Oct. 23, 2014
Beijing

DAWN OF HIGH ENERGY SPIN PHYSICS

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Early History of High Energy Physics with Polarized Target

Discovery of Dynamic Polarization of Protons

Static Polarization (thermal equilibrium) → High Proton Polarization (non-thermal)

by **solid effect** (microwaves)

$$P_n = \tanh\left(\frac{\mu_n H}{kT}\right)$$

$$= \tanh\left(1.02\times 10^{-7}\,\frac{H}{T}\right)$$

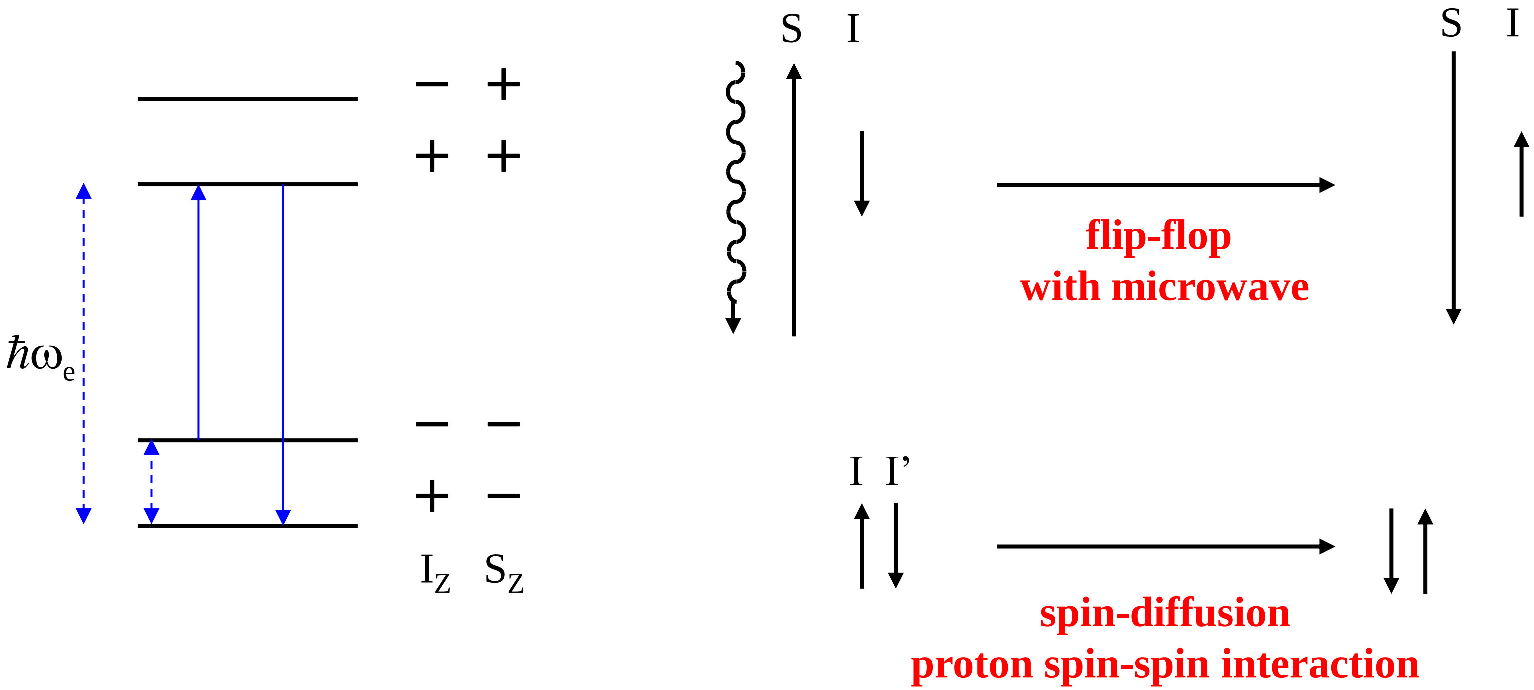
$$\sim 10^{-3}$$

at 1 K , 1T

80 ~ 90 % polarization

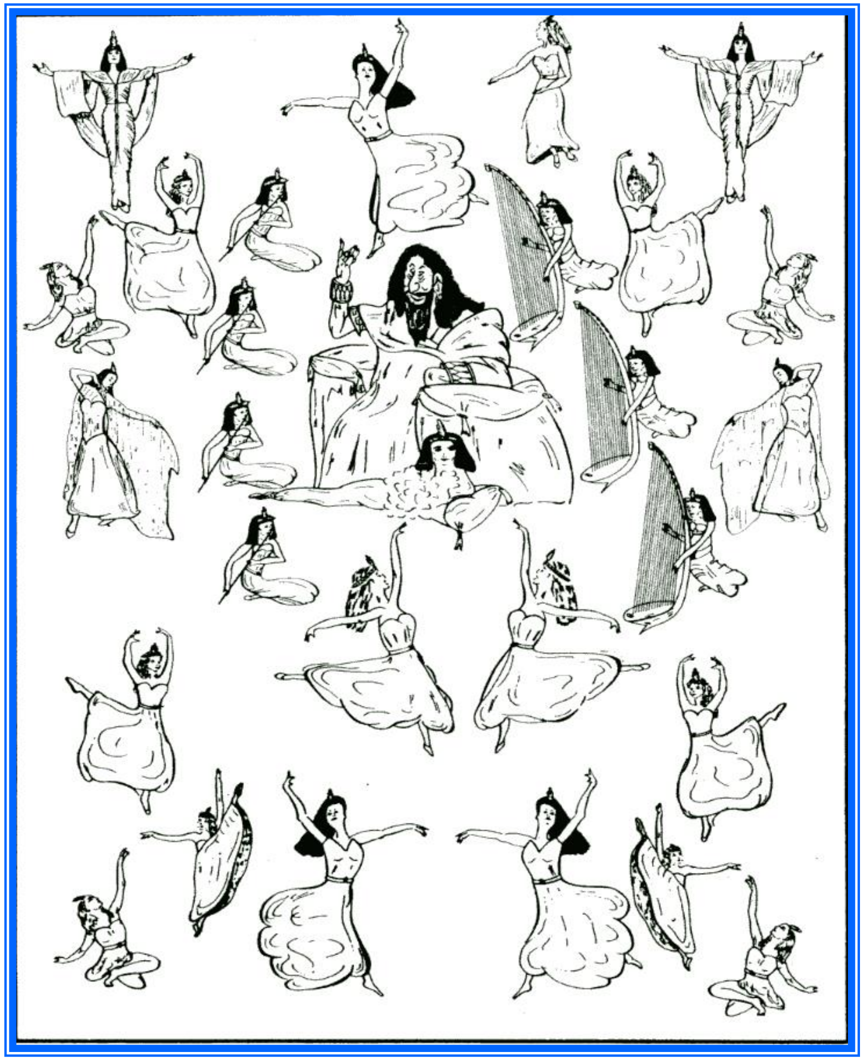
at 1 K, 1.8 T

Solid Effect : A. Abragam (Saclay), C. D. Jeffries(Berkeley)



The proton spin can be polarized with microwaves by means of spin flip-flop effect coupled with the electron spin, followed by spin diffusion due to proton spin-spin dipole interaction, if

$$N_I/T_{1n} << N_s/T_{1e} \qquad (N_I/N_s \sim 700)$$



Comics
by A. Abragam *

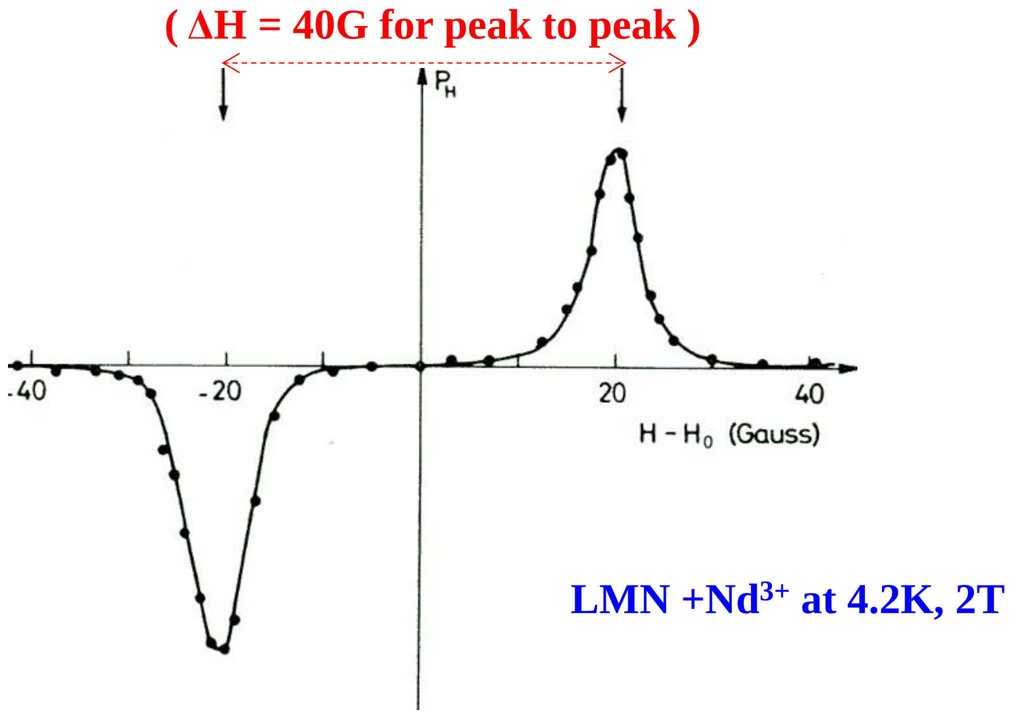
«Le Roi Salomon eut donc sept cents femmes princesses…..»
(I Rois XI)

$(N_I / N_S) = 700$

One electron has 700 protons like The King Solomon had 700 princesses.

•The solid effect works quite well in **$La_2Mg_3(NO_3)_{12}\cdot 24H_2O$ (LMN)** doped with **Nd^{3+}**, where the NMR signals for positive and negative polarizations are well-resolved.

• Protons in the crystal could be polarized up to 80 % at 1 K and 1.8 T.



NMR Amplitude vs. Magnetic Field for LMN

T. J. Schmugge, C. D. Jeffries

High Energy Physics with Spin as a Probe

- The success of the dynamic polarization was really amazing-event for all the high energy physicists in the world, since it promised to open a new field of elementary particle physics.

- I was chocked about the discovery of the solid effect in France and the US.
- Soon after we got the news, we started to construct a polarized proton target in Nagoya University, Japan, for high energy scattering experiments.

- At the International Conference on Polarized Targets held at Saclay, in 1966, many theoretical physicists, including R. H. Dalitz, J. D. Jackson and M. Jacob showed that polarized targets are extremely powerful devices for high energy physics.

Dalitz : Polarized Target Experiments for Elementary Particle Physics

Symposium at Saclay, 1966

The polarized target gives us especially important information on three major areas of particle physics.

1. Very high energy scattering where the contributions of Regge-pole exchange is dominant.

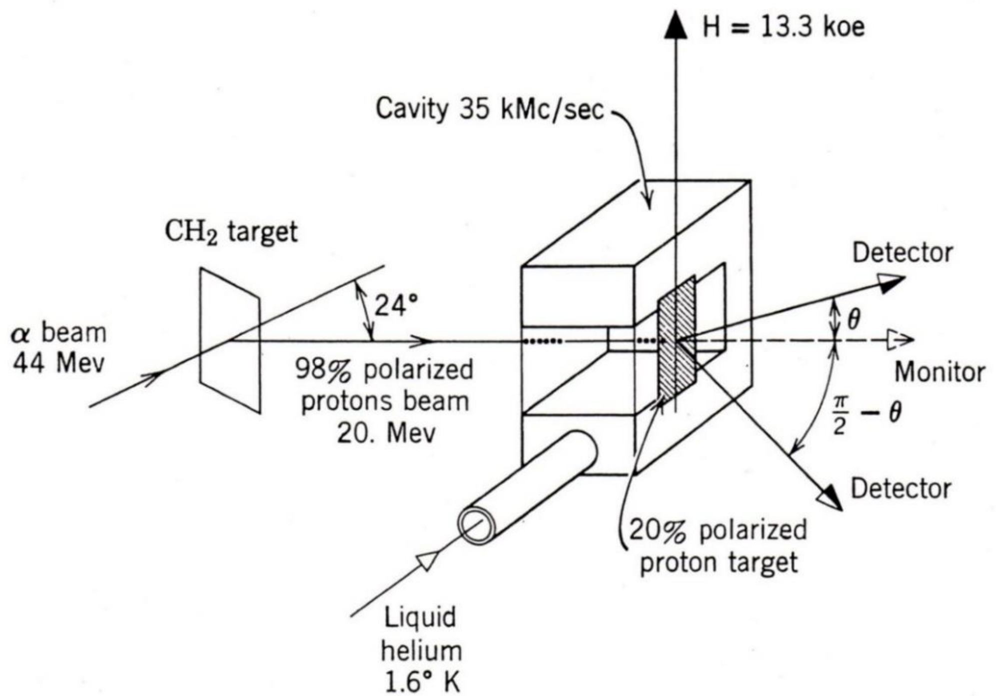
2. Tests of time-reversal invariance.

3. Hadron spectroscopy.
   Many resonant states had been observed for baryonic states. The first need was the determination of the spin and parity for each state.

First Experiment using a Polarized Target

- The first experiment with polarized target was performed by A. Abragam, M. Borghini, P. Catillon, J. Coustham, P. Roubeau and J. Thirion to measure the parameter Cnn for proton-proton scattering at 20 MeV in 1962 at Saclay.

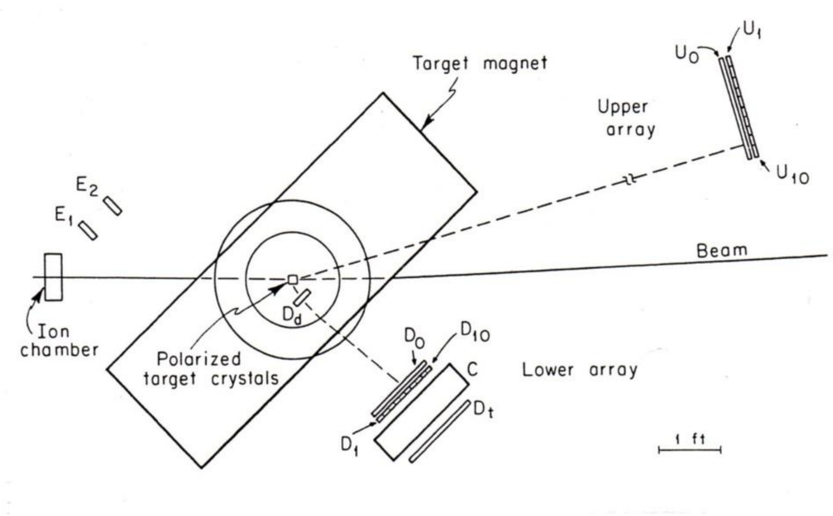
- The experiment was done with a polarized beam on a polarized proton in a crystal of LMN.



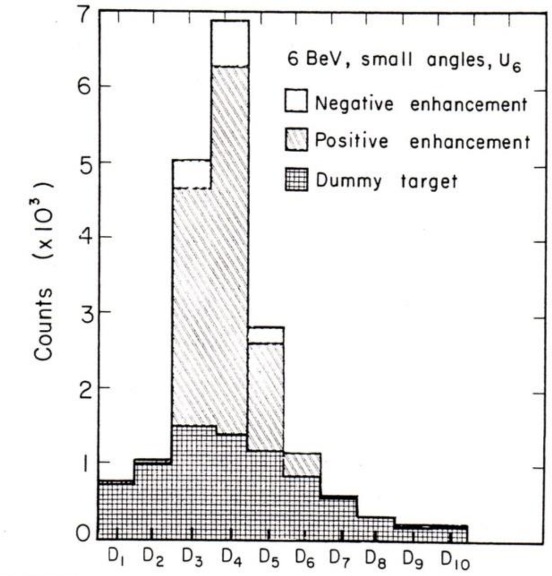
Apparatus of a p-p experiment with a polarized target at Saclay

First High Energy Experiment with Polarized Target

- The first high energy experement was performed at Berkeley (Bevatron) in 1963.
- A pion-proton scattering experiment with a polarized target was done by O. Chamberlain, C. Jeffries, C. Schutz, J. Shapiro, L. van Rossum.
- It was necessary to measure the angles of poins and protons in order to check the coplanarity, since the background from complex nuclei were enormous.



Experimental setup for π-p scattering at Berkeley



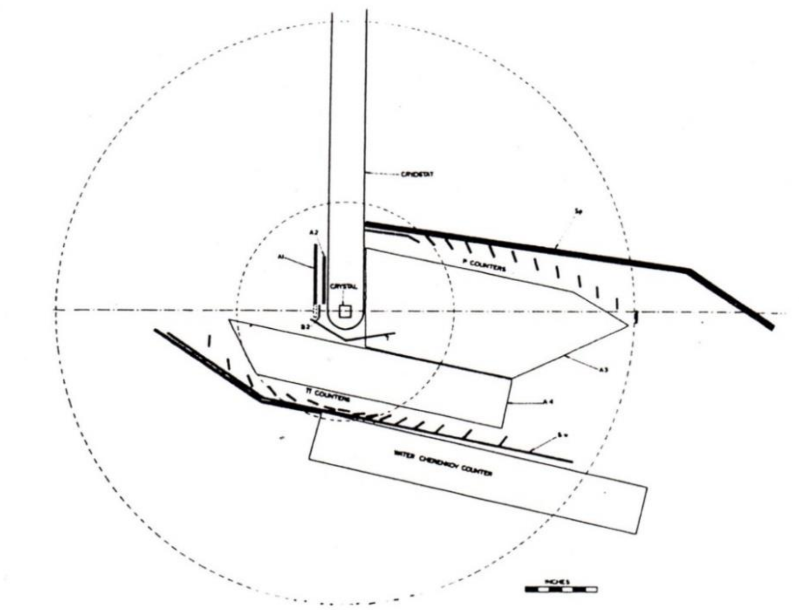
Histogram of coincidence events between π-minus and proton

Pion- proton scattering at Rutherford

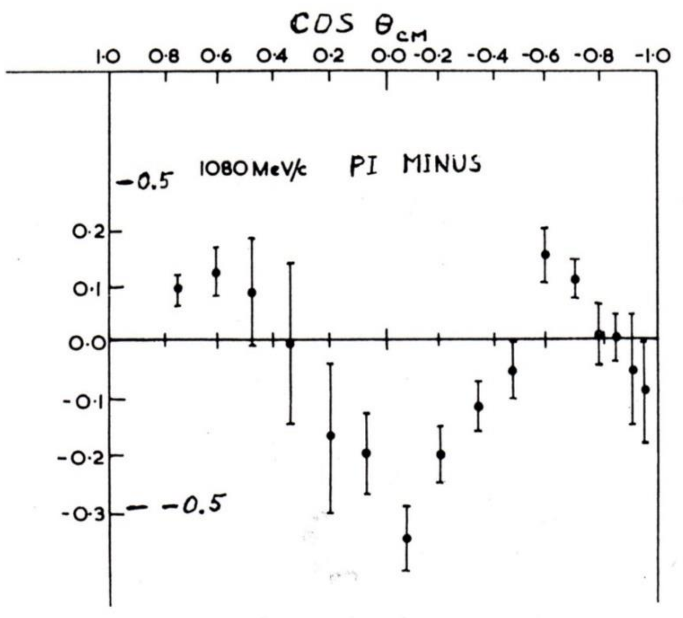
by H.H. Atkinson, C.R. Cox, P.J. Duke, K.S. Heard, D.P. Jones, M.A.R. Kemp, P.G. Murphy, J.D. Prentice and J.J.Thresher with ~1GeV pions (1965).

- It indicated three nucleon resonances, which were not previously seen.

[N* (1674) 5/2 $^-$ N*(1688) 5/2 $^+$ N* (1920) 7/2 $^+$]



Apparatus for polarization measurement at Rutherford



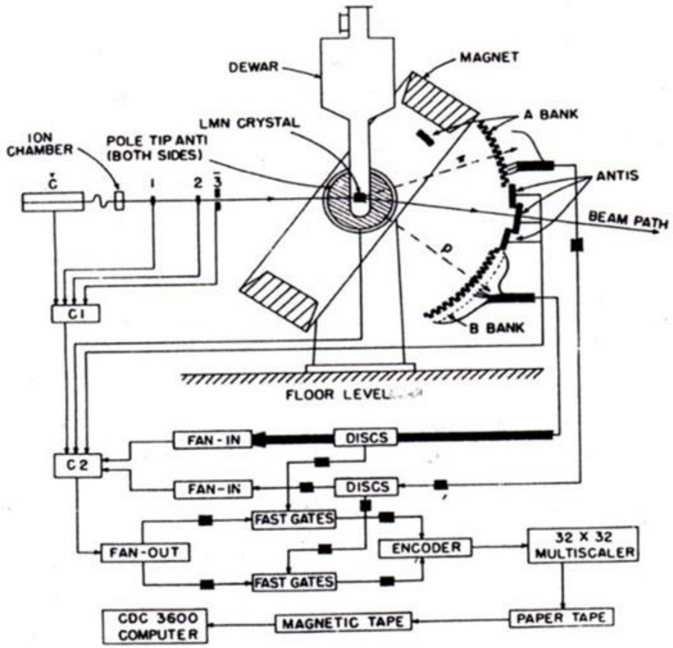
Asymmetry of π-minus proton scattering at 1.08 GeV/c

Pion- proton Scattering at Argonne

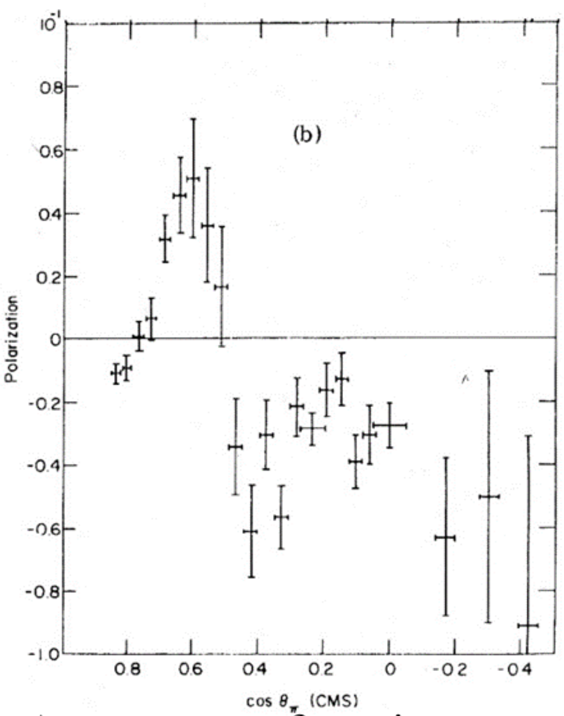
with ˜2GeV pions (1965)

by S. Suwa, A. Yokosawa, N.E. Booth, R.J. Esterling, R.J. Hill

- The result shows a higher nucleon resonance $N^{*}(2190)\ 7/2^{-}$



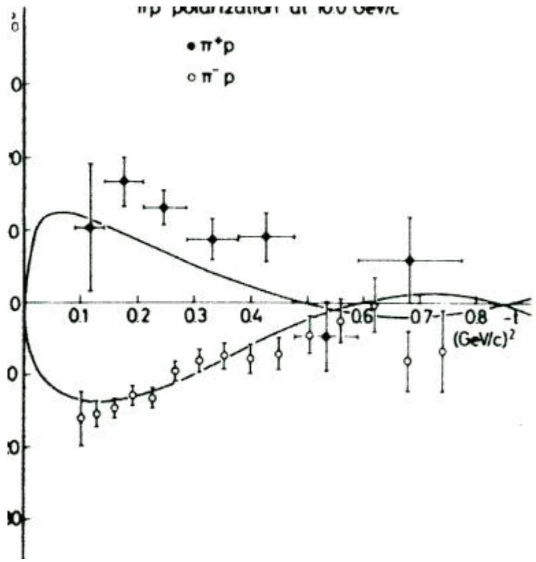
Polarized target arrangement at Argonne



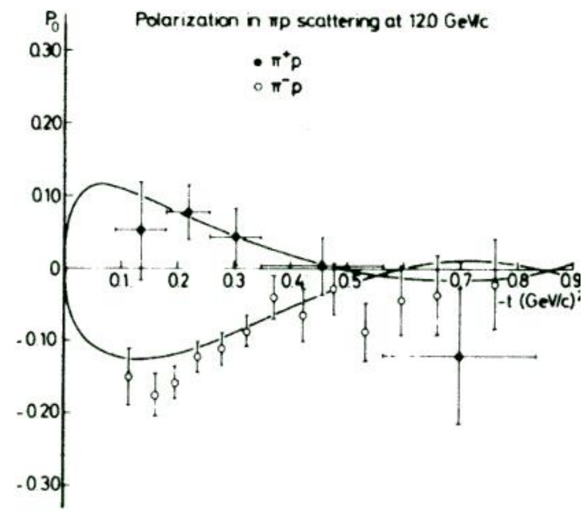
Asymmetry of π-minus proton scattering at 2.08 GeV/c

Asymmetries of Pion-Proton Elastic Scattering

10.0 GeV/c



12.0 GeV/c



M. Borghini et.al at CERN （1966）

- LMN targets were successfully operated for elastic scattering with π, K, p and n beams.
- The polarized target became a very important tool for particle physics.

In the mid-1960s LMN polarized targets were constructed in most of the high energy laboratories.

CERN
Saclay (France)
Rutherford (UK)
Liverpool (UK)

Dubna (USSR)
Protvino (USSR)

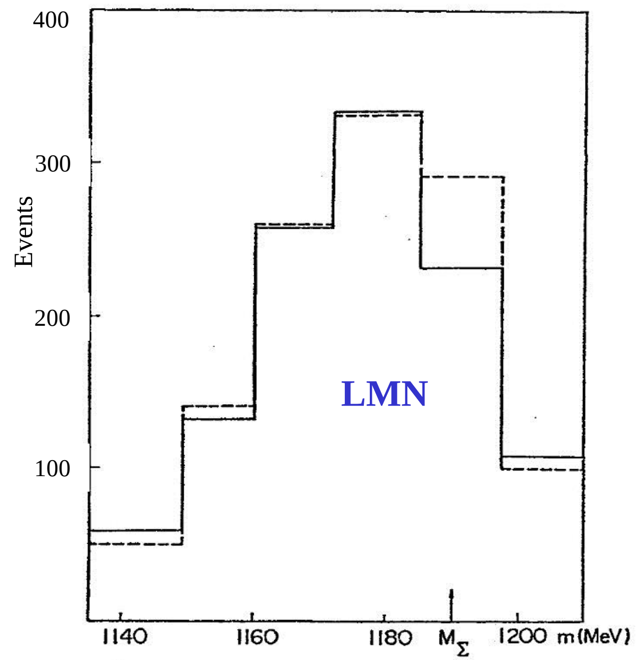
Berkeley (USA)
Argonne (USA)
Los Alamos (USA)
Harvard (USA)

Nagoya-INS (Japan)

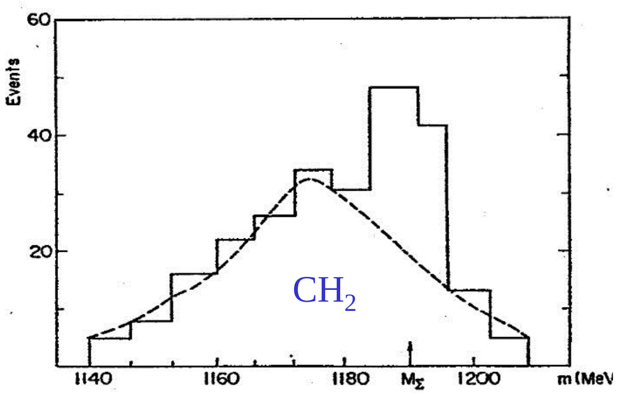
- There was no polarized target in SLAC and DESY, because the radiation damages of LMN crystal with electron and γ-beams was very serious.

Comparison of the data with LMN and CH_2 in $\pi^+ + p \rightarrow K^+ + \Sigma^+$ Reactions

- Although LMN was an excellent polarized target material for elastic scattering experiments, it was not convenient for other experiments, e.g. backward scatterings, inelastic scatterings, rare decays, and for electrons and photons.
- The experiment of $\pi^+ + p \rightarrow K^+ + \Sigma^+$ reaction was tried to carry out at CERN and Berkeley (1964) in order to test the parity conservation using LMN, but it was difficult to identify events from free protons, since the ratio of free protons to bound protons was 1 : 15.



(a)



(b)

Test of Polarization in Organic Materials in the late 1960s

- Dynamic polarizations of protons had been tested with organic materials with free radicals in many laboratories in the last half of the 1960s, since these materials have higher concentration of free protons, and are strong for radiation damage.
- LMN is damaged with relativistic particles of $2\times10^{12}/cm^2$, but diol is damaged with particles of $5\times10^{14}/cm^2$.

- At CERN more than 200 organic materials with many kinds of free radicals had been tested to be polarized by M. Borghini et al..
- However, most of them were not polarized higher than 20 %.

Materials tried to polarize at CERN (in 1965 ~ 1971)

by M. Borghini, S. Mango, O. Runolfsson,
K. Sheffler, A. Masaike, F. Udo

Benzene
Toluene
Ethanol
Methanol
Propanol
Polyethylene
Polystyrene
LiF
Wax
Para Wax

Polexiglass
M-xylol
Mylar
$C_6H_5CF_3$
Diethylether
Tetracosane
Octacosane
$LiBH_4$
Cyclododecan

Palmitin acid
Polyphene
Thanol
Prophlbenzol
Phenylethylether
Phenylethyl-alcohol
$NaBH_4$
Prehnitene
Durol

- **Free Radicals**

DPPH
PAC
BPA
Shape BPA
Violanthrene
Porphyrexide
TEMPO
Ziegler
Anthracene Na^+
TMR
PB
PR

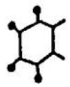
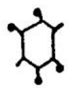
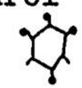
BPA + DPPH
BPA + Cob. Oleale
Ziegler + DPPH
Ziegler + Cob. Oleale
Ziegler + BPA
TMPD
Tri-tetra-bythlphenyl
Tetramethyl 1,3 cyclobutadien
DTBM

etc.

- **Neutron Irradiation**
- **^{60}Co-γ Irradiation**
- **X-ray Irradiation**

Some of the Test Results of Dynamic Polarization with Organic Materials at CERN (1965~1971)

from unpublished memorandum by A. Masaike (1971)

Sample	Radical	Conc. of Rad.	Polarization	T_{1n}	Note
Benzene (C_6H_6)	DPPH		2%	4^m	
Prehnitene 	DPPH	4%	0%	-	
Isodurol 	DPPH	4%	5.5%	2^m	
Durol 	DPPH	4%	0%	-	
Benzen + other BPA		?	30%	$\sim 100^s$	
Diethylether	DPPH	4% (Sat.)	9%	70^s	
(C_2H_5-O-C_2H_5)	"	2%	2%	20^m	
"	TMR	3%	1.5%	125^s	
"	PB	2%	2%	2^m	
Anthracene + Na^+		-	0	12^h	
Methanol (CH_3OH)	DPPH	2%	1.5%	75^s	
"	PB	<2%(Sat.)	4%	13.5^m	

2^{nd} Generation of Polarized Target

Breakthrough was made in 1969

Protons in **butanol** with small amount of water doped with **porphyrexide** were polarized up to 40 % at 1 K and 2.5 T at CERN by S.Mango, O.Runolfsson and **M. Borghini**, .

At the same time, protons in **diol solutions with Cr^{5+} complex** were polarized up to 45 % at 1 K and 2.5 T at Saclay by M. Odehnel and H. Glättli.

High Polarization in ^{3}He Cryostat / High Field

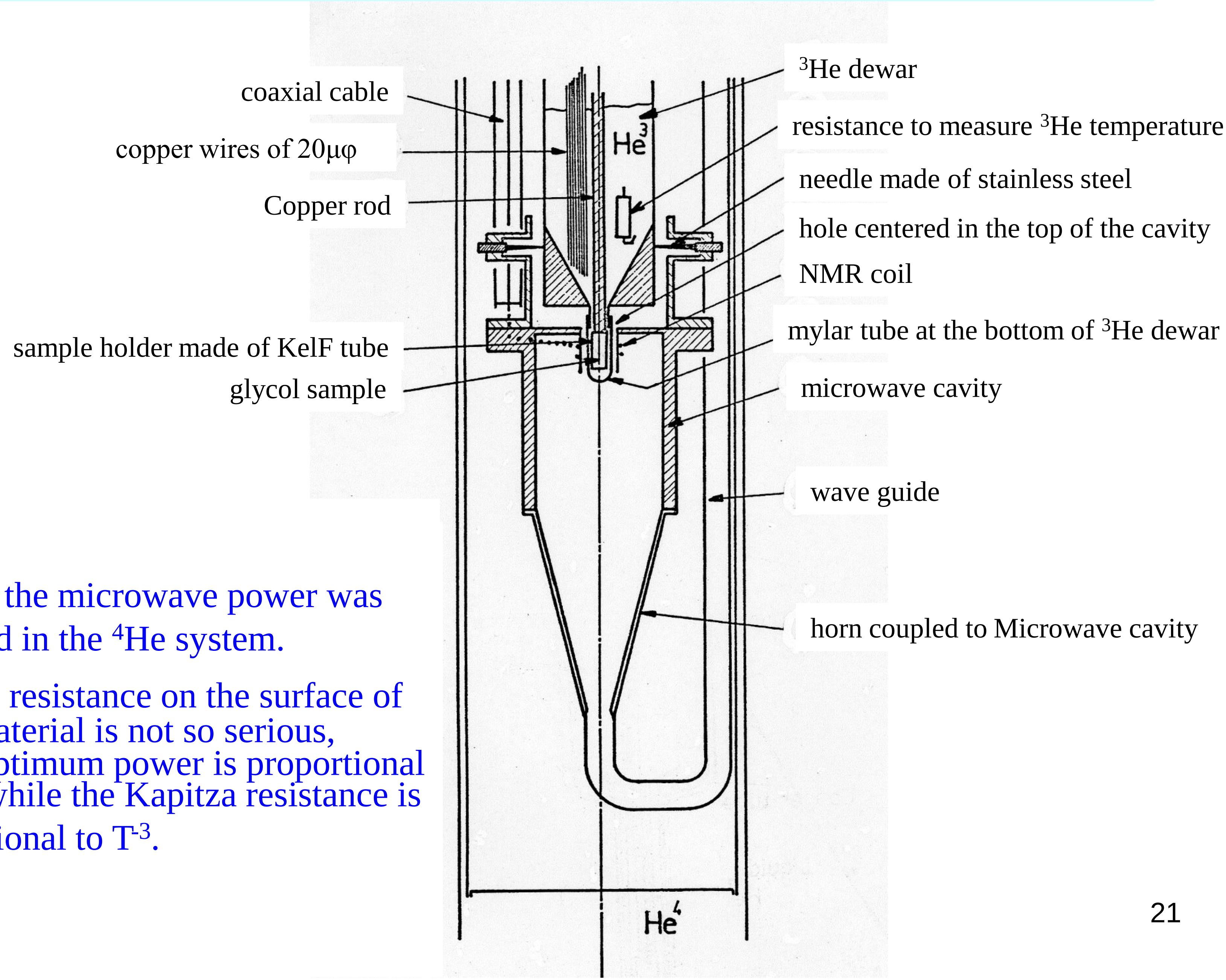
- In 1969, polarizations of 80 % and 70 % were obtained for diol and butanol in ^{3}He cryostats at Saclay (A. Masaike) and Argonne (D. Hill) , respectively.

- These values are surprisingly large, since it had been believed that the polarization in lower temperature than 1 K would be less than that at 1 K.

- In 1970, butanol and diol in ^{3}He cryostats became standard targets in almost all the laboratories.

- In 1971, the polarization at 1 K and 5.0 T was tried at CERN (A. Masaike and M. Borghini), and the similar polarization as at 0.5 K and 2.5 T was obtained.

^{3}He Cryostat used for Dynamic Polarization at Saclay



Most of the microwave power was absorbed in the ^{4}He system.

Kapitza resistance on the surface of solid material is not so serious, since optimum power is proportional to T^4, while the Kapitza resistance is proportional to T^{-3}.

Theories beyond Solid Effect

Spin Temperature Theory (Abragam and Redfield)

A spin system isolated from the lattice and subjected to spin-spin interactions proceeds toward an equilibrium such that the probabilities of finding the system on the energy levels are given by a Boltzmann distribution

$$\exp(-E_i/kT_s),$$

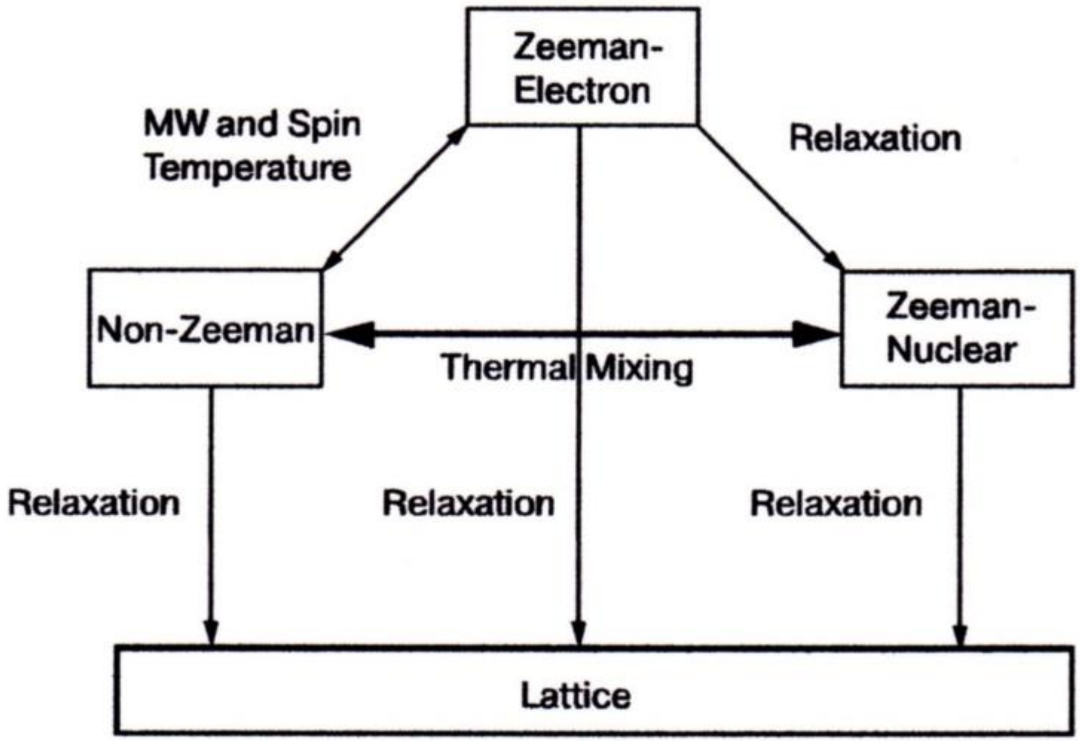
where T_s is the spin temperature of the state.

In this hypothesis, spin systems behave in the same way as the systems usually considered in thermodynamics, for which the approximations leading to the concept of temperature have proved to be valid experimentally.

Application of Spin Temperature Theory to DNP (by M. Goldman and A. Landesman and I. Solomon (Saclay) in 1963)

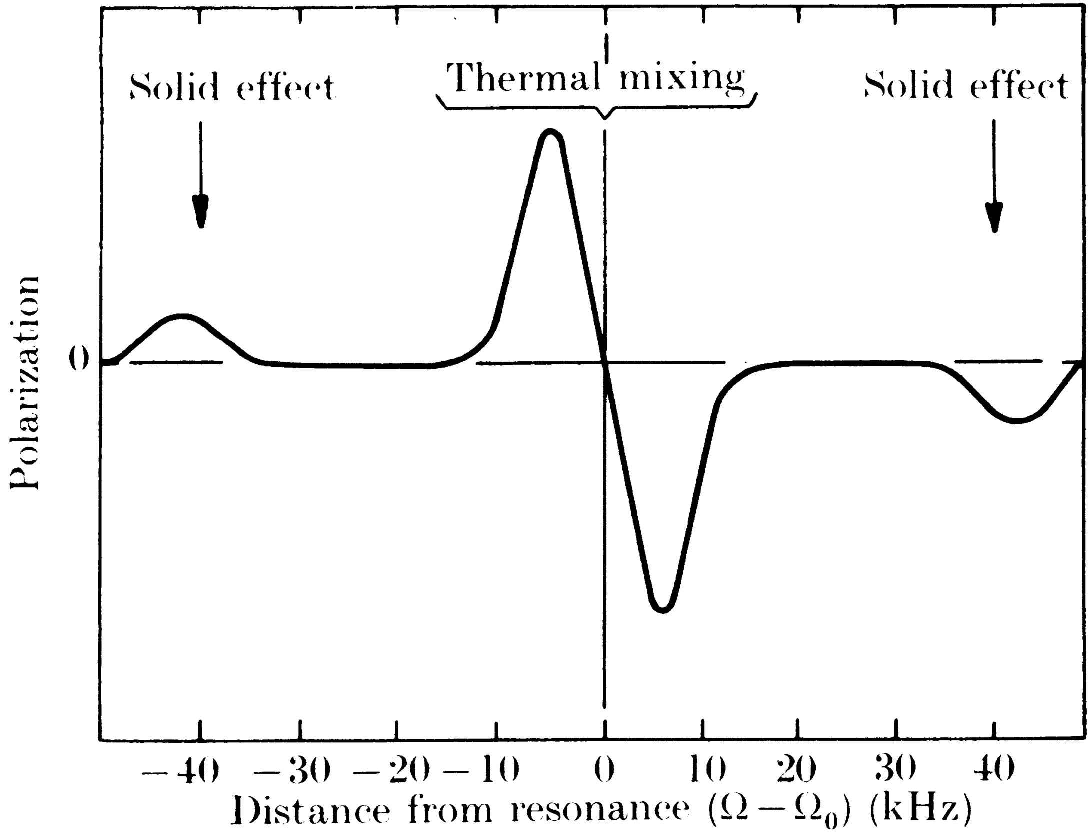
The electron non-Zeeman reservoir is in close thermal contact with the nuclear Zeeman reservoir through thermal mixing, and their common spin temperature evolves towards a high nuclear polarization.

Spin Temperature Model by M. Goldman et al.



Thermal couplings between 4 thermal reservoirs:
electronic Zeeman, electronic non-Zeeman and nuclear Zeeman and lattice, in the course of DNP under microwave irradiation

Solid Effect and Thermal Mixing



Microwave Frequency

The mechanism of polarization by thermal mixing in spin temperature theory is completely different from the solid effect.

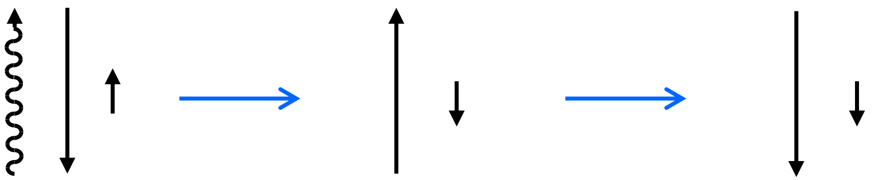
Contribution of Borghini to Spin Temperature Model

- M. Borghini reinvented the spin temperature model independently in 1968 and presented the idea to particle physicists at Berkeley Conference on Polarized Target (1971).

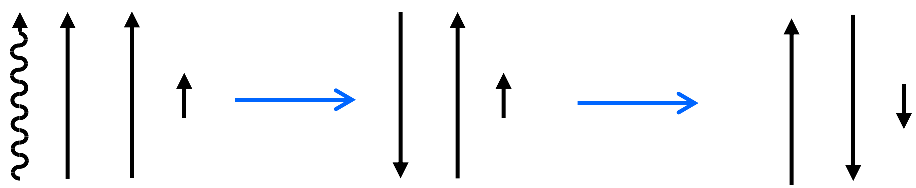
- He made an important contribution by introducing a model of spin packets for the inhomogeneous broadening of the electronic resonance lines, whereas he could compute the nuclear polarization under microwave irradiation in low temperature.

- He named the spin temperature model the 'Donky Effect'.

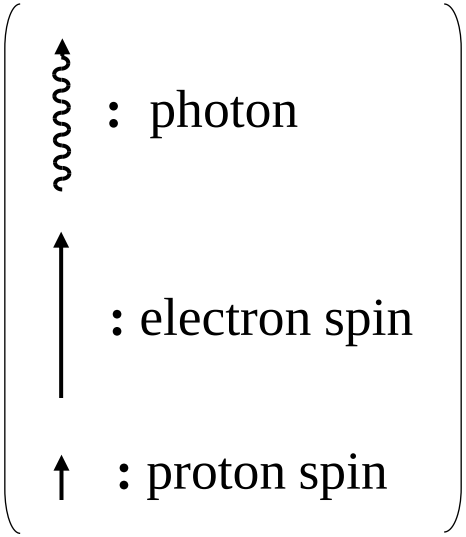
The phenomena were explained with a model of exchange of energy quanta between a nuclear Zeeman energy reservoir and an electron spin-spin interaction reservoir. (Donkey Effect: M. Borghini)



Solid Effect



Cooling by Electron Spin-spin Interaction *
(Donkey Effect)



In the donky effect more than two electrons are involved in the proton polarization.

Seminar at Berkeley

- In the summer of 1969, Owen Chainberlain invited Michel and myself to Berkeley to give talks on the new ideas toward the next generation of high energy spin physics.

- At that time, measurements of spin parameters in several particle reactions were an urgent issue in high energy physics.

- However, it was a hot summer for students in Berkeley against the Vietnam War.

- Therefore, it was impossible to have a seminar in the university.
- Finally, Owen invited us to his home.

- After dinner, he asked us to give talks to only one audience, Owen.
- I showed the latest results of high polarization of proton in the temperature lower than 1 Kelvin.
- After my talk, Michel explained his idea on the spin temperature model in a quiet manner, since he was kind of shy.

- I remember that our talks at Owen’s home implicated the new generation of the high energy spin physics.

Several new ideas were proposed in the 1960s

^{3}He-^{4}He dilution refrigerators with high power were developed toward higher polarization in lower temperature than 0.1K.

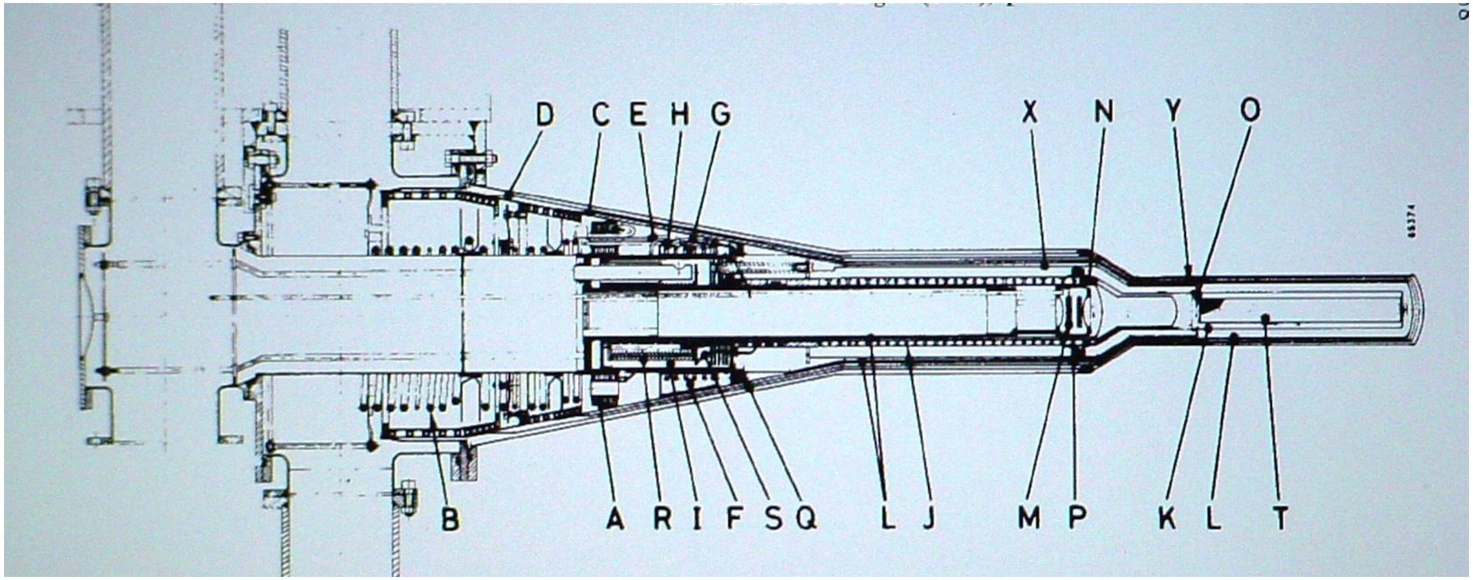
1966 B. Neganov (Dubna): high power dilution refrigerator
E. Varoquaux (Orsay): proposal for polarized target
K. Nagamine (Tokyo): static polarization of nuclei

1974 T. Niinikoski (CERN): spin frozen target with very high power dilution refrigerator
1974 A. Masaike (KEK) : polarized deuteron target with dilution refrigerator

Spin Frozen Target

- The target polarization can be maintained without microwave irradiation for long time, if the nuclear spin relaxation time is long enough.
- It is advantageous because of large access angle around the target region in less homogeneous and lower magnetic field than in the polarizing condition.
- Such targets"Spin Frozen Targets" with dilution refrigerators were constructed at **CERN** in 1974, few months later at KEK, then Saclay, Dubna and Bonn.

Beam →



The Dilution Refrigerator for Spin Frozen Target at CERN

T. Niinikoski

T < 50 mK   B = 2.5 T   Propanediol polarization : 80 ~ 90 %

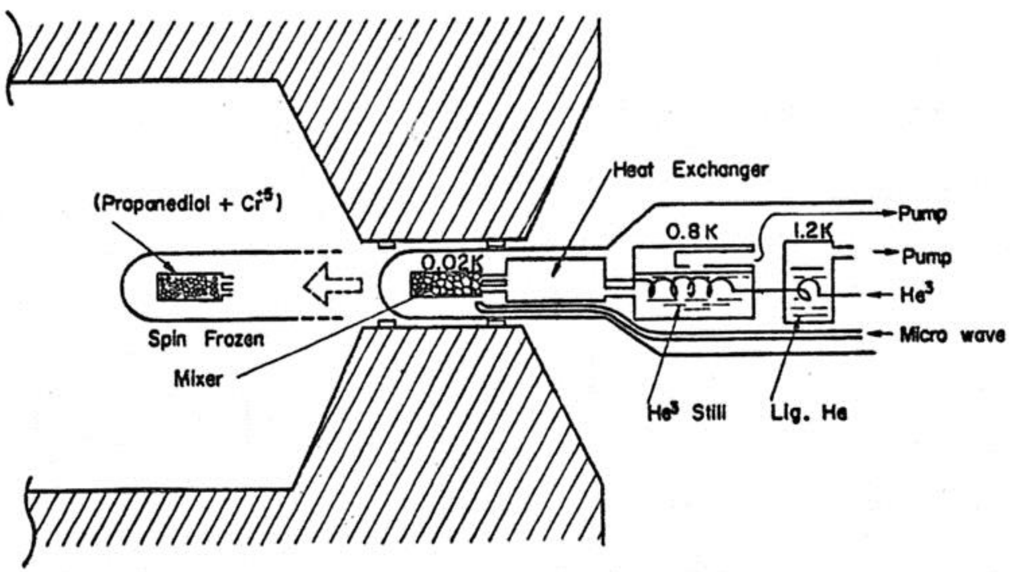
Schematic View of the Spin Frozen Target at KEK

- In 1974 a spin frozen deuteron target was constructed at KEK for the reactions

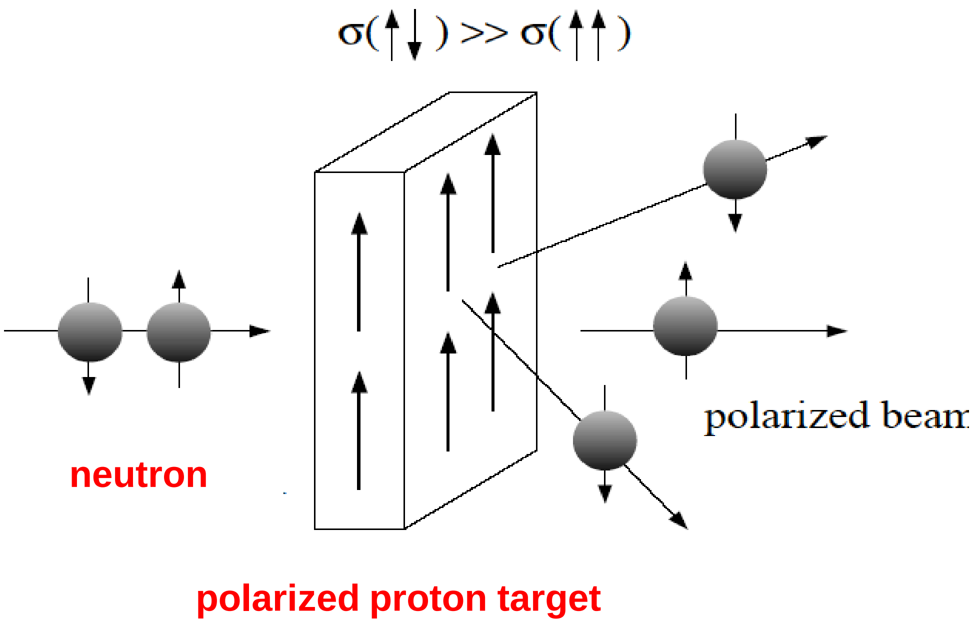
$$K^+ + n \rightarrow K^+ + n\ ,\ K^0 + p$$

in order to search for 5 quark systems.

- Target material was deuterated propanediol doped with Cr^{5+} complex.
- The target could be used for various experiments with wide access angle.



Neutron Polarization with Proton Filter



- Neutron transmitted through polarized protons are polarized, since neutrons with spin anti-parallel to the proton spin are scattered away.

1966: F. L. Shapiro, V. I. Lushchikov at Dubna (LMN)

1975: S.Ishimoto A. Masaike et al. at KEK (propanediol in ^{3}He cryostat)

1980s: KEK, TIT and Los Alamos polarized neutrons of 0.02 ~ 1 eV, for parity violation experiments

Summary

- High energy spin physics was born in the early 1960s.
- Polarized proton targets in **$La_2 Mg_3(NO_3)_{12} \cdot 24H_2O$** was developed by dynamic nuclear polarization in Saclay and Berkeley at the beginning of the 1960s.
- Many high energy experiments with polarized targets were performed soon after the discovery of the dynamic polarization method.
- In 1969 protons in organic materials were found to be polarized.
- High polarization was obtained in lower temperature than 1K.
- The spin temperature model was proposed for nuclear polarization.
- Dilution refrigerators were found to be useful for spin physics.

Michel Borghini opened new field of physics
'High Energy Spin Physics'

Back Up Slide

Talk in memory of Michel Borghini

*

- It is a great honor for me to give a talk on the dawn of high energy spin physics in memory of Michel Borghini.

- First of all, we would like to express our deepest condolences to Michel's family.

- Michel was an outstanding physicist who played an important role in progress of high energy spin physics.

- His contribution in this field has been highly appreciated.

- He had been a good friend to many of us throughout his life.

- He has been missed.

- Michel was born in 1934.
- He comes from a noble family in Monaco.
- After graduation from Ecole Polytechnique in Paris in 1955, he joined the group of Prof. Anatole Abragam at Saclay.
- In Saclay, he studied the nuclear magnetic resonance, in particular, dynamic polarization of proton, then he joined the first nucleon-nucleon scattering experiment with a polarized target.
- It was in the mid-1960s when I had been acquainted with Michel.
- And then sometimes we worked together, sometimes separately.
- Michel was the first particle physicist who came originally from the field of solid state physics.
- He opened the new field in particle physics, "**high energy spin physics**", the dawn of which I briefly would like to look at.

Beginning of High Energy Spin Physics

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- High energy spin physics was born in the early 1960s.
- It was pointed out that studying the spin dependent forces is one of the most important issues for particle physics.
- Therefore, it became an urgent need to measure the spin parameters of particle reactions.
- In order to realize such experiments, it was indispensable to polarize the target protons.
- The dynamic methods which transfer the electronic polarization to protons using magnetic couplings between electronic and nuclear spins was proposed by Antole Abragam in Saclay and Carson Jeffries in Berkeley.
- I am going to explain the origin of the idea of dynamic polarization, progress of the polarized target, and high energy physics using it in the1960s.
- Michel was certainly a main player for all of them.

*

- The static polarization at 1 K and 1T is 10 to -3.
- It is impossible to use static polarization for polarized targets.
- Using the solid effect with microwaves, we can get much higher polarization.

High Energy Physics with Spin as a Probe

*・

・ In the 1960s, it was pointed out that the spin was a very important probe to study the property of elementary particles.

(At that time, the quark model had not been established yet.)

・Information concerning spin could be obtained only by scattering experiments using the polarized target.

・At the International Conference on Polarized Targets held at Saclay, France, in 1966, so many theoretical physicists, including R. H. Dalitz (Oxford) J. D. Jackson (Univ. Illinois, Urbana) and M. Jacob (Saclay) showed that polarized targets are extremely powerful devices for high energy physics.

Anthracene
Hexanol
Water
Propanol
Methylcyclohexan
Isodurol
Tetrahydrofuran
O-xylol
2,5 Dimethyltetrahydrofuran
1-Hexadecarol
Dioxan
Oppanol
$(CH_3)_4NBH_4$
$(CH_3CH_2)_4NBH_4$
NH_4BH_4
Tetramethylbenzene
Tritetra-butylphenol

Benzen + Ether
Propanol + Ethanol
Ethanol + Water
Ethanol + Methanol
Ethanol + Propanol
Ethanol + Diethylether
Butylalcohol + Methanol
Methanol + Propanol
$NaBH_4 + NH_4F + NH_3$

Maximum Polarization and Reproducibility

It was a very serious problem that the reproducibility of polarization was not satisfactory.

Sometimes the proton polarization was about 15%, and sometime it was about 20%, even though the recipes for cooking the target materials (mixture of organic material, water and free radicals) were the same.

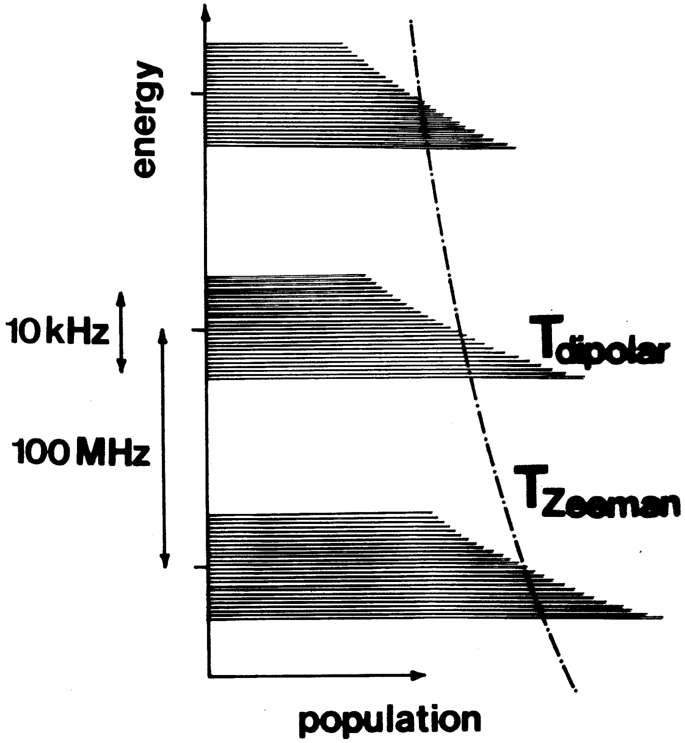
Moreover, the polarization at CERN was always slightly higher than that at Berkeley and Argonne.

They said that water in Lac Leman and water in Lake Michigan in the US are different.

In any case, 20% polarization of proton in organic material had not had better figure of merit than 80% polarization in LMN.

Therefore, all the high energy physicists had been waiting for a break-though in the solution.

Two constants of the motion



Two temperatures :

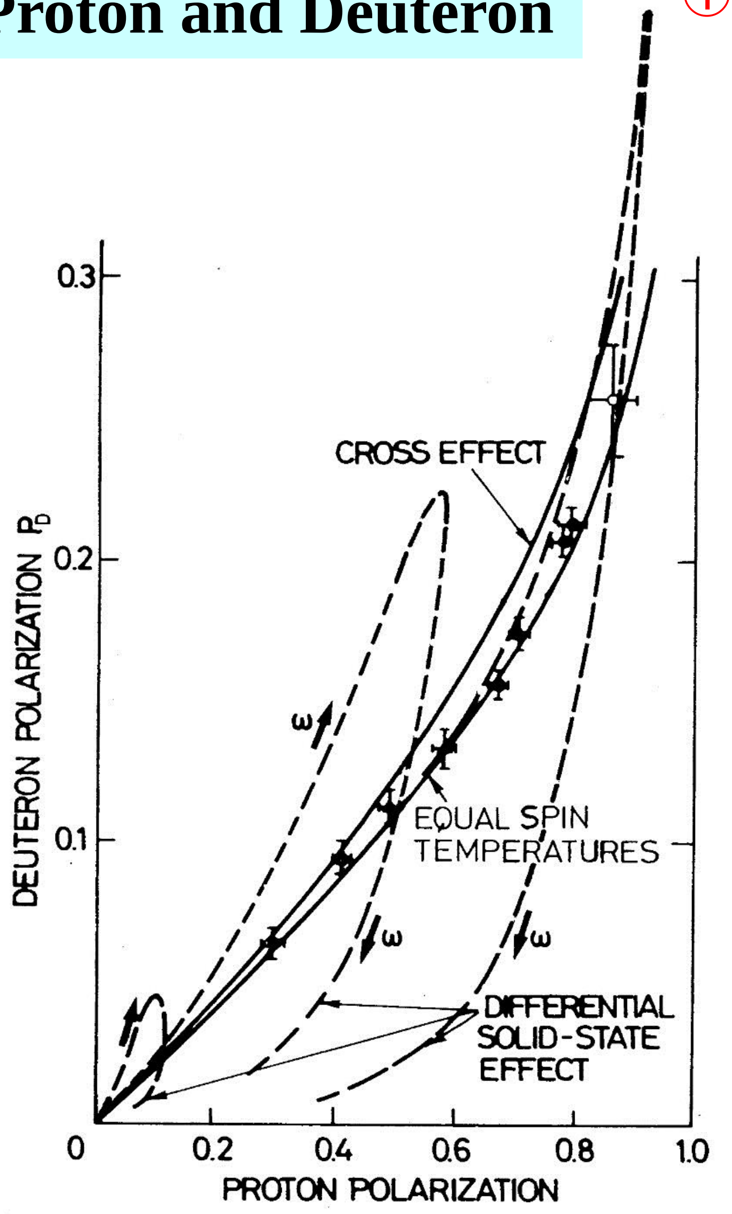
- Zeeman temperature,
- Dipolar temperature

The two thermal reservoirs can be coupled by rf irradiation close to Larmor frequency.

Relation between Polarizations of Proton and Deuteron

①

- The relation between polarizations of protons and deuterons can be interpreted as the **Equal Spin Temperature**, which is applicable also to polarizations of several nuclei as **^{1}H, ^{2}D, ^{6}Li, ^{7}Li, ^{11}B, ^{13}C, ^{14}N, ^{15}N, ^{17}O, ^{19}F, ^{27}Al, ...^{139}La, ...**



Comparison of Proton and Deuteron Polarizations measured in Ethanol and 1-Butanol Doped with Porphyrexide at 2.5 T and 5.0 T and at 0.5 K, 1.1 K

(M.Borghini, K.Scheffler, A.Masaike and F.Udo)

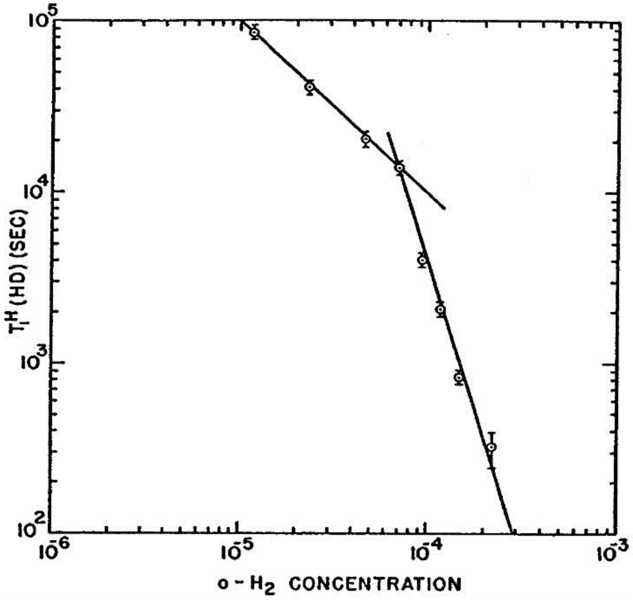
Hydrogen Deuteride (HD)

- W. N. Hardy (1966) and A. Honig (1967) proposed the HD polarization by a brute force method. (Static Polarization)

- They measured the relaxation properties of proton and deuteron at 0.5 K, which depend on the ortho-para and para-ortho conversions of H_2 and D_2, respectively .

- Honig pointed out that p's and d's can be polarized with a little ortho H_2 at 0.5 K, then polarization is kept for long time at ^{4}He temperature after ortho H_2 converts to para H_2 in 2 months (relaxation switch).

- H. M. Bozer and E. H. Graf measured T_{1n} in a dilution refrigerator.

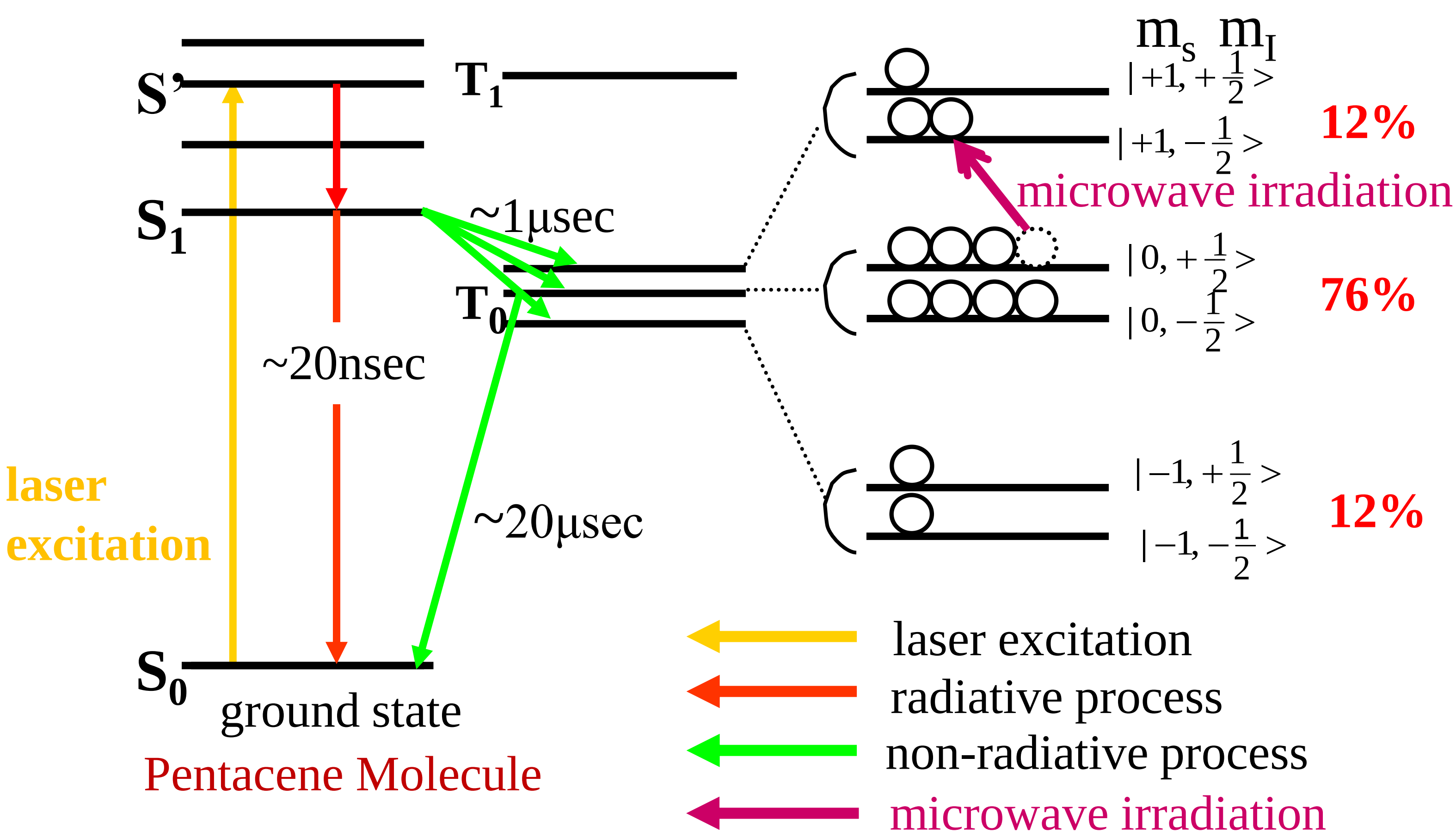


Proton spin relaxation time vs. ortho-H_2 consentration at 0.5 K (Honig)*

Polarization of naphthalene at High Temperature

by M. Iinuma et al.

Pentacene molecule is excited to triplet states by laser irradiation, where protons are polarized dynamically.
The polarization is diffused to naphthalene at the ground state in room temperature.



Summary 2

Several new ideas were proposed in 1960s

- Dilution Refrigerator with High Power for Spin Physics :
  toward lower temperature than 0.3K

- HD polarization was proposed by a brute force method.
  (Static Polarization)

- Slow neutrons can be polarized through a polarized filter.

Future applications of polarized target to wide field of science is expected.

Michel Borghini opened new field of physics
'High Energy Spin Physics'

Foundation of High Energy Spin physics

High energy spin physics was born early 1960s, when the principle of dynamic polarization was found in France and the United States.

Needless to say, the contribution of Michel to the naissance of the ideas of dynamic polarization of proton and it’s application to high energy physics was enormous.

I am going to explain the origin of the idea of dynamic polarization, progress of the polarized target, and high energy physics using it in 1960s.

Michel was a main player for all of them.

I remember that he took lunch together with the king of Monaco and his wife Greass Kelly quite often.

After his retirement from CERN, at the beginning of the $21^{st.}$ Century
he became the representative of Monaco for United Nations.

(He was a quite but strong man.)

Early History of High Energy Physics with Polarized Target

1962 ~ : 1st Generation

$La_2\ Mg_3(NO_3)_{12} \cdot 24H_2O$ in ≥ 1 K and 1.8 T

1969 ~ : 2nd Generation

Butanol & Diol at 0.5 K and 2.5 T

1974 ~ : 3rd Generation

Spin Frozen Target with Dilution Refrigerator

1980 ~

- **NH_3 , LiD, LiH***
- **HD**
- **Neutron Polarization using Polarized Target**
- **Polarization in High Temperature**

- **I will cover the first and second generations, and I hope the following speakers would cover the rest.**

How to Polarize the Proton for Spin Physics

Orientation of Nuclear Spin

Polarization : $P_n = < I_Z > / I$

Alignment : $A_n = < I_Z^2 - I(I+1)/3 > / I^2$

$= 0$ for proton

I : Nuclear spin

I_Z: Magnetic substate of nuclear spin

P(-m) = P(m) $\rightarrow$ $P_n = 0$

P(m): Population on the substate with magnetic quantum number m

Static Polarization

Paramagnetic Salts

Salts have poor thermal conductivities

Solutes in Ferromagnets

Hyperfine magnetic fields at nuclei of solutes in ferromagnetic lattice are used.

Rare gases, halogens, and alkali metals are put into a metallic iron lattice.

Ferromagnetic Compounds

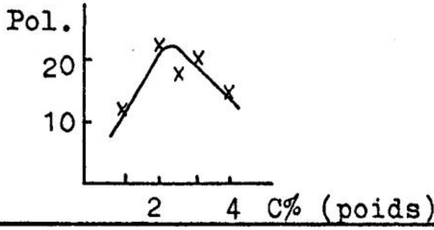
BiMn compounds : orientation of ^{205}Bi, ^{206}Bi

Direct Polarization

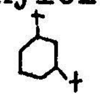
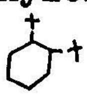
Brute force methods for polarization

Some Results of Dynamic Polarization with Organic Materials at CERN (1965~1971)

from unpublished memorandum (18 pages) by A. Masaike (1971)

Sample	Radical	Conc. of Rad.	Polarization	T_{1n}	Note
Toluene ($C_6H_5CH_3$)	DPPH	2%	23% (max)	$90^s, 140^s$	
$C_6H_5CF_3$	DPPH	2%	H: 5% F: 5%	12^m 11.5^m	
$C_6H_5CH_3$	PAC	2%	2%	16.5^m	(PAC: Picryl -amino-Carbazyl)
	violanthrone	Sat.	0	—	
	BPA	4%(0.3%DPPH)	max 19.5%	93^s	tried 3~4.2% BPA with 0~2%DPPH
	TMPIO	3%	~+8%		
	Ziegler	10%	-44%	1^h	
	Ziegler	~ 10% + DPPH 1%	-44%	1½ h	
	Ziegler	~ 3% + CoOl 2.5%	5%	30^m	
	Ziegler	~ 8% + " 1%	20%	10^m	
	Ziegler	~ 10% + BPA 3%	~-39%	24^m	
$C_6H_5CH_3$	Ziegler	5%	+31% -33%	1^h20^m	
$C_6H_5CH_3$	Ziegler	Sat.	36%	115^m	

Some of the Test Results of Dynamic Polarization with Organic Materials at CERN (1969~1971)

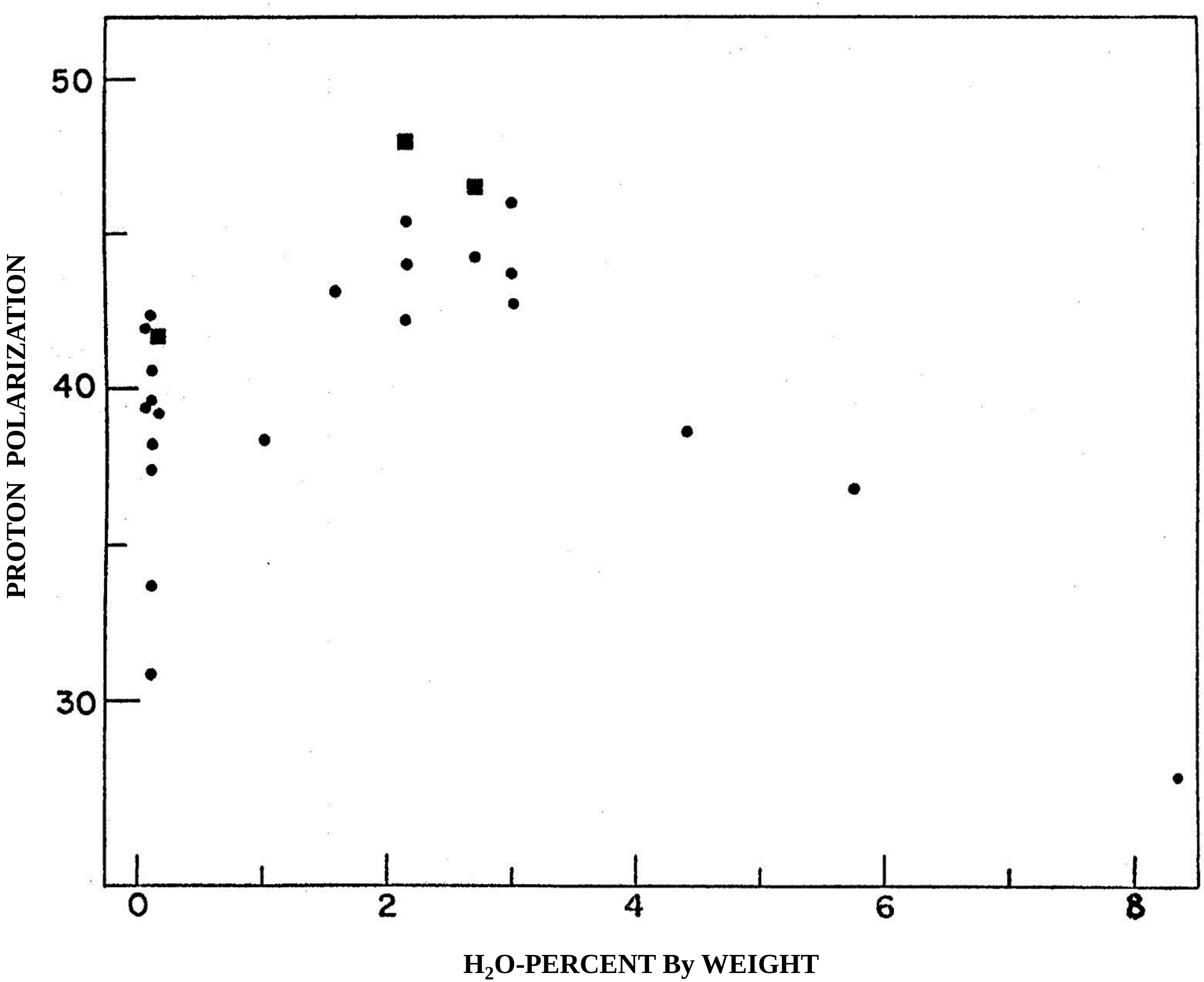
Sample	Radical	Conc. of Rad.	Polarization	T_{1n}	Note
M-Xylol	BPA	5.5%	-10%	-	
	"	1.2%	9%	-	
O-Xylol	BPA	3.2%	15~18%	32^m	
	"	3.9%	15%	40^m	
	"	5.3%	12%	22^m	
"	BPA + DPPH	3.9% + 0.5%	<13%	100^m	
	BPA + Cob. Oleale	3% + 1.4%	12%	30^m	$\tau_n \sim 300^s$, $T = 1.08^oK$
2,5 Dimethyltetrahydrofuran					
	BPA	2% (Sat.)	17%	-	
1 Hexadecarol					
	BPA	4%	~0	$\sim1^h$	
Dioxan	BPA	low	0.5%	-	

Some of the Test Results of Dynamic Polarization with Organic Materials at CERN (1969~1971)

Sample	Radical	Conc. of Rad.	Polarization	T_{1n}	Note
Methanol	PR	Sat (4%?)	10%	14^s	
		2%	8.5%	255^s	
		3%	13%	30^s	
Hexanol	DPPH	1~2% (Sat.)	+0	-	
"	PR	4% (Sat.)	+1%	142^s	
			+2.5%	700^s	
Tetrahydrofuran	DPPH	2%	+4%	8^m	
"	Ziegler		+4%	$\sim 40^m$	$T_{pol} \sim 20^m$
			~+6%	$\sim 24^m$	
Water	PB	2%	0	$\sim 30^m$	
Propanol	PR	3%	~20%	120^s	$T \sim 1.02^o K$
Propanol + 5% Ethanol	PR	4%	~15%	$\sim 135^s$	
Methylcyclohexan	DPPH	3%	<1%		
	TMPD	4.2%	~1%		

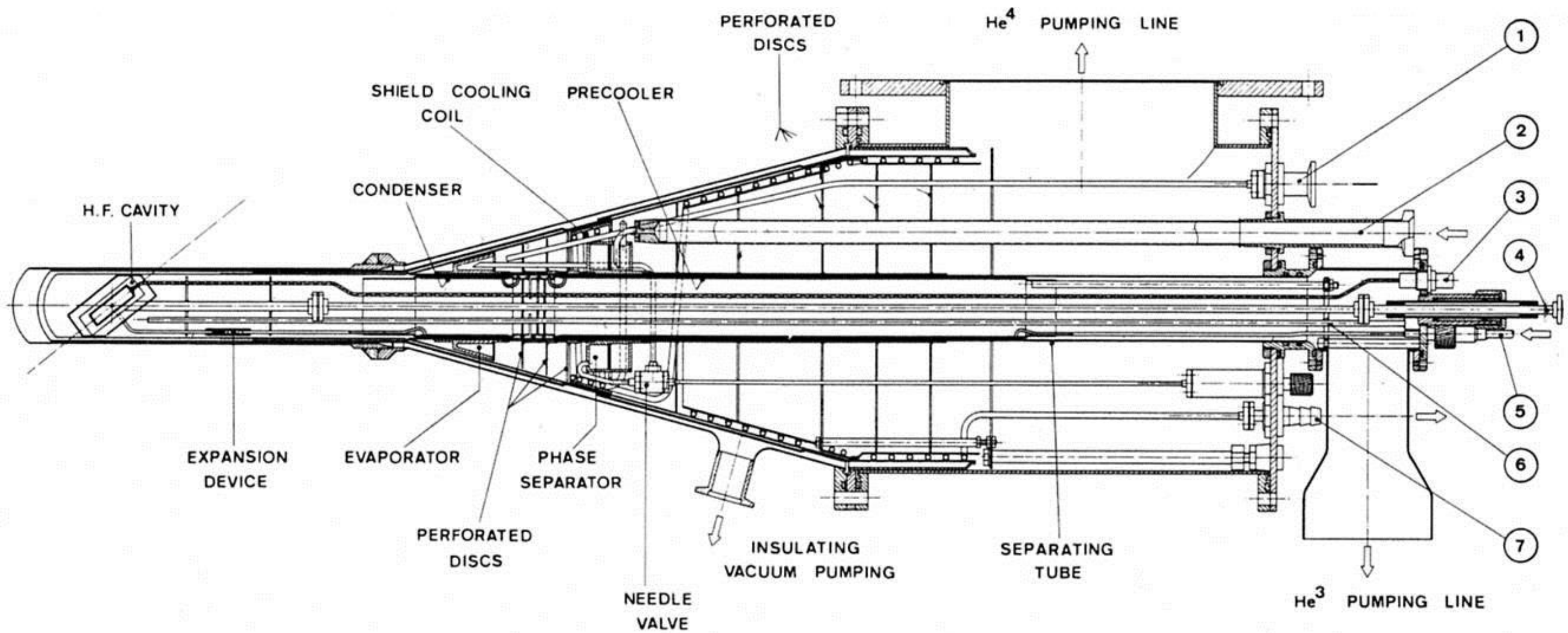
B

Proton Polarization in the Eutectic Mixture of Ethylene Glycol-Water with Cr^{5+} at 1 K and 2.5 T (H. Glattli)



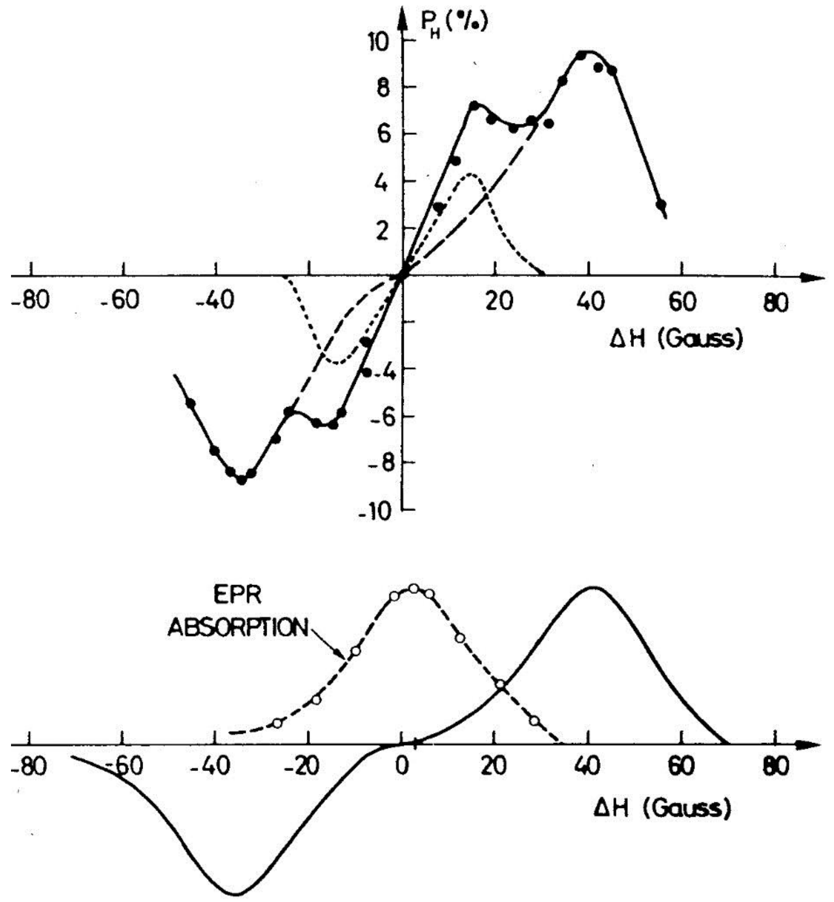
B

Horizontal ^{3}He cryostat for Polarized Target at Saclay and CERN by P, Roubeau (1970)



- ^{3}He system is completely separated from ^{4}He-precooling system.
- Target material can be put in the position without warming up the ^{4}He system.

B



P_H (°/.)
10
8
6
4
2
-4
-6
-8
-10
-80
-60
-40
20
40
60
80
ΔH (Gauss)
EPR
ABSORPTION
-80
-60
-40
-20
0
20
40
60
80
ΔH (Gauss)

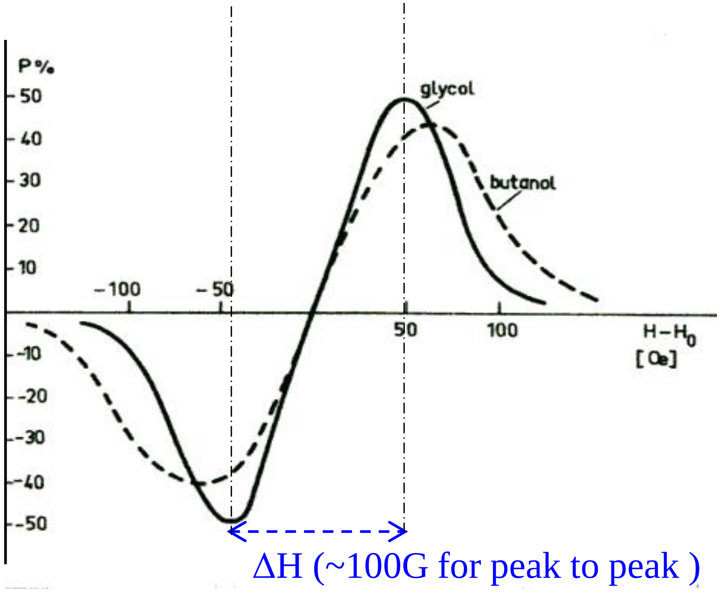
(P. Weymuth) *

B

Beyond Solid Effect

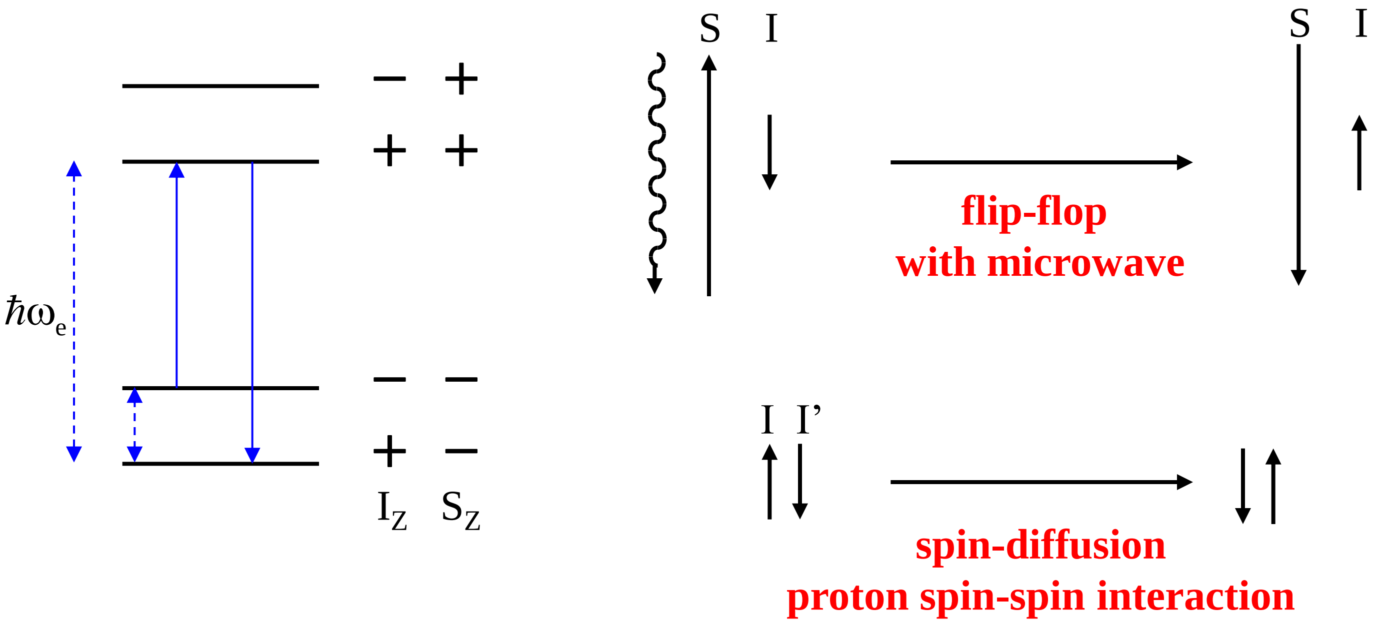
- High polarization of proton in organic materials was amazing.
- Electron spin resonance lines are so broad in polycrystal compared to the nuclear Larmor frequency, and it is difficult to separate the contributions from positive and negative solid effects.

- Microwave frequencies for positive and negative polarizations are far more separated than expected from the solid effect model.??
- Therefore, the phenomena could not be explained with the simple solid effect.



NMR Amplitude vs. Magnetic Field

Solid Effect : A. Abragam (Saclay), C. D. Jeffries(Berkeley)



The proton spin can be polarized with microwaves by means of spin flip-flop effect coupled with the electron spin, followed by spin diffusion due to proton spin-spin dipole interaction, if

$$N_I / T_{1n} \ll N_s / T_{1e} \qquad (N_I / N_s \sim 700)$$

Dynamics

Thermal mixing between two thermal reservoirs (Zeeman and spin-spin):

by rf irradiation,

Mixing through off-diagonal spin-spin interactions at high temperature:

cause an evolution towards a common spin temperature

Borghini CERN) : at Berkeley Conference on Polarized Target (1971)

The existence of "spin-spin interaction reservoir" separates from the other energy reservoirs of the overall system (lattice vibrations, Zeeman energy systems rf-field system etc.).

It is possible to "cool" this spin-spin reservoir, in particular the electron spin-spin interaction reservoir, by means of rf-transitions; then it is possible that this "cooling" be transmitted to nuclear spins present in the sample, thus increasing their polarization when they are located in strong magnetic fields.

That is named "dynamic orientation of nuclei by cooling of electron spin-spin interactions" (Donky effects).

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Borghini (cont'd)

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The cooling of the electron spin-spin interaction reservoir can occur in two ways;

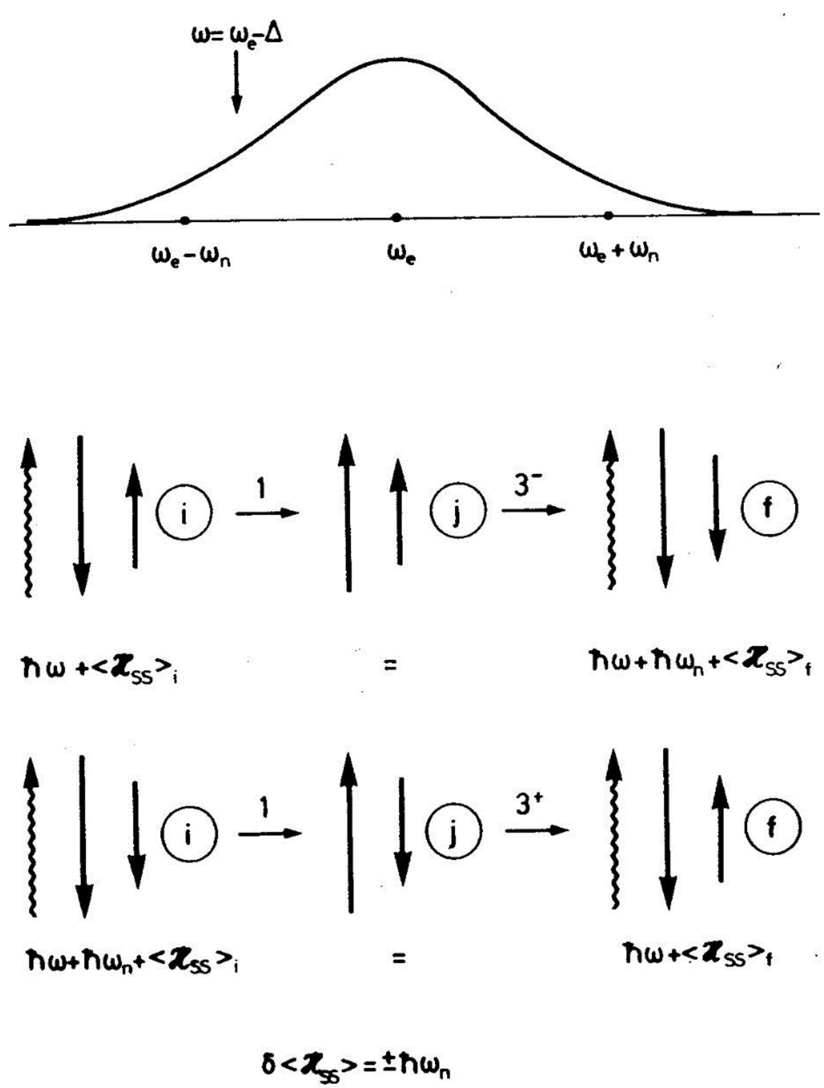
1. by saturation of one electron resonance line, slightly off-resonance ; then energy conservation in the rf field induced transitions requires a change in the electron spin-spin interaction reservoir temperature.

2. in systems with two (or more) discrete resonance line, by saturation one of these lines at exact resonance, while energy conservation in electron cross –relaxation, slightly off-resonance, requires a change in the spin-spin interaction reservoir temperature.

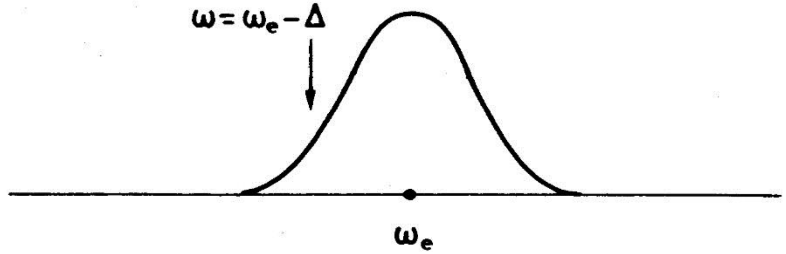
The thermal contact between the spin-spin interaction reservoir and the nuclear spins can occur through two different mechanisms:

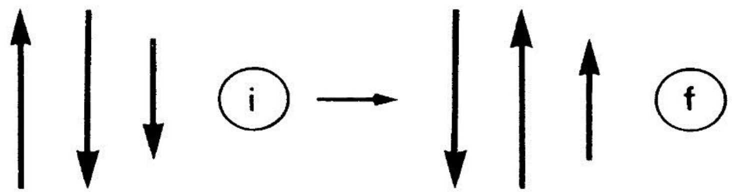
1.by forbidden transitions induced by the rf field responsible for the cooling of the spin-spin interactions.

2. by forbidden cross relaxation transitions, in which one nuclear spin flips together with two electron spins, as shown.

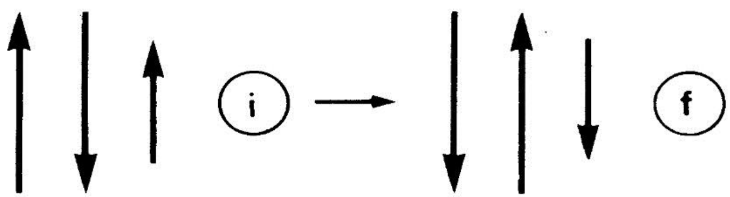


Mechanisms of Dynamic Orientation of Nuclei by Solid Effect
and
Cooling of Electron Spin-Spin Interaction



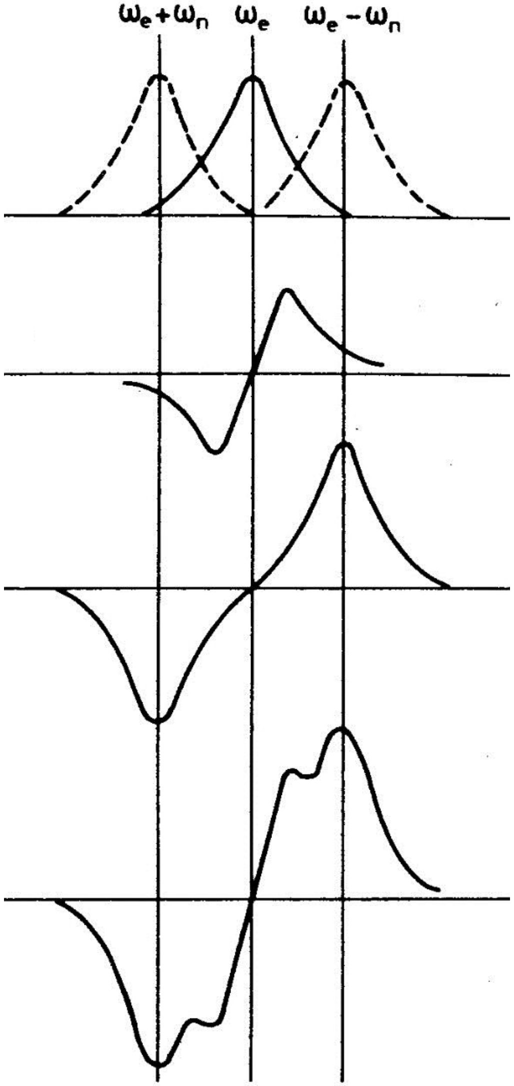


$$\hbar\omega_e + \hbar\omega_n + \langle \mathcal{H}_{SS} \rangle_i = \hbar\omega_e + \langle \mathcal{H}_{SS} \rangle_f$$



$$\hbar\omega_e + \langle \mathcal{H}_{SS} \rangle_i = \hbar\omega_e + \hbar\omega_n + \langle \mathcal{H}_{SS} \rangle_f$$

Mechanisms of Dynamic Orientation of Nuclei by Solid Effect
and
Cooling of Electron Spin-Spin Interaction



Transition probabilities

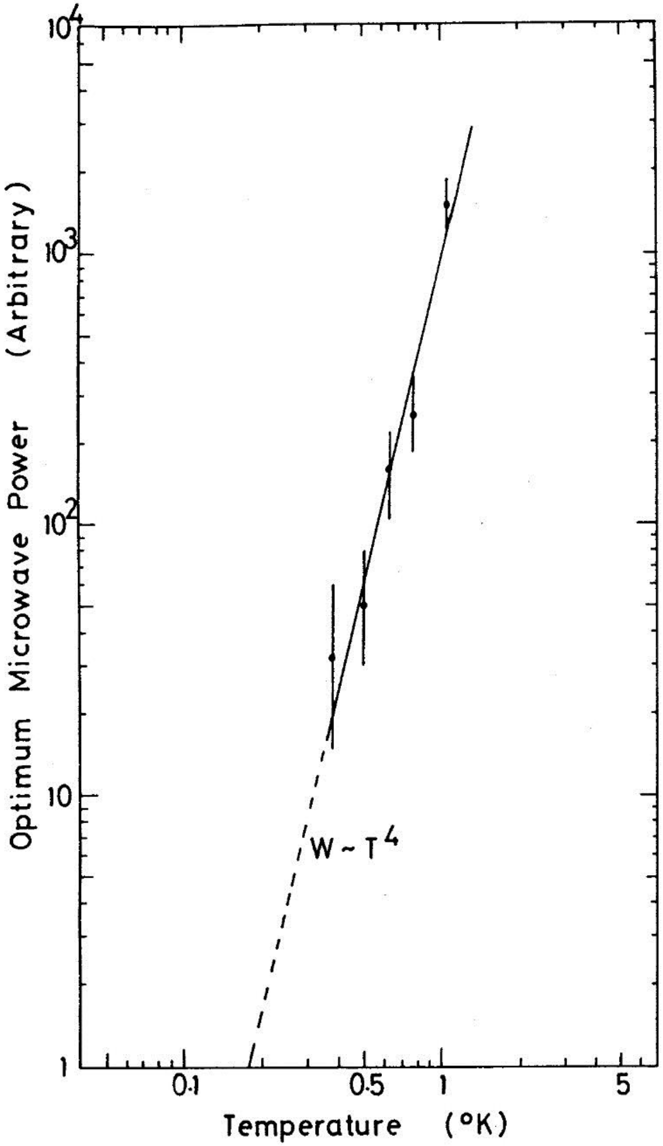
Cooling by Spin-Spin Interaction
(Donkey Effect)

Solid Effect

Solid Effect + Donkey Effect

Schematic View of the Mixture of Solid Effect and Donkey Effect

B

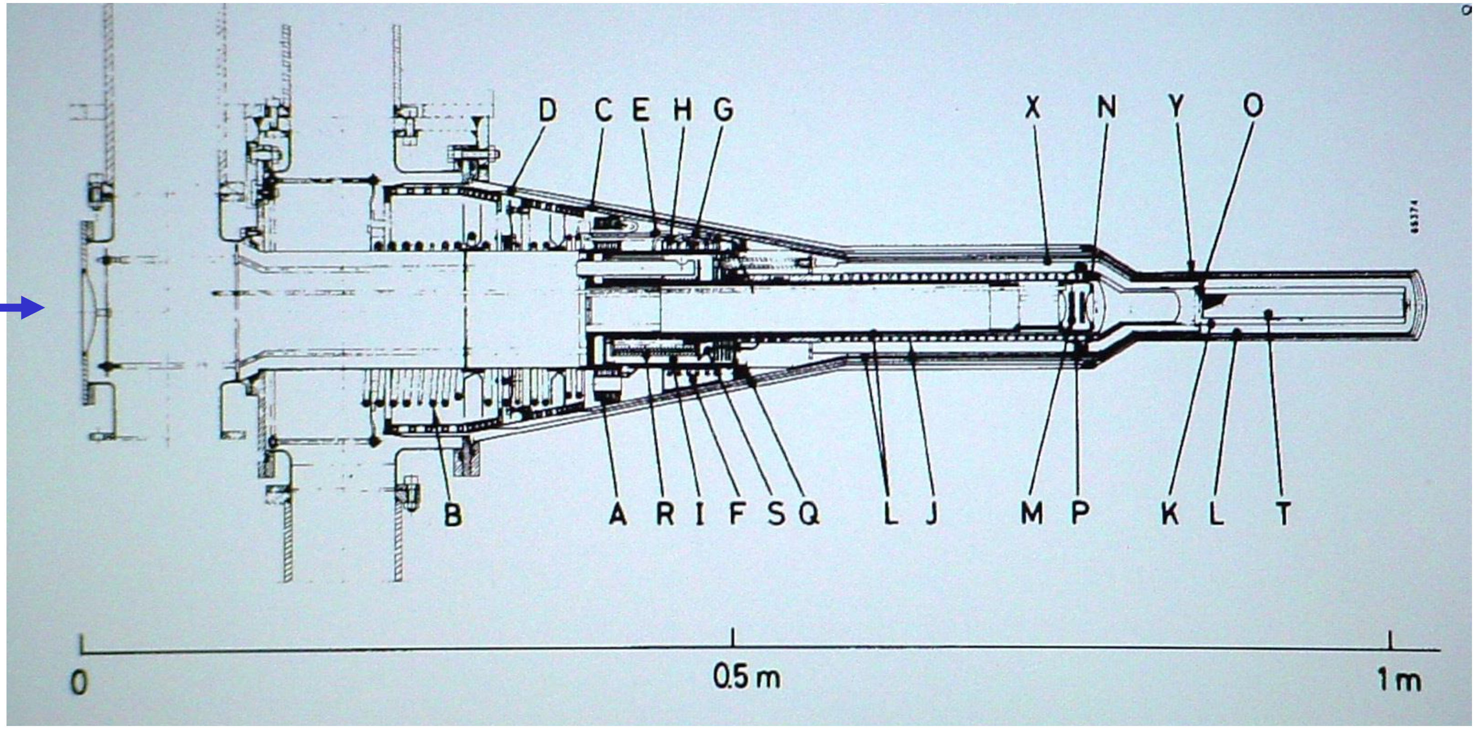


Temperature dependence of the microwave power absorbed in ethylene glycol

- The optimum microwave power at 0.4 K is 40 times less than that at 1 K.
- The optimum power is proportional to T^4, while the Kapitza resistance is proportional to $\sim T^{-3}$.
- Therefore, Kapitza resistance on the surface of solid material is not so serious even at 0.4 K.

• Spin frozen targets with dilution refrigerators were constructed at **CERN and KEK in 1974**, then Saclay, Dubna and Bonn a few years later.

Beam



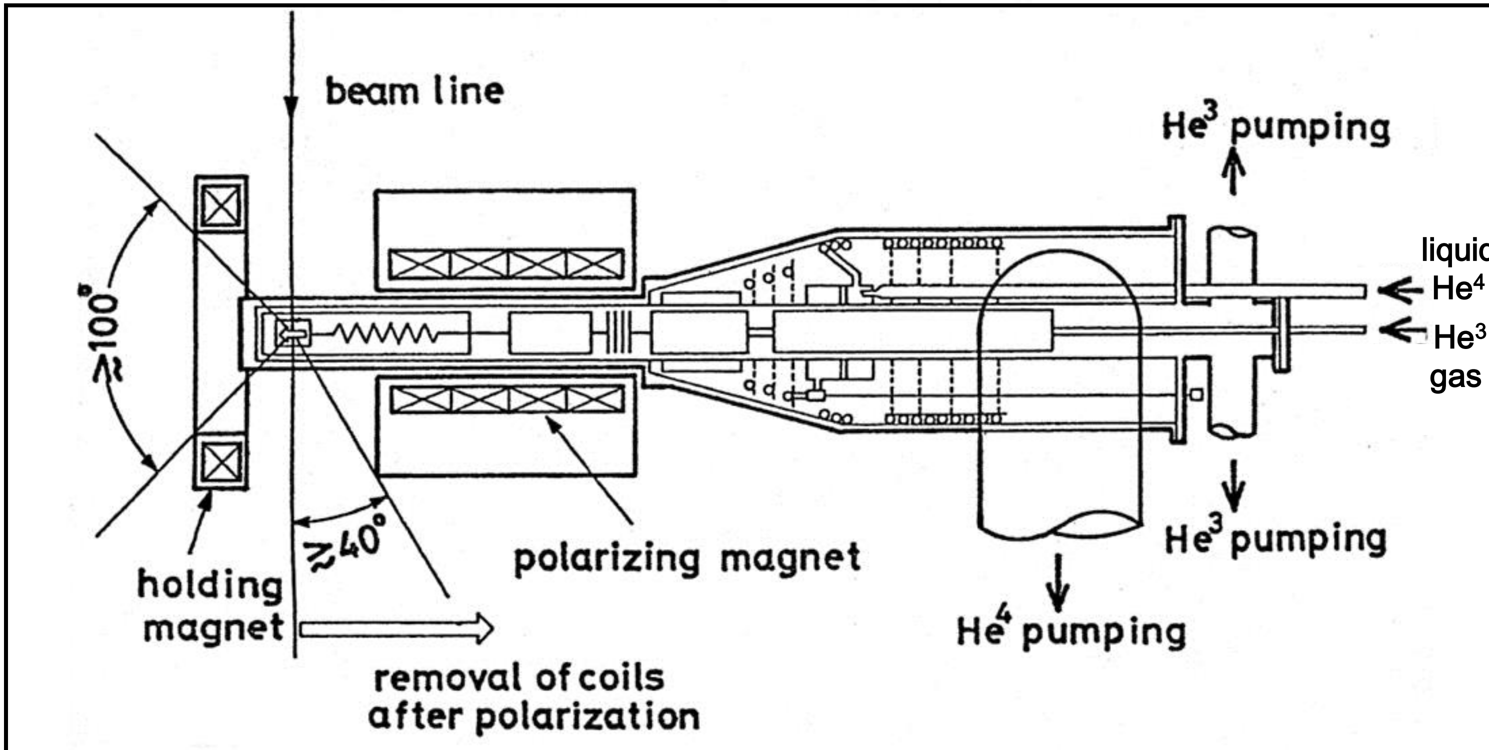
The Dilution Refrigerator for Spin Frozen Target at CERN T. Niinikoski

T < 50 mK B = 2.5 T **Propanediol polarization : 80 ~ 90 %**

CERN Courier Vol. 14 No. 11 (1974) 393-394

KEK
Frozen spin target

Hot (or cold) on the heels of the news of the successful operation of the frozen spin target at CERN, we have information from the National Laboratory for High Energy Physics (KEK) in Japan, where a frozen spin target is nearing completion. It is to be used for the direct measurement of scattering amplitudes of elastic and inelastic



Schematic View of the Spin Frozen Target at KEK

In 1974 a spin frozen deuteron target was constructed at KEK
for the reactions

$$K^+ + n \rightarrow K^+ + n\ ,\ K^0 + p$$

in order to search for 5 quark systems.

Target material was deuterated propanediol doped with Cr^{5+} complex.

Typical deuteron polarization was 40 % with frequency modulation.

The temperature to keep the polarization was less than 60 mK and
the relaxation time was more than 20 days.

Such targets could be used for various experiments with wide access angle.

Actual Hydrogen Deuteride (HD) Targets

~ 2000: Grenoble - Orsay:

- ♦ Static Polarization at 10 mK and 13.5 T
  - ▪ Proton polarization : $\geq$ 60 %
  - ▪ Deuteron Polarization : $\geq$ 14 %
  - ▪ Small concentration of ortho H_2 and para D_2

~2000: BNL (LEGS): Polarizing proton to 70% and deuteron to 17%
at 17mK and 15T, and holding at 1.25K and 0.7T

~2002: (Bochum): Trial of DNP with free radicals of ~10^{18} spins/cm^3
produced by ^{90}Sr.

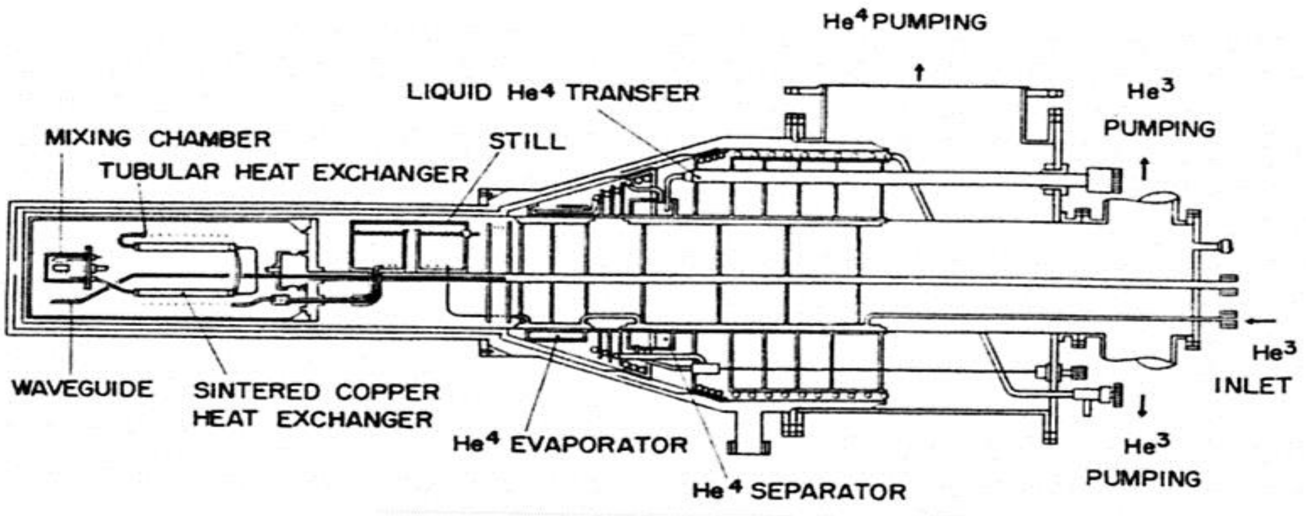
scattering up to large momentum transfers (the measurement of spin rotational parameters as well as polarization parameters) on the 10 GeV synchrotron now under construction. The target work is led by A. Masaike who invented the helium 3 cryostat for polarized targets.

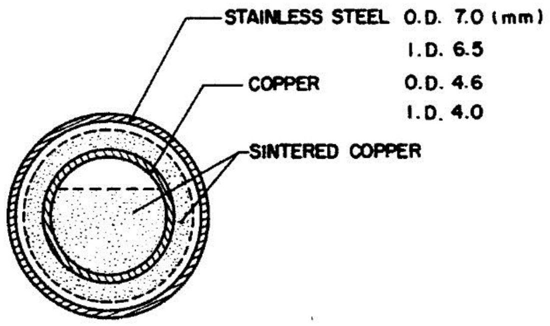
In the KEK frozen spin target, protons and deuterons in organic materials are polarized in a magnetic field of 2.5 or 5 T with good homogeneity at about 0.3 K and are then used in a beam while at a lower temperature of 0.1 K and in a lower magnetic field. The spins of the proton and the deuteron can be kept polarized at such a low temperature for a long time. For example, the relaxation time of protons in propanediol is about two weeks at 0.1 K in 1.5 T and therefore, a field of 1.5 T with a homogeneity of 2×10^{-2} is adequate for holding the polarization. It is then comparatively easy to make such a magnetic field with a large access angle. Moreover, the lower magnetic field makes it easier to bring in the beam and to detect the scattered particles.

The frozen spin target at KEK is designed to be used in horizontal or vertical configurations. The target material is polarized in a solenoid coil giving a field of 2.5 T for the proton and 5 T for the deuteron in a horizontal dilution refrigerator. The polarizing coil, which is 35 cm long, is moved horizontally about 25 cm to free the target for the beam. The holding field is the fringe field of the polarizing coil plus a small auxiliary coil adjusted to give a homogeneous field at the target. This gives good access for studying scattering and the auxiliary holding coil can be changed to meet the requirements of different experiments.

A high power horizontal dilution refrigerator of He^3-He^4 and a horizontal superconducting solenoid coil were successfully operated in August. The precooler and the still of the refrigerator are larger versions of the cryostat built by T. Niinikoski for CERN (see vol. 11, p. 353). There is enough cooling power near 0.1 K for holding, and 0.3 K for polarizing, by means of special heat exchangers. The flow rate of He^3 is about 3×10^{-4} mol/s or higher at about 0.1 K and the cooling time from 1 K to 0.1 K is about 15 min. Detail of the cryostat design is to be published by K. Amako, S. Ishimoto, A. Masaike and K. Morimoto.

Schematic View of Horizontal Dilution Refrigerator for Spin Frozen target at KEK





Coaxial Double Heat Exchanger
Filled with Sintered Copper

B

LiH and LiD : high dilution factor (f ~ 0.5)

1959 : Abragam used ^{6}LiF to validate the Overhouser Effect.

1969 (Saclay) : $^{6}Li^{19}F$ was used for studying anti-ferromagnetism.*

1980 (Saclay) : ^{6}Li polarization of 70 % in ^{6}LiD at 6.5 T, 200mK irradiated with 10^{17} – 10^{18} e^{-}/cm^{2} in liquid Ar.

1988 (PSI) : ^{6}Li polarization of 53 % in ^{6}LiD at 2.5 T.

~ 1990s (Bonn) : The best condition for ^{6}LiD was found.

Ammonia Polarization*

- Ammonia is advantageous because of its high dilution factor.

  f : polarizable nucleons / to total number of nucleons

  f (LMN) : 0.03

  f (butanol) : 0.135

  f (NH_3) : 0.176

- Handling of ammonia is not easy.

- In early trial of dynamic polarization, 40 % polarization was obtained in NH_3 doped with glycerol Cr^{5+} complex at 1 K and 2.5 T at CERN. (K. Scheffler, M. Borghini)

- Polarization of 70 % was obtained with ethanediol Cr^{5+} complex in 0.5 K and 2.5 T.

- Difficulties were in reproducibility of DNP and slow growth of polarization.

Radiation-doped Ammonia

1979 (CERN): Irradiation of 0.95×10^{15} protons/cm^2 in liquid N_2

- Polarization of 90~93 % was obtained at < 0.5 K and 2.5T.
  Further proton irradiation led to explosions.

1980 (Bonn) : Irradiation of 10^{17} e^-/cm^2 in liquid argon (~ 90 K).
(W. Meyer)

proton : ≥ 90 % at 0.3 K and 2.5 T

deuteron : ~ 40 % with additional irradiation at 1 K.

- Short polarization build up time
- Very strong for radiation damage
- Chemically stable for more than a year
- No problem of explosion

Ammonia irradiated in liquid argon became a standard polarized target.

COMPASS Target

^{6}LiD : 350 gr. (largest)

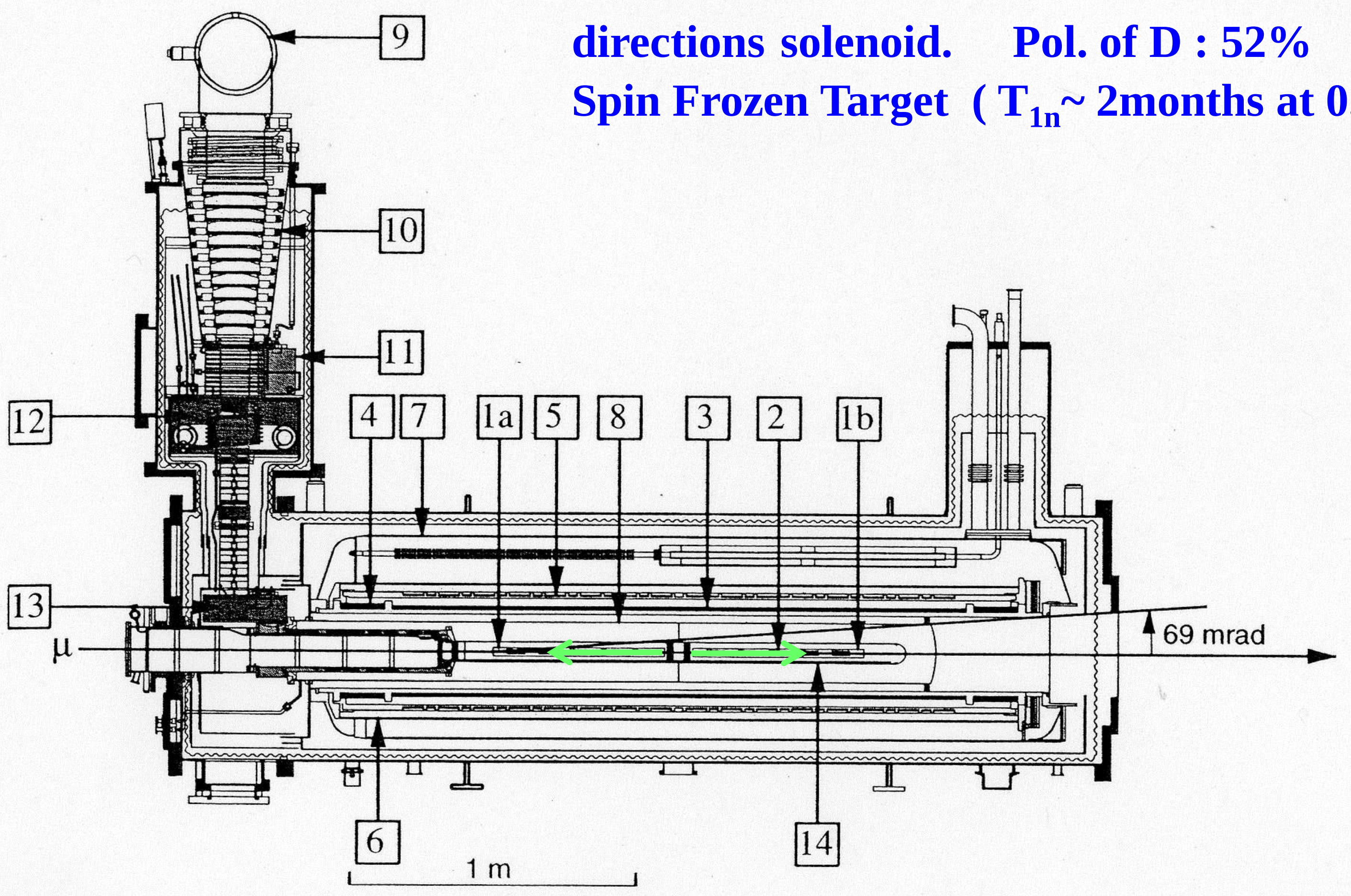
- Irradiated with 20 MeV electrons in Bonn
- Microwave frequency : 70 GHz
- Polarizing at ≤ 300 mK in a dilution refrigerator
- Polarization of deuteron is ~ 52 % with frequency modulation.
- Two targets were polarized in the opposite directions, in order to cancel the false asymmetry.
- holding at ~ 60 mK
- T_p = 1400 hrs. at 0.42 T

PSI Polarized Target for n-p Scattering

- butanol-water with Cr^{5+} : 100 gr
- polarizing at 5T in a dilution refrigerater
- holding at 0.8T and 50mK

COMPASS Target at CERN (^{6}LiD : 350 gr.)

Irradiated with 20 MeV electrons in Bonn
Two targets were polarized in the opposite directions solenoid. Pol. of D : 52%
Spin Frozen Target (T_{1n}~ 2months at 0.42T)



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Hydrogen Deuteride (HD) Target

~ 2000: Grenoble - Orsay:

♦Brute Force Method

- Proton polarization : ≥ 60 %
- Deuteron Polarization : ≥ 14 %
- Polarizing at 10 mK and 13.5 T
- Small concentration of ortho H_2 and para D_2 are necessary for polarization.

~2000: BNL (LEGS): Polarizing proton to 70% and deuteron to 17% at 17mK and 15T, and holding at 1.25K and 0.7T

~2002: (Bochum): Trial of DNP with free radicals of ~10^{18} spins/cm^3 produced by ^{90}Sr.

LiH and LiD : high dilution factor (f : 0.5)

1959 : Abragam used ^{6}LiF to validate the Overhouser Effect.

1969 (Saclay) : ^{6}Li^{19}F was used for studying anti-ferromagnetism.

1978 : LiH polarization irradiated with electrons was proposed.
Polarizations of 80 % for **^{7}Li** and 35 % for **^{6}Li** were obtained at 6.5 T, 200 mK, in agreement with the spin temperature model.

1980 (Saclay) : ^{6}Li polarization of 70 % in ^{6}LiD at 6.5 T irradiated with 10^{17} – 10^{18} e^-/cm^2 in liquid Ar.

1988 (PSI) : ^{6}Li polarization of 53 % in ^{6}LiD at 2.5 T.

~ 1990s (Bonn) : The best condition for ^{6}LiD was found.

Neutron Polarization with Polarized Proton Filter

E. Fermi in "Nuclear Physics"

- Since $\sigma_{\uparrow\downarrow} > \sigma_{\uparrow\uparrow}$ for n-p scattering, slow neutron with spin down (anti-parallel to proton) will be scattered away, leaving a transmitted beam which is predominately spin up (parallel).

- However, this method is **not feasible**, since temperatures on the order of 0.01 K would be necessary to line up the nuclear spin.

$$P_n \cong \tanh(P_p\, n\, \sigma_p\, t)$$

$$T_n \cong T_0 \cosh(P_p\, n\, \sigma_p\, t)$$

- In spite of Fermi's comment, slow neutrons could be polarized by the polarized proton target, thanks to the dynamic polarization.

- Very low energy neutron beam can be polarized by transmission through polarized target, since $\sigma_{np}(\uparrow\downarrow) \geq \sigma_{np}(\uparrow\uparrow)$ for low energy neutrons. It is applicable to particle physics.

- Spin dependence of the interaction of proton and unstable nuclei has been measured for the first time.

- Applicable to quantum computing using NMR because of the high temperature and low magnetic field.

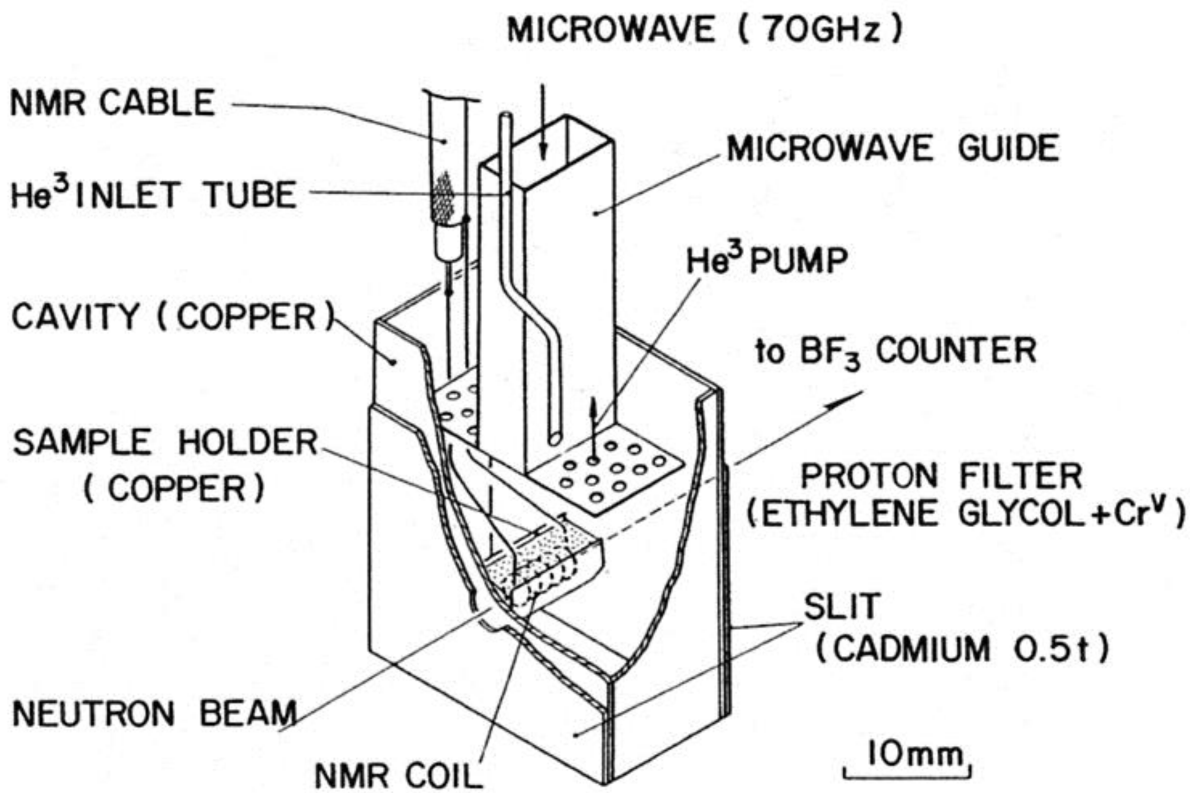
- Useful to study a high resolution NMR system; It could allow nuclear magnetic ordering in a low field at high temperature.

- Possible study of the structure of biopolymers.

1960s: F. L. Shapiro, then V. I. Lushchikov obtained
a polarized neutron beam with LMN at Dubna.

1970s: KEK group got thermal neutron polarization of more than
85% with propanediol using a ^{3}He cryostat.

1980s: KEK, TIT and Los Alamos groups used polarized neutron
beams in wide energy range (0,02 ~ 1 eV), using propanediol with
Cr^{5+}, which have contributed to the parity violation experiments
at KEK and Los Alamos in 1980s ~ 1990s.



at KEK

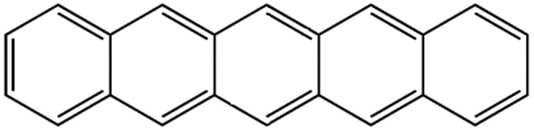
Schematic view of a microwave cavity containing propanediol which was used as polarized proton filter for slow neutron beams*

B

DNP at high temperature and low field

DNP on a photo-excited triplet state of **pentacene molecules**

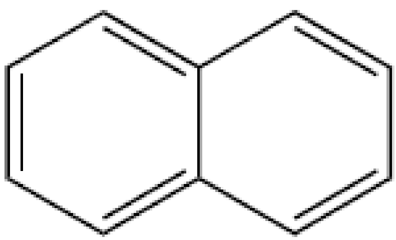
- Electron spins are spontaneously aligned on the triplet state **independent of temperature and magnetic field.**
- Relaxation time of proton is very long on the ground state.



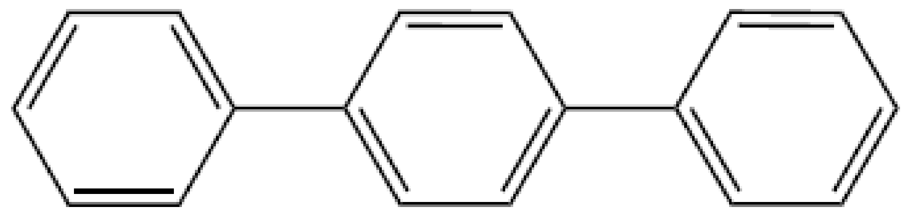
Efficient polarization transfer (Integrated Solid Effect (ISE))

in low magnetic field at high temperature
by sweeping the magnetic field

Pentacene molecules are doped in the crystals of naphthalene and p-terphenyl with concentrations of 0.001~0.018 mol% and 0.1 mol%, respectively.



(a) naphthalene



(b) p-terphenyl

	$C_{10}H_8$	$C_{18}H_{14}$
molecular weight :	128.2 g / mol	230.3 g / mol
density :	1.16 g / cm^3	1.24 g / cm^3

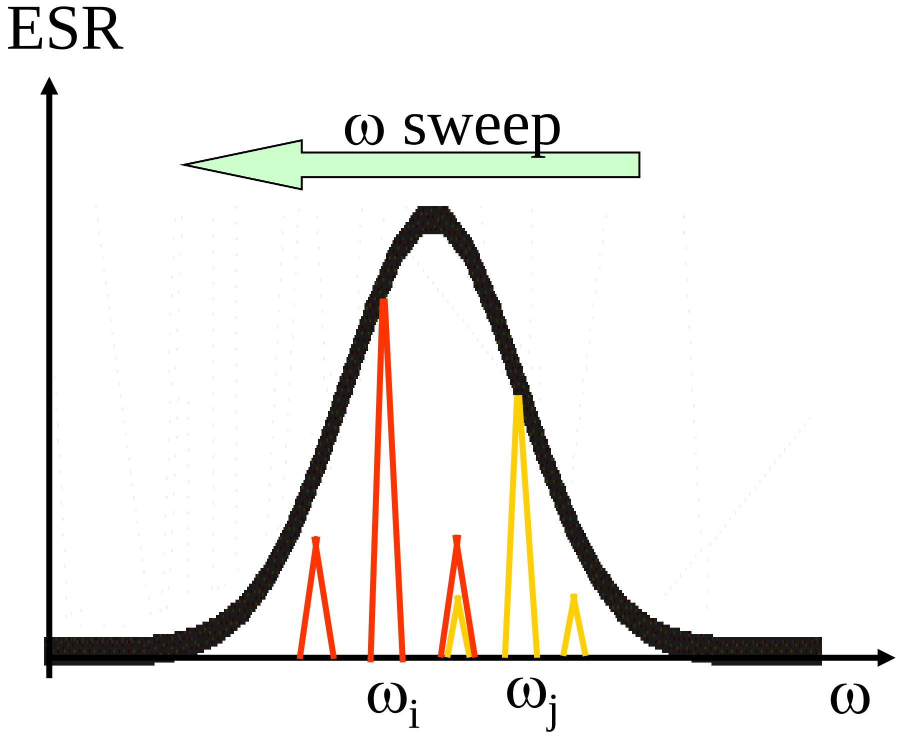
- Van Kesteren obtained proton polarization of 42 % in a crystal of fluoranthene ($C_{16}H_{10}$) involving the photoexcited triplet state of the guest phenenthrene molecule by means of MIONP at 1.4 K and 2.7 T.

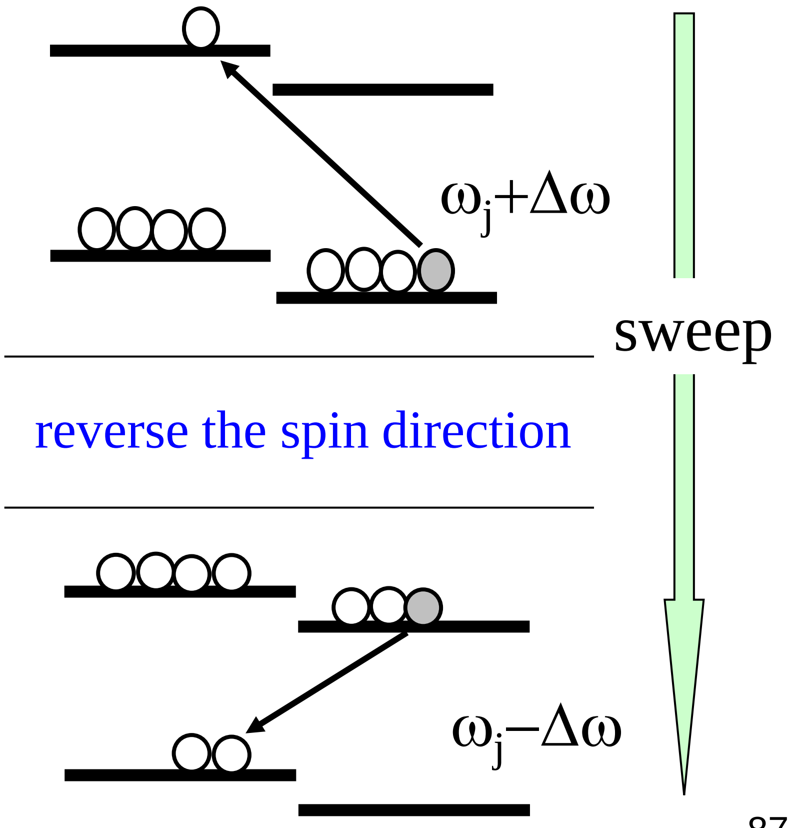
- However, polarization was significantly reduced above 1.4 K.

- DNP efficiency is significantly reduced, when width of ESR is larger than the energy difference between magnetic sublevels of the nucleus in low magnetic field.

Integrated Solid Effect (ISE)

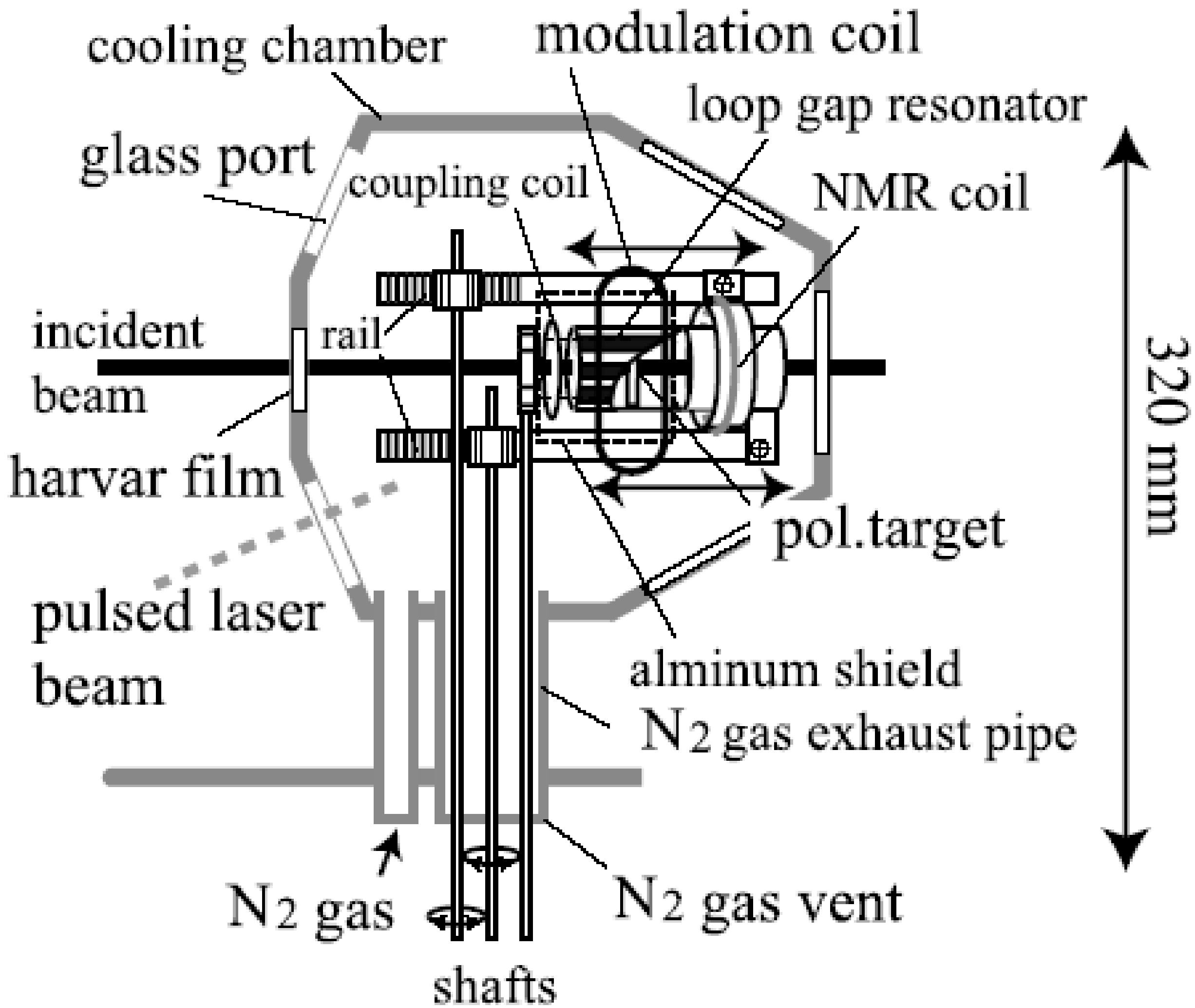
Magnetic field is swept during the microwave irradiation in order to integrate all the solid effects.





Polarized Naphthalene Target for ^{6}He + p Scattering

(Riken-Univ. of Tokyo)



B

High Energy Polarized Proton Beam

There were no high energy polarized beam in 1960's .

Polarized ion sources were developed for low energy nuclear physics experiments at the beginning of 1960's.

Atomic beam type $\rightarrow$ Adiabatic Passage

Lamb shift type for negative ions

First polarized proton beam was successfully accelerated up to 12 GeV/c in Argonne National Laboratory (US) in 1970, using of fast passage method through the depolarization resonances.

Several experiments of the di-baryon search and p-p scattering at high pt were performed with the polarized beam and polarized targets from the beginning of 1970s.

The Siberian snake method was proposed at the end of 1970's for passing through depolarization resonances.

- He was the representative of Monaco to the United Nations in New York at the beginng of 21st Century, when I was the representative of JSPS, Japan.