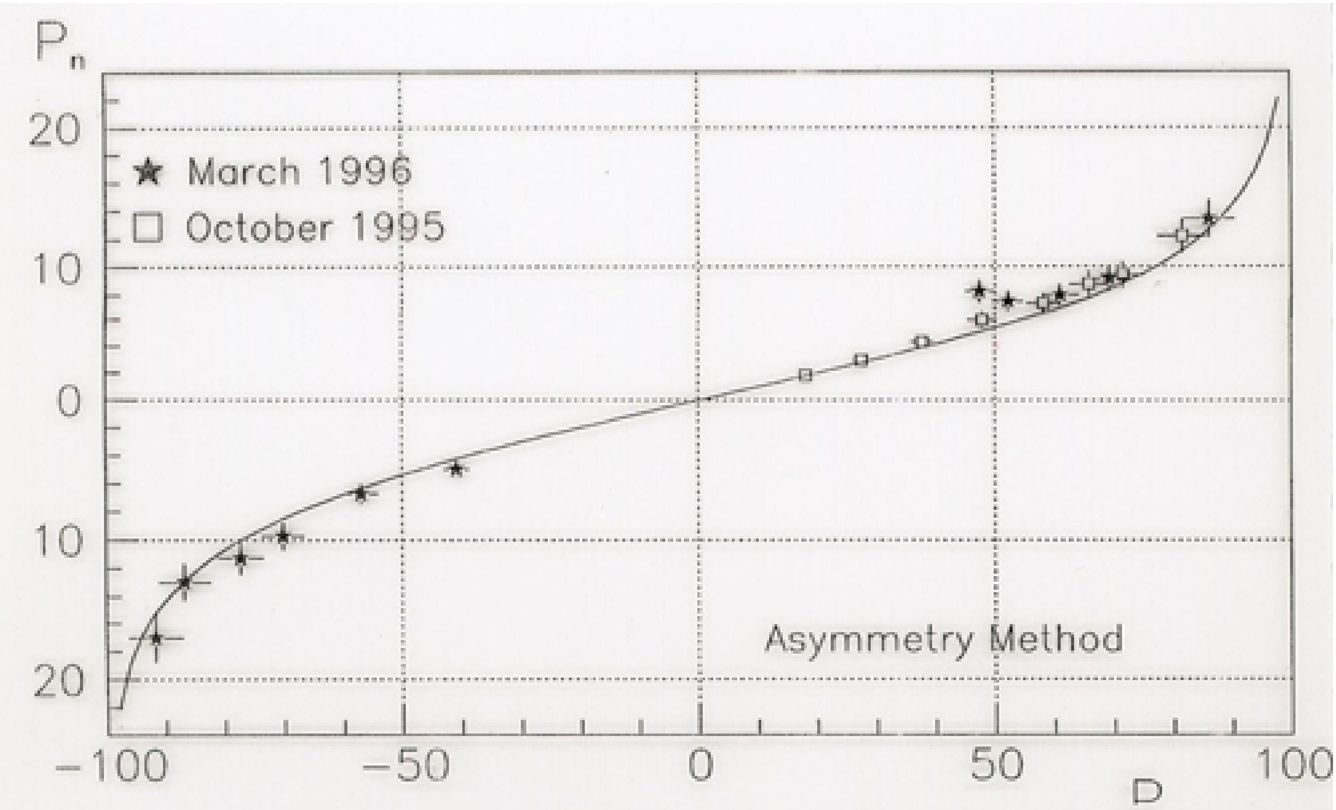






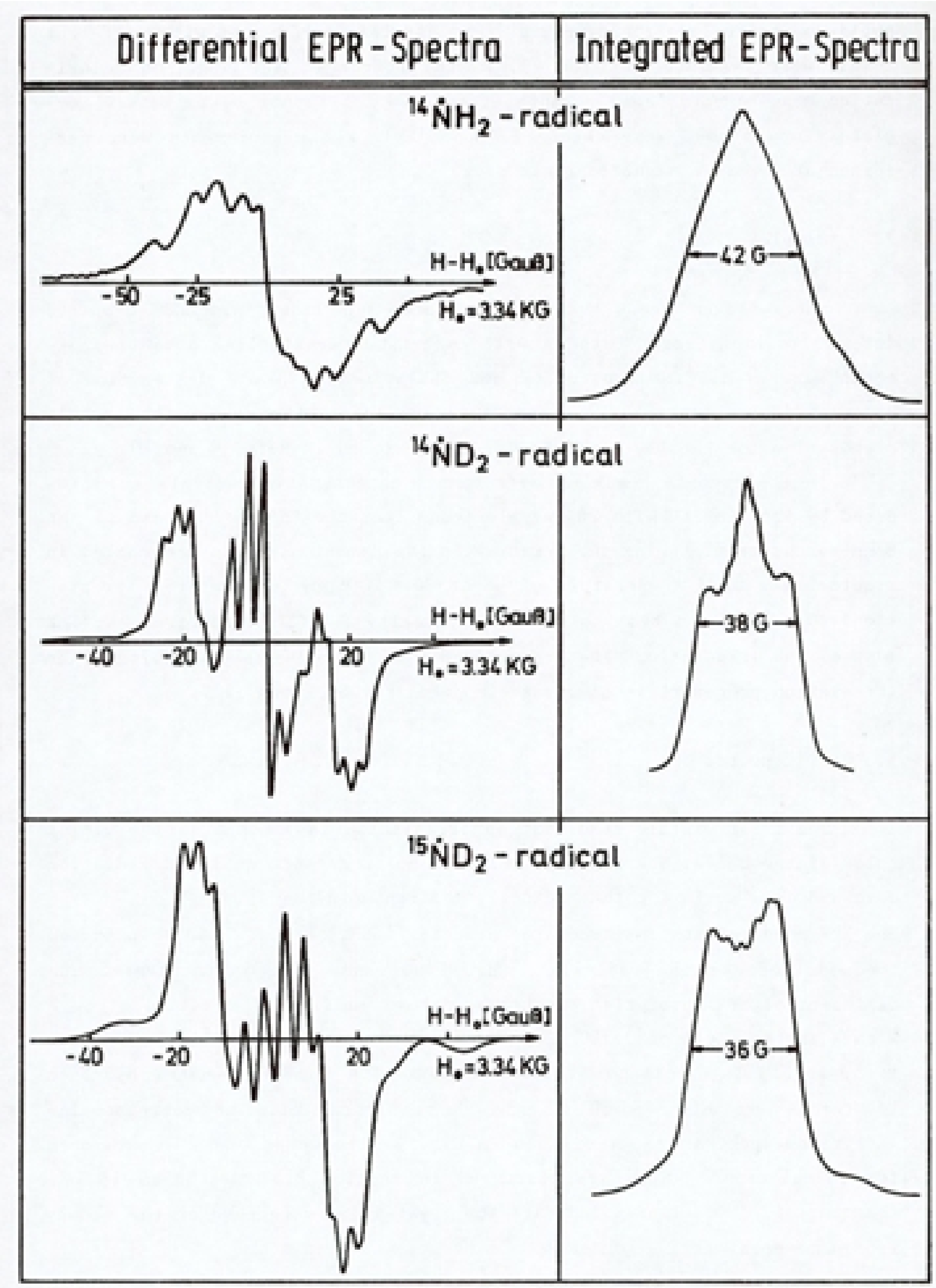


P_n
March 1996
October 1995
Area Method
D



P_n
March 1996
October 1995
Asymmetry Method
D

Radicals in high-temperature irradiated AMMONIA



Questions: Polarization mechanisms in deuterated Ammonia ?
High-temp. irradiated material : Differential solid state effect ?
W. Meyer et al. NIM 227 (1984) 35

Low temp. irradiated material ?

Needed: In situ EPR
→ High field EPR (→ J. Heckmann)

Potassium doped NH_3

1982 : H Riechert (Bonn Univ.) Diploma work, BONN-IR-82-33

NH_3 as good solvent for potassium → solved electrons move freely
excited states → blue colour

2 samples : 0.12 mol solution $\triangleq 7 \cdot 10^{19}$ K-atoms/cm^3
1.2 mol solution $\triangleq 7 \cdot 10^{20}$ K-atoms/cm^3

Solution solidified in liq. N_2 → after rapid freezing metallic shine on the surface
→ dismixture of the solution ?

$P_- \approx 0.2$ % after 2 h build-up for both samples at 1 K and 2.5 T
T.E signal build-up time also very long
→ probably small number of active paramagnetic centers ?
→ electrons from K-atoms recombine to pairs?
→ low P values probably due to impurities?

1979 : 2nd target workshop Abingdon

New chemical doping agents:

M. Krumpole and J. Rozek, J. Am Chem. Soc. 101 (1978) 32

HMBA (2-hydroxy-2-methylbutric acid)
BHHA (2-butyl-hydroxyhexanoic acid
EHBA (2ethyl-2-hydroxybutyric acid)

– stable powders at room temperature
– soluble in many polar solvents (water, alcohols, glycol)
– solutions undergo negligible decomposition during the preparation time of target materials
– deuterated version also available nowadays

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

DNP in High-Hydrogen Glasses

1980-84 *D. Hill and M. Krumpolc*
Proc. 3nd workshop (Brookhaven) p. 479
Proc. 4th workshop (Bad Honnef) p. 94
Proc. 4th workshop (Bad Honnef) p. 84

Concentration on glasses based on amines and ammonia

1. high hydrogen content → 16.0 – 18.6 ωt %
2. polar solvents facilitates Cr(V) solubility

→ 2 families of glasses

1. borane-amine complexes
2. ammonium borohydride

} as ‚vitrifying' additives

Selected polarization results for samples prepared at 200K (exception: sample 16 was prepared at 225K). Each result represents a single sample, except for sample 1 which represents an average over eight samples. Meaning of symbols: ammonium borohydride (AB), borane-ammonia (BA), borane-dimethylamine (BDMA), borane-methylamine (BMA), dimethylamine (DMA), ethylamine (EA), methylamine (MA).

Sample No.	Composition By Wt.	EHBA-Cr^V 10^{19}/mℓ	Wt.%-H	Polarization (%) (+)	Polarization (%) (-)
1	EA(.85)BA(.15)	4 nom.	16.0	83	77
2	EA(.90)AB(.10)	4 nom.	16.3	68	
3	EA(.88)AB(.12)	4 nom.	16.5	65	
4	DMA(.85)BA(.15)	4 nom.	16.0	74	
5	MA(.85)BA(.15)	5 nom.	16.4	84	72
6	MA(.85)BA(.15)	4 nom.	16.4	79	67
7	MA(.80)BA(.20)	4 nom.	16.6	74	66
8	NH_3(.55)EA(.25)BA(.20)	4.5	17.2	76	65
9	NH_3(.44)EA(.18)BA(.38)	4.5 nom.	17.7	66	
10	NH_3(.17)MA(.56)BA(.27)	5 nom.	17.0	72	
11	NH_3(.55)MA(.25)BA(.20)	5	17.3	76	66
12	NH_3(.26)MA(.40)BA(.34)	5 nom.	17.4	79	70
13	NH_3(.32)MA(.30)BA(.38)	5 nom.	17.6	73	
14	NH_3(.43)MA(.18)BA(.39)	5 nom.	17.7	77	65
15	NH_3(.49)MA(.12)BA(.39)	5 nom.	17.9	66	
16	NH_3(.62)BDMA(.16)BA(.22)	4	17.7	63	53
17	NH_3(.65)BMA(.15)BA(.20)	6 nom.	17.6	56	
18	NH_3(.65)BMA(.15)BA(.20)	4 nom.	17.8	58	
19	NH_3(.50)BMA(.20)BA(.30)	5 nom.	17.9	61	
20	NH_3(.79)AB(.12)BA(.09)	4 nom.	18.4	39	37
21	NH_3(.75)AB(.12)BA(.13)	4 nom.	18.4	38	
22	NH_3(.52)AB(.08)BA(.40)	4 nom.	18.6	49	

Materials not longer considered

Reasons

1. radiation resistance comparable to butanol-porhyrexide sample
   i.e. more than factor 10 worse compared to that of irradiated ammonia
   – measured with ethylamine (85 ωt %) + borane-ammonia (15 ωt %) + EHBA-Cr(V) ($4 \cdot 10^{19}$ spins/ml) $\triangleq C_2NH_7BH_3NH_3 \rightarrow f = 16.5$
   D. Crabb and D. Hill
   Proc. 4th workshop (Bad Honnef) p. 100

2. low polarization in borane-ammonia (BH_3NH_3: f = 19.6 %) and ammonium borohydride (NH_4BH_4: f = 24.6 %) in case of radiation doping
   M.L. Seely et al.
   Proc. 3rd workshop (Brookhaven) p. 525
   G.R. Court and W.G. Heyes
   Proc. 4th workshop (Bad Honnef) p. 60

3. More problems with spin of the spectator nuclei
   ^{10}B (spin 3)
   ^{11}B (spin $^3/_2$)
   in addition to ^{14}N (spin 1)

Conclusions

After 5 years (1979 – 1984) work on irradiated AMMONIA:

Most practical preparation method: High temperature irradiation

Highlight No 1:

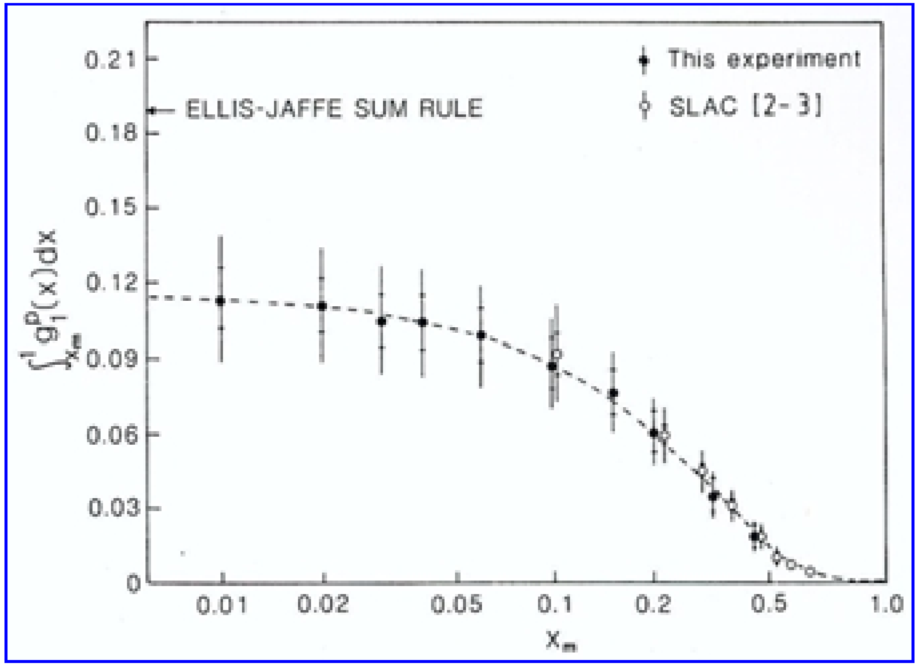
EMC-results from CERN deep inelastic polarized muon-polarized proton scattering experiments with NH_3 $\rightarrow$ spin problem ! *J. Ashman et al., Nucl. Phys. B328(1989)1*

– 3 ltr NH_3 irradiated in Bonn by Bonn-CERN-Liverpool activities

S. Brown et al., Proc. 4th workshop (Bad Honnef) p. 66

– P = 80 %

Important: high dilution factor



Highlight No 2:

SLAC measurements in the 1990s in deep inelastic polarized electron – polarized proton (deuteron) scattering

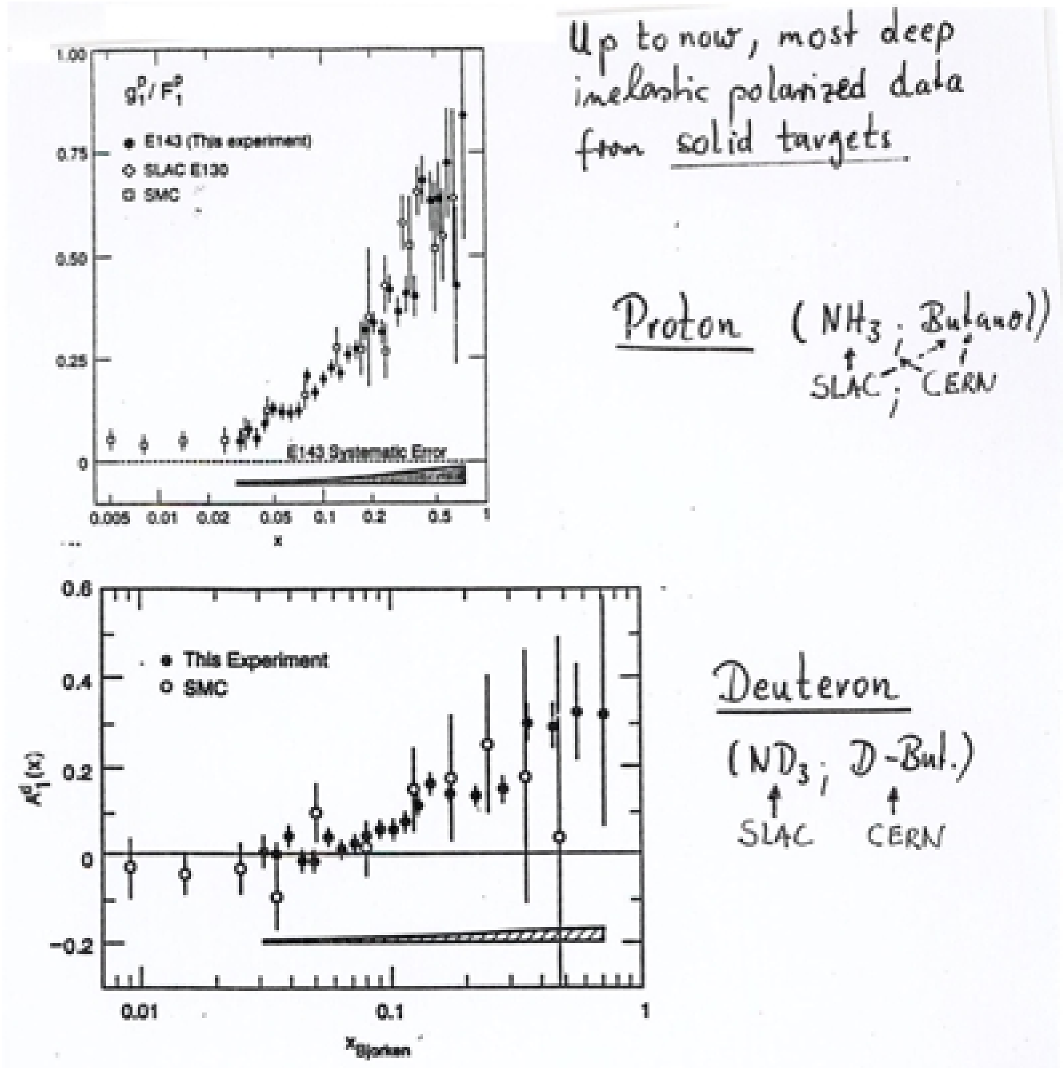
→ Confirmation of the EMC result

→ Bjorken sum rule

D. Crabb et al.,
Phys. Rev. Lett. 65 (1990) 3241
D. Crabb et al.,
NIM A356 (1995) 9
D. Adams et al.,
Phys. Lett B396 (1997) 338
B. Adeva et al.,
Phys. Rev. D58 (1998) 112001
B. Adeva et al.,
Phys. Lett. B420 (1998) 180

– $P_{^{15}NH_3} = 90\ \%$; $P_{^{15}ND_3} = 42\ \%$

Important: high polarization resistance



Highlight No 3:

SMC measurements in 1996 at CERN

→ Confirmation of the EMC result

→ Bjorken sum rule

K. Abe et al.,
Phys. Rev. Lett. 74 (1995) 346
K. Abe et al.,
Phys. Rev. Lett. 74 (1995) 25
K. Abe et al.,
Phys. Rev. Lett. 65 (1995) 587

– $P_{^{14}NH_3} = 90\ \%$