Polarized Targets, Polarizable Materials and Dynamic Nuclear Polarization

Donal Day
University of Virginia

HUGS 2007

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Newport News, VA

Motivation

- hadron-hadron interactions

- with polarized beams, nucleon-nucleon interactions, time reversal invariance

- pions to study the spin-dependent part of interactions and discriminate among theories

- perturbative and non-perturbative regimes

- DIS to measure the spin content of the proton carried by quarks

- with real photons to study the electromagnetic properties of the baryon resonances
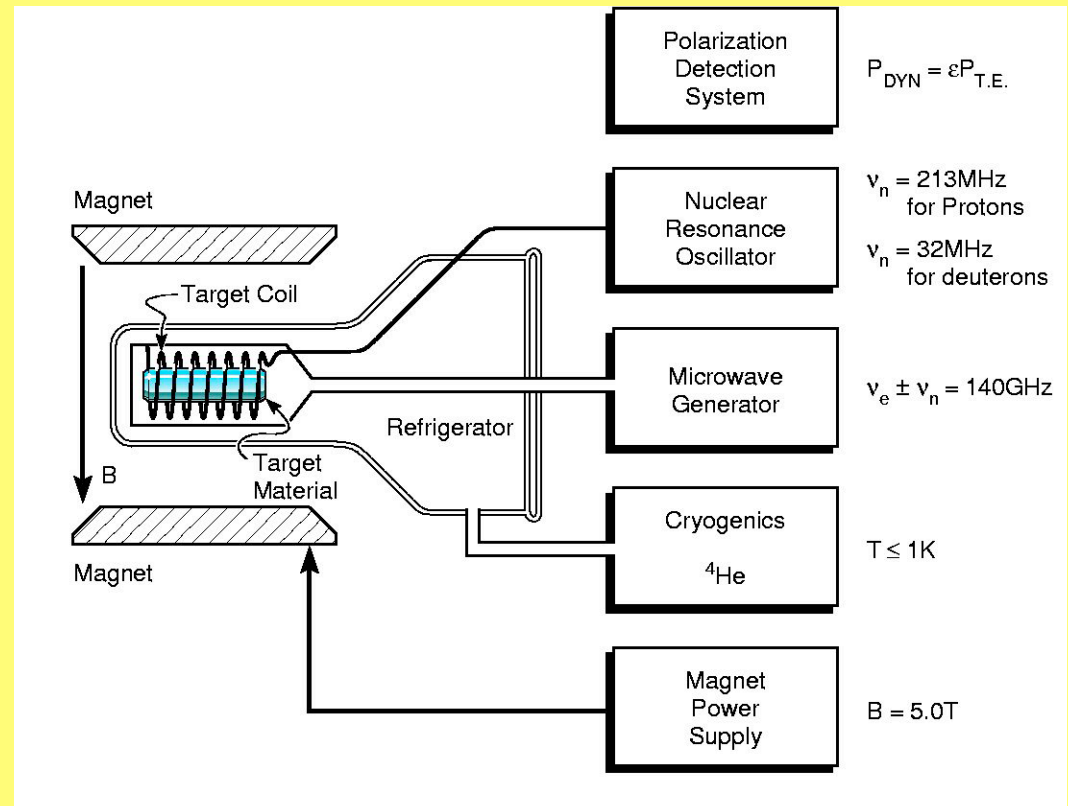
- nucleon form factors

Bonn, Mainz, MIT-Bates, NIKHEF, JLAB, CERN

Techniques

- Dynamic Nuclear Polarization

- Frozen Spin (FROST)

- Brute Force (HD)

- ^{3}He targets

Dynamic Nuclear Polarization

- Refrigerator – 0.5K to 1 K

- Magnetic Field – 2 T to 6 T

- Microwaves – 55 GHz – 165 GHz

- NMR system

- DAQ

Magnet

Target Coil

B

Target Material

Magnet

Refrigerator

Polarization Detection System

$P_{DYN} = \varepsilon P_{T.E.}$

Nuclear Resonance Oscillator

$\nu_n = 213 MHz$ for Protons

$\nu_n = 32 MHz$ for deuterons

Microwave Generator

$\nu_e \pm \nu_n = 140 GHz$

Cryogenics ^4He

$T \leq 1K$

Magnet Power Supply

$B = 5.0T$

Polarization

protons 70% – 100%
deuterons 20% – 50%
and higher

- Crystal – LMN

- Alcohols – propane-diol, ethane diol, butanol

- Ammonia

Paramagnetism

Substances which possess a permanent magnetic dipole moment

a) atoms, molecules possessing and odd number of electrons, i.e. $S \neq 0$ NO gas, free Na atoms, organic free radicals

b) Free atoms & ions with partly filled inner shell; transition elements; rare earth & actinide elements Mn^{2+}, Gd^{3+}, U^{4+}

c) Metals

d) Few miscellaneous compounds O_2, organic biradicals

Assembly of paramagnetic atoms (one unpaired electron).
Dilute enough for no magnetic interaction between atoms

Apply external magnetic field
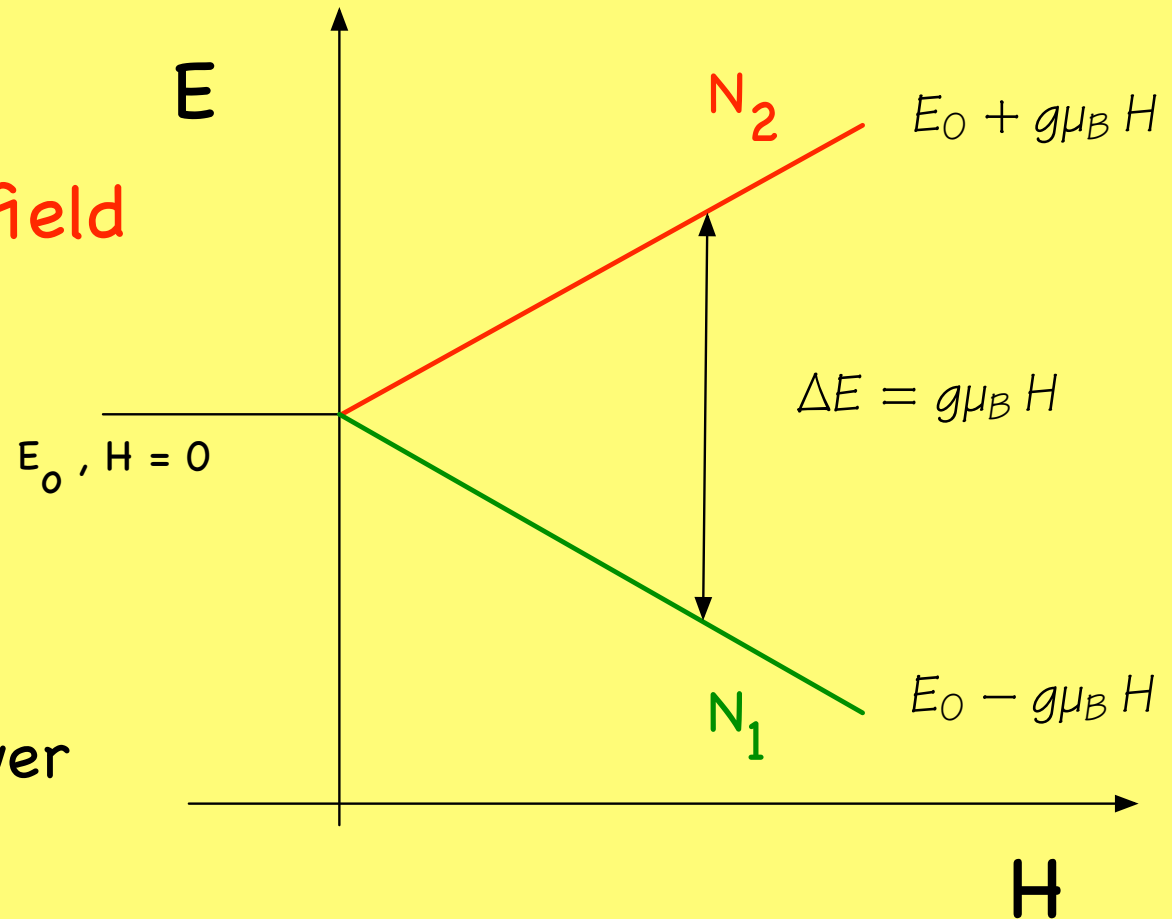
Zeeman Splitting

N_1 atoms || magnetic field (lower
energy state) ($m_s = -1/2$)

N_2 atoms anti-|| magnetic field (higher
energy state) ($m_s = +1/2$)

E

N_2

$E_O + g\mu_B H$

$\Delta E = g\mu_B H$

E_o , H = 0

N_1

$E_O - g\mu_B H$

H

$N_1 > N_2 \therefore$ net magnetization || field

Thermal agitation $\Rightarrow$ random orientation

Lower temperature $\Rightarrow$ less agitation

$\Rightarrow N_1 > N_2 \Rightarrow$ larger magnetization

If we let H >> and T <<, eventually $N_2 \Rightarrow 0$

At thermal equilibrium (spin 1/2)

$$\frac{N_2}{N_1} = exp\left[-\frac{g\mu_B H}{kT}\right]$$

$$Polarization \;=\; \frac{N_1 - N_2}{N_1 + N_2}$$

$$=\; tanh\left[\frac{\mu_B H}{kT}\right]$$
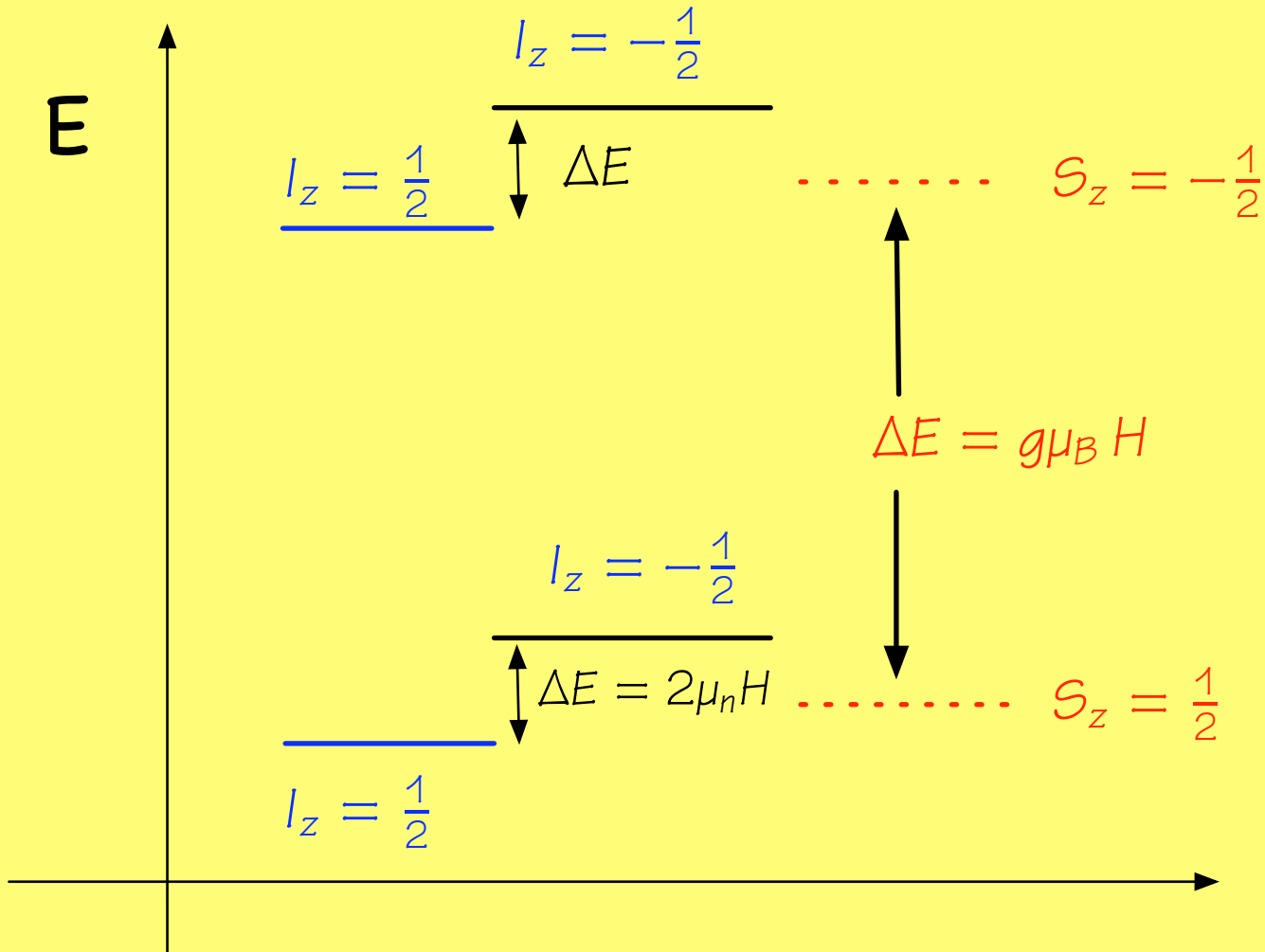
Proton

(g=2)
For H = 2.5T, T = 0.5 K
P = 0.998

Nucleons and nuclei have magnetic moments too

However $\dfrac{\text{nuclear moments}}{\text{electron moments}} \simeq 10^{-3}$

Include nuclear moments $\Rightarrow$ hyperfine splitting in magnetic field

$I_z = -\dfrac{1}{2}$

E

$I_z = \dfrac{1}{2}$ ΔE $\cdots\cdots$ $S_z = -\dfrac{1}{2}$

$\Delta E = g\mu_B H$

$I_z = -\dfrac{1}{2}$

$\Delta E = 2\mu_n H$ $\cdots\cdots\cdots$ $S_z = \dfrac{1}{2}$

$I_z = \dfrac{1}{2}$

Analogous to the electron

$$Polarization = tanh \left[\frac{\mu_n H}{kT} \right]$$

μ_n = proton magnetic moment

Again for H = 2.5T, T = 0.5 K, P = 0.005!

So very small polarizations for the static case.

$\Downarrow$

Solution is: RF Induced Transitions

E

I_z S_z

-1/2 +1/2

+1/2 +1/2

ν_n

ν_e

ν_e

ν_e

ν_n

-1/2 -1/2

+1/2 -1/2
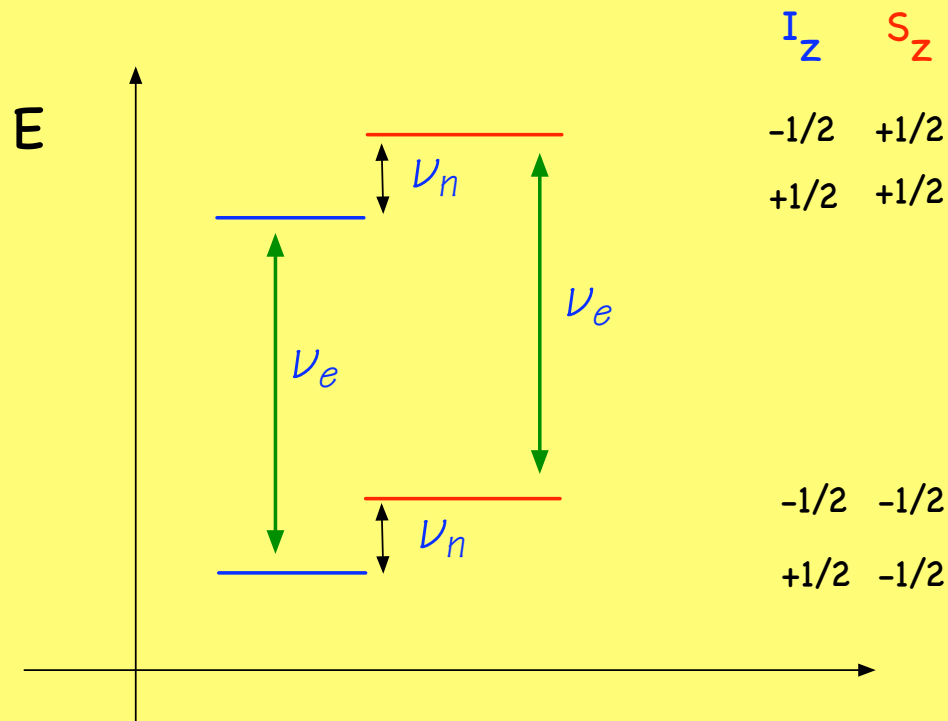
RF fields can be used to induce transitions between spin states

Allowed Transitions

ΔI_z, $\Delta S_z = 1$

1) $\nu_{RF} \sim \nu_e$ get transitions:

$-- \leftrightarrow -+$

$++ \leftrightarrow +-$

where $\Delta E = g\mu_B H = h\nu_e$ and at 2.5 T ν_e = 70 GHz

Pure electron spin transitions $\uparrow \rightarrow \downarrow$ is electron paramagnetic resonance EPR (or ESR)

E

	I_z	S_z
	-1/2	+1/2
	+1/2	+1/2
	-1/2	-1/2
	+1/2	-1/2
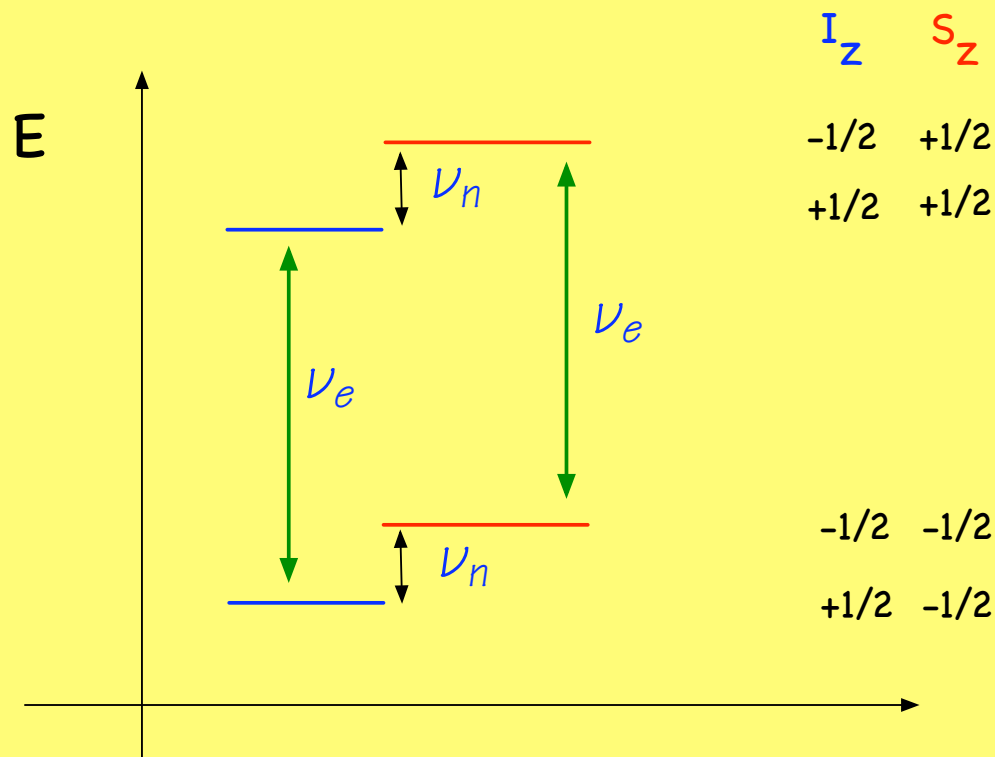
2) $\nu_{RF} \sim \nu_n$ get transitions: +- ↔ --

++ ↔ -+

Here $\Delta E = 2 \mu_n H = h\nu_n$

and $\nu_n = 106.5$ MHz at 2.5 T

Pure nuclear spin transitions

Nuclear Magnetic Resonance ↑ → ↓

(NMR)

But the population of these states are essentially equal and the transitions do not modify them.

Forbidden Transitions

Transitions between -- and ++ and +- and -+ are forbidden. Unfortunately these are the transitions we want.

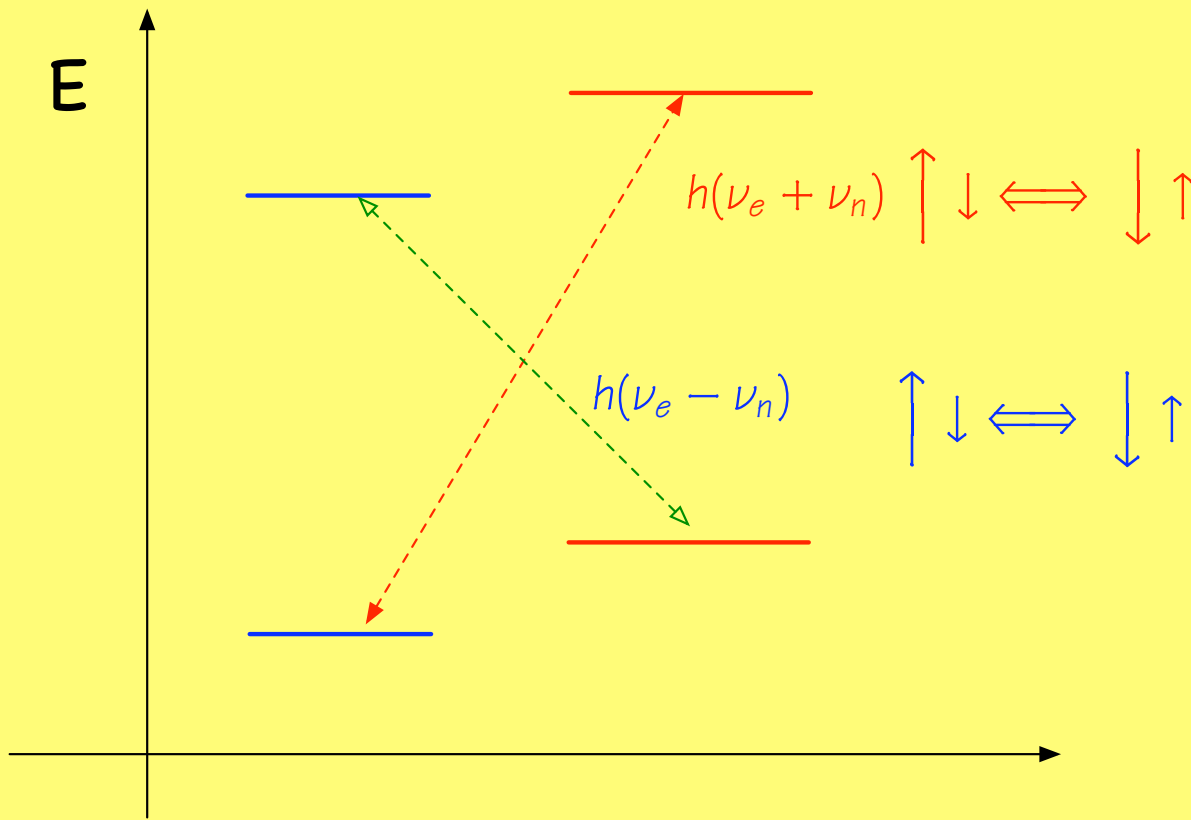## However there exists the dipole-dipole interaction

I_z S_z

E

NMR

EPR

EPR

Forbidden

Forbidden

NMR

-1/2 +1/2

+1/2 +1/2

-1/2 -1/2

+1/2 -1/2
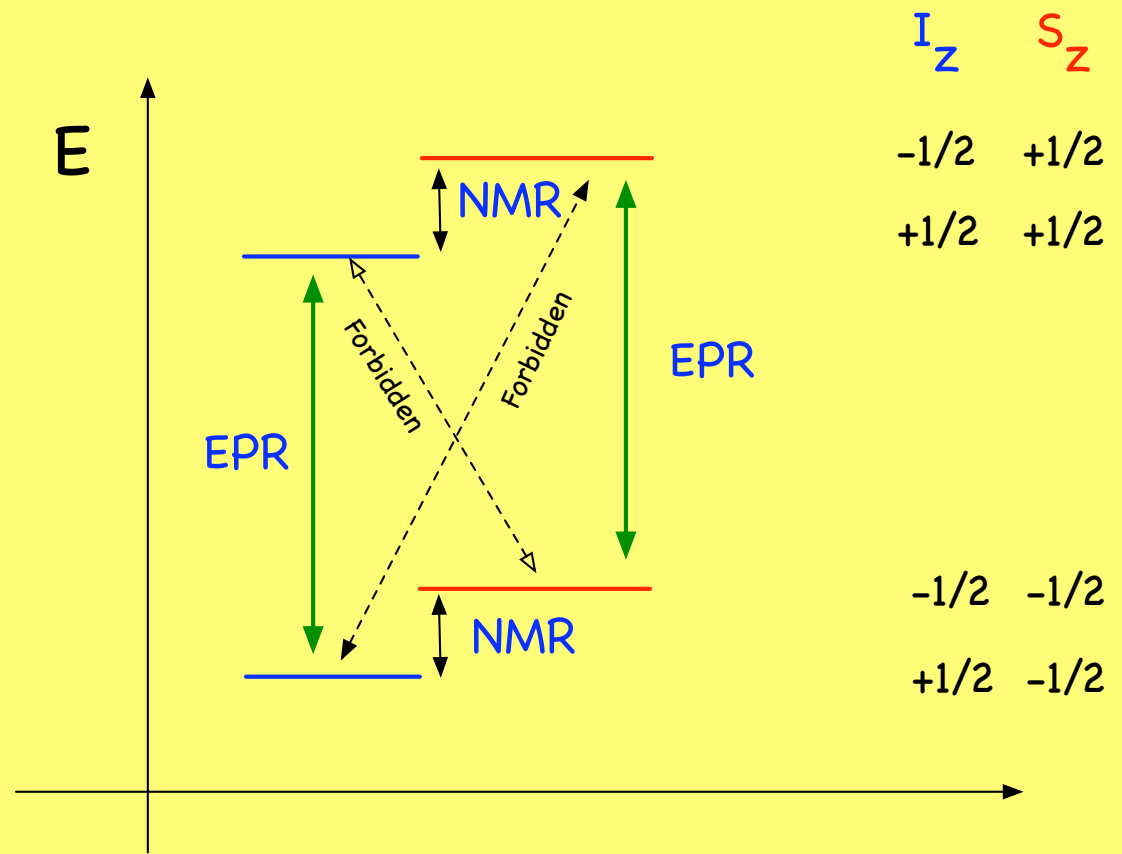
Two distant magnetic moments → dipole-dipole interaction << Zeeman.

This leads to a slight mixing of the nuclear states which then allows transitions of the type +- ↔ -+ and ++ ↔ -- though with a probability much less (10^{-4}) than allowed transitions.

E

$$h(\nu_e + \nu_n) \quad \uparrow \downarrow \Longleftrightarrow \downarrow \uparrow$$

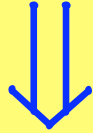$$h(\nu_e - \nu_n) \quad \uparrow \downarrow \Longleftrightarrow \downarrow \uparrow$$

The rf field can drive the forbidden transitions which can be visualized as

flips-flops $\quad \uparrow \downarrow \Longleftrightarrow \downarrow \uparrow$

flips-flips $\quad \uparrow \uparrow \Longleftrightarrow \downarrow \downarrow$

$$\Delta(S_z + I_z) = 0, 2$$

An objection could be raised that there are only a small number of electron spins – when they all have been pumped up the polarization is still small

$\Downarrow$

Spin-Lattice relaxation

A radiationless process by which energy is exchanged between the spin and the lattice (thermal motions of the solid)

Spin polarization -> value corresponding to TE at lattice temperature

Electrons Spins

Strong interaction with the lattice TE achieved very rapidly. The processes amount to electron spin flips of the type $\uparrow \longrightarrow \downarrow$
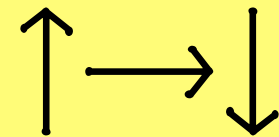
These are fast: ~ one transition / millisecond

Nuclear Spins

Generally weakly coupled to lattice - therefore slow
~ one transition / minute

The difference in the relaxation rates between electron spins
and nuclear spins is crucial to the polarization process

Real Target Material

We have the beginnings of a mechanism for polarizing nuclear (nucleon) spins but ... we have to consider what targets are useful for particle physics

1. Interactions with nucleons are investigated in most experiments

2. Need a high density of nucleons

   a) polarized atomic hydrogen – low density

   b) molecular hydrogen – high density but no polarization

   c) Hydrogenous materials, eg CH_2, NH_3, containing as high a ratio of H/other as possible

3. We still must polarize

In general the materials of (2c) are NOT paramagnetic but

Paramagnetic centers can be doped into bulk target material (chemical or by radiation doping)

So if we have some target material containing free protons with a "dilute" doping of paramagnetic centers we can consider a mechanism by which the protons can be polarized.

The (Resolved) Solid-State Effect

The first polarized target: hydrated Lanthanum Magnesium Nitrate (LMN) $La_2Mg_3(NO_3)_{12} \cdot 24H_2O$ + 0.2% neodymium

Initial conditions: low temperature, strong magnetic field, then P_e = -100% and P_n = 0.

Apply rf at frequency $\nu = \nu_e + \nu_n$. It produces flip-flops but no flip-flips (energy not conserved)

Apply rf at frequency $\nu = \nu_e + \nu_n$. It produces flip-flops but no flip-flips (energy not conserved)

$$\downarrow \uparrow \Longleftrightarrow \uparrow \downarrow$$

but the electron spin rapidly goes back to $\downarrow$ while the nuclear spin with its much longer relaxation time stays $\uparrow$ and is no longer affected by the rf field.
Eventually $P_n = P_e = -100\%$

In fact P_e may not be 100% because of other relaxation effects and $P_n \leq P_e$.

Similarly: If $\nu = \nu_e - \nu_n$, then $P_n = +100\%$.

One could object that this reasoning applies only to those nuclei in the immediate vicinity of the paramagnetic centers. Forbidden transition probability decreases very rapidly with distance -> most nuclei would not become polarized in a finite time.

Spin Diffusion

Neighboring nuclei coupled by dipole-dipole interaction which produces energy conserving processes such as

↑ ↓ ⟺ ↓ ↑
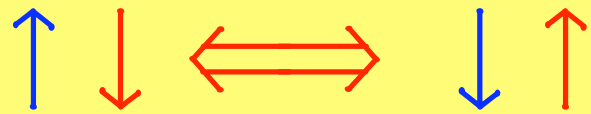
Very frequent ~ 10^4/sec and nuclear polarization is transported throughout the sample

or nuclear ordering near nuclear spins is transmitted to all nuclei

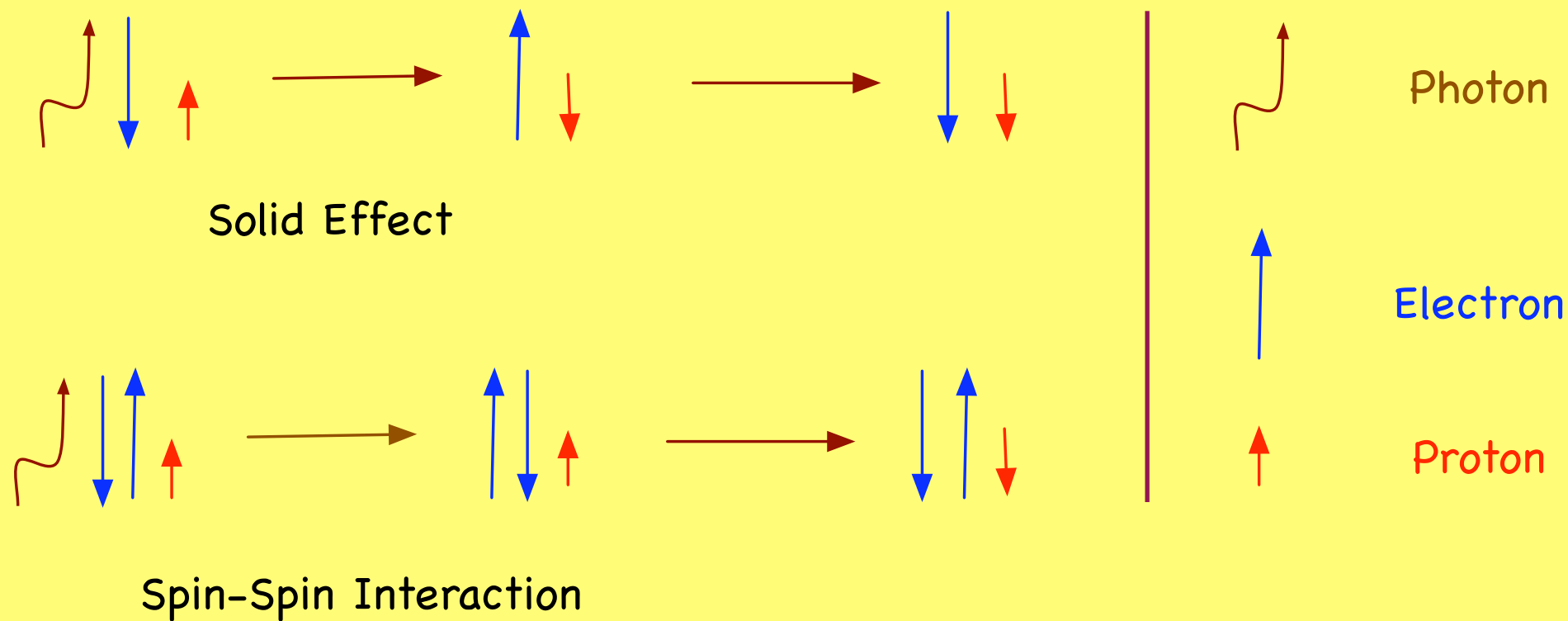Assumption for this process to work is that the line widths must be narrow

$\nu_e - \nu_n$ ν_e $\nu_e + \nu_n$

P_n
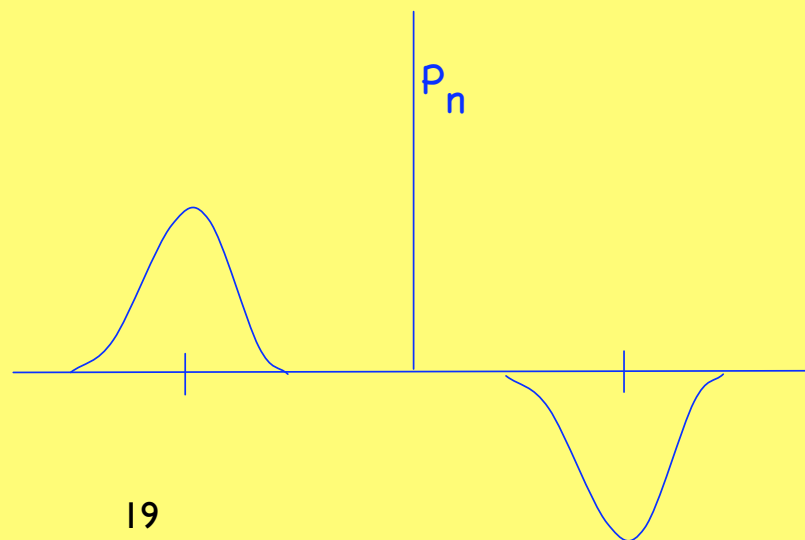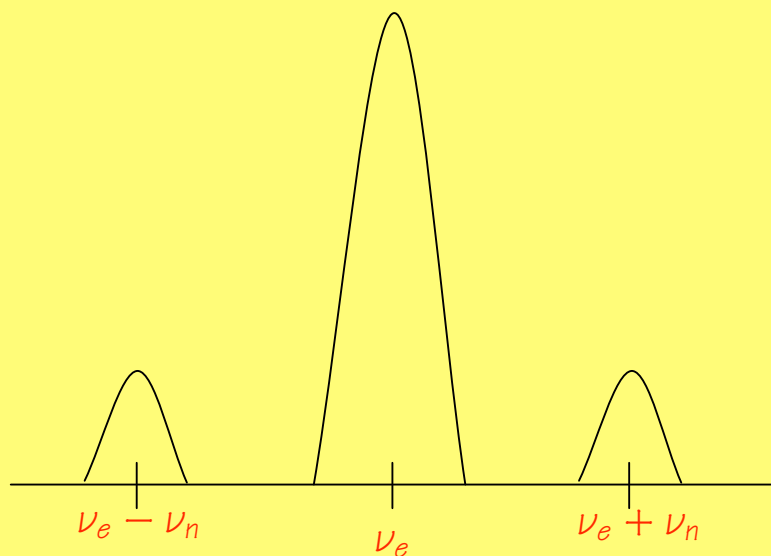
For other nuclear species (deuteron) the enhancement frequencies ($\nu_e \pm \nu_n$) will be different from that of the proton because of the different Lamor frequency.

For present-day PT materials (alcohols, ammonia) are not single crystals but rather glassy or amorphous materials. They do not have discrete energy levels and require another mechanism to describe the polarizing process. This is called the Equal Spin Temperature Theory

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

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Solid Effect

Spin–Spin Interaction

Photon

Electron

Proton

Materials that were studied for polarization at CERN (1965 – 1971)

Benzene
Toluene
Ethanol
Methanol
Propanol
Polyethylene
Polystyrene
LiF
Wax
Para Wax

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

Plexiglass
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

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

Free Radicals - Dopants

DPPH

PAC

BPA

Shape BPA

Violanthrene

Porphyrexide

TEMPO

Ziegler

Anthracene Na$^+$

TMR

PB

PR

TMPD

Tri-tetra-bythlphenyl

Tetramethyl 1,3 cyclobutadien

DTBM

etc.

BPA + DPPH

BPA + Cob. Oleale

Ziegler + DPPH

Ziegler + Cob. Oleale

Ziegler + BPA

etc.

neutron irradiation

^{60}Co-γ irradiation

γ - irradiation

Polarization Detection System

$P_{DYN} = \varepsilon P_{T.E.}$

Nuclear Resonance Oscillator

$\nu_n = 213MHz$ for Protons

$\nu_n = 32MHz$ for deuterons

Microwave Generator

$\nu_e \pm \nu_n = 140GHz$

Cryogenics ^4He

$T \leq 1K$

Magnet Power Supply

$B = 5.0T$

Magnet

Magnet

Target Coil

Target Material

Refrigerator

B
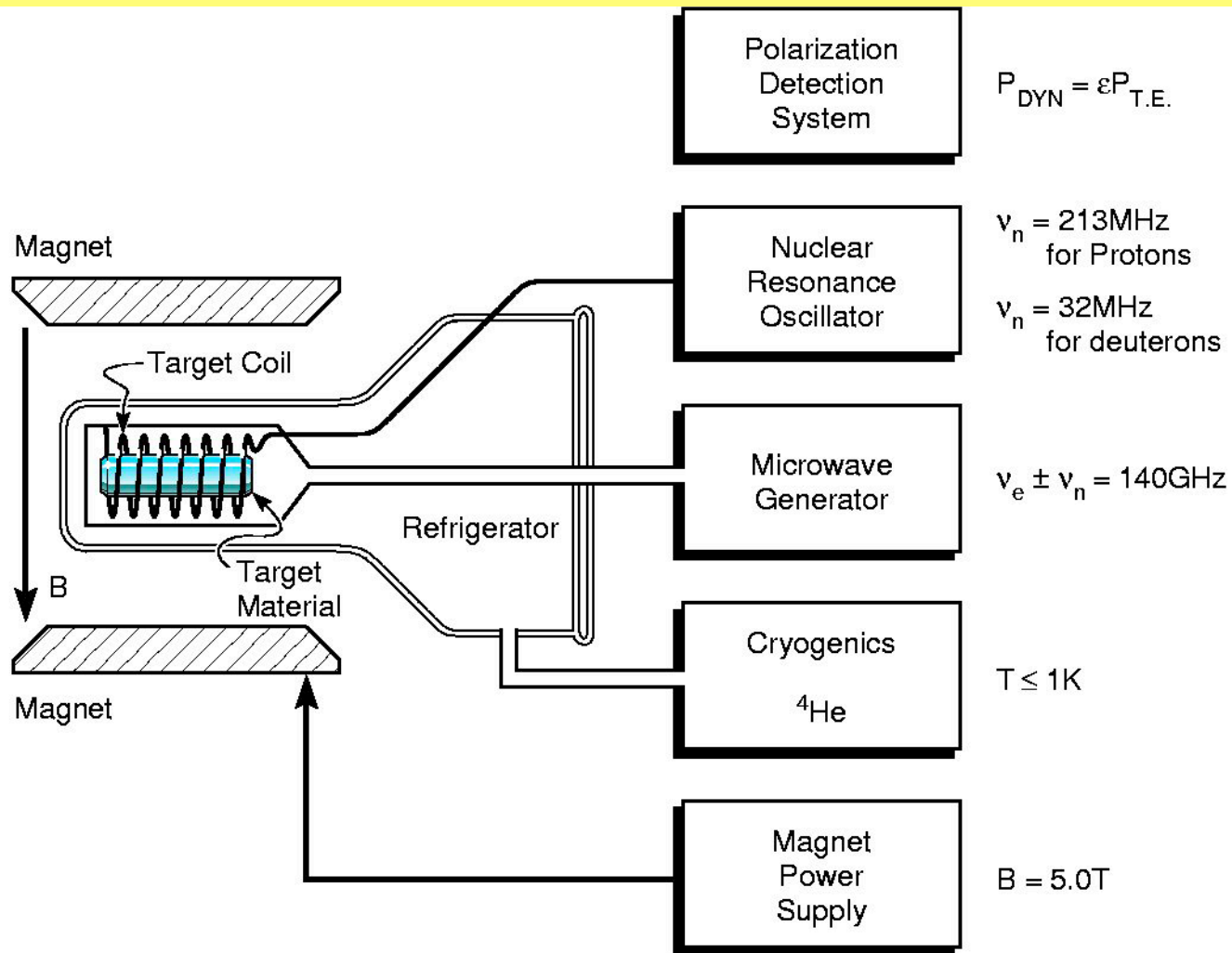
Cryogenics: ^{3}He and ^{4}He Evaporation Refrigerators

Here, liquid He is fed into a separator pot where the liquid phase is separated from the vapor phase by a sintered copper plate. The cold vapor is pumped away and used to cool the radiation shields and baffles that intercept the radiation heat load. Liquid helium flows through the separator plate into a heat exchanger and then is metered into the target holder (or evaporator) via a needle valve. The pool of liquid in the target holder is pumped on by large capacity Roots pumps to reduce the temperature to ≤1 K. As the cold vapor is pumped away, it exchanges heat with and cools the incoming warm liquid. Services such as microwaves and NMR are also brought into the target cavity. Because of the thermal properties of liquid helium, cooling powers of ≈2 W can be achieved with sufficiently large pumps and can withstand high heat input from particle beams.

Figure 7 Schematic of the ^{4}He refrigerator described in [Ref. 28]. siphon de transfert = transfer siphon; vers la pompe à He = to the He pump; separateur = separator; écran = shield; exchangeur = exchanger; enceinte à vide = vacuum enclosure; vanne à He liquide = liquid helium (needle) valve; sortie He de gaz du separateur = exit for the separator helium gas; commande de la vanne = valve control; cavité UHF = UHF microwave cavity; guide d'onde UHF = UHF microwave guide.

^{3}He Evaporation Refrigerators

A ^{4}He is wrapped around and mechanically isolated from the ^{3}He section. T ≈ 0.5T. Since the ^{3}He is expensive, it is circulated through a sealed set of pumps.
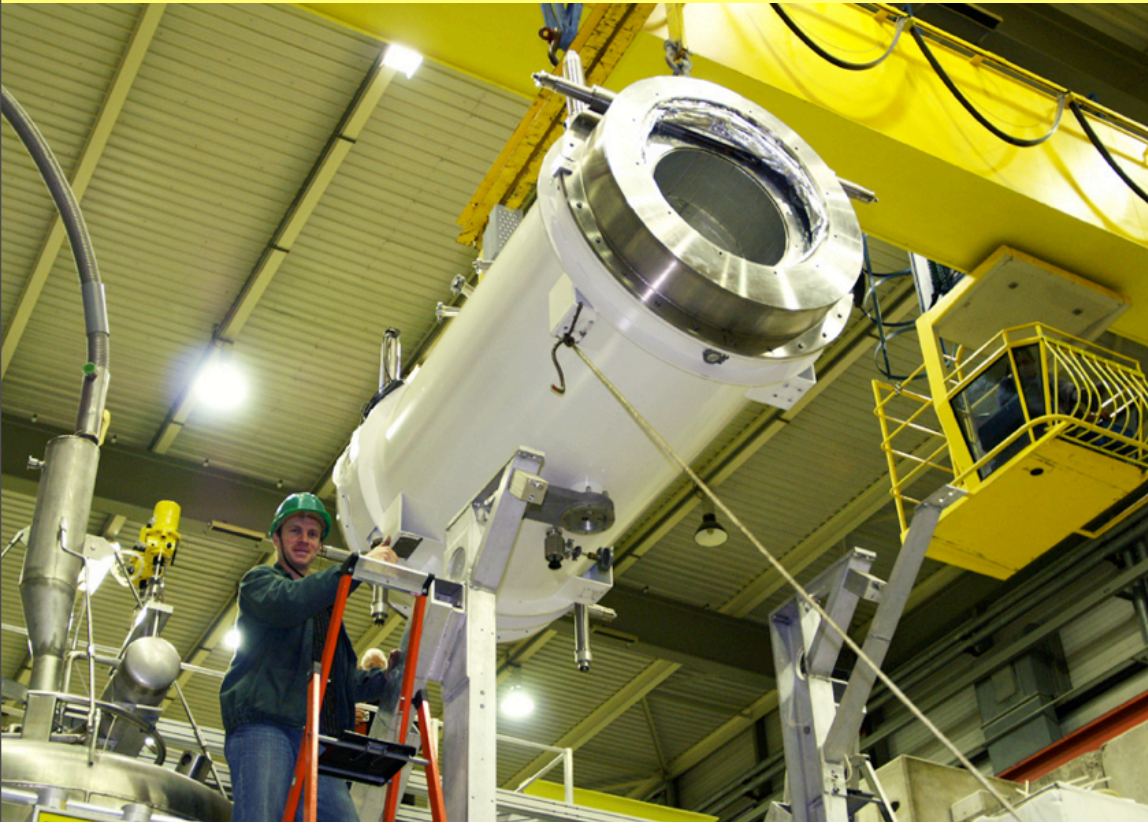
³He/⁴He Dilution Refrigerators

Depends on the special properties of mixtures of ³He and ⁴He. Two phases (below 700 mK) of the mixture: diluted and a rich phase. Cooling occurs when when a ³He atom is removed from the concentrated phase to the dilute phase: 1mW at 50 mK, 15mW at 100 mK, 400mW at 300 mK and 1.3 W at 500mK.
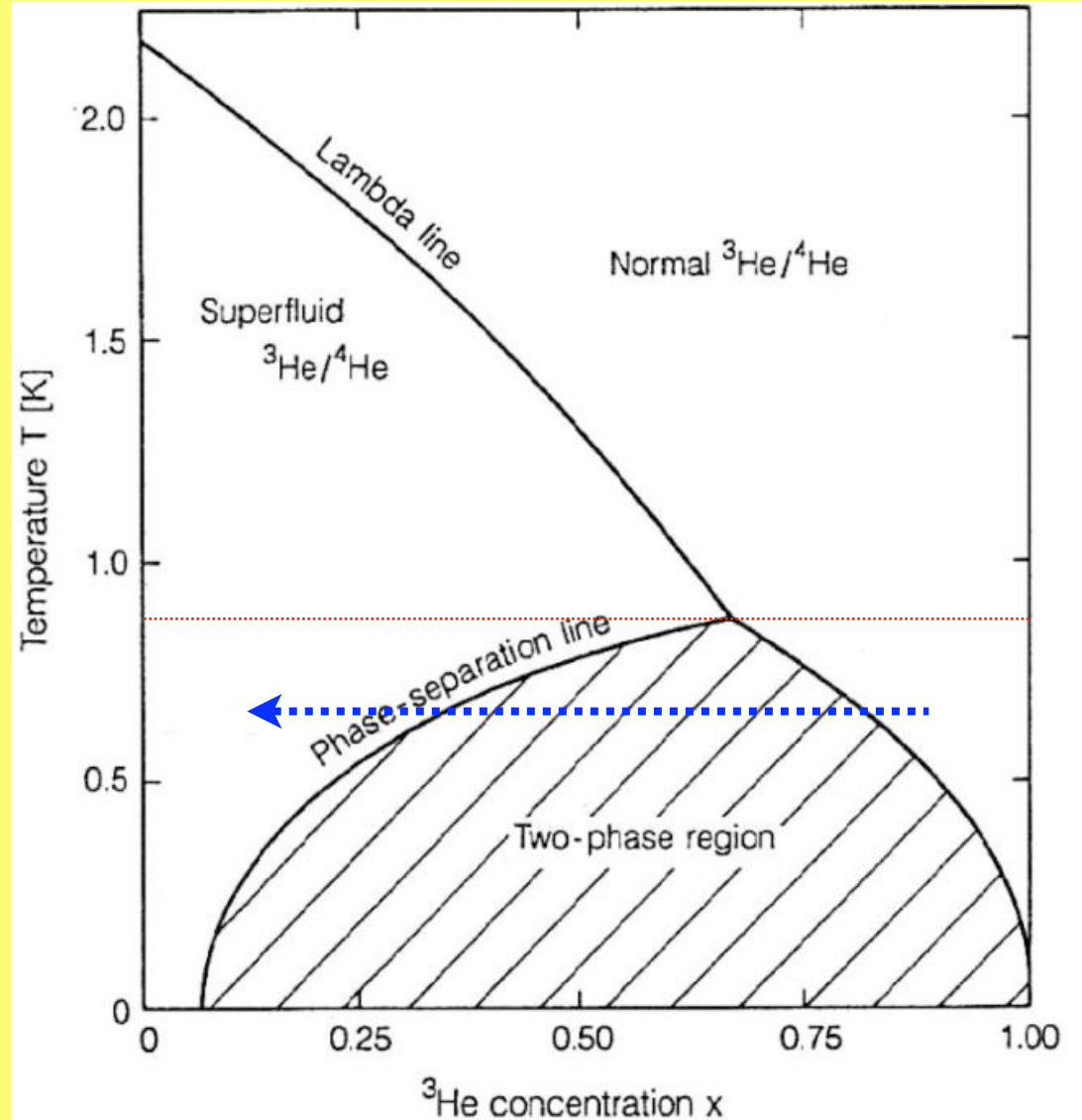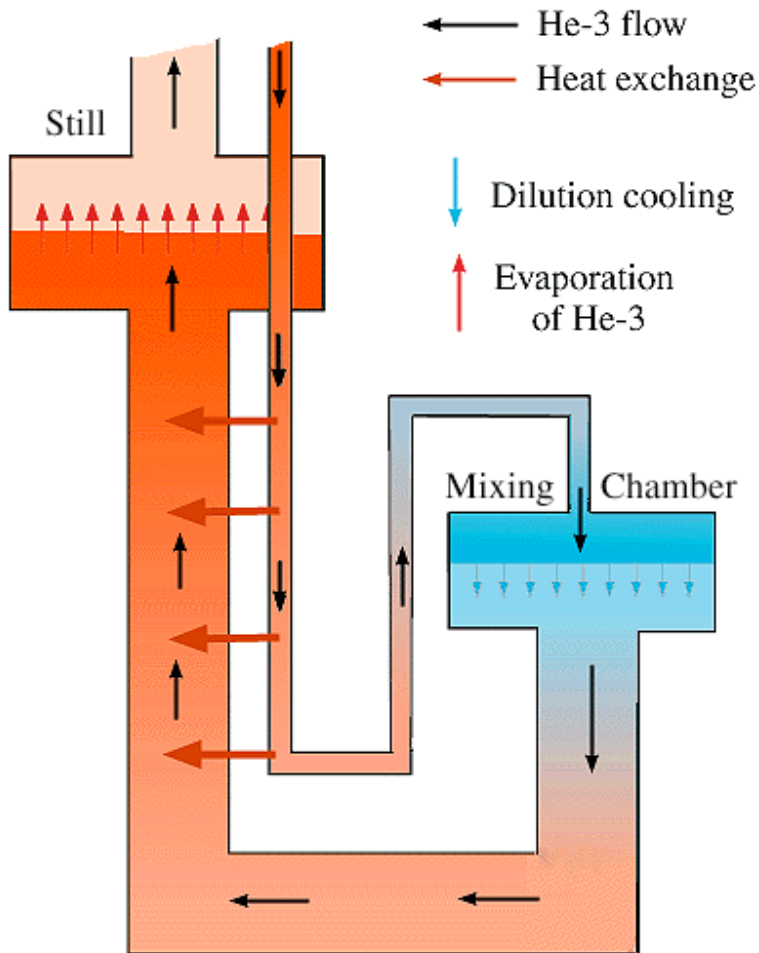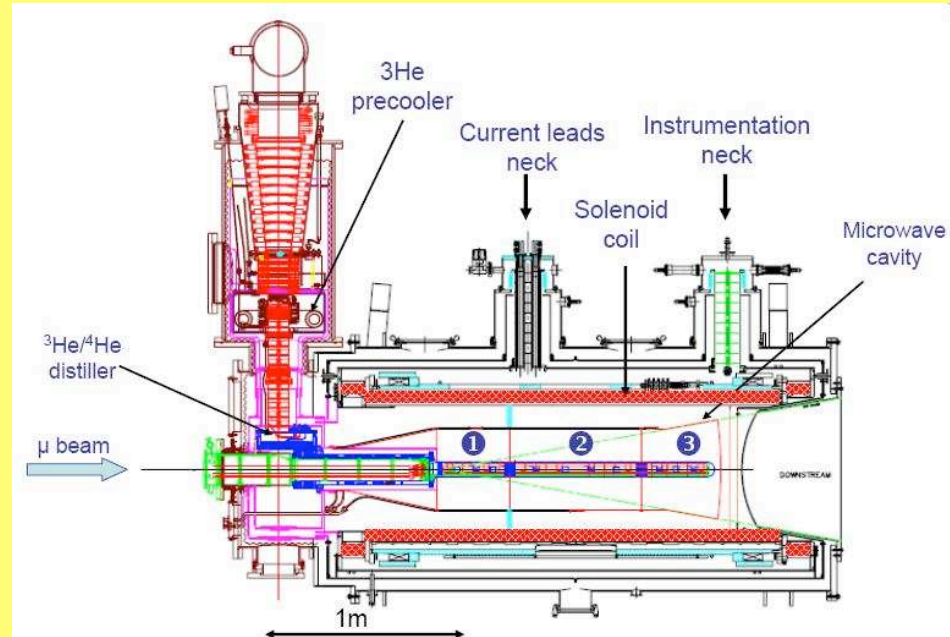
Polarizing Magnets – Solenoids

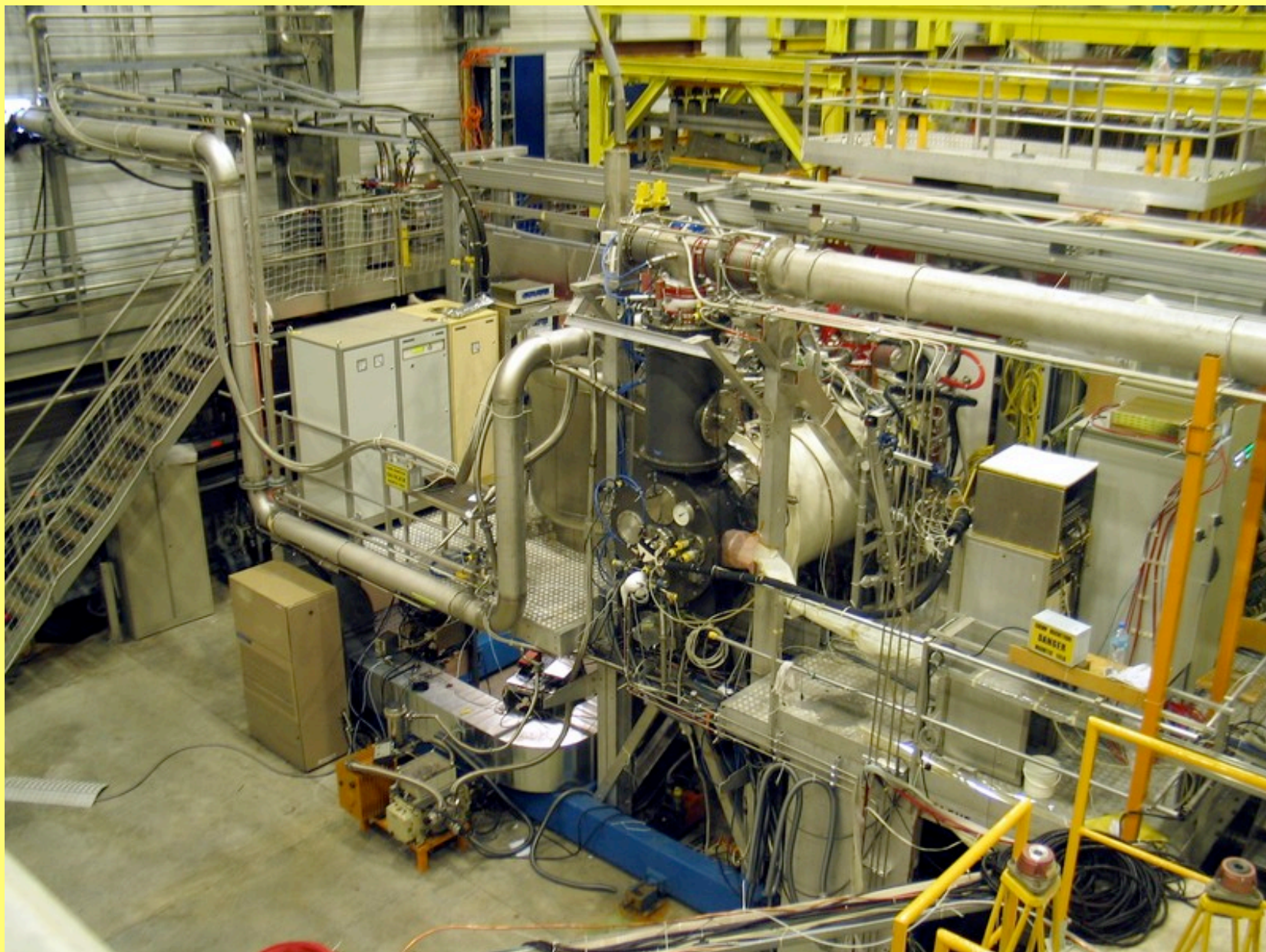DNP works best when
B/T ≈ 5 – 10

Modern magnets are 2.5 – 7.5T

COMPASS – CERN

COmmon Muon Proton Apparatus
for Structure and Spectroscopy

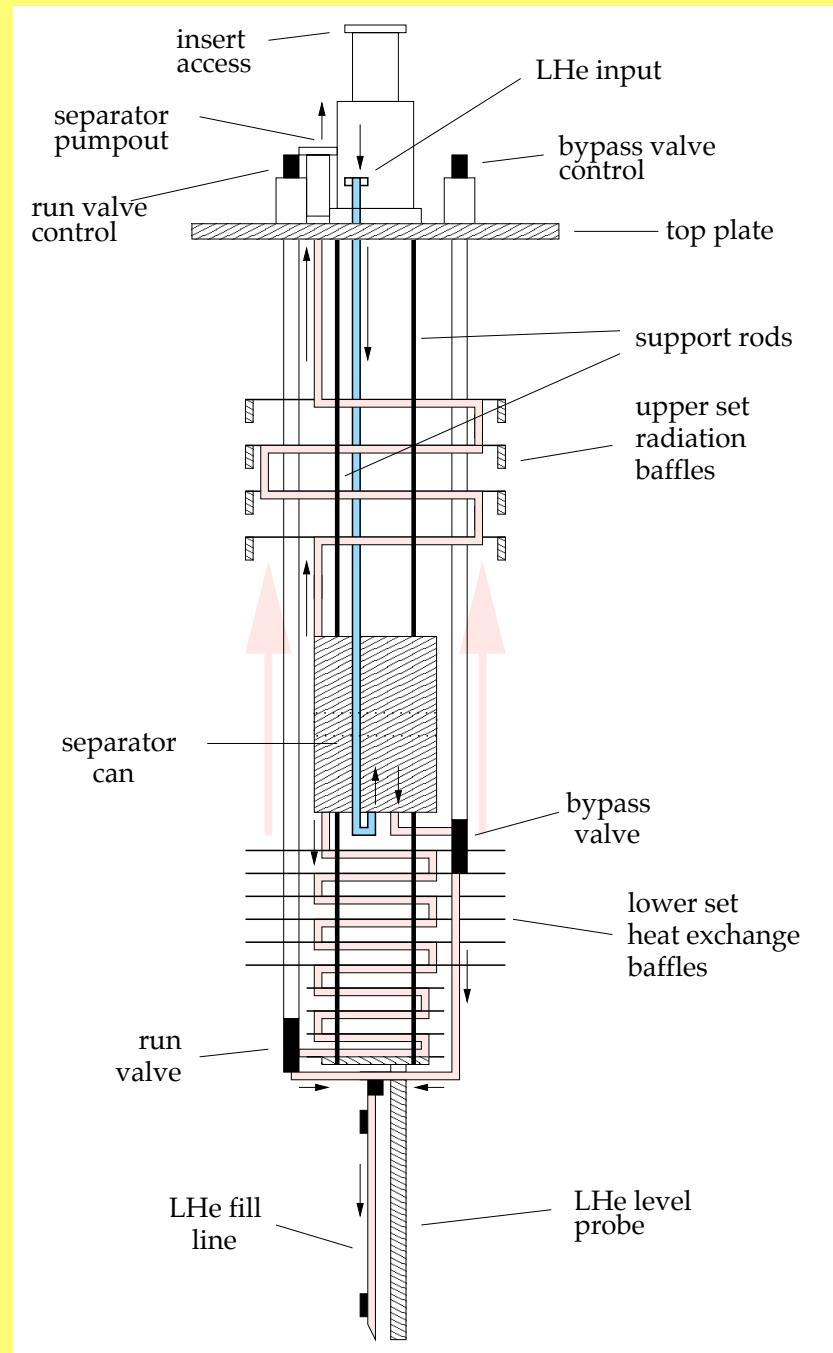# Polarizing Magnets- Split Pair UVA/SLAC/JLAB Target

Microwave Input

NMR Signal Out

Refrigerator

Signal

Frequency

To Pumps

To Pumps

LN₂

LN₂

Liquid Helium

Liquid Helium

Magnet

NMR Coil

B
5T

e⁻ Beam

Target (inside coil) 1° K

insert access

LHe input

separator pumpout

bypass valve control

run valve control

top plate

support rods

upper set radiation baffles

separator can

bypass valve

lower set heat exchange baffles

run valve

LHe fill line

LHe level probe
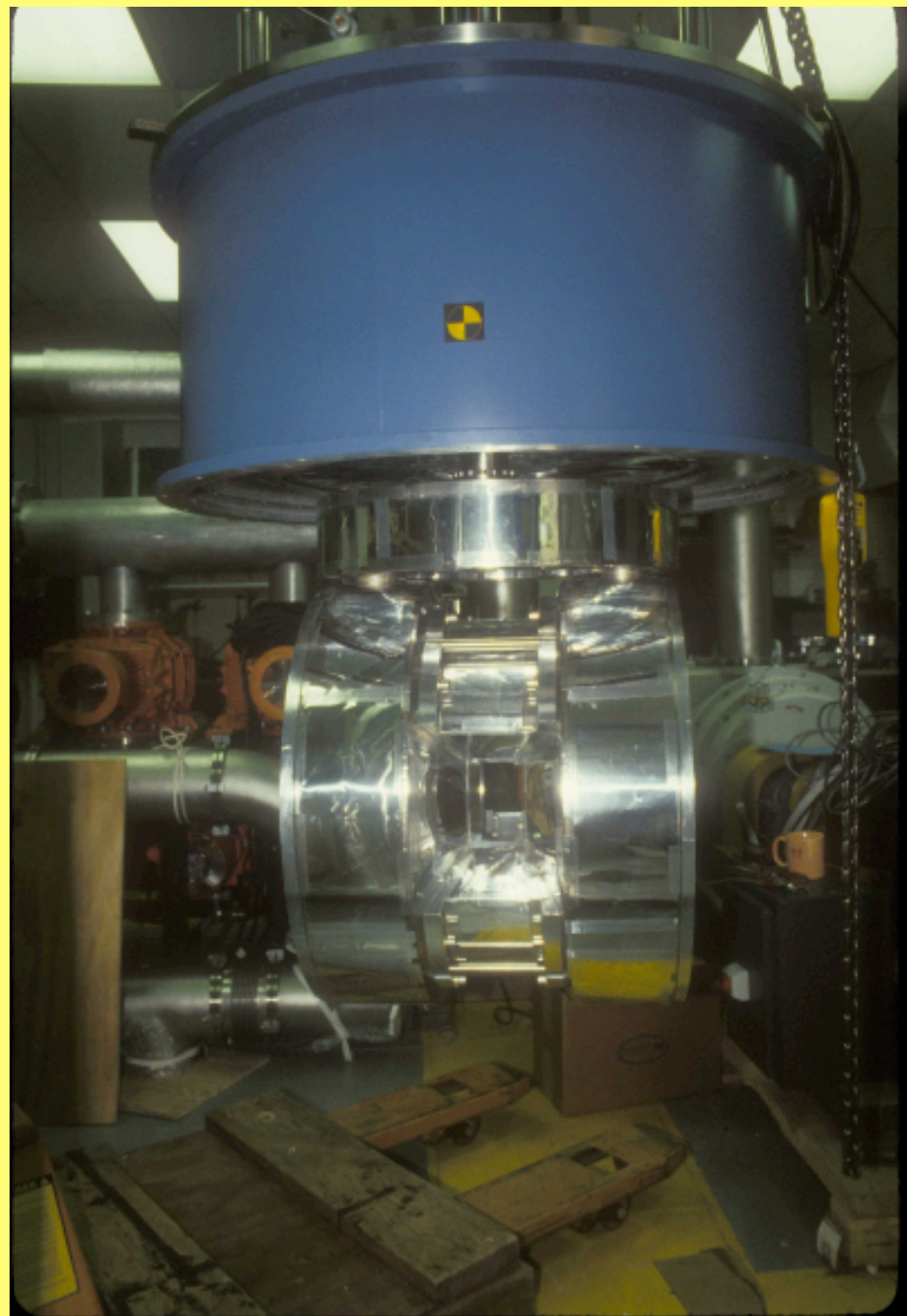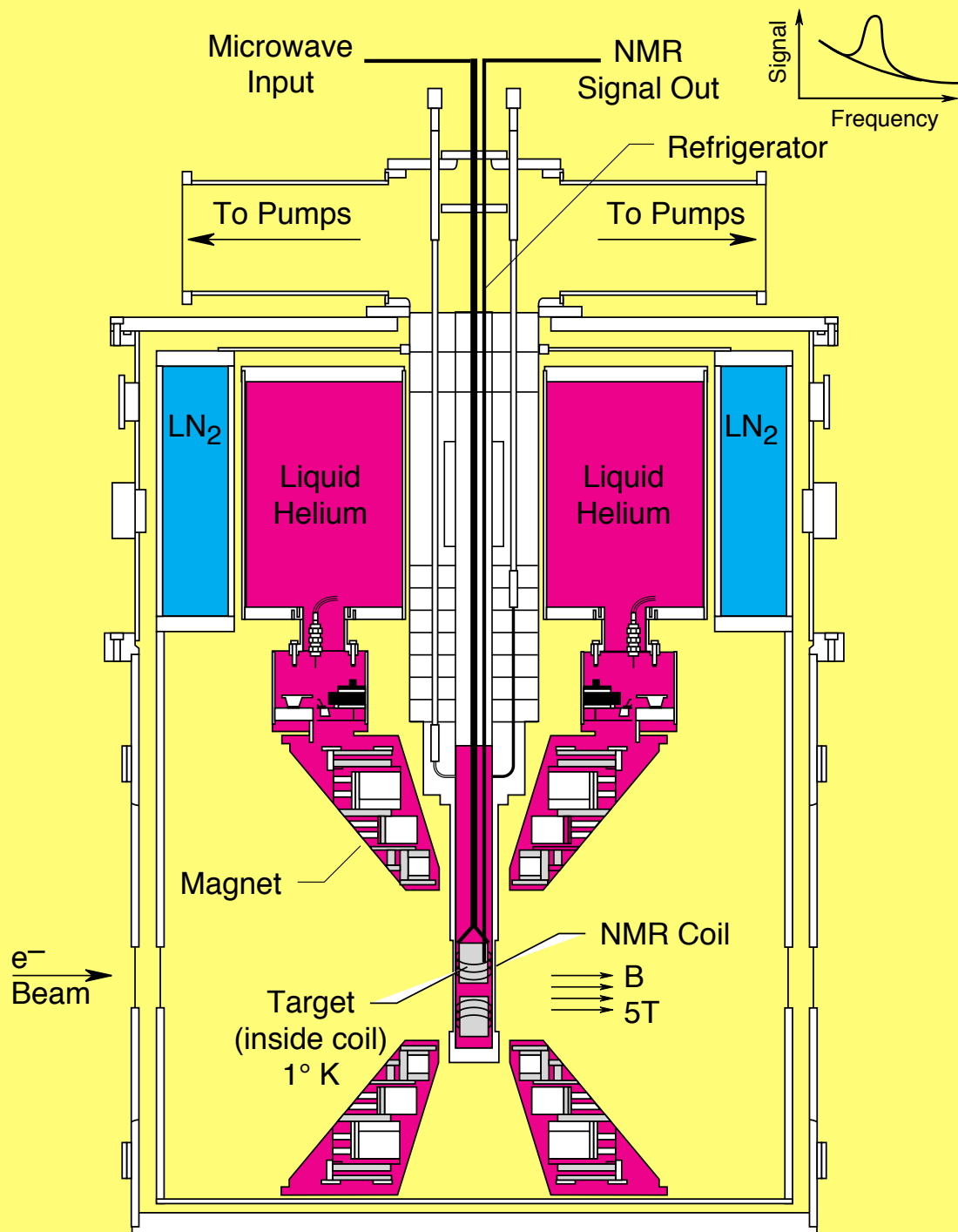
Polarization Measurement

Measuring the polarization is equivalent to measuring the net nuclear magnetization of the material **Nuclear Magnetic Resonance (NMR)**
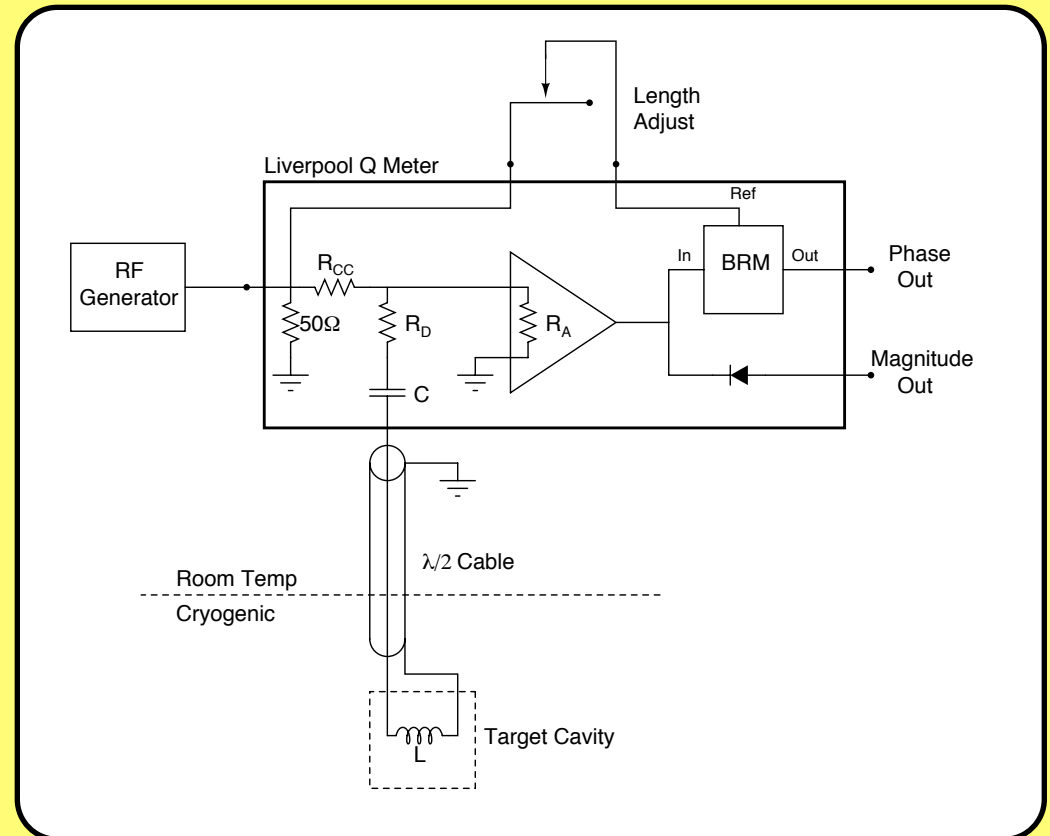
Exposed to rf field at the Lamor frequency, a spin system in a magnetic field either absorbs or emits energy. The response is described by the the magnetic susceptibility

$$\chi(\omega) = \underbrace{\chi'(\omega)}_{dispersive} + \underbrace{\chi''(\omega)}_{absorptive}$$

$$P = K \int_0^\infty \chi''(\omega)d\omega$$

Series Q-meter connected to NMR coil with inductance L_c and resistance r_c that is embedded in target material

$$Z_c = r_c + i\omega L_c(1 + 4\pi\eta\chi(\omega))$$

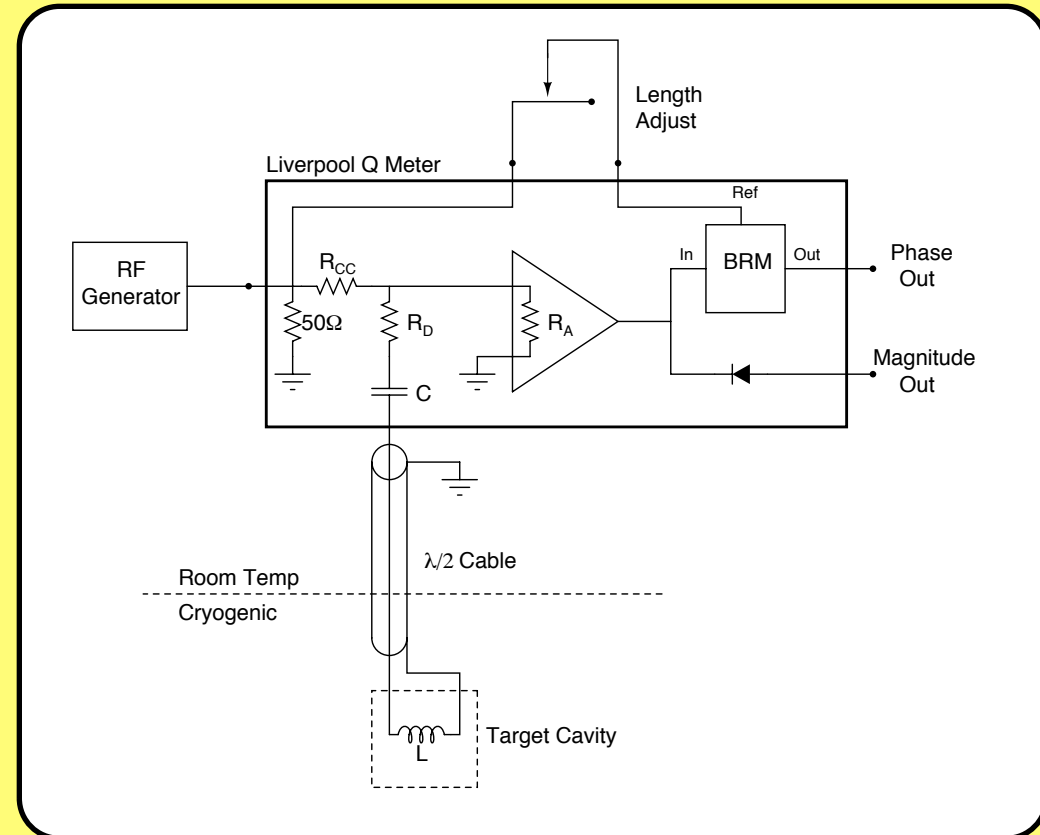$$Z_c = r_c + i\omega L_c(1 + 4\pi\eta\chi(\omega))$$

Inductance (and impedance) changes when the material absorbs or emits energy and thus the voltage, V(ω,χ)
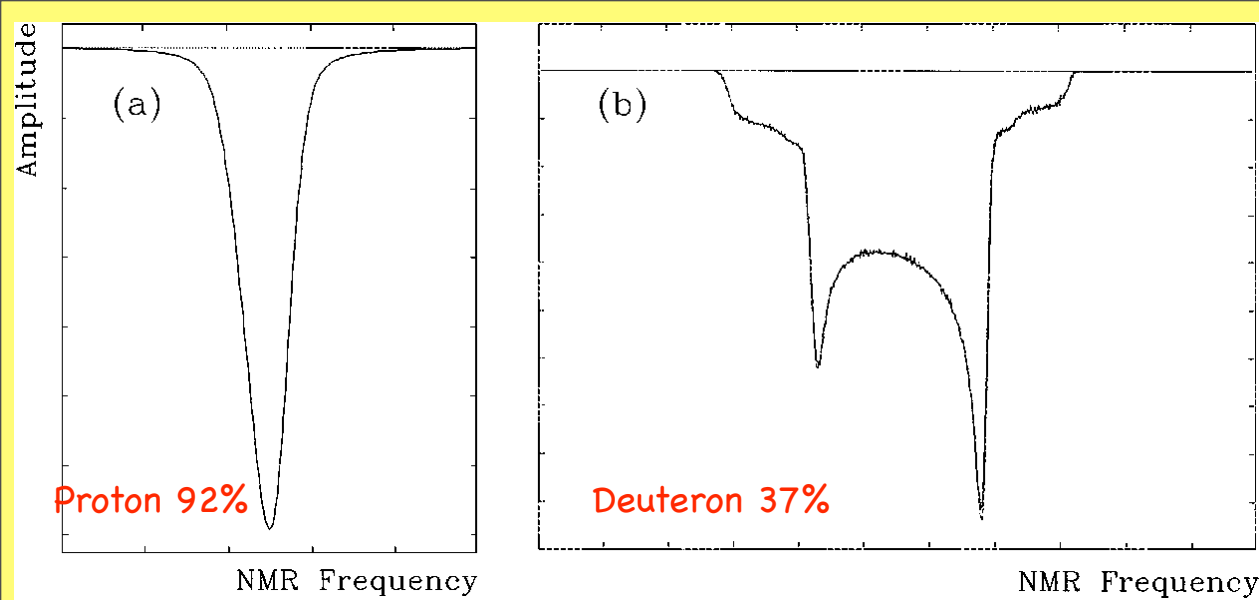
$$S(\omega) = Re(V(\omega, \chi) - V(\omega, 0)) \simeq \chi''(\omega)$$

Polarization is calibrated by using the calculable polarization P_{TE}

$$P_{TE} = tanh\left[\frac{\mu_n H}{kT}\right]$$

$$P = \frac{\int S_{enh}(\omega)d\omega}{\int S_{TE}(\omega)d\omega}$$

(a)

Proton 92%

NMR Frequency

(b)

Deuteron 37%

NMR Frequency

Important criteria

(a) the degree of polarization P

(b) the dilution factor f, which is the ratio of free polarizable nucleons to the total number of nucleons.

$$A = \frac{\sigma^{\uparrow} - \sigma^{\downarrow}}{\sigma^{\uparrow} + \sigma^{\downarrow}} \qquad \epsilon = \frac{N^{\uparrow} - N^{\downarrow}}{N^{\uparrow} + N^{\downarrow}} \qquad A = \frac{1}{Pf} \frac{N^{\uparrow} - N^{\downarrow}}{N^{\uparrow} + N^{\downarrow}}$$

$$f = \frac{f_A \sigma}{(1 - f_A)\sigma_0 + f_A \sigma}$$

$$\sigma = \sigma_0 (1 \pm PA)$$

f_A = fraction of polarized nuclei

where P and f correct for the fact that the target is not 100% polarized and contains other materials

Beam time t necessary to achieve a certain statistical error ΔA has the following dependency

$$t^{-1} \propto \rho(f \cdot P)^2 \qquad \text{important to optimized f and P} \qquad \rho \text{ is density}$$
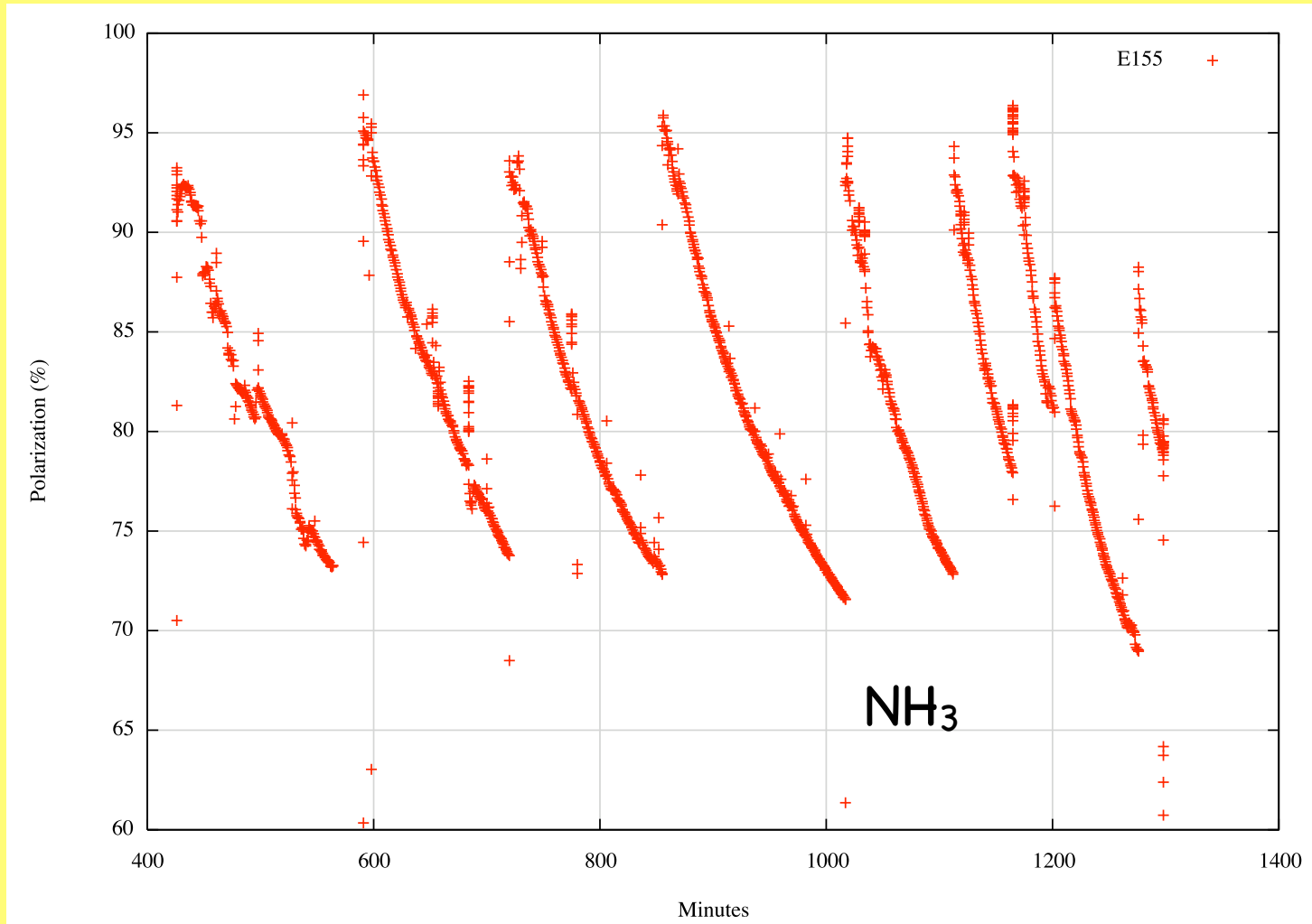
33

Target materials
Performance and Experience
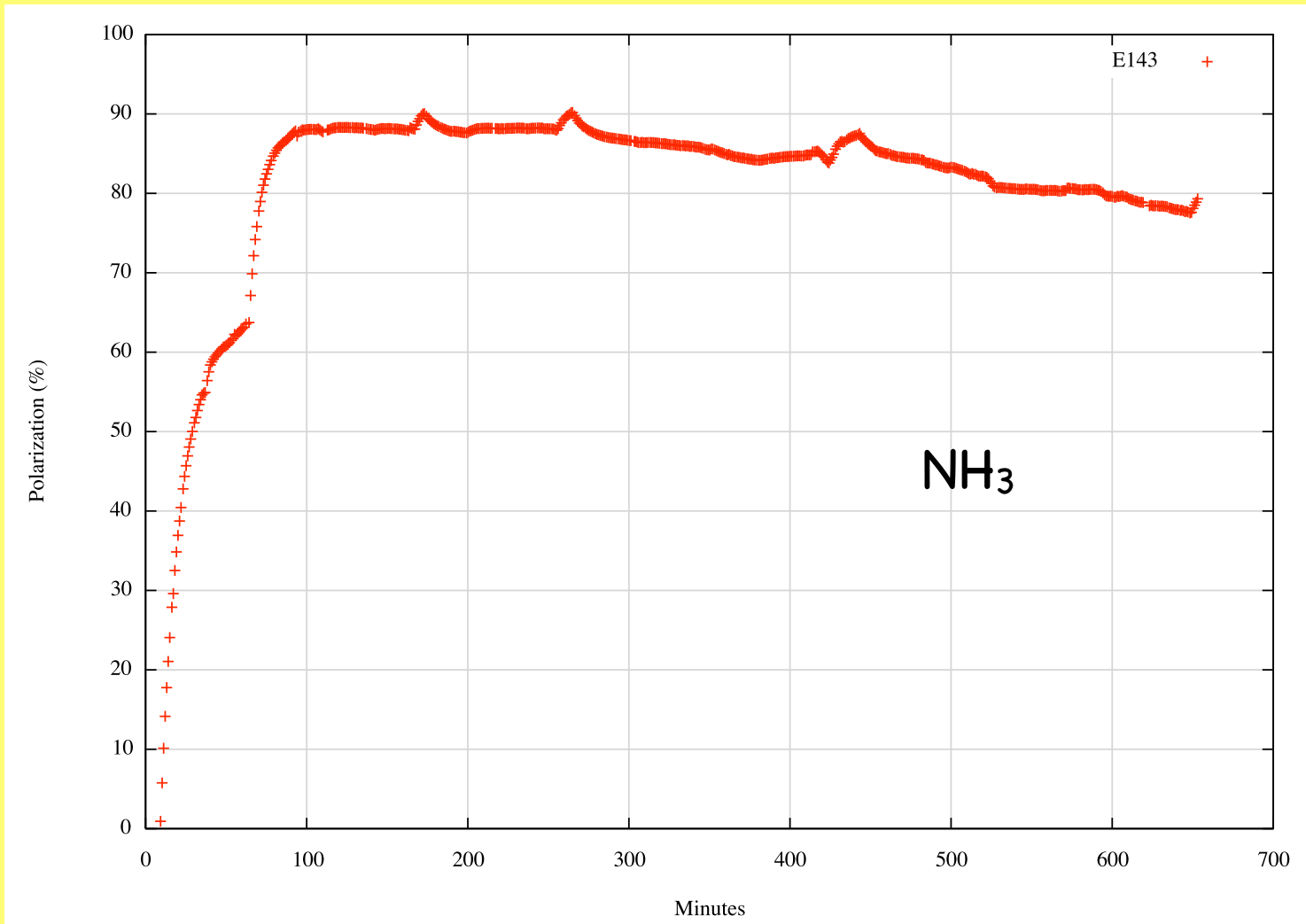
Polarization growth, mistuning, beam on/off, and decay

Target materials
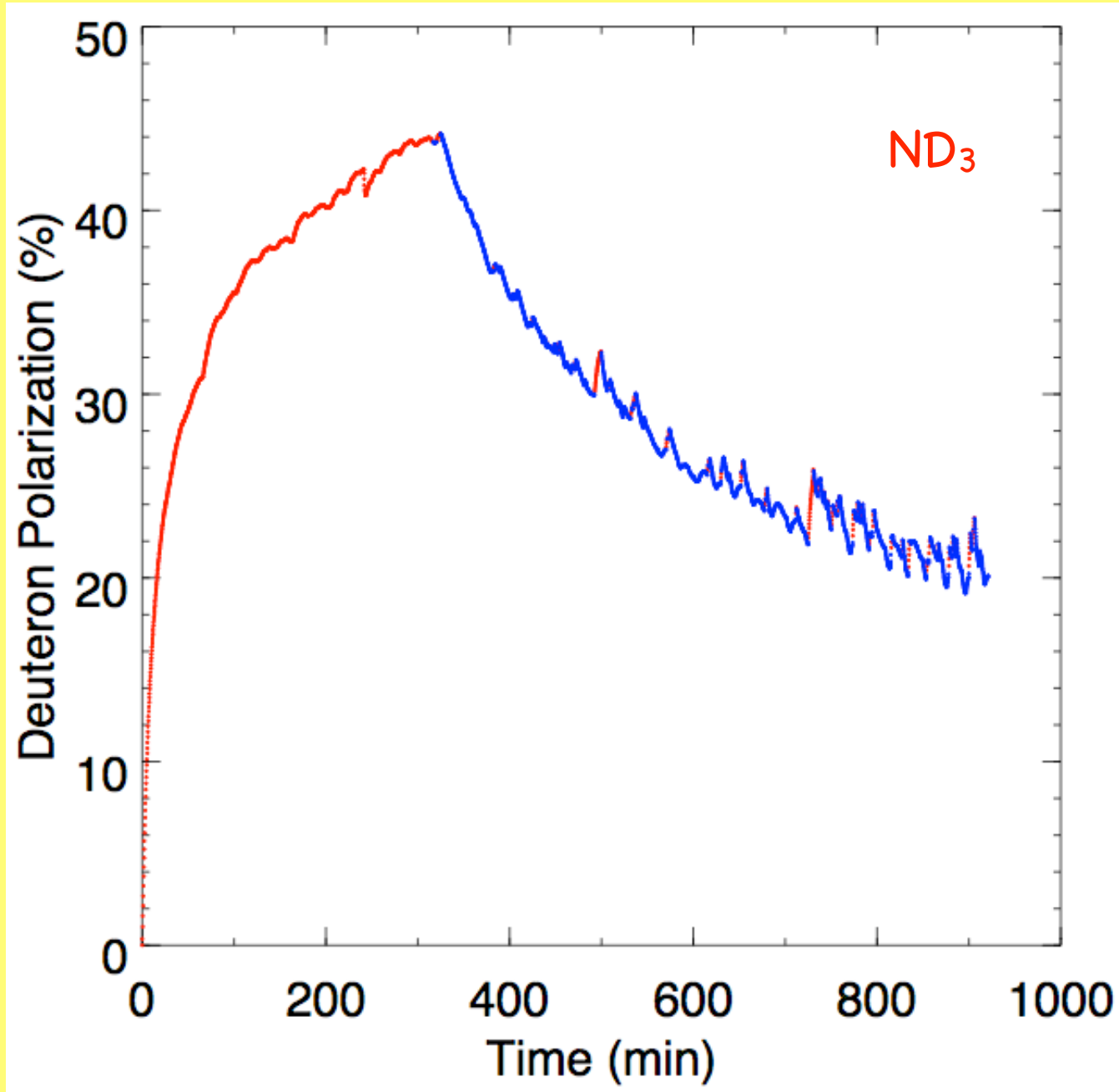
Performance and Experience

Polarization growth, radiation damage, decay of material
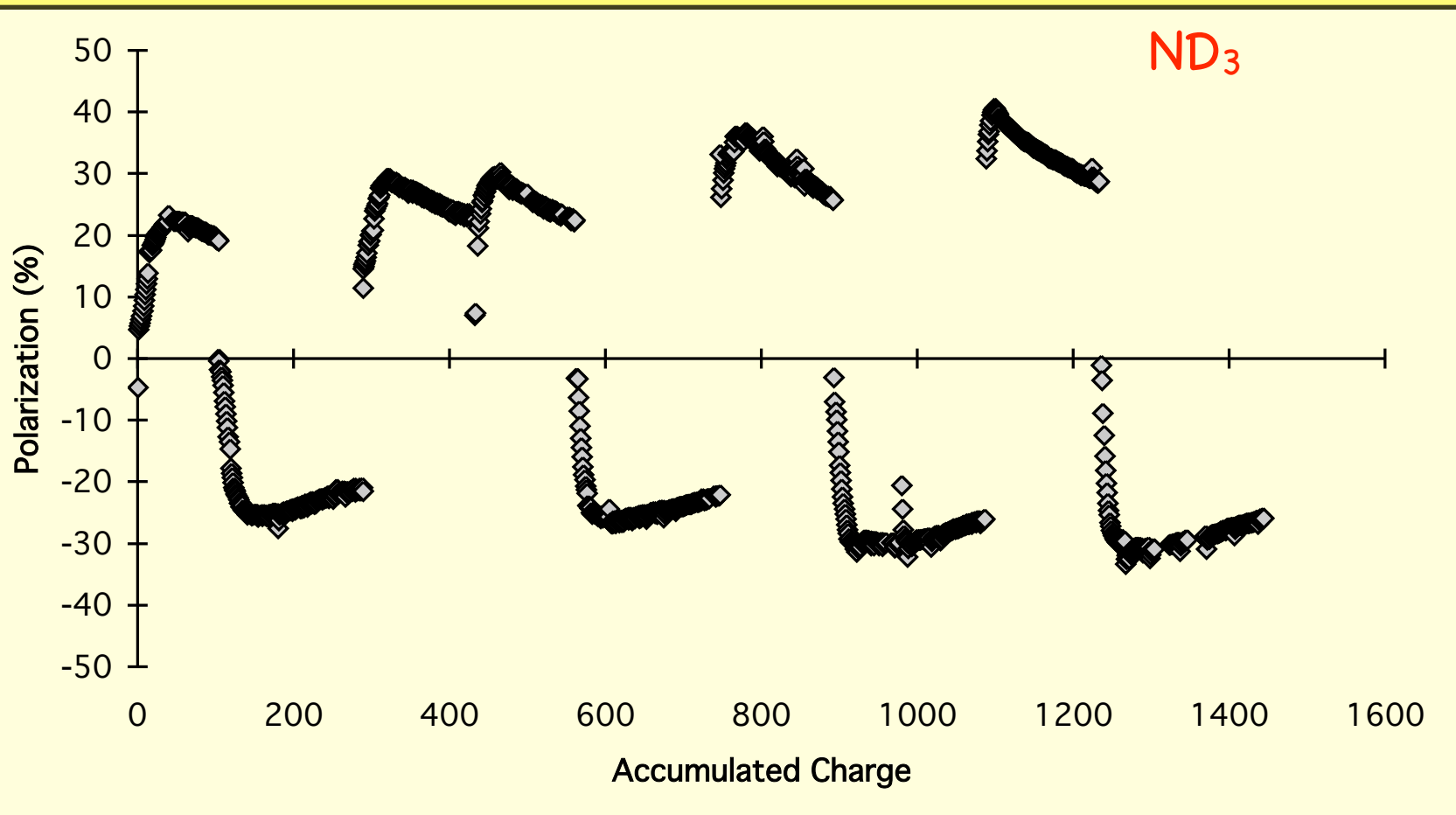
Target materials

Performance and Experience

Target materials
Performance and Experience

Polarization growth, Radiation damage, anneal, reverse sign

UVA/SLAC/JLAB Target

Microwaves

For DNP the frequency needed is about 28 GHz/T, (140 GHz at 5T) and required power is, at 1K, 1-mW/g target material at 2.5 T (70GHz) and 20 mW/g at 5T (140 GHz)
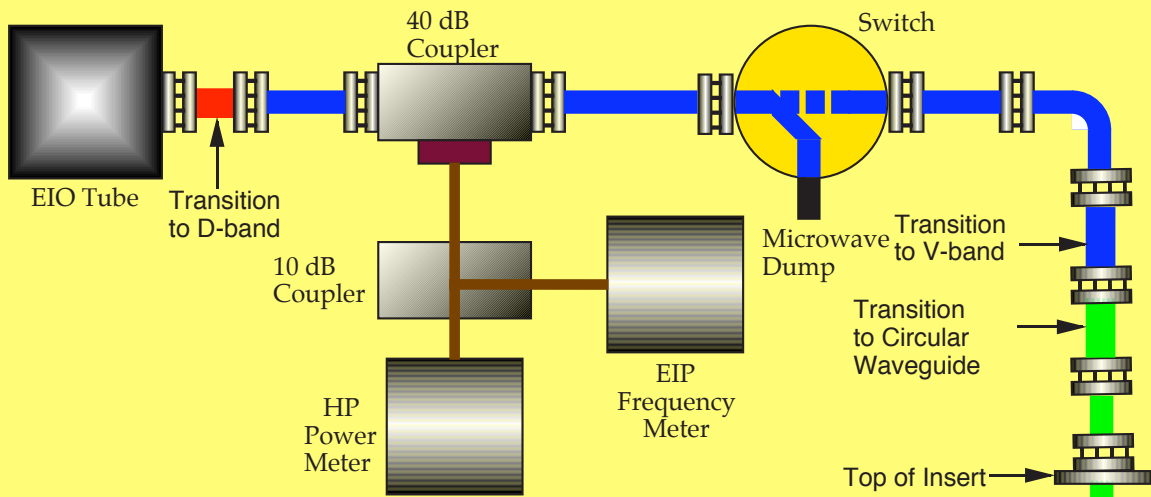
140 GHz Extended Interaction Oscillator (EIO)

Power is inversely related to frequency and power absorption in microwave components increases with frequency ==> a practical limit at 210 GHz, corresponding to 7.5T.
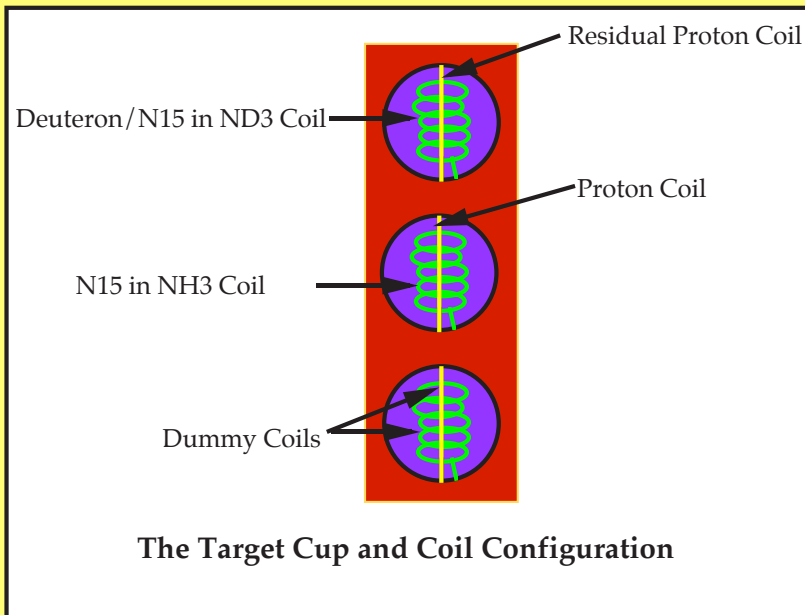
Also: klystrons, IMPATT and Gunn diodes
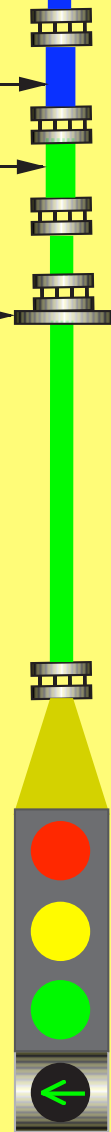
IMPact ionization Avalanche Transit-Time
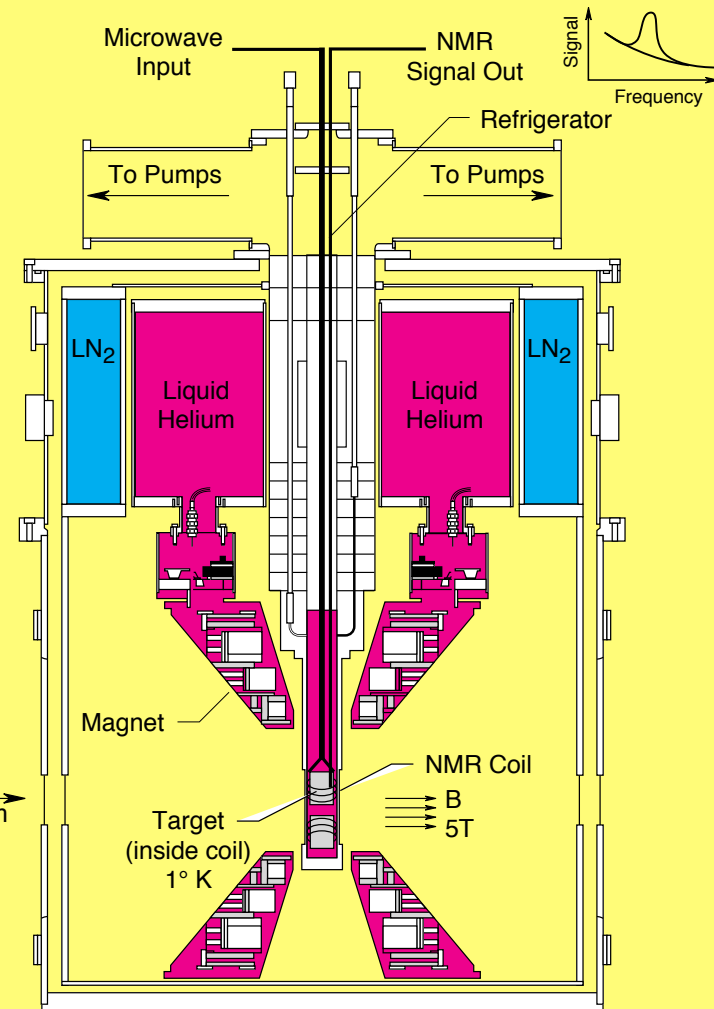
UVA/SLAC/JLAB Target

40 dB
Coupler

Switch

EIO Tube

Transition
to D-band

10 dB
Coupler

Microwave
Dump

Transition
to V-band

Transition
to Circular
Waveguide

HP
Power
Meter

EIP
Frequency
Meter

Top of Insert

— F-band Waveguide (90-140 GHz)
— D-band Waveguide (110-170 GHz)
— V-band Oversized Waveguide (50-75 GHz)

Residual Proton Coil

Deuteron/N15 in ND3 Coil
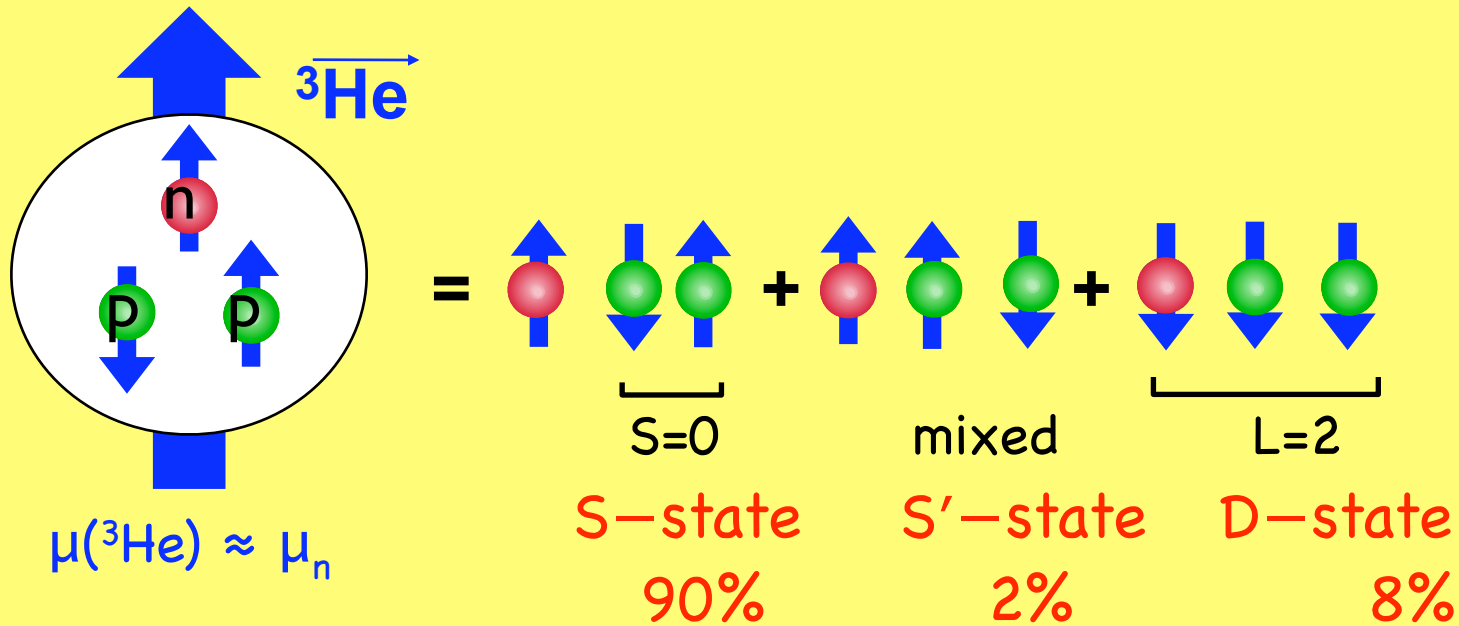
Proton Coil

N15 in NH3 Coil

Dummy Coils

The Target Cup and Coil Configuration

Microwave Horn

Deuteron Cup

Proton Cup

Empty Cup

Carbon

40

Microwave Input

NMR Signal Out

Signal

Frequency

Refrigerator

To Pumps

To Pumps

LN₂

LN₂

Liquid Helium

Liquid Helium

Magnet

NMR Coil

e⁻ Beam

Target (inside coil) 1° K

B 5T

Polarized ³He as polarized n-target

$\vec{^3He}$

$\mu(^3He) \approx \mu_n$

=   S=0   mixed   L=2

S−state   S′−state   D−state
90%   2%   8%

Spin-dependent momentum distribution $N(p_m)$

$$\frac{\rho(\uparrow) - \rho(\downarrow)}{\rho(\uparrow) + \rho(\downarrow)}$$

R.W. Schulze, P.U.Sauer:
Phys. Rev. C 48, 38(1993)

Neutron

D-state

Proton

$N^N(p_m)$

p_m /
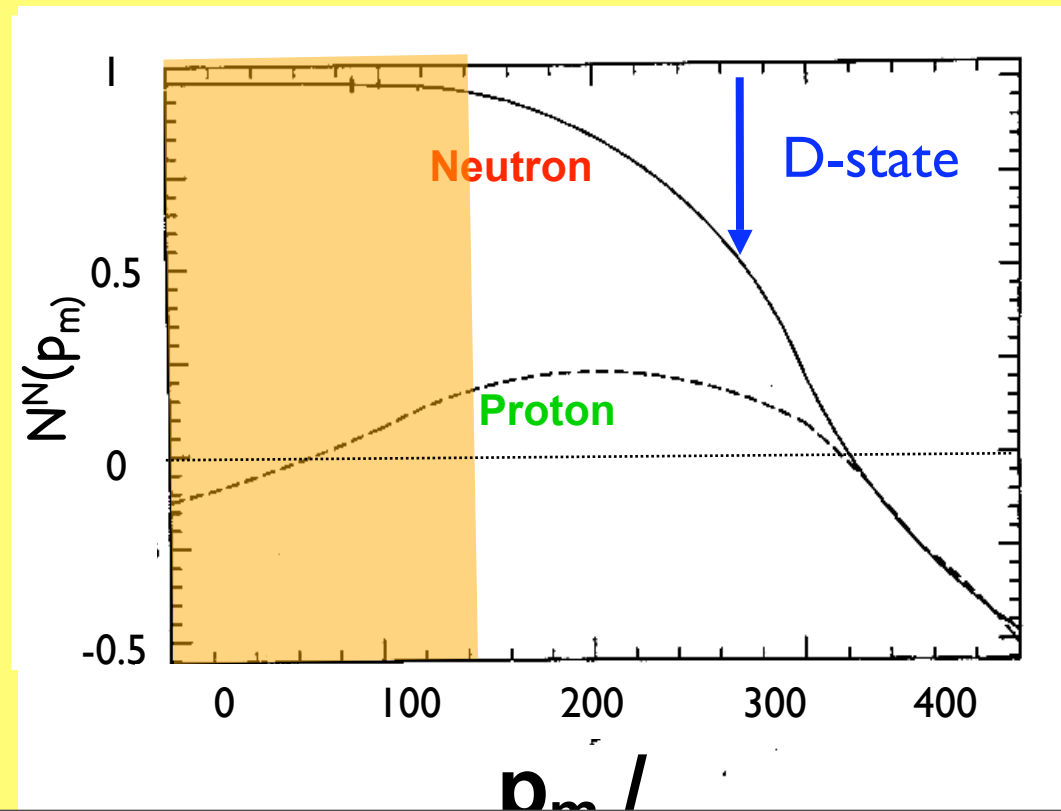
Polarized ³He Targets

Physics interests

- few-body structure

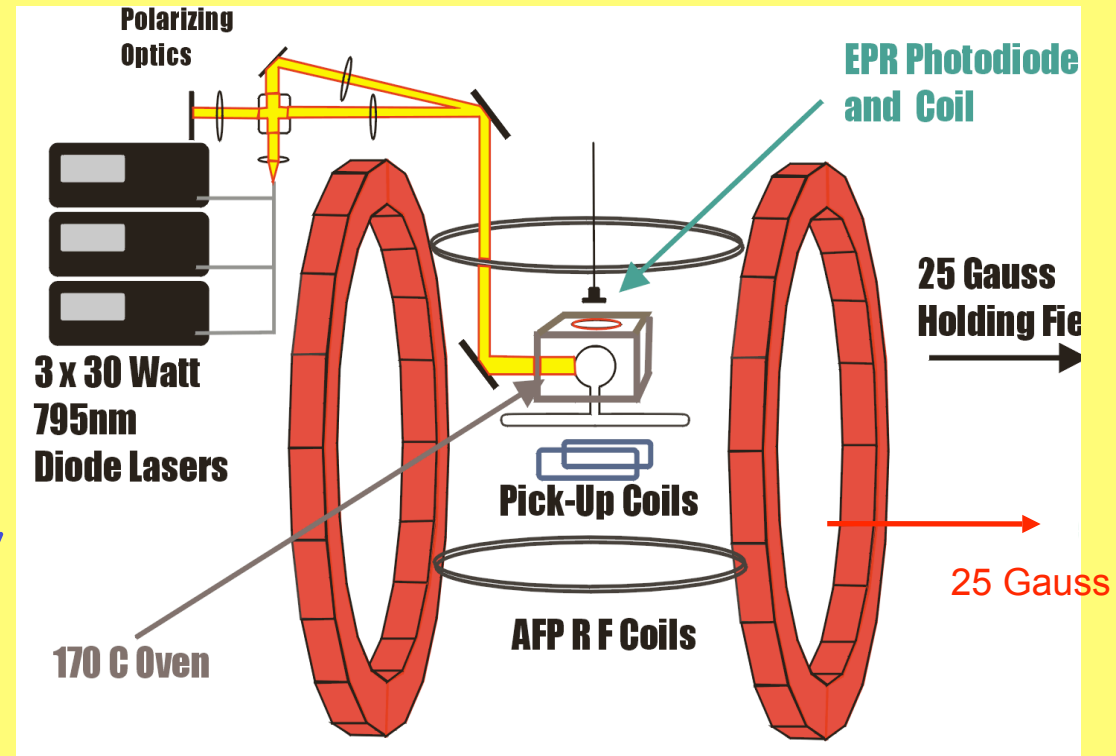- good approximation for polarized free n (P_n=87 % and P_p=2.7 %), requires corrections for nuclear effects

Standard technique:

- optical pumping of Rb vapor, followed by polarization transfer to ³He through spin-exchange collisions

- target polarization measured by EPR/NMR

Performance

- 40cm long target (10atm, I_e=12μA)

- luminosity ~$2 \cdot 10^{36} cm^{-2} s^{-1}$

- average polarization 42%

Hall A ³He target
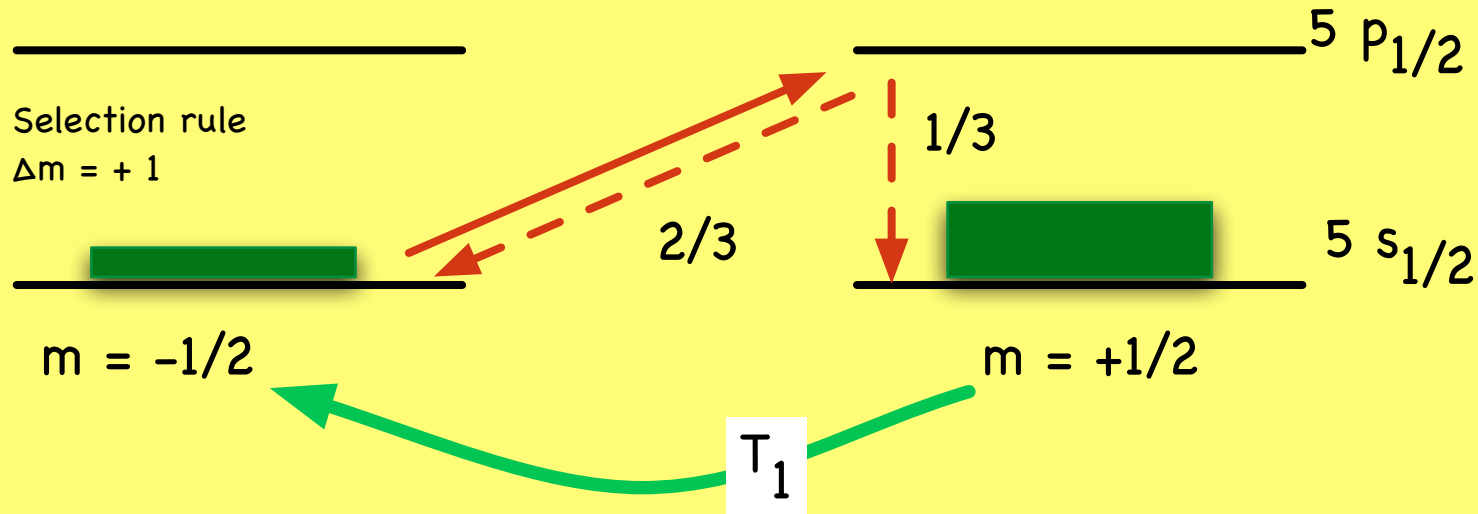

Polarizing Optics

EPR Photodiode and Coil

3 x 30 Watt 795nm Diode Lasers

25 Gauss Holding Fie

Pick-Up Coils

25 Gauss

170 C Oven

AFP R F Coils

Latest development:

- optical pumping of Rb/K mixture

Optical pumping of Rubidium

$5\ p_{1/2}$

Selection rule
$\Delta m = +1$

1/3

2/3

$5\ s_{1/2}$

m = –1/2

m = +1/2

T_1

Spin–exchange between Rb and ³He due to Hyperfine interaction
small cross section

Rb

Rb

3He

3He
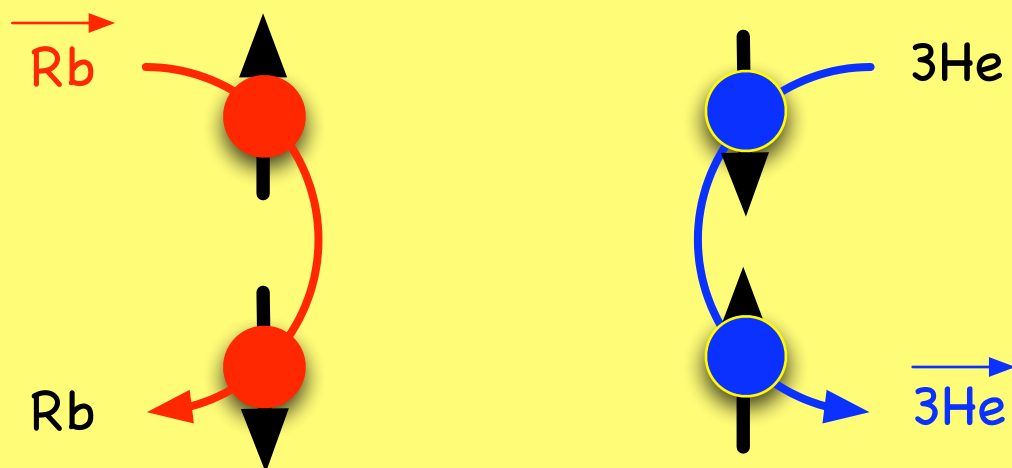
new at JLAB : hybrid cells
contain potassium, build–up
time: 4.2 h

3.5 in

40 cm

45