Polarized Targets, Polarizable Materials and Dynamic Nuclear Polarization

Donal Day
University of Virginia

HUGS 2007

June 2007

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

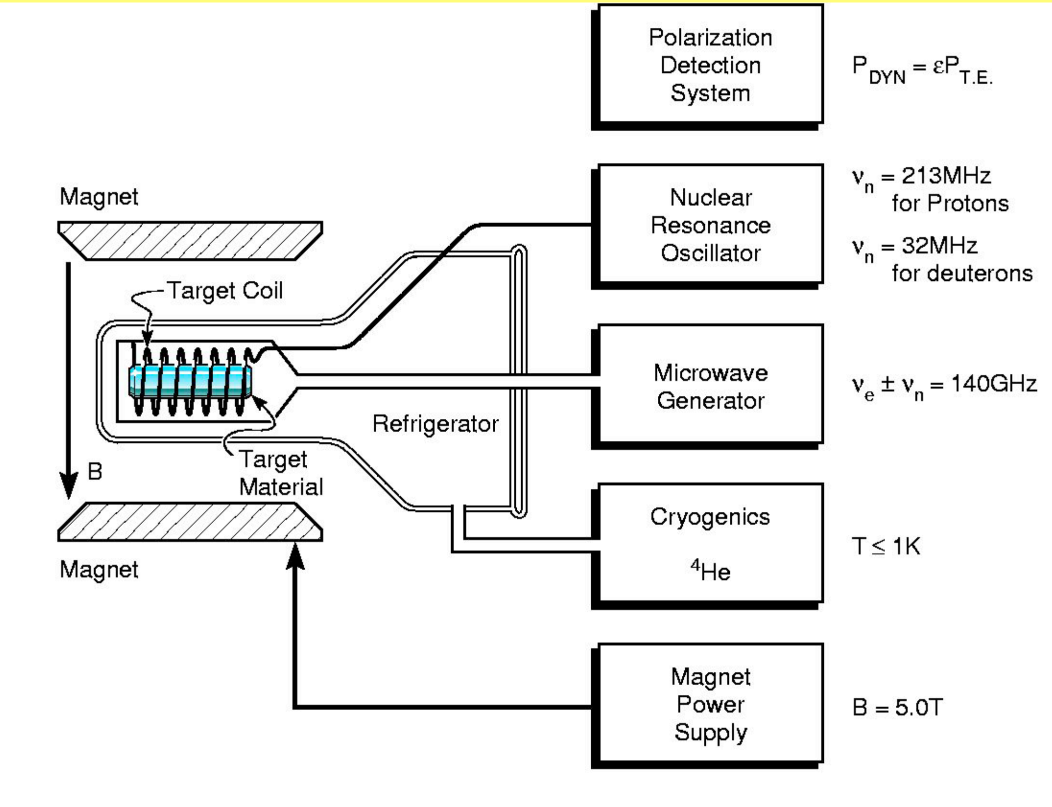
Bonn, Mainz, MIT-Bates, NIKHEF, JLAB, CERN

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



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

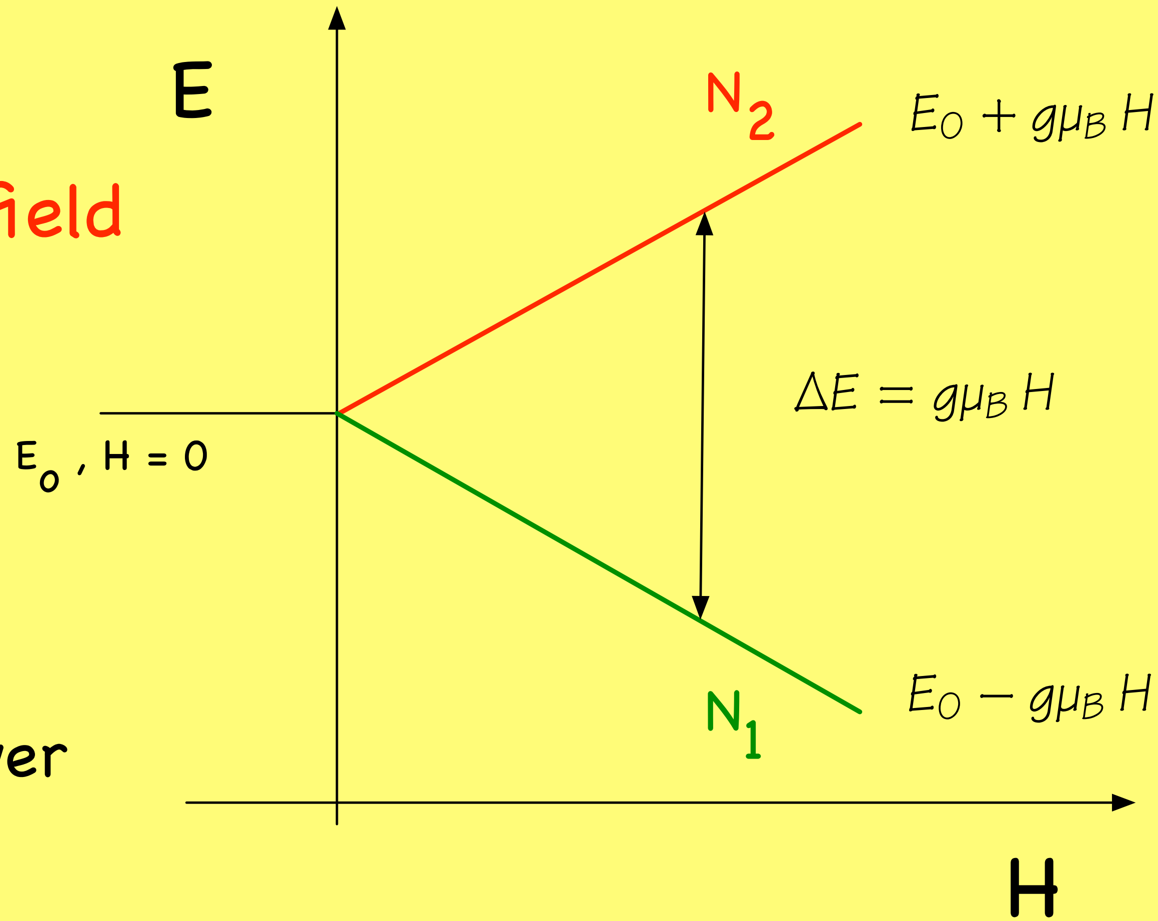
b) Free atoms & ions with partly filled inner shell; transition elements; rare earth & actinide elements Mn^{2+}, Gd^{3+}, U^{4+}

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)

$N_1 > N_2$ ∴ net magnetization || field

Thermal agitation ⇒ random orientation

Lower temperature ⇒ less agitation

⇒ $N_1 > N_2$ ⇒ larger magnetization

If we let H >> and T <<, eventually N_2 ⇒0

At thermal equilibrium (spin 1/2)

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

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

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

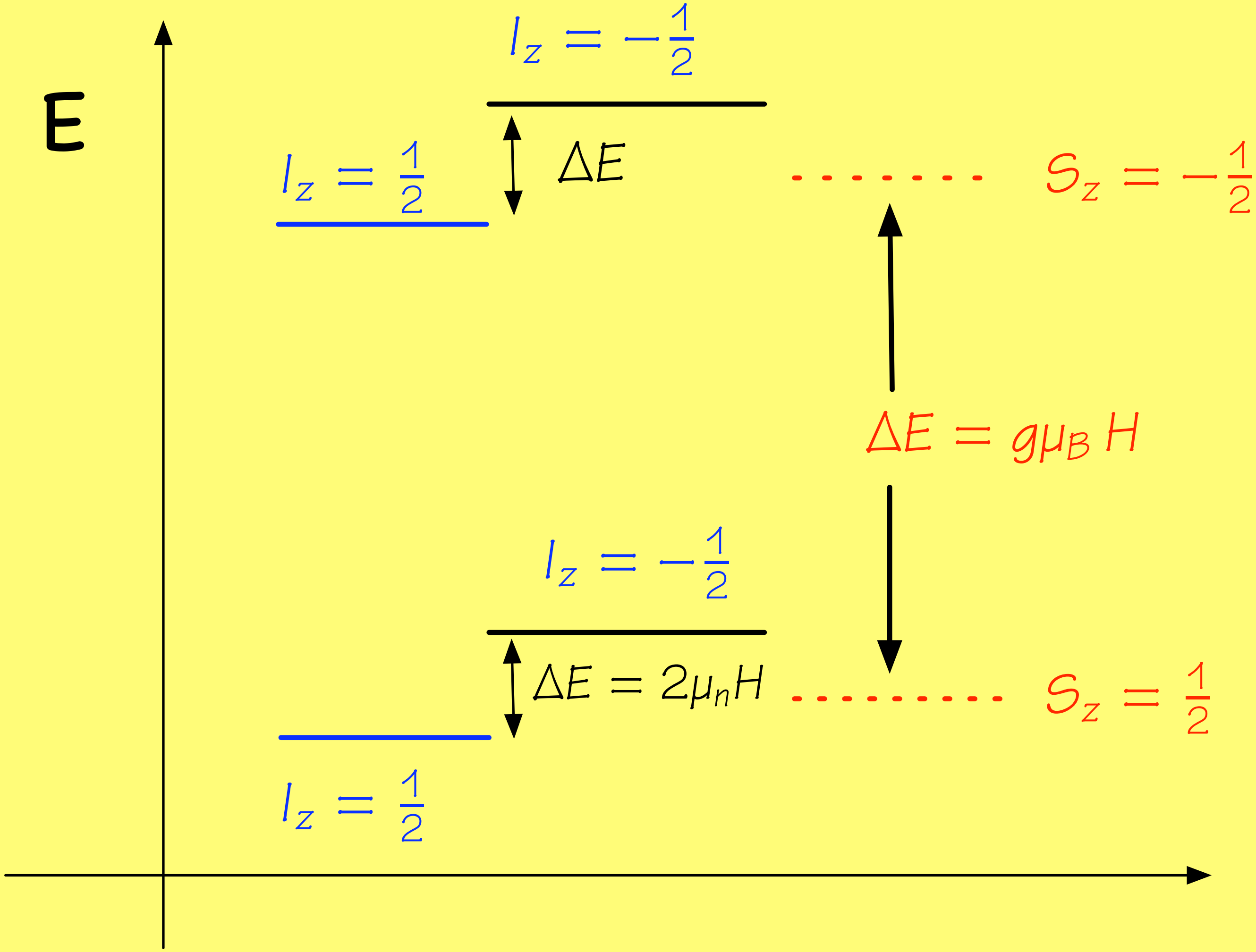
Proton

(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 ⇒ hyperfine splitting in magnetic field



Analogous to the electron

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

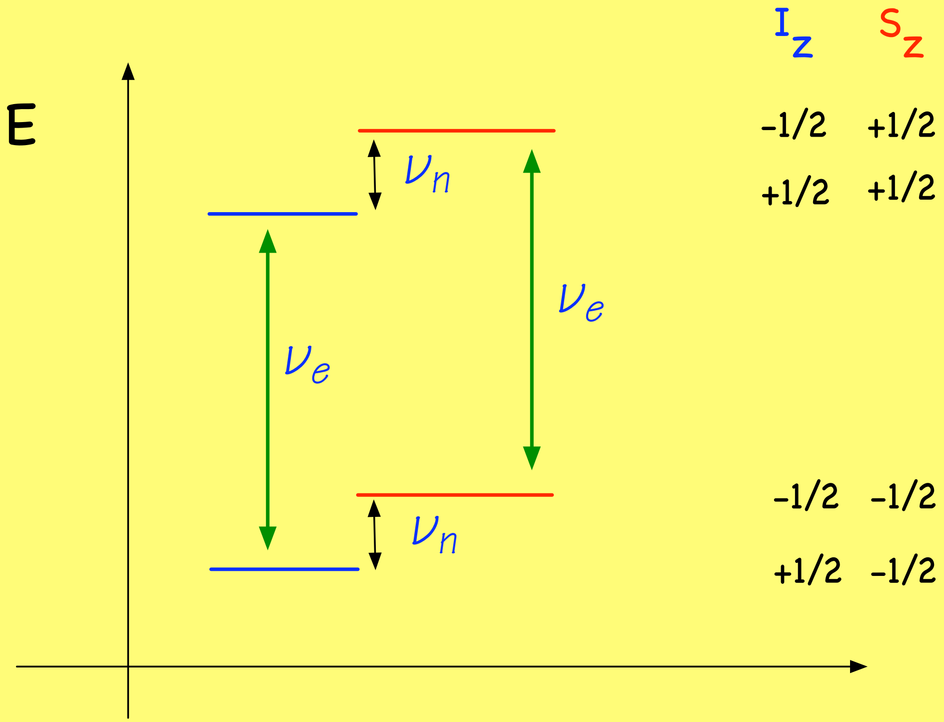
μ_n = proton magnetic moment

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



RF fields can be used to induce transitions between spin states

Allowed Transitions

$\Delta I_z, \Delta S_z = 1$

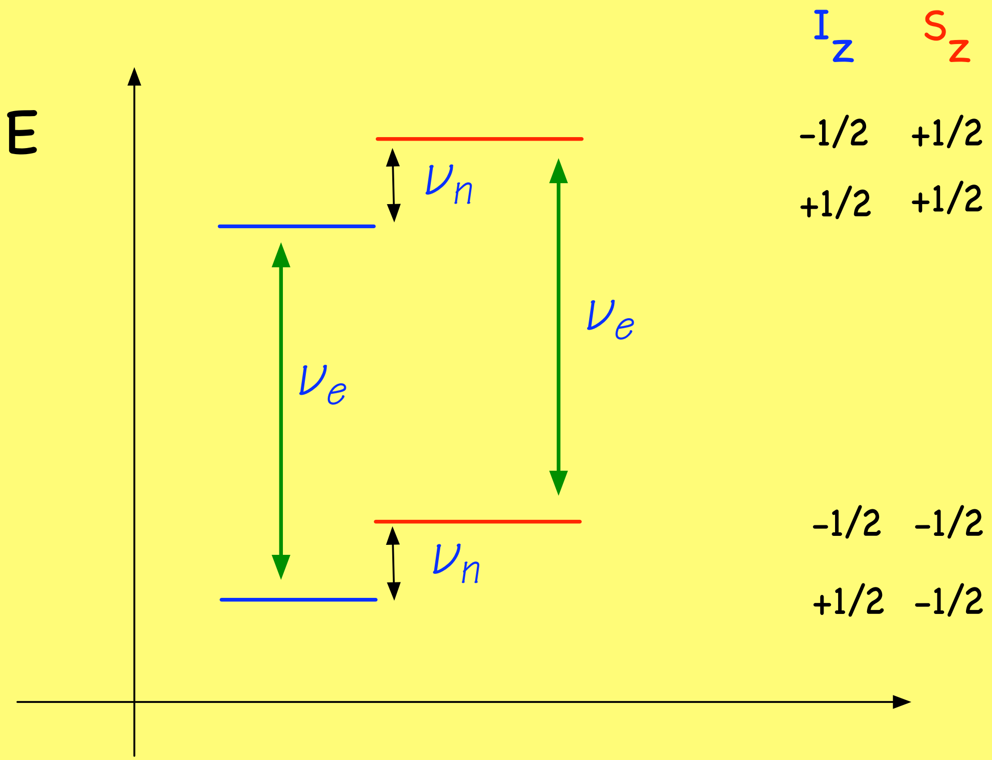
1) $\nu_{RF} \sim \nu_e$ get transitions:

-- ↔ -+

++ ↔ +-

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

Pure electron spin transitions ↑ → ↓ is electron paramagnetic resonance EPR (or ESR)



2) $\nu_{RF} \sim \nu_n$ get transitions: $+- \leftrightarrow --$

$++ \leftrightarrow -+$

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

and ν_n = 106.5 MHz at 2.5 T

Pure nuclear spin transitions

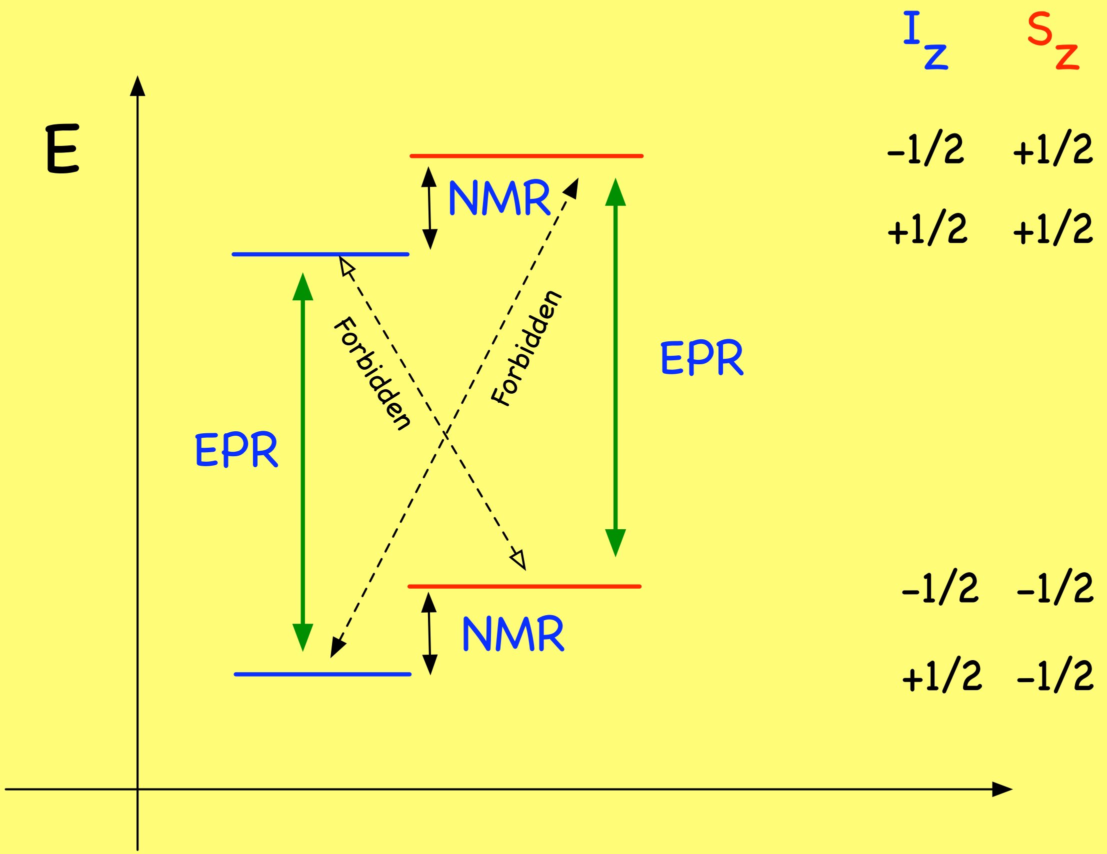
Nuclear Magnetic Resonance $\uparrow \rightarrow \downarrow$

(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.

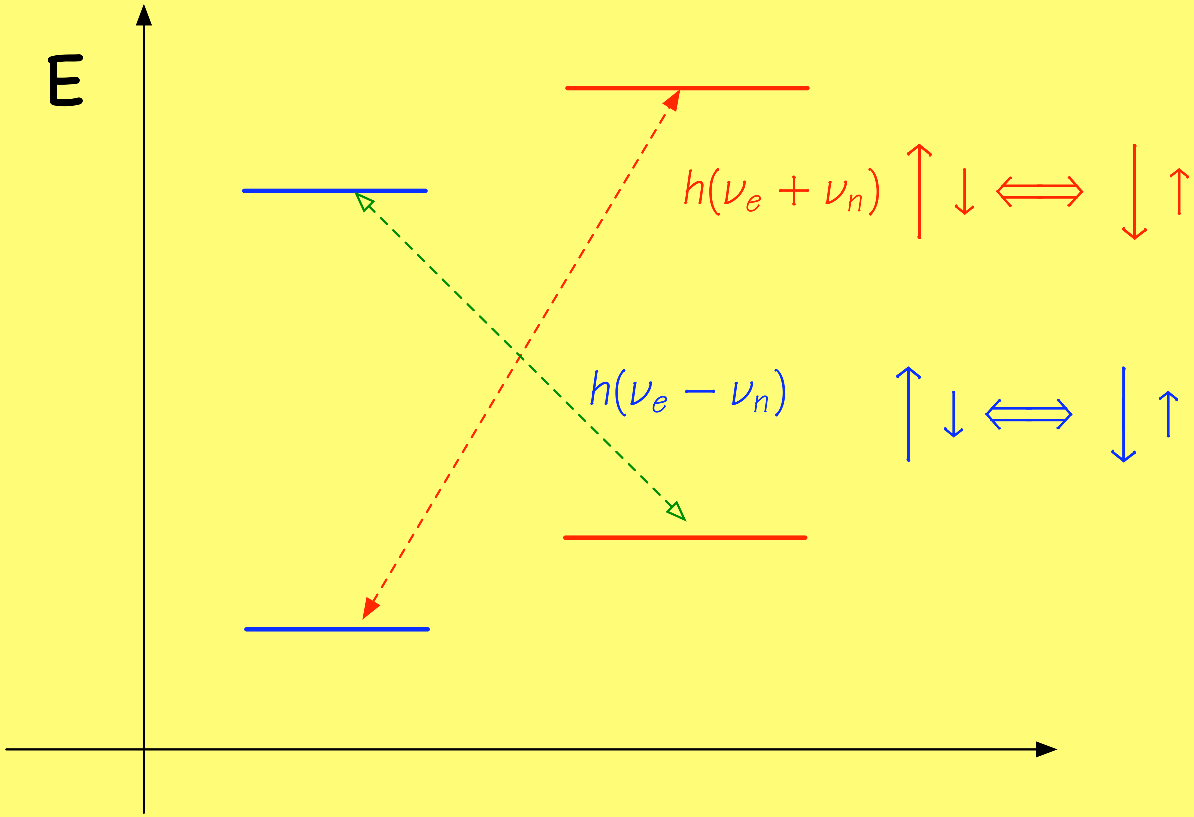


However there exists the dipole-dipole interaction

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.

Forbidden Transitions



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

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

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

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

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 $\uparrow \rightarrow \downarrow$

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)

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

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

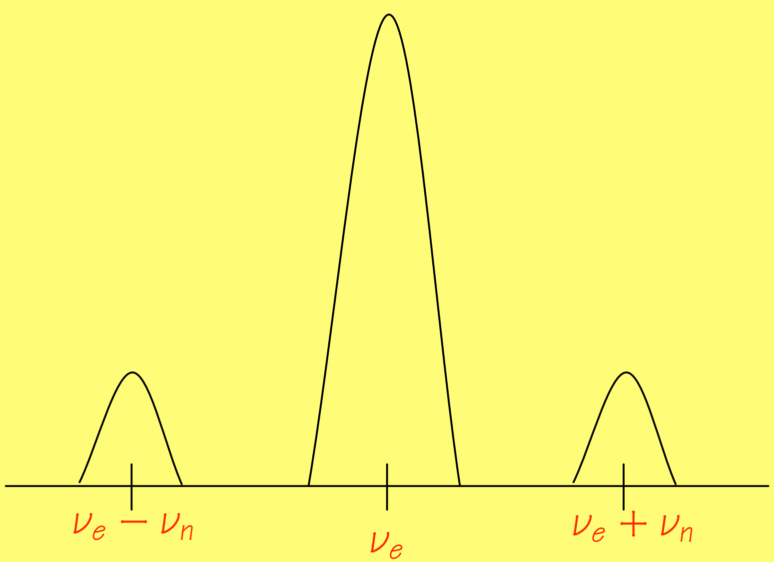
Neighboring nuclei coupled by dipole-dipole interaction which produces energy conserving processes such as

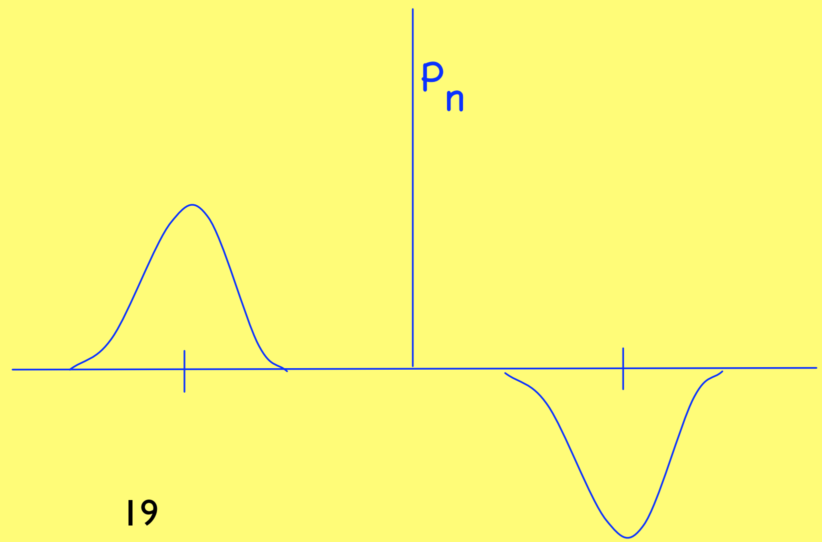
$$\uparrow \downarrow \Longleftrightarrow \downarrow \uparrow$$

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



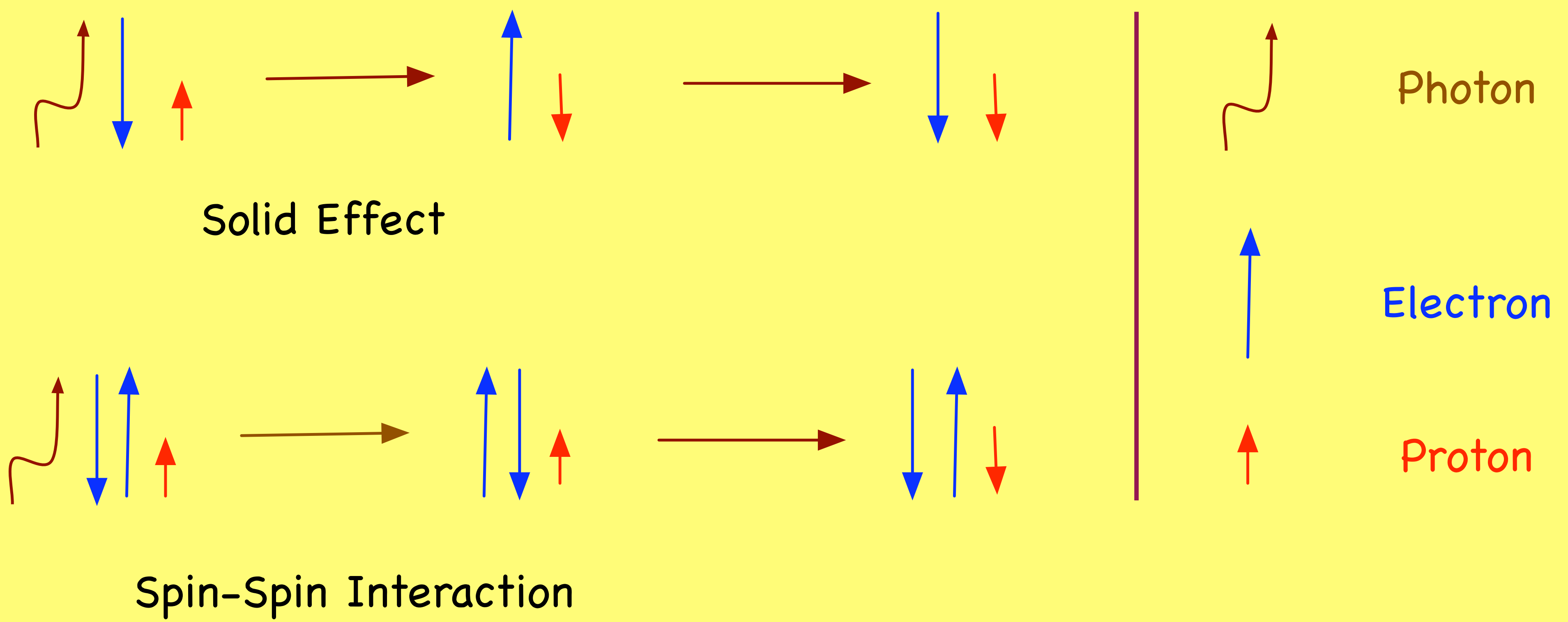


For other nuclear species (deuteron) the enhancement frequencies ($\nu_e \pm \nu_n$) will be different from that of the proton because of the different Lamor frequency.

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

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



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

Benzene
Toluene
Ethanol
Methanol
Propanol
Polyethylene
Polystyrene
LiF
Wax
Para Wax

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

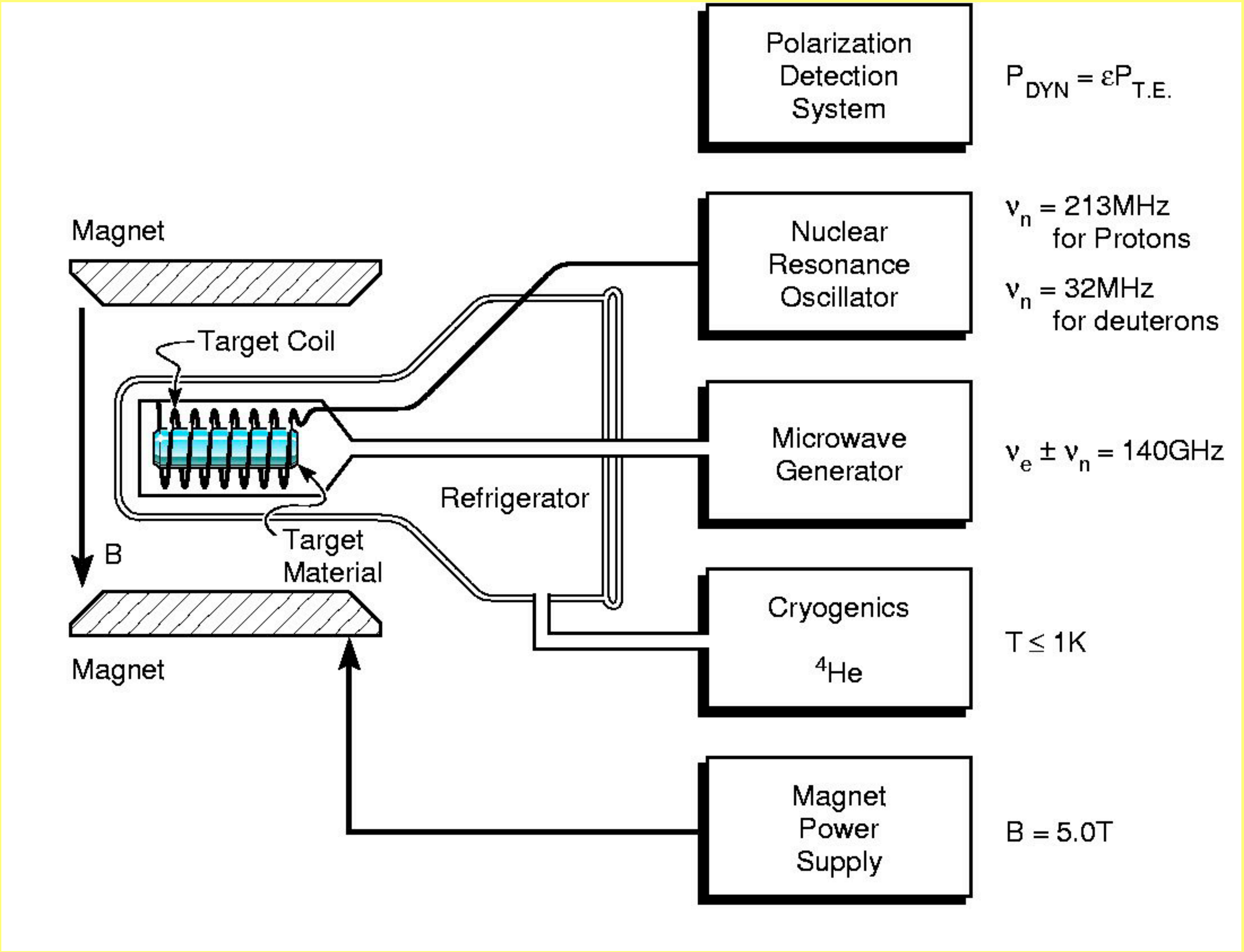
Benzene+ Ether
Propanol + Ethanol
Ethanol + Water
Ethanol + Methanol
Ethanol + Propanol
Ethanol + Diethylether
Butylalcohol + Methanol
Methanol + Propanol
$NaBH_4 + NH_4F + NH_3$

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
ν_n = 213MHz for Protons
ν_n = 32MHz for deuterons
Microwave Generator
$\nu_e \pm \nu_n$ = 140GHz
Cryogenics
^{4}He
$T \leq 1K$
Magnet Power Supply
B = 5.0T
Magnet
Target Coil
Refrigerator
B
Target Material
Magnet

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

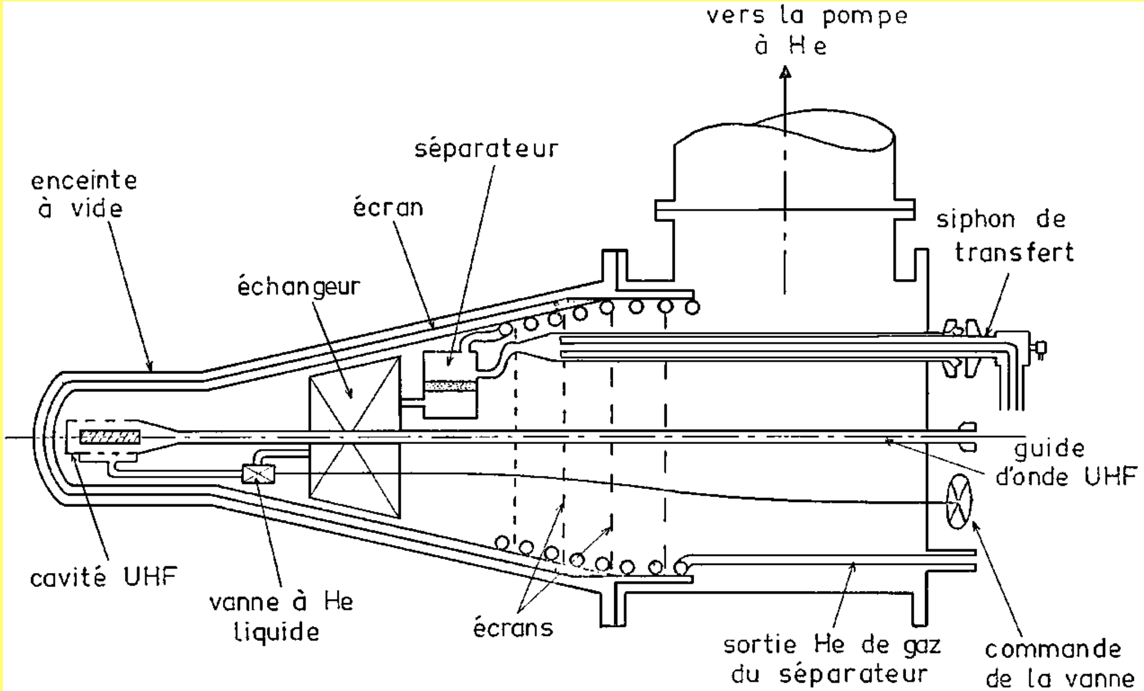


Figure 7 Schematic of the ^{4}He refrigerator described in [Ref. 28]. siphon de transfert = transfer siphon; vers la pompe à He = to the He pump; separateur = separator; écran = shield; exchangeur = exchanger; enceinte à vide = vacuum enclosure; vanne à He liquide = liquid helium (needle) valve; sortie 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.

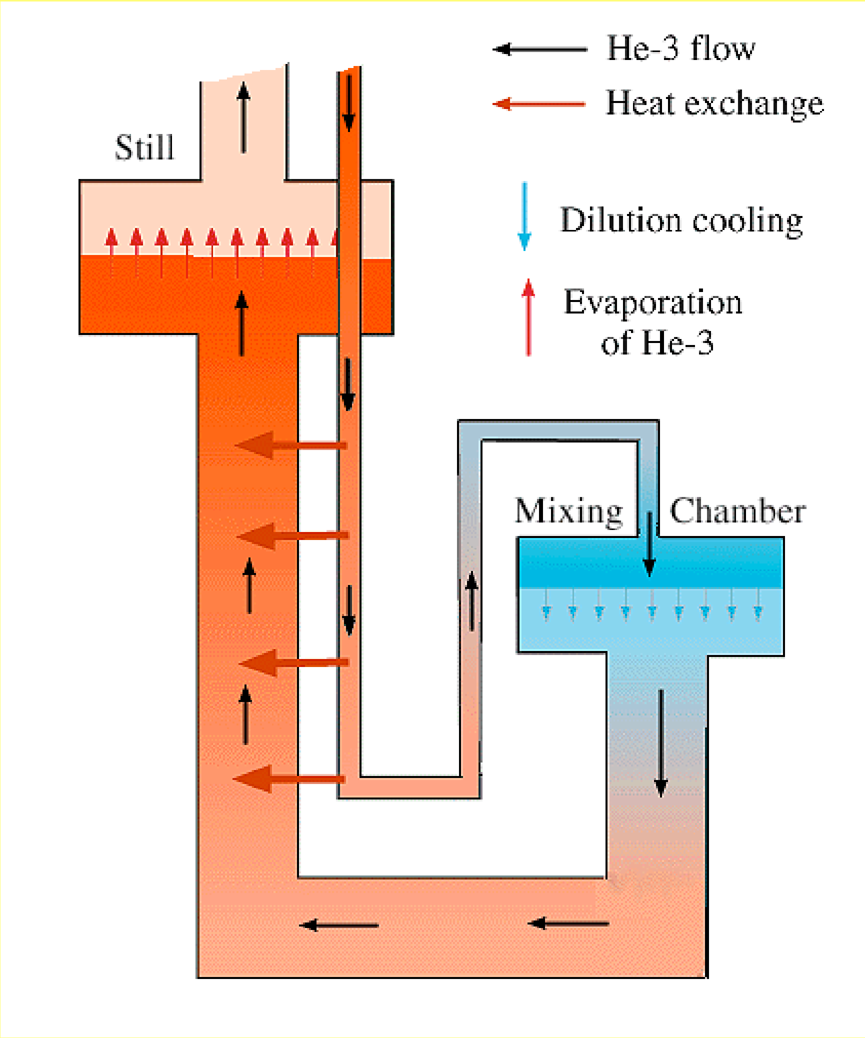
Here, liquid He is fed into a separator pot where the liquid phase is separated from the vapor phase by a sintered copper plate. The cold vapor is pumped away and used to cool the radiation shields and baffles that intercept the radiation heat load. Liquid helium flows through the separator plate into 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.

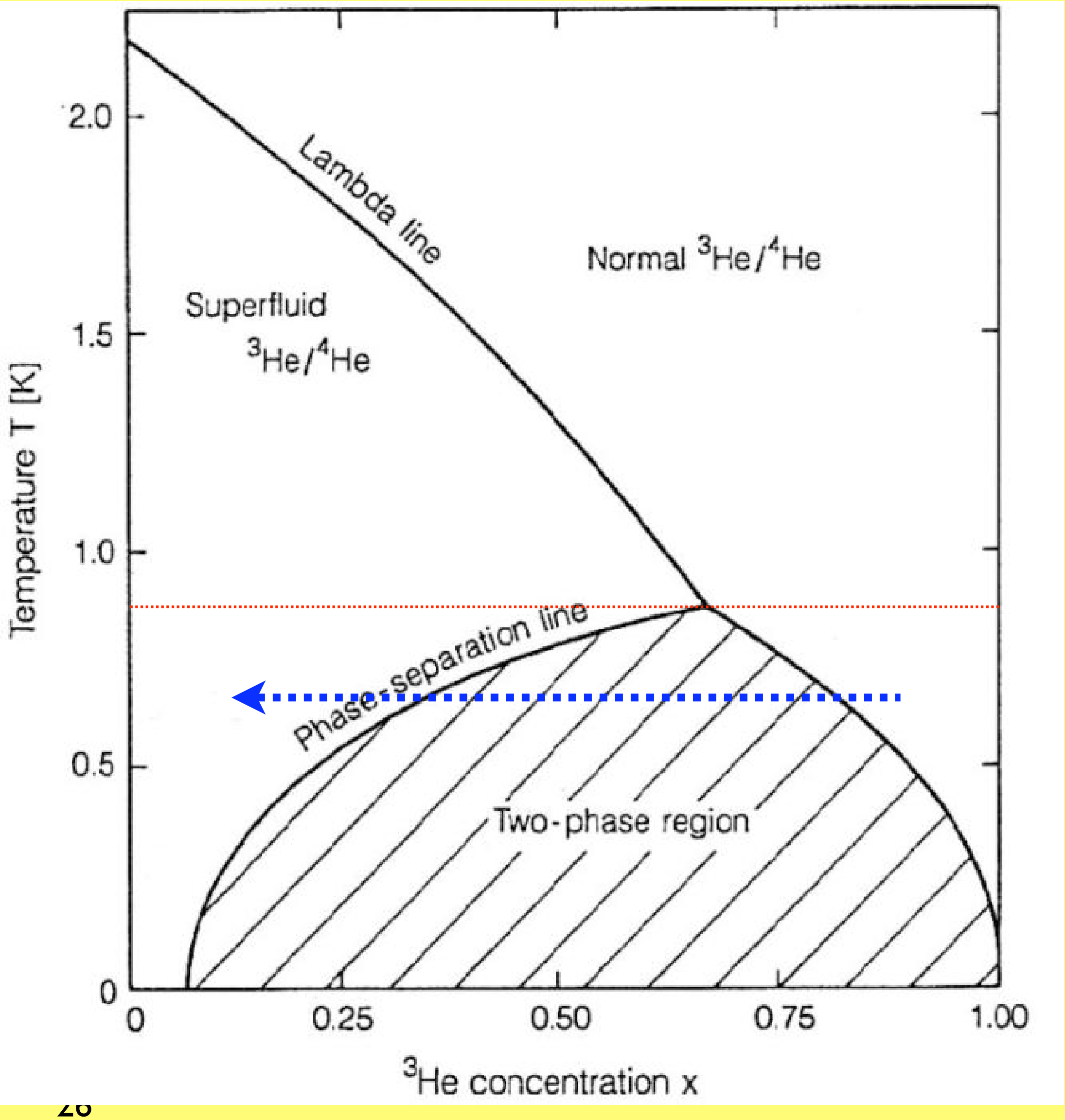
^{3}He Evaporation Refrigerators

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.





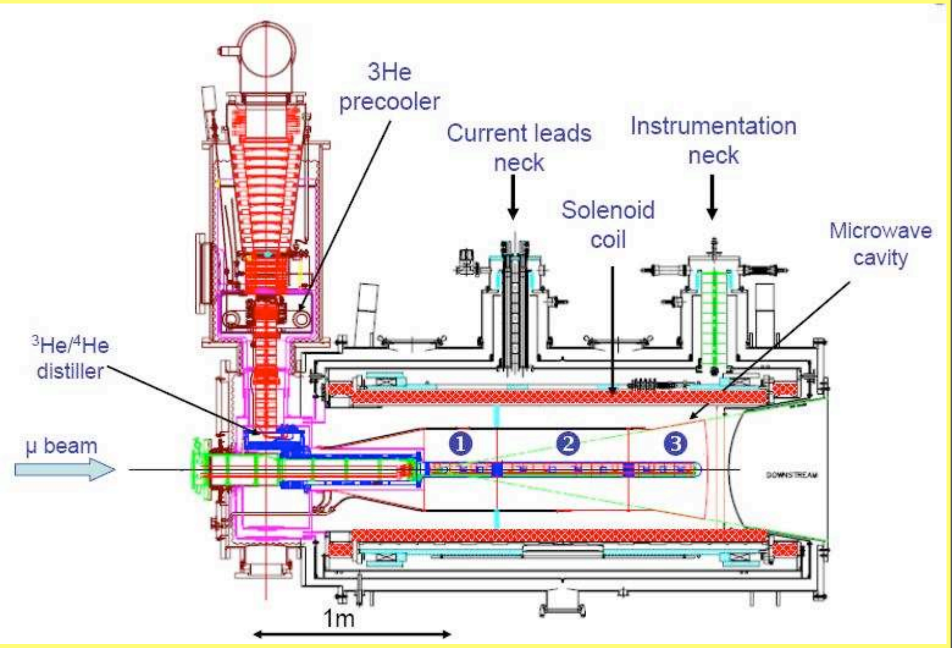
Polarizing Magnets – Solenoids

DNP works best when
B/T ≈ 5 – 10

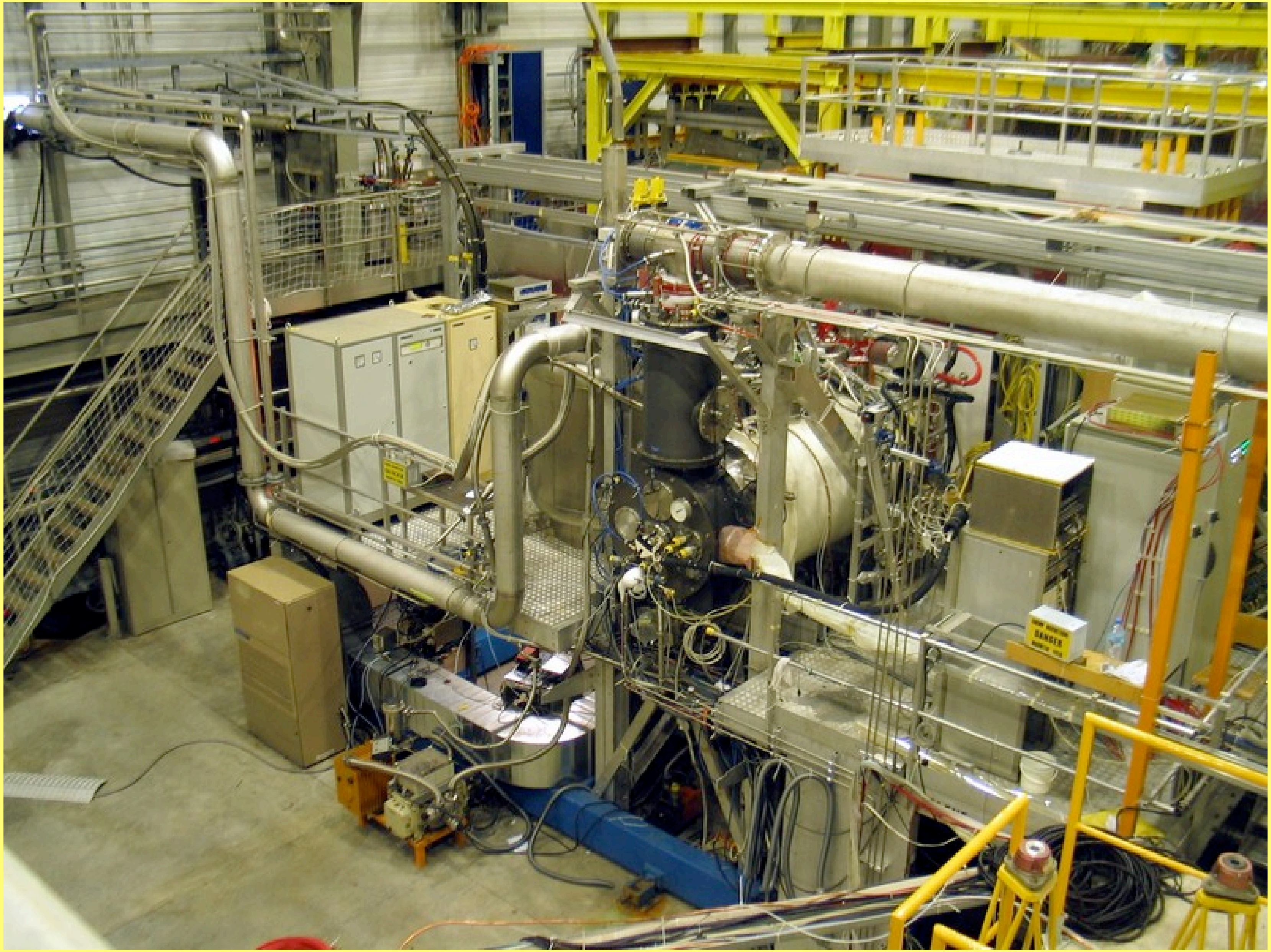
Modern magnets are 2.5 – 7.5T

COMPASS – CERN

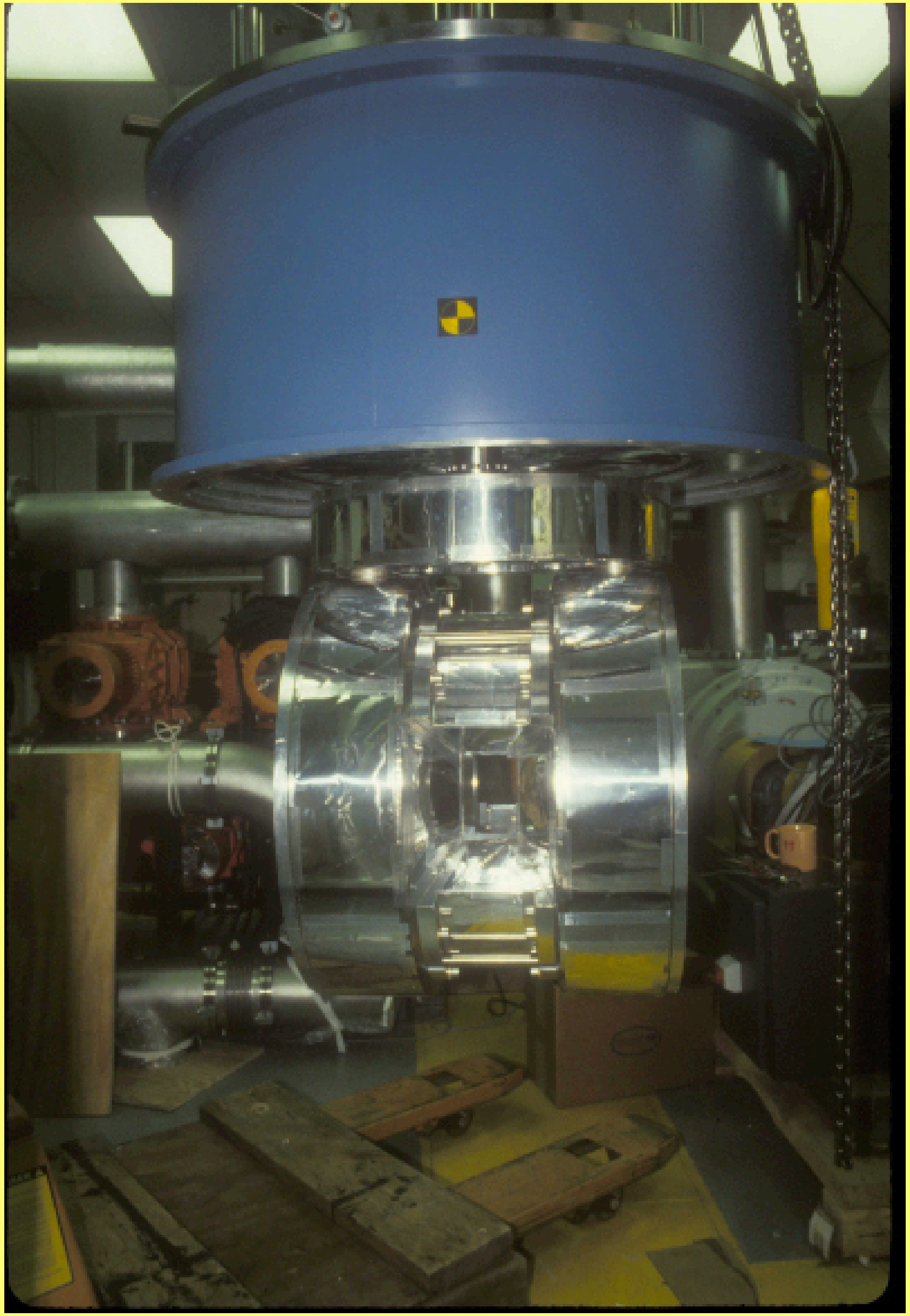
COmmon Muon Proton Apparatus
for Structure and Spectroscopy



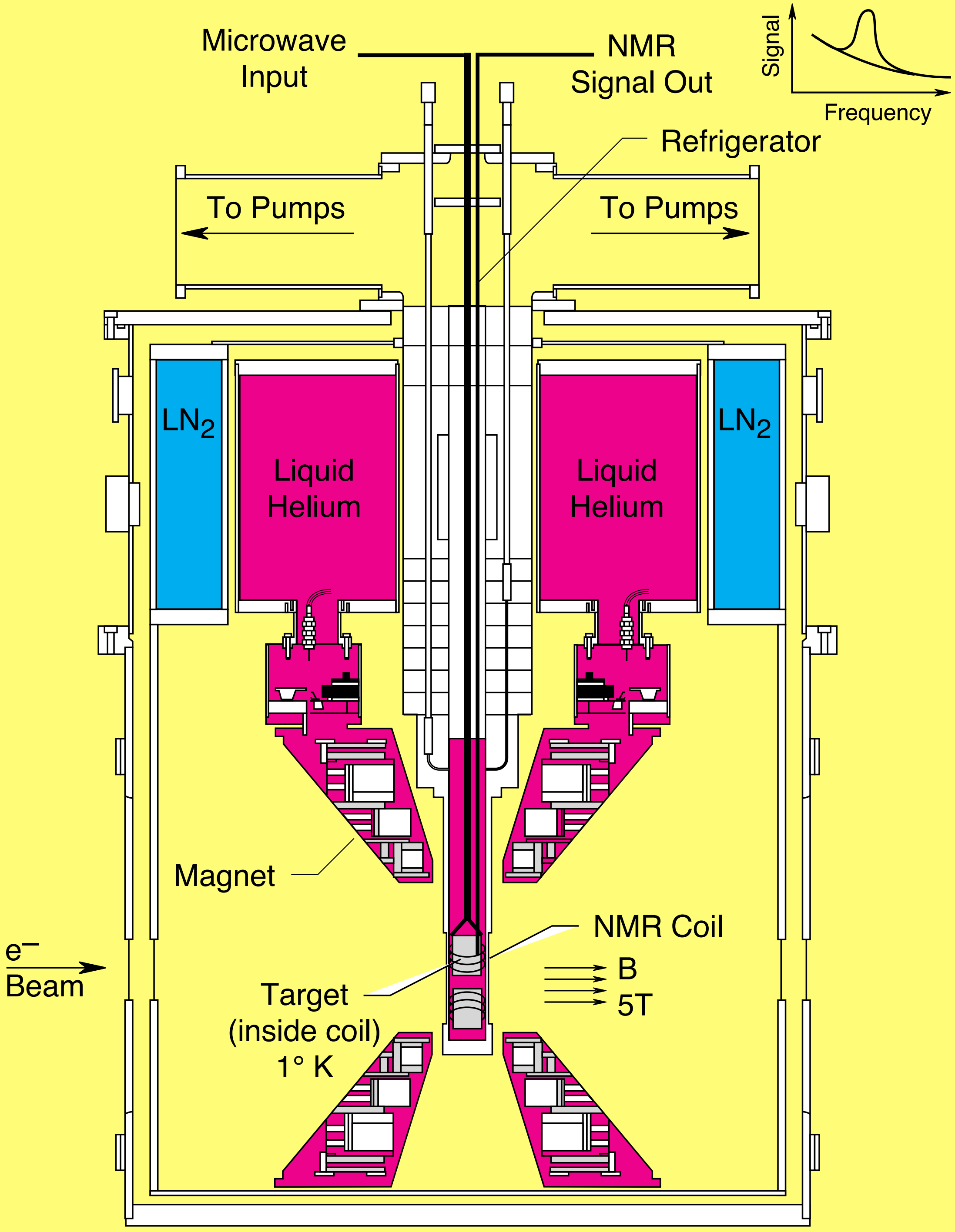
COMPASS - CERN

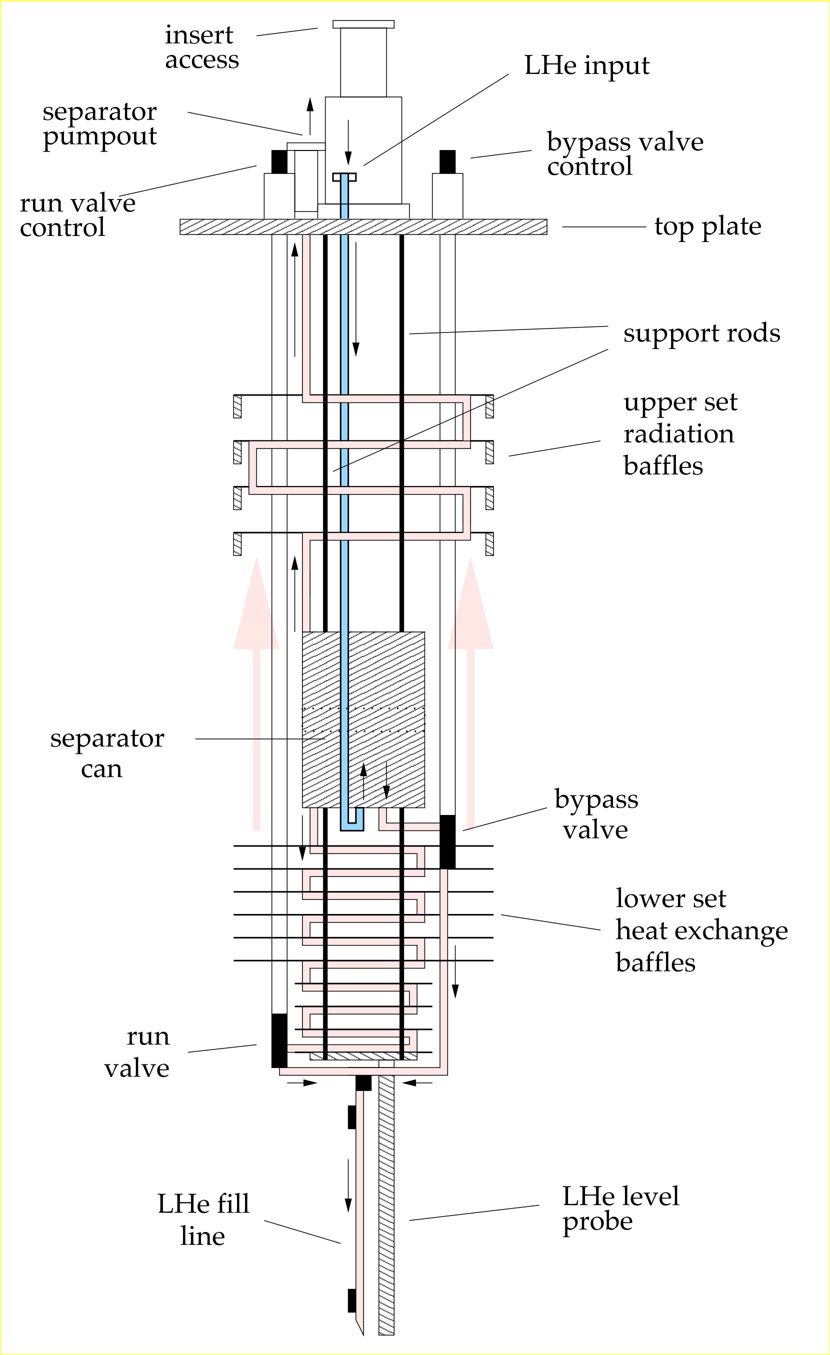


Polarizing Magnets- Split Pair UVA/SLAC/JLAB Target



UVA/SLAC/JLAB Target





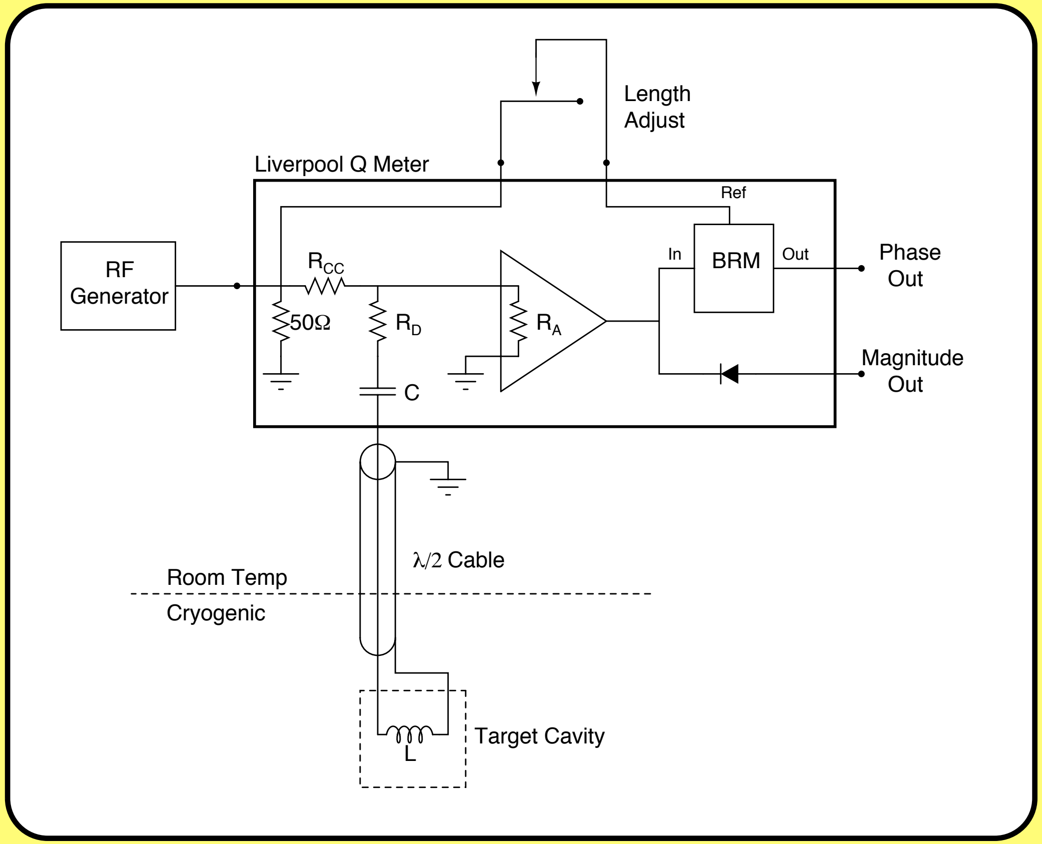
Polarization Measurement

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

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

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



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

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

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

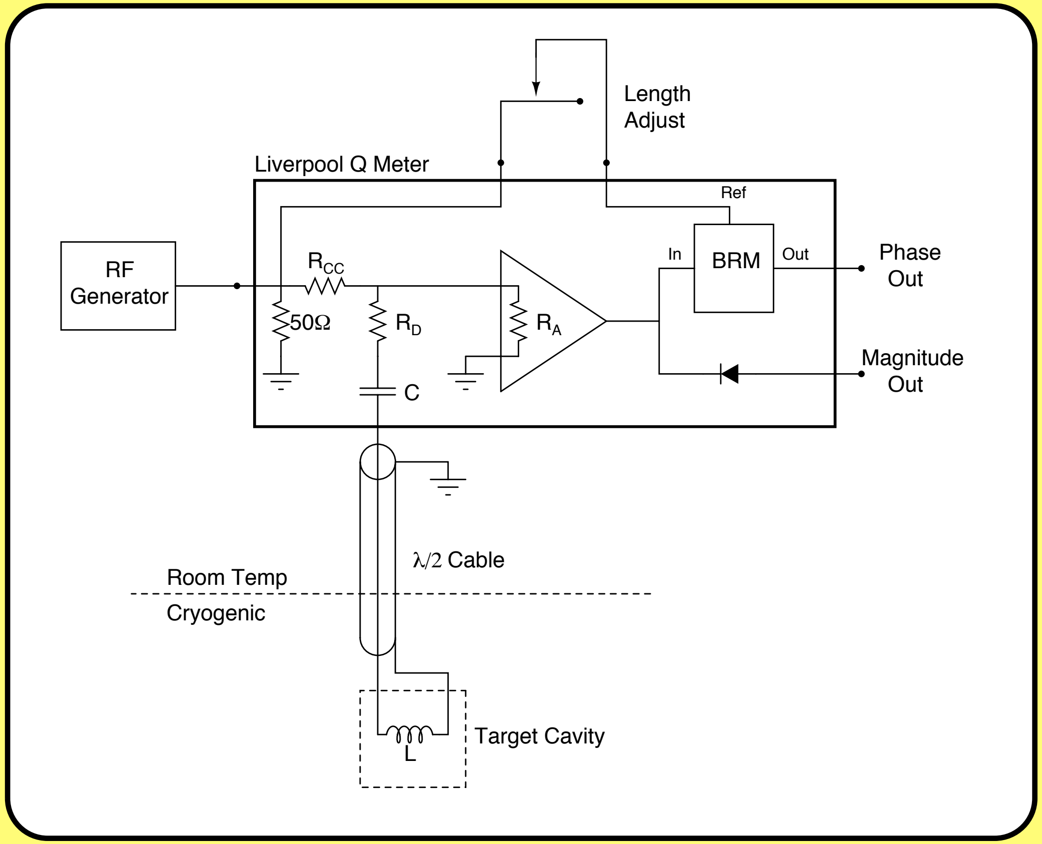
Inductance (and impedance) changes when the material absorbs or emits energy and thus the voltage, $V(\omega,\chi)$

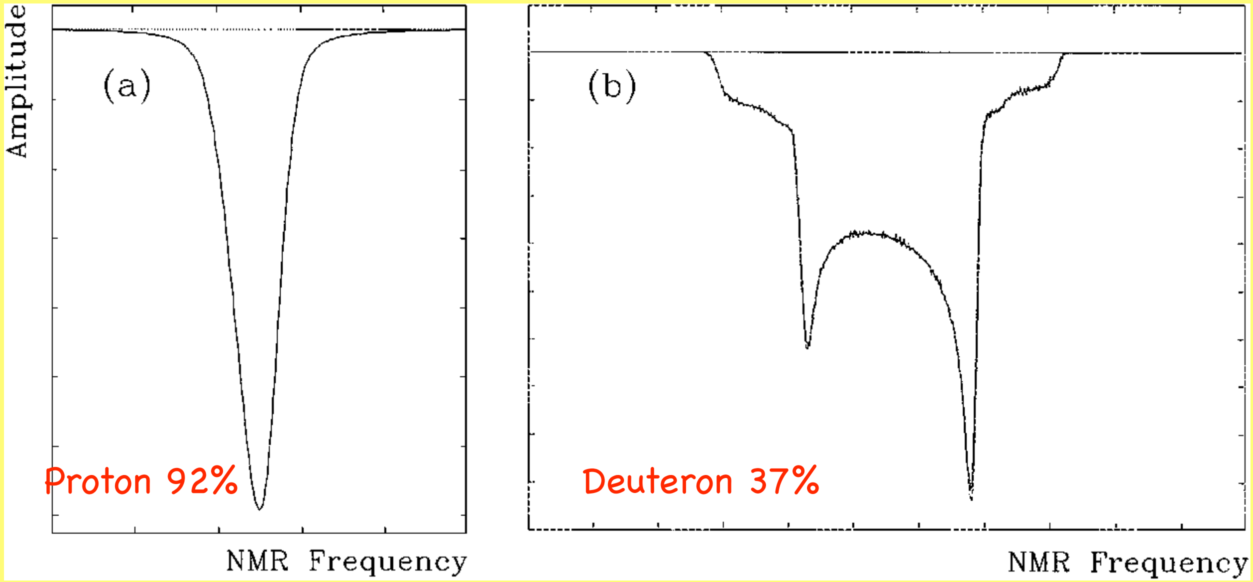
$$S(\omega) = Re(V(\omega,\chi) - V(\omega,0)) \simeq \chi''(\omega)$$

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

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

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





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.

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

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

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

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

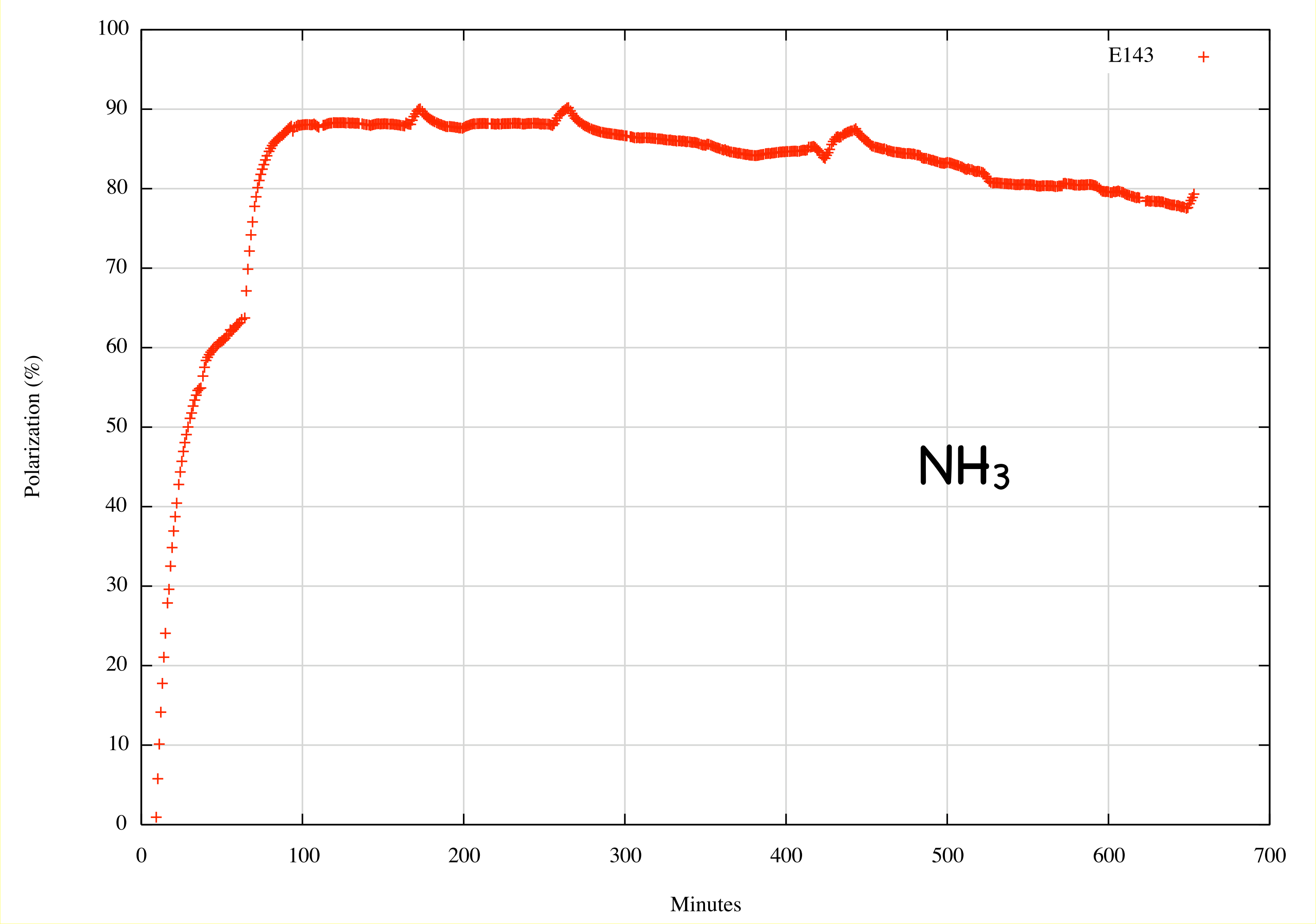
$$t^{-1} \propto \rho(f \cdot P)^2$$

important to optimized f and P

ρ is density

Target materials

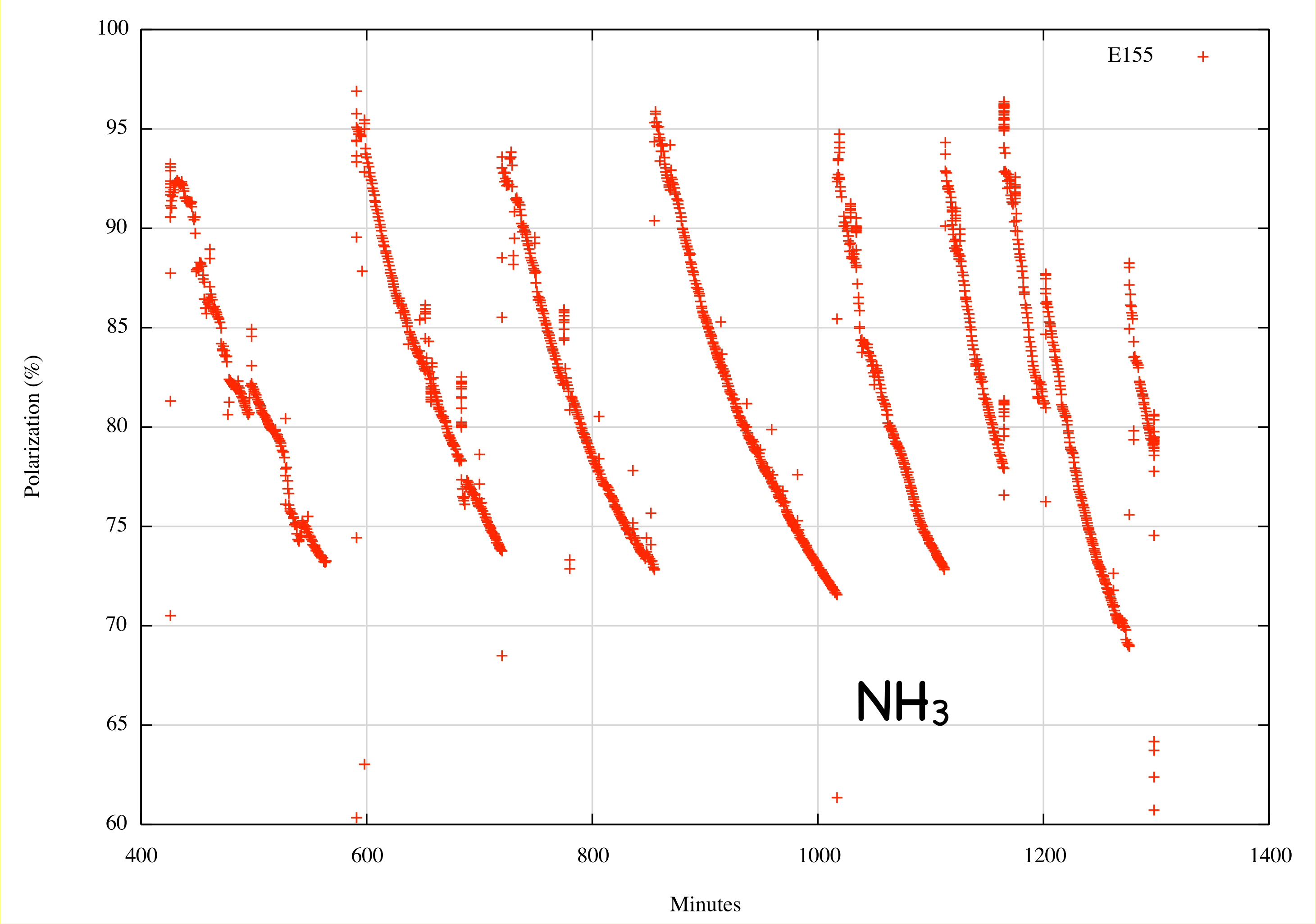
Performance and Experience



Polarization growth, mistuning, beam on/off, and decay

Target materials

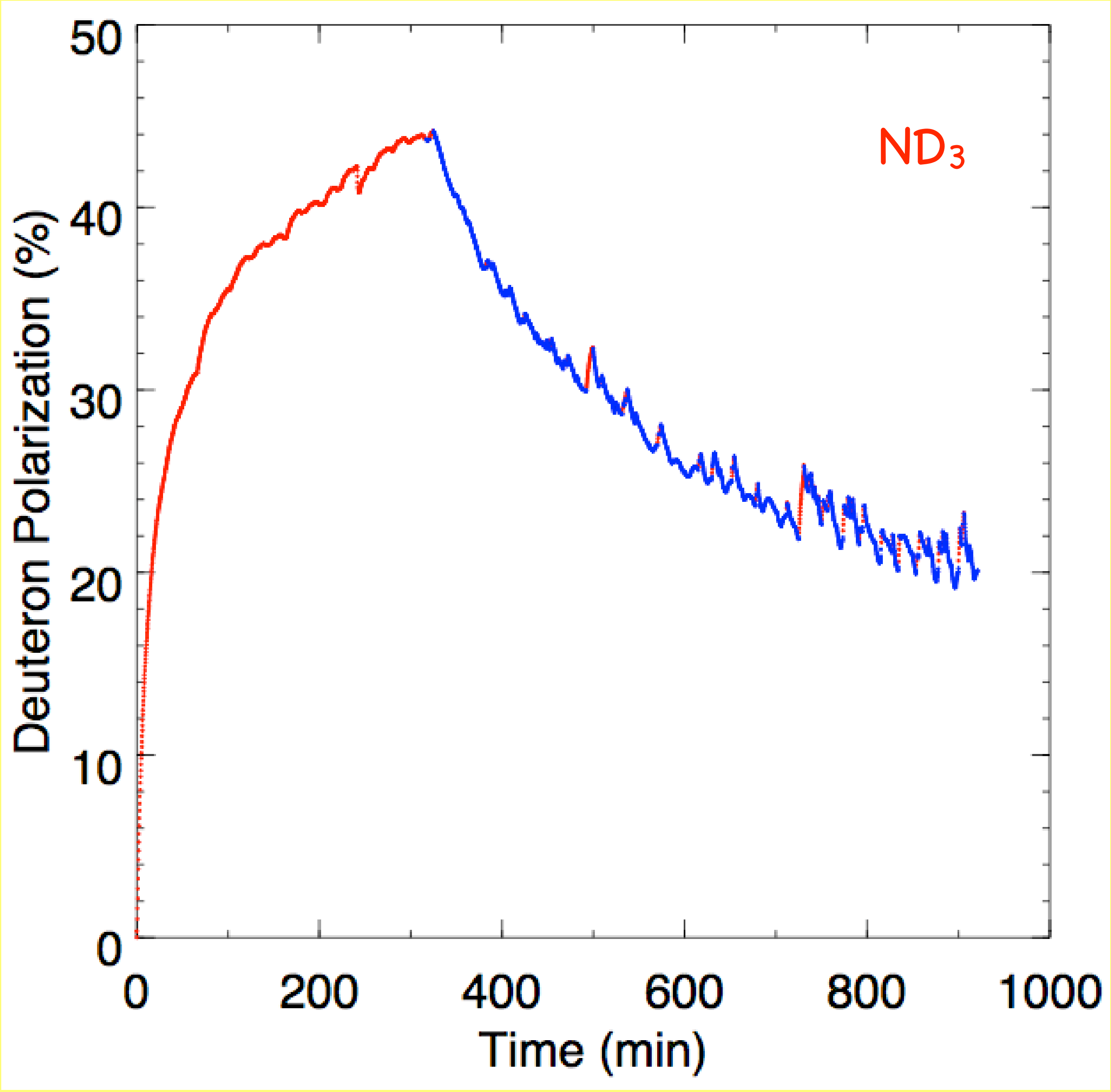
Performance and Experience



Polarization growth, radiation damage, decay of material

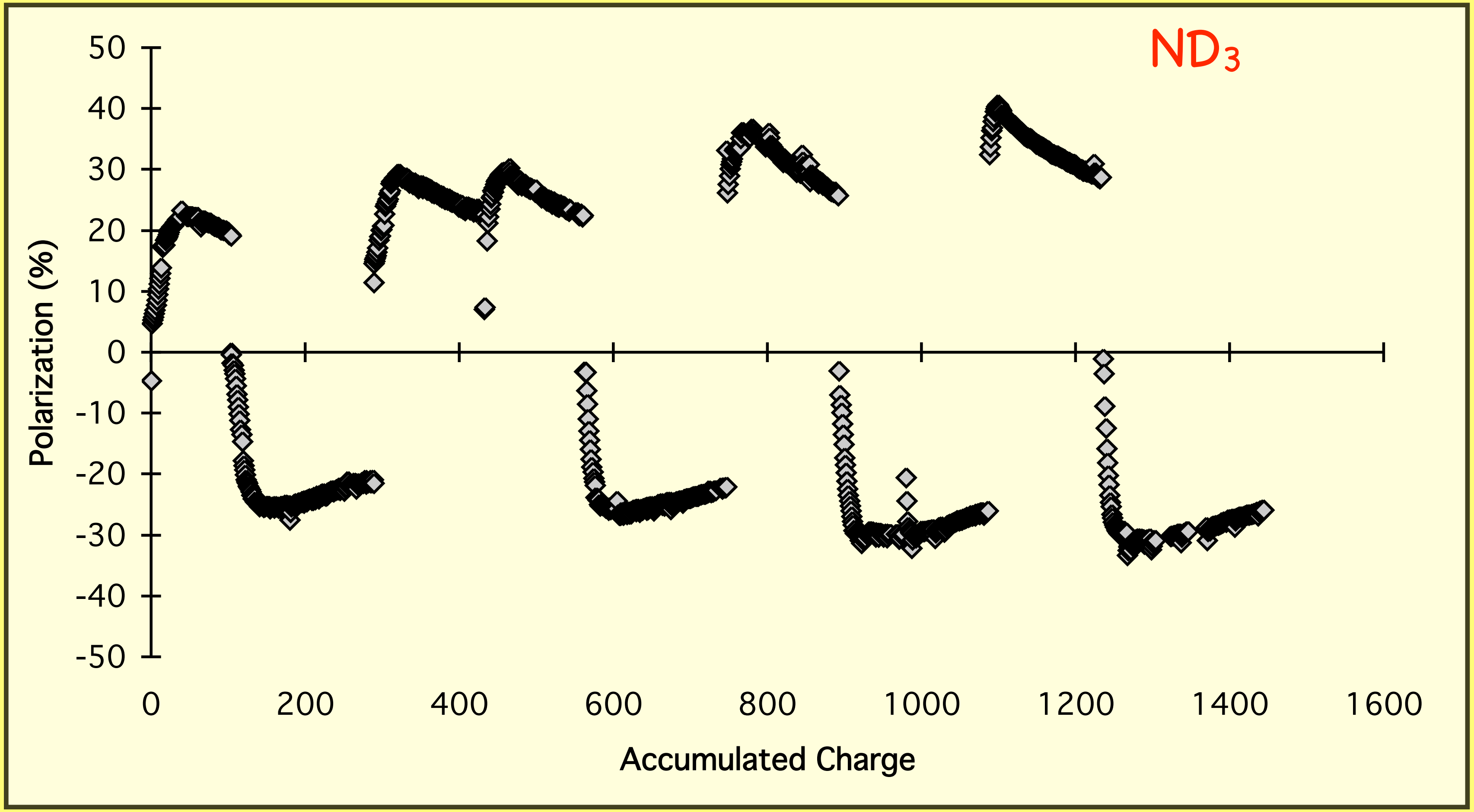
Target materials

Performance and Experience

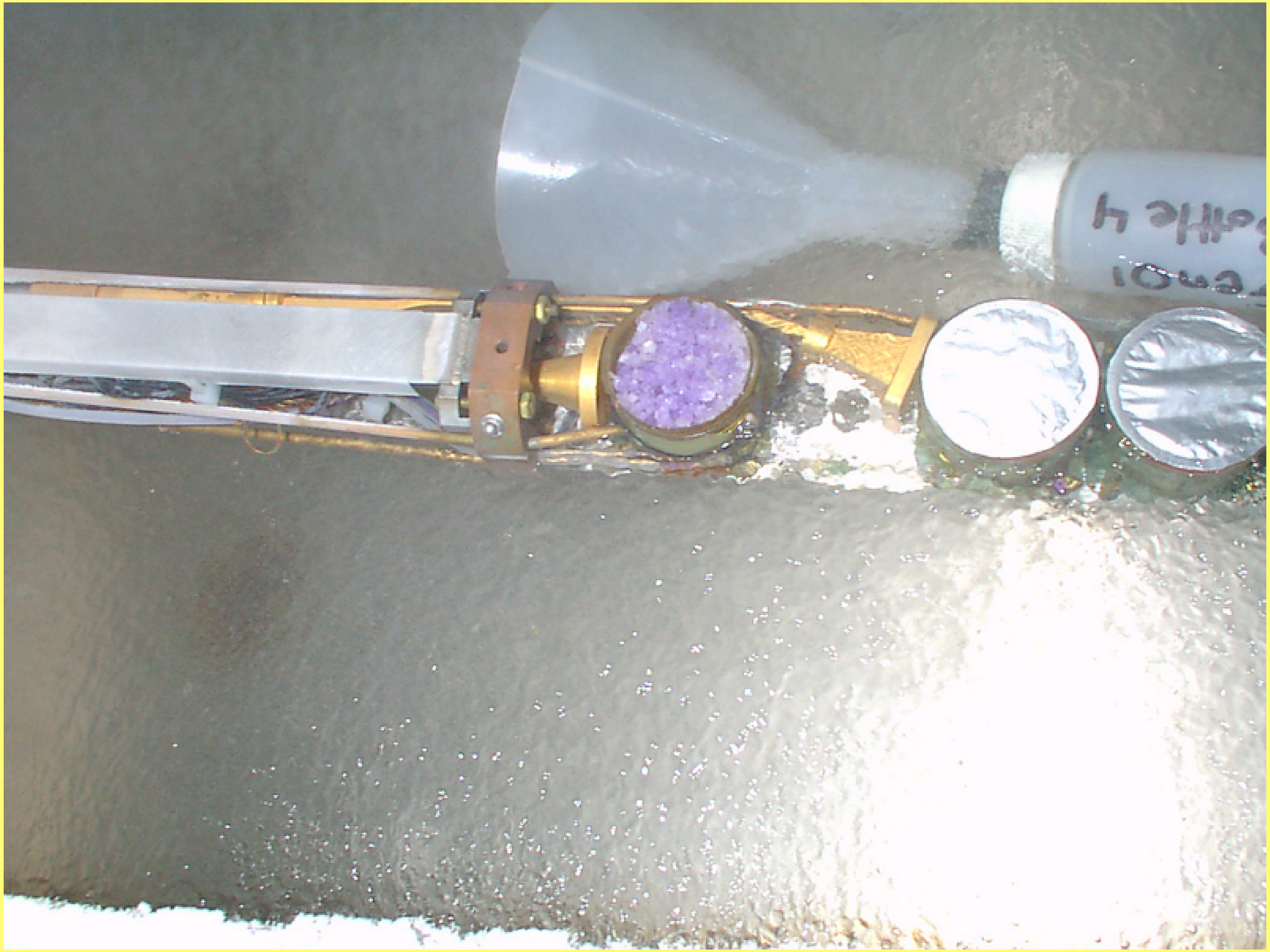


Target materials

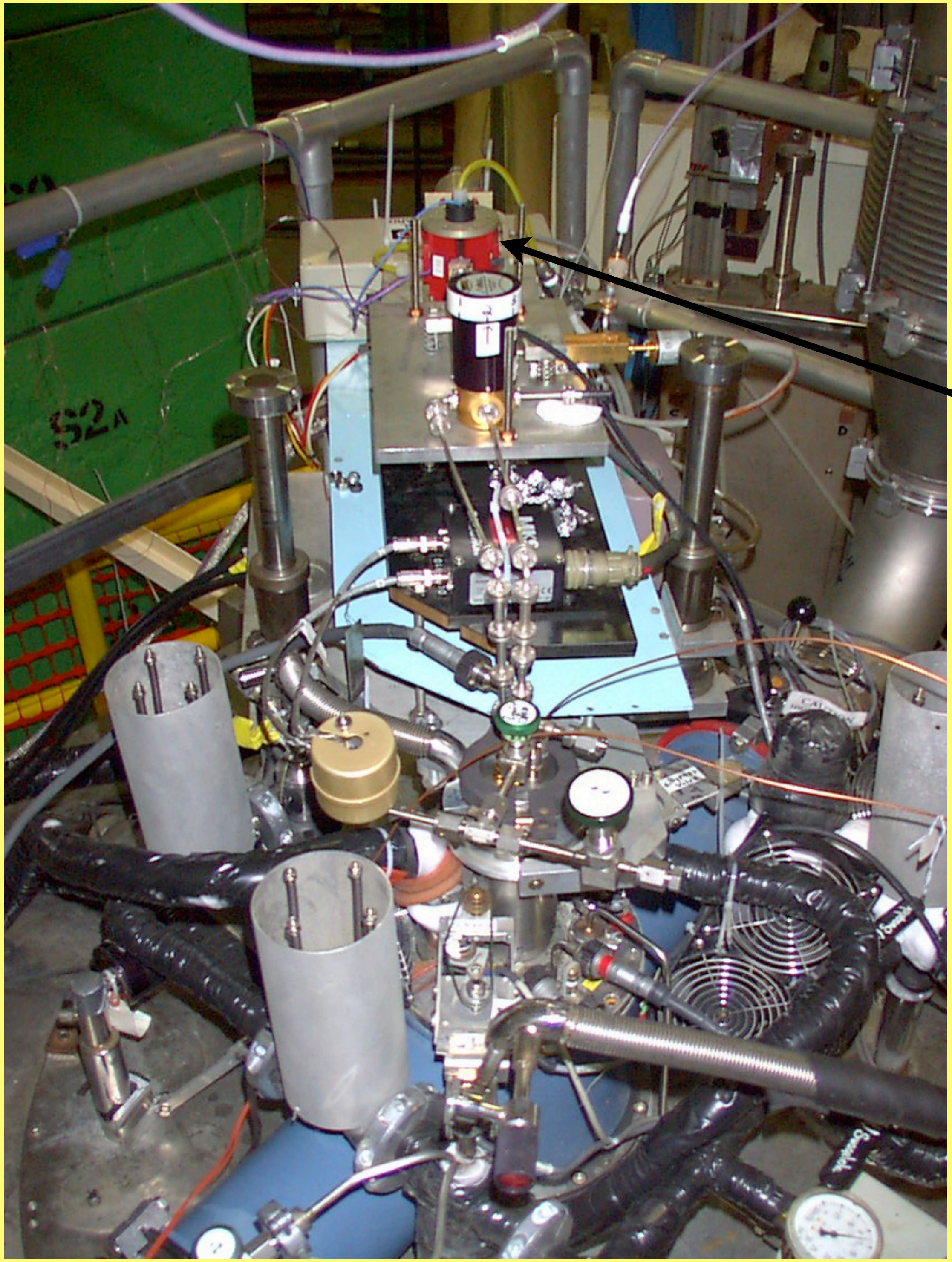
Performance and Experience



Polarization growth, Radiation damage, anneal, reverse sign



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)

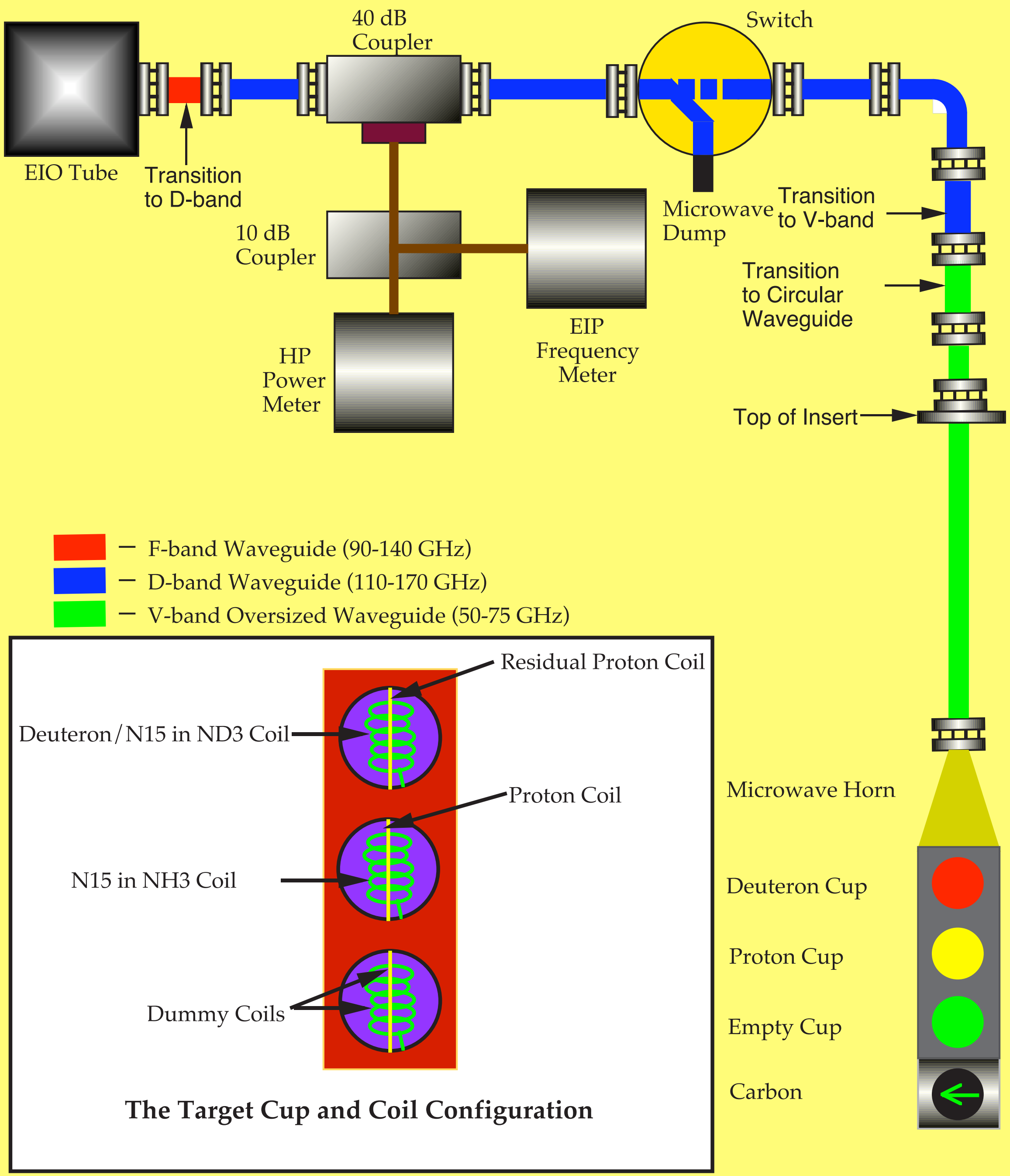
140 GHz Extended Interaction Oscillator (EIO)

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.

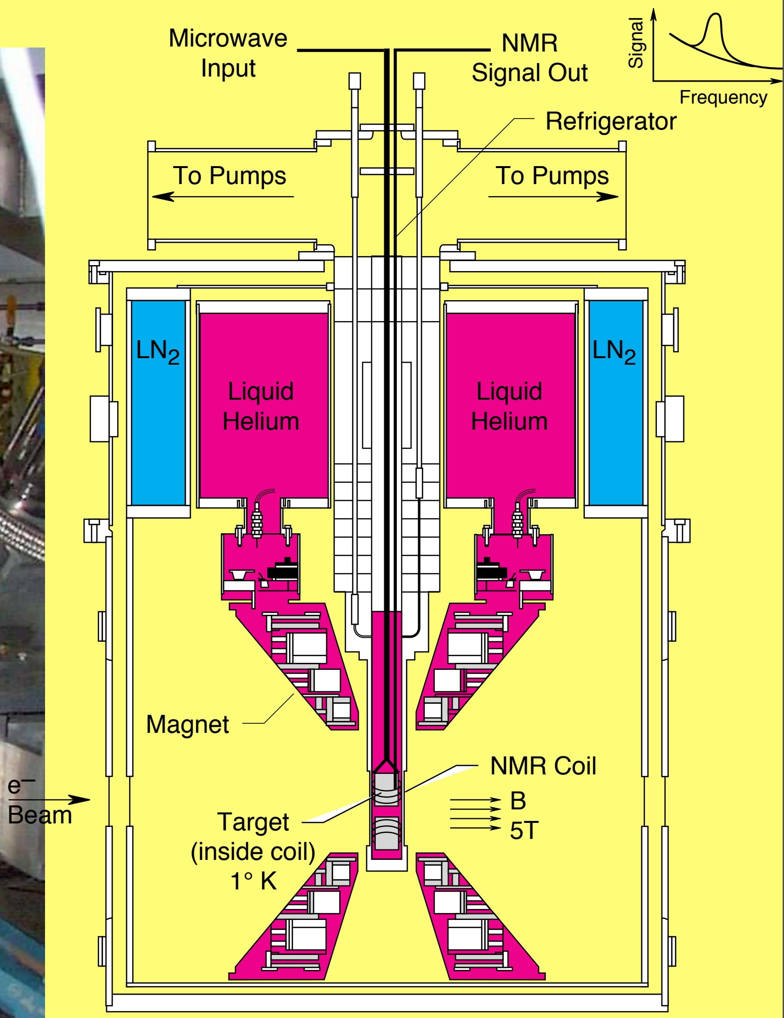
Also: klystrons, IMPATT and Gunn diodes

IMPact ionization Avalanche Transit-Time

UVA/SLAC/JLAB Target

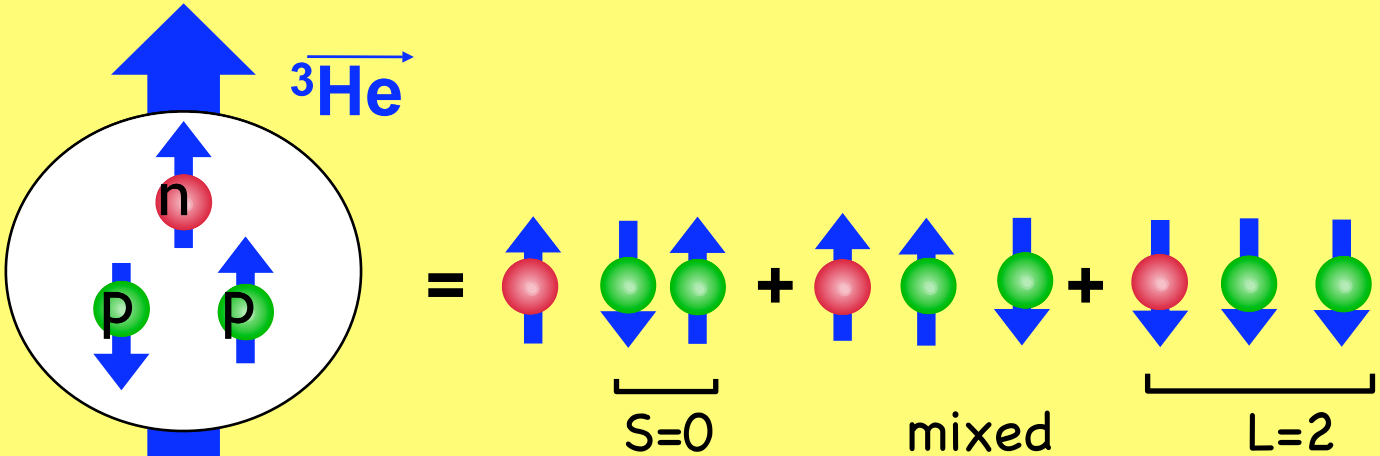


EIO Tube
Transition
to D-band
40 dB
Coupler
Switch
Microwave
Dump
Transition
to V-band
10 dB
Coupler
Transition
to Circular
Waveguide
HP
Power
Meter
EIP
Frequency
Meter
Top of Insert
— F-band Waveguide (90-140 GHz)
— D-band Waveguide (110-170 GHz)
— V-band Oversized Waveguide (50-75 GHz)
Residual Proton 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



Microwave Input
NMR Signal Out
Signal
Frequency
Refrigerator
To Pumps
To Pumps
LN_2
Liquid Helium
Liquid Helium
LN_2
Magnet
NMR Coil
e⁻ Beam
Target (inside coil) 1° K
B 5T

Polarized ^3He as polarized n-target



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

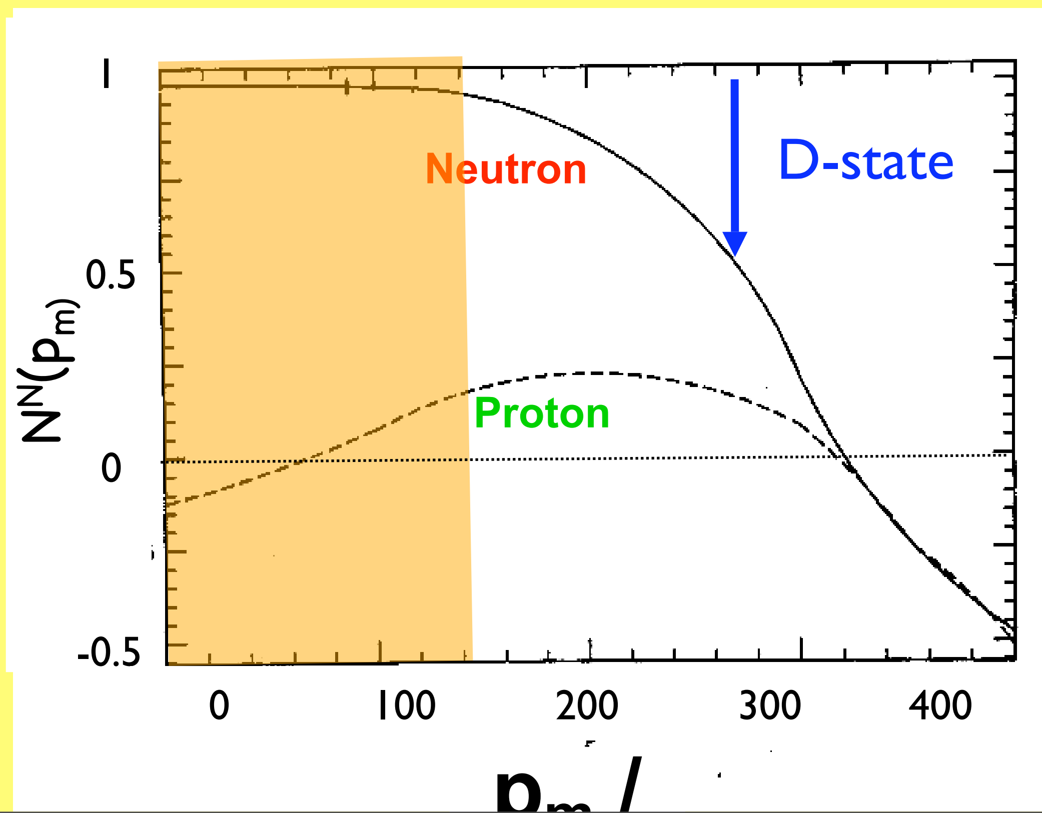
S–state 90%

S′–state 2%

D–state 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)

Polarized ^{3}He 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

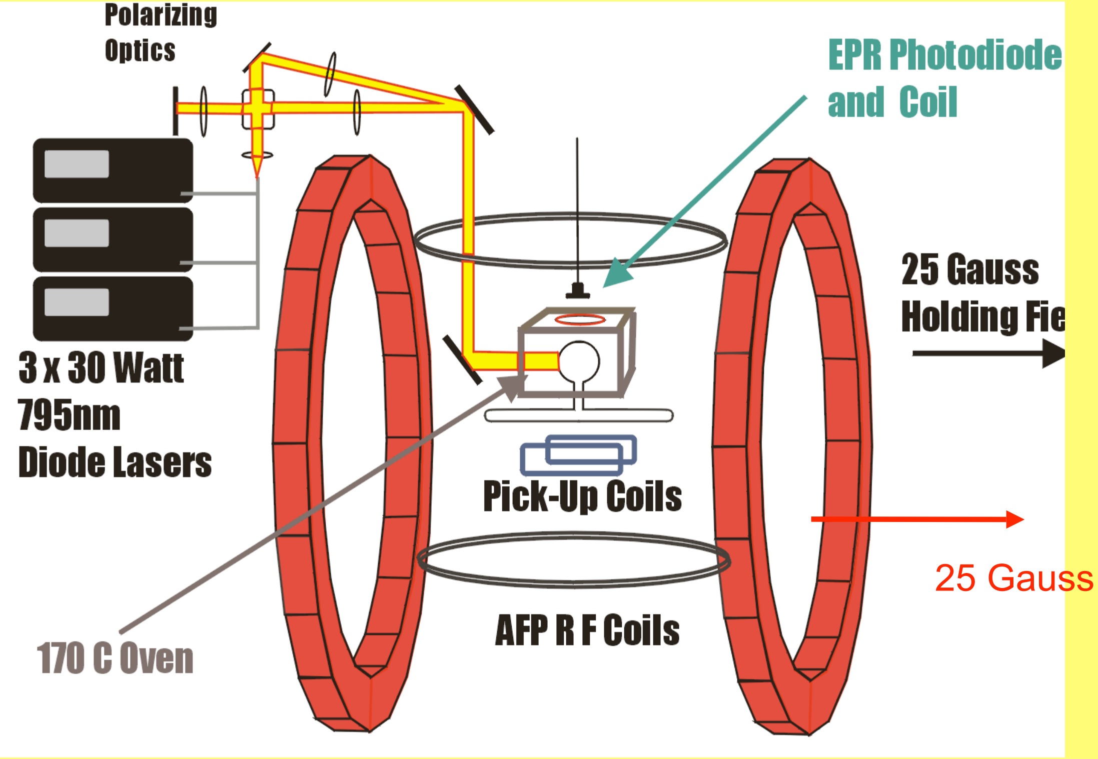
Standard technique:

- optical pumping of Rb vapor, followed by polarization transfer to ^{3}He 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%

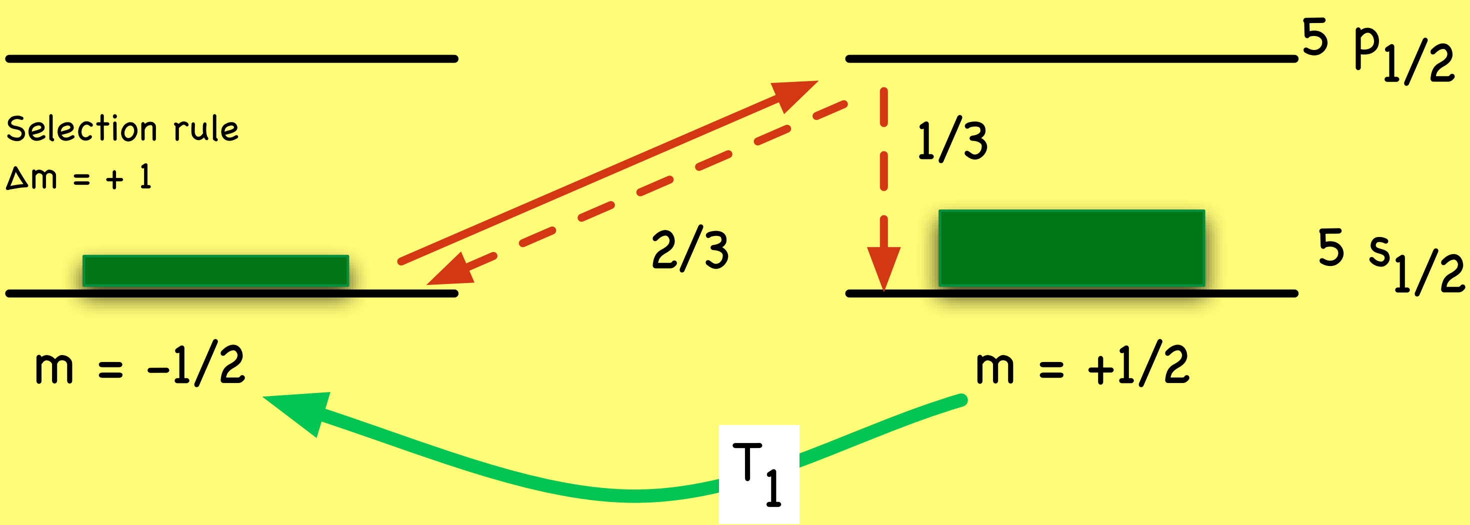
Hall A ^{3}He target



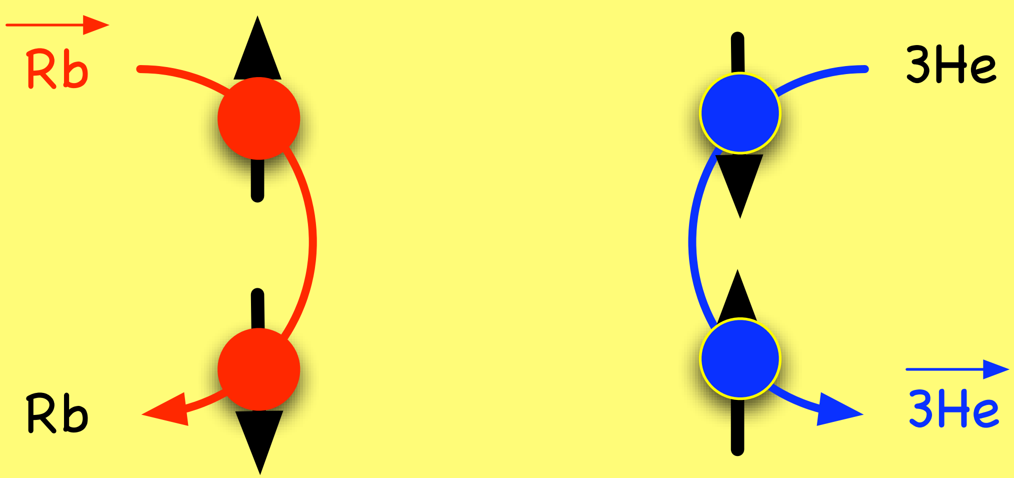
Latest development:

- optical pumping of Rb/K mixture

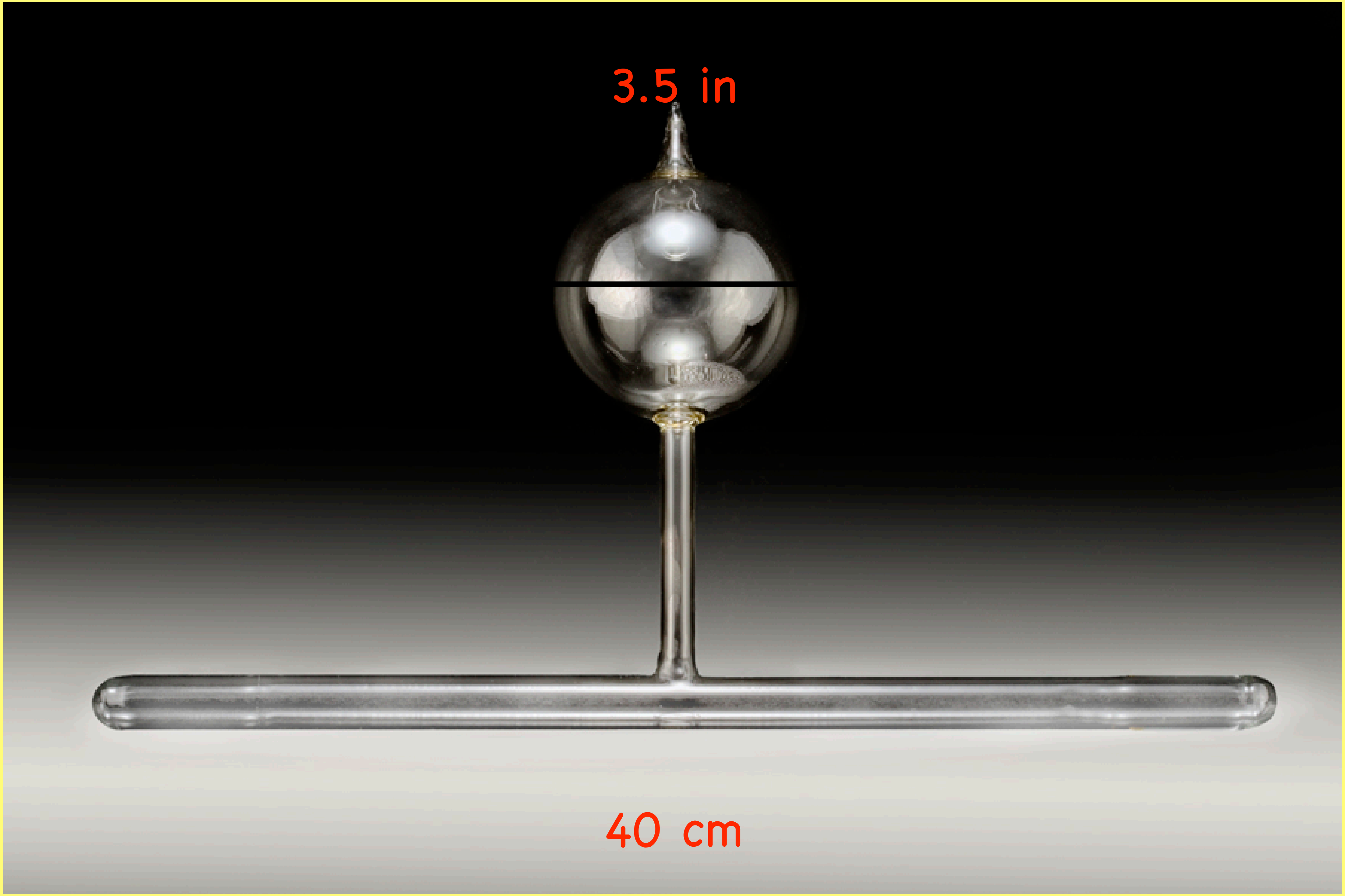
Optical pumping of Rubidium



Spin-exchange between Rb and ^{3}He due to Hyperfine interaction
small cross section



new at JLAB : hybrid cells contain potassium, build-up time: 4.2 h


3.5 in
40 cm