BOA meeting, PSI, February 22, 2013

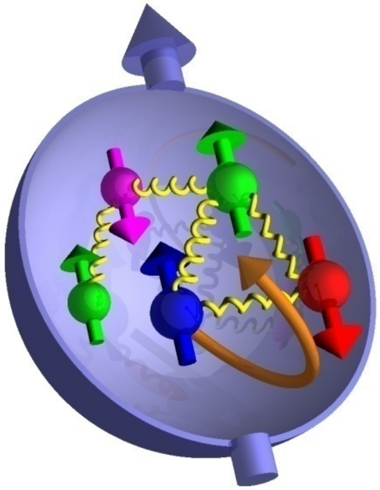
Neutron spin filtering with polarized protons

using photo-excited triplet states

P. Hautle

Paul Scherrer Institut
CH-5232 Villigen
Switzerland

Outline



WHY
Neutron spin filtering with polarized protons

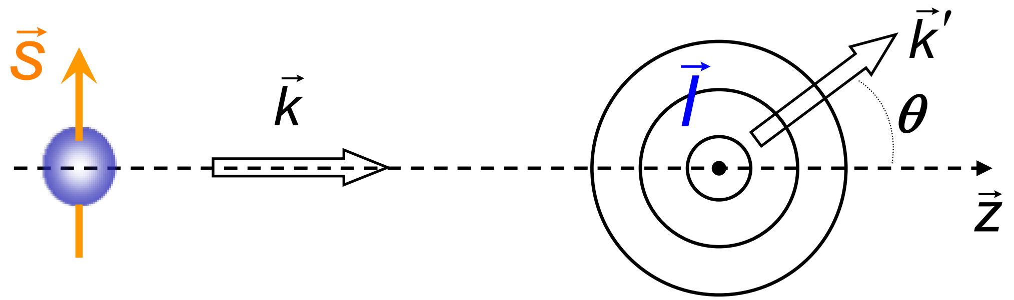
HOW to polarize protons :
DNP using photo-excited triplet states

RESULTS
2 beam times @ BOA

FUTURE

Neutron Nuclear Scattering

Cold neutrons → s-wave scattering



$$\psi = e^{i\vec{k}\cdot r} - f(\theta)\frac{e^{ikr}}{r}$$

$$\rightarrow e^{i\vec{k}\cdot r} - b\frac{e^{ikr}}{r}$$

Scattering length operator

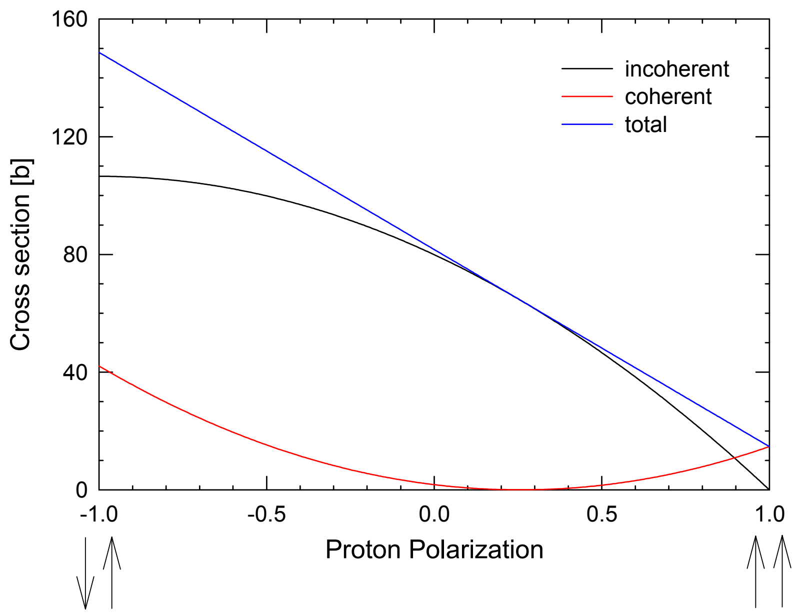
$$\hat{b} = b + b_N \, \vec{s}\cdot\vec{I}$$

→ ***depends on spin orientation***

proton:

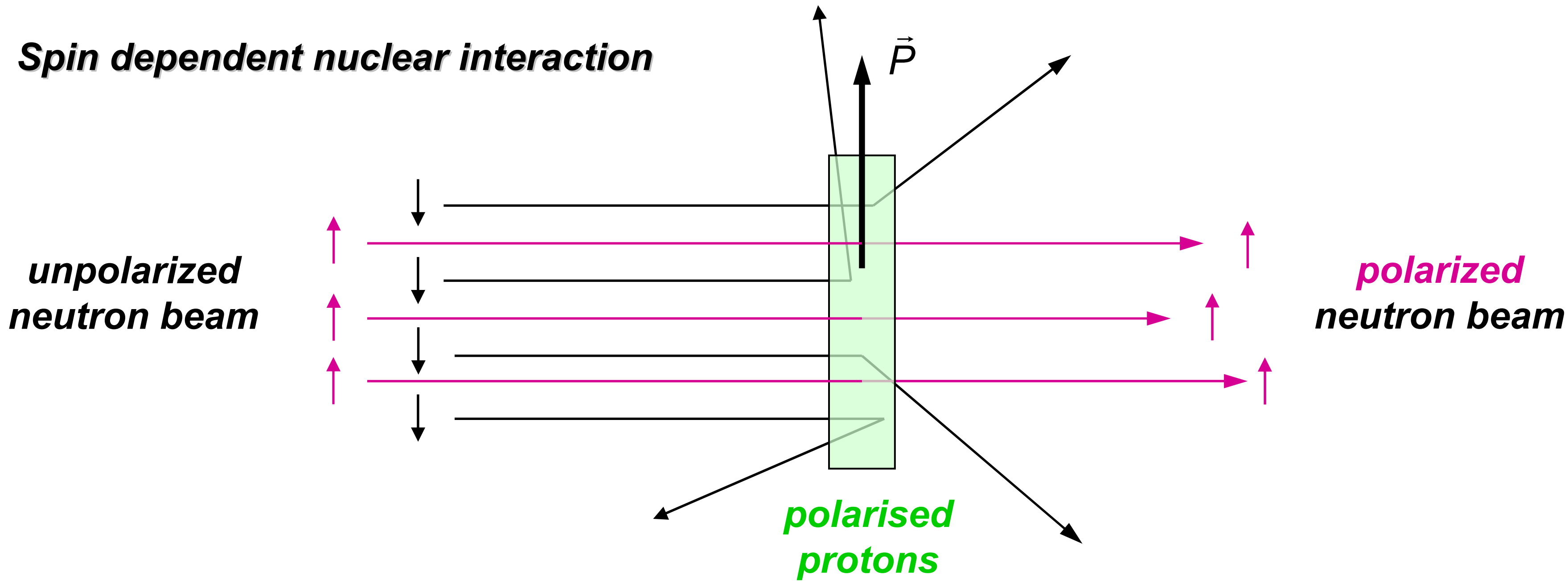
$b = -3.74$ fm

$b_N = 58.254$ fm



Concept of a spin filter – polarized protons

Spin dependent nuclear interaction



polarized protons : **spin dependent scattering**

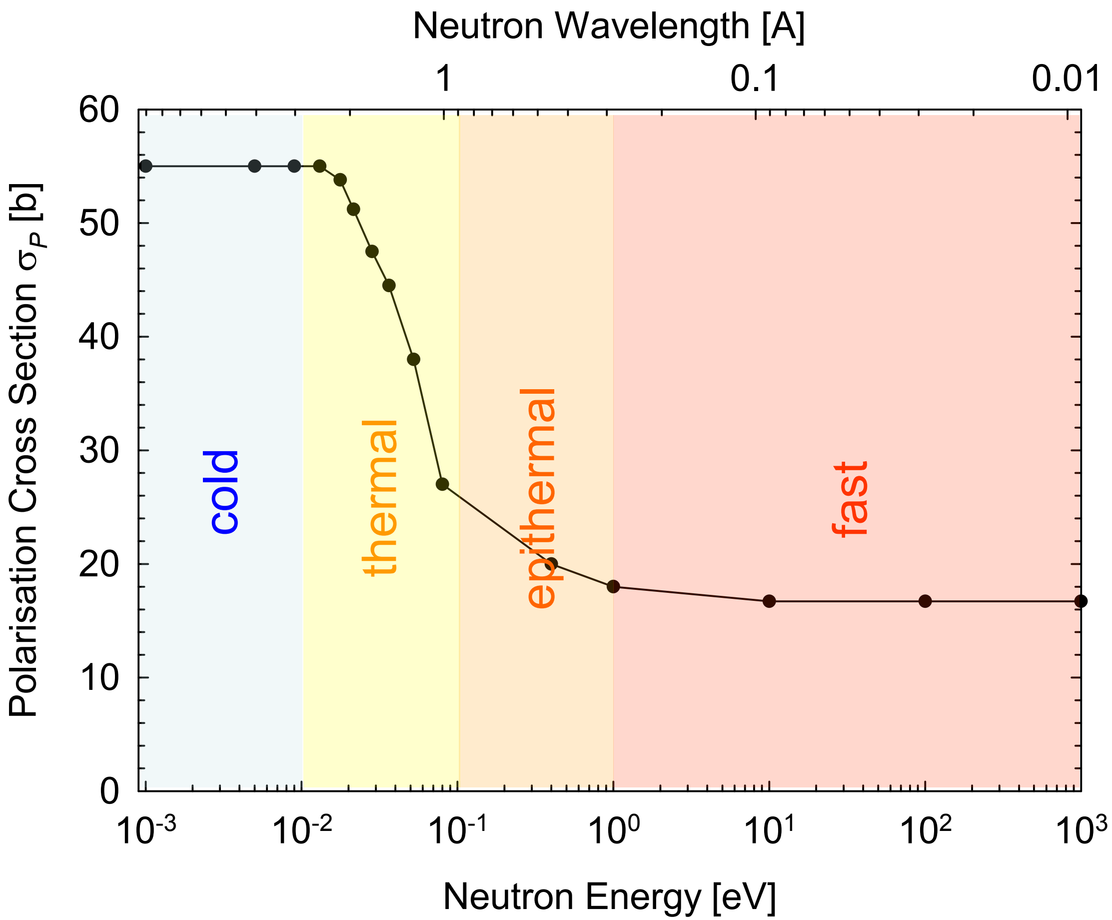
difference between triplet & singlet scattering

effctive cross section : $\sigma_{\pm} = \sigma_0 \pm \sigma_P(\lambda)P$

Polarized protons are the ultimate broad-band spin filter

[Lushikov, Taran, Shapiro, Sov. J. Nucl. Phys. 10 (1970) 699]

Polarization Cross Section $\sigma_P(\lambda)$



E < E_{lim} for Bragg scattering :

elastic incoherent scattering
+ absorption on bound nuclei

transition region

inelastic scattering
interference…

isolated free nuclei

Opaque spin filters / A-power / Transmission

[Zimmer, Müller, Hautle, Heil, Humblot, Phys. Lett. B 455 (1999) 62]

Effective cross section

$$\sigma_{\pm} = \sigma_0 \pm \sigma_P P$$

Intensity of beams behind the spin filter

$$N_+ = \frac{I_0}{2} \exp\left[-(\sigma_0 + \sigma_P P) N d\right] \qquad N_- = \frac{I_0}{2} \exp\left[-(\sigma_0 - \sigma_P P) N d\right]$$

$$p_N = \frac{N_- - N_+}{N_- + N_+} \quad \Rightarrow \quad A = \left| \tanh(\sigma_P P N d) \right| = P_F(\lambda)$$

Choose **opacity** $x = \sigma_P P N d$ ⇒ $A \to 1$

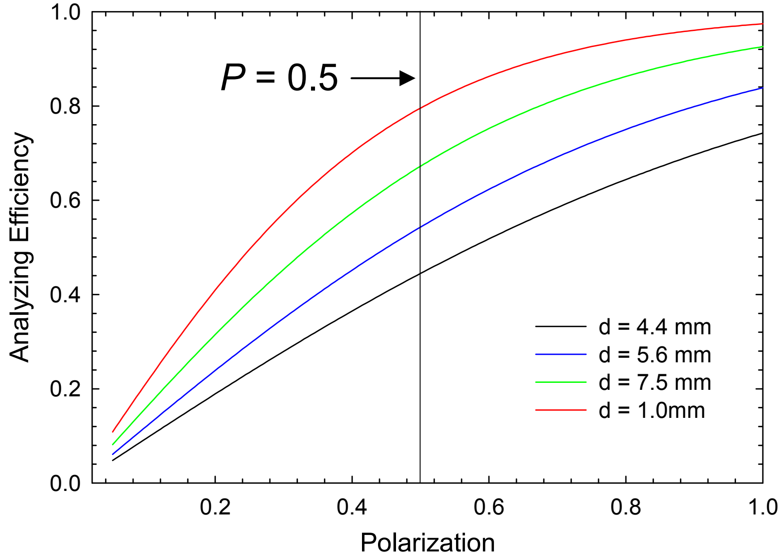
Example: polarized naphthalene sample:

$N = 4.3 \times 10^{22}\ \mathrm{cm}^{-3}$, $d = 1$ cm , $P = 50$ % ⇒ A = 0.82

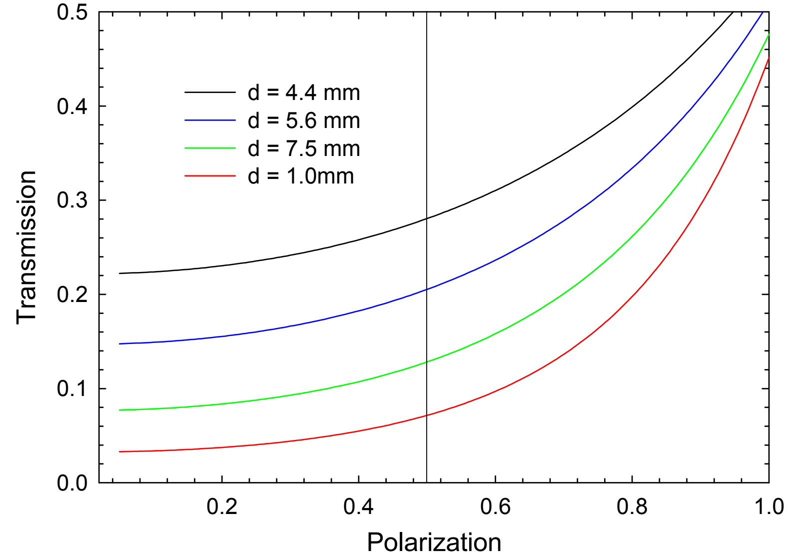
Transmission

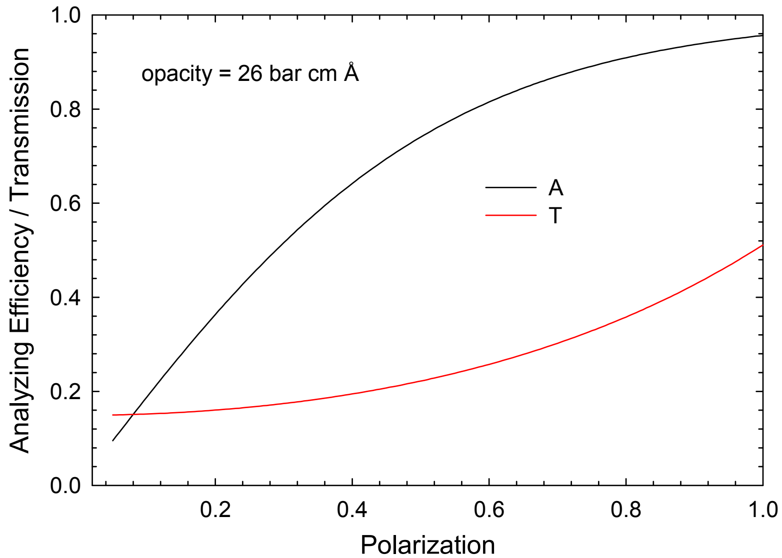
$$T(\lambda) = \frac{N_+ + N_-}{I_0} = \exp(-\sigma_0 N d) \cosh(\sigma_P P N d)$$

Proton spin filter @ λ > 2 Å



polarized naphthalene sample $N = 4.3$ ´ 10^{22} cm^{-3}





^{3}He spin filter

polarized ^{3}He gas (optimum opacity)

Historical remarks

1953 **Overhauser**
nuclear polarization via polarization of conduction electrons in metals (theory)

1958 **Abragam and Proctor**
first experimental DNP in dielectrics (LiF)

1962 **Abragam Borghini** et al.
polarized proton target for low energy particle beam (polarised!)(0.12 cm slab LMN, 2 mg, P = 20%)

1963 **Chamberlain**
polarized proton target for high energy particle beam (1 inch cube LMN, 26 g, P = 20%)

1971 **PSI (SIN) …** Salvatore Mango


Chamberlain, Berkley 1962

Since then:

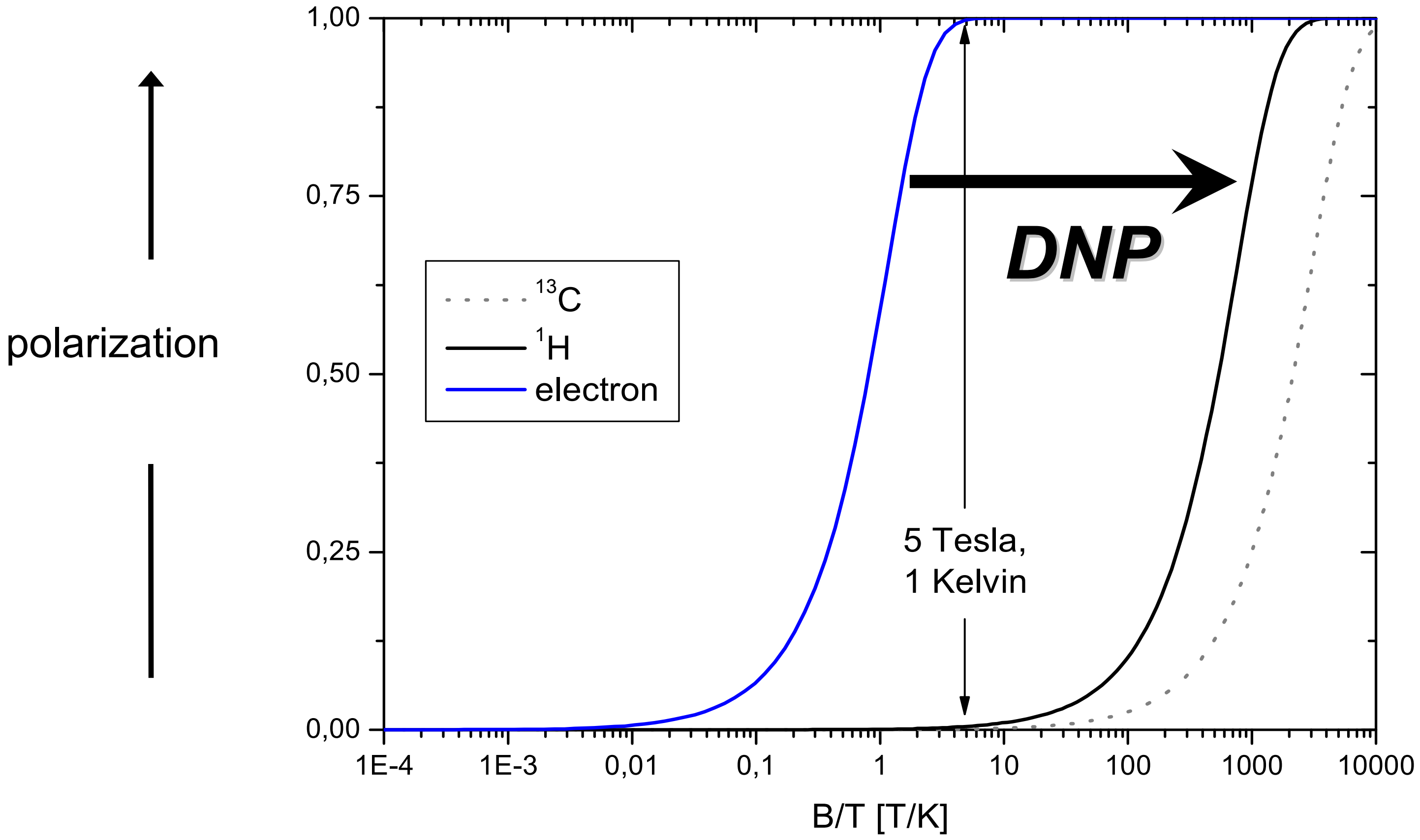
DNP @ PSI

- more than 70 particle physics experiment on pion, proton, neutron beams with polarized targets designed and constructed in our lab
  polarised nuclei: p, d, ^{6}Li, ^{7}Li, ^{10}B, ^{13}C, ^{15}N, ^{19}F, ^{27}Al, ^{139}La, …
- more than 20 neutron scattering experiment with polarized targets
- developed dissolution DNP („Hyperpolarization”) for NMR / MRI
- DNP with photo-excited triplet states

Static Polarization

spin polarization for spin 1/2 – particles:

$$P = \frac{N^+ - N^-}{N^+ + N^-} = \tanh\left(\frac{\mu B}{2kT}\right)$$

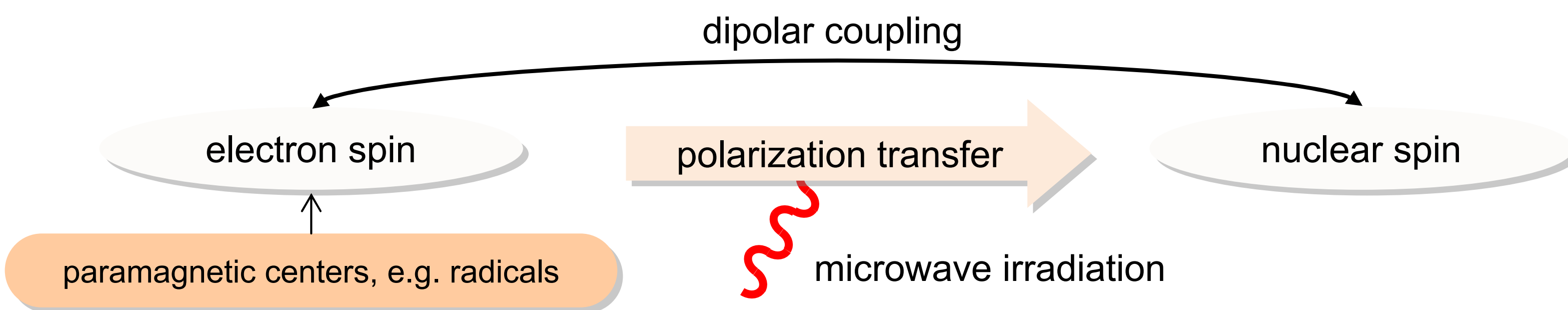


Dynamic Nuclear Polarization (DNP)

polarization transfer from electron spins to surrounding nuclei

via dipolar coupling

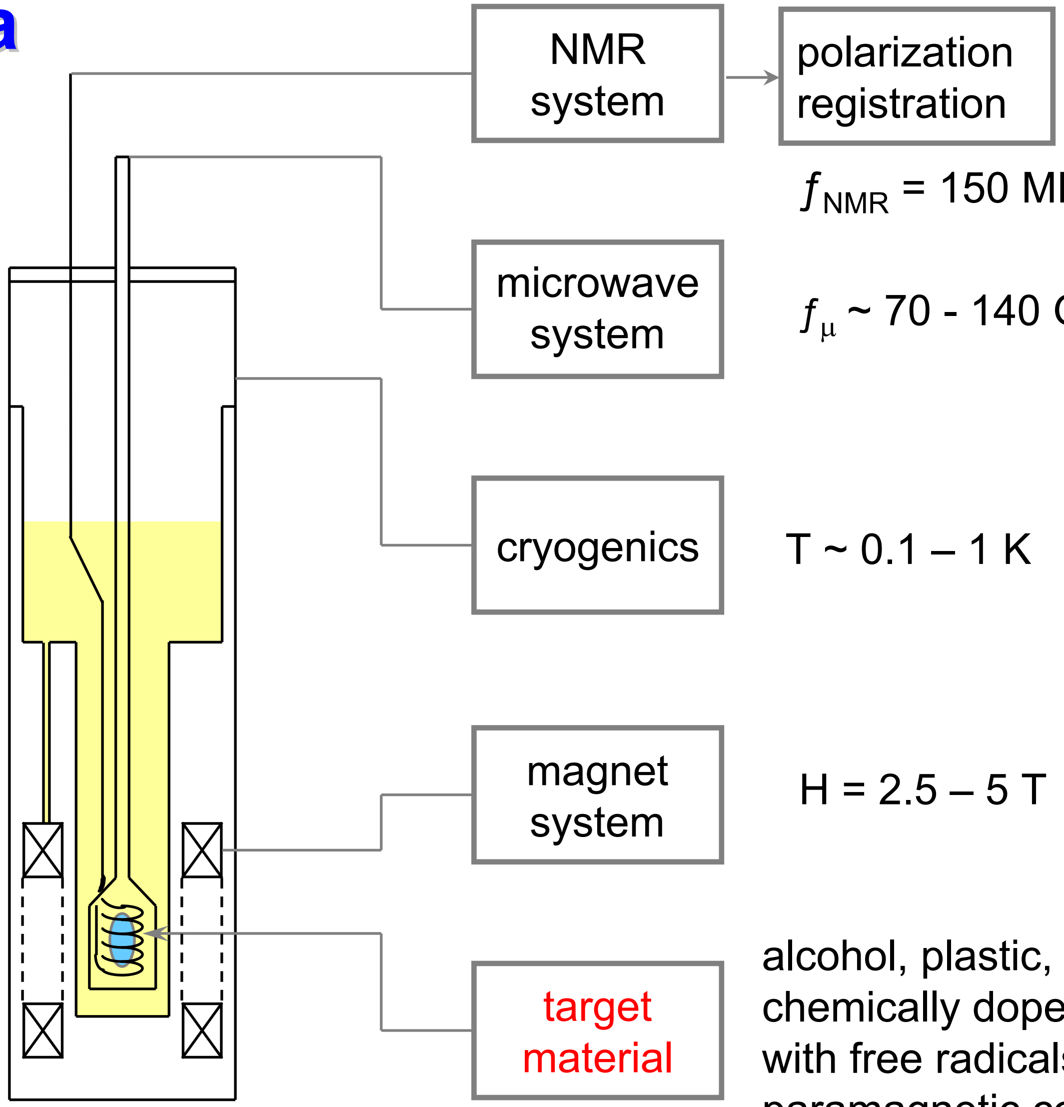
through microwave irradiation (driving mutual spin flips)



prerequisite for “classical” DNP:

- **low temperature** (typically around 1 K)
- **high magnetic fields** (typically 2.5 – 5 Tesla)

Components of a classical DNP system



f_{NMR} = 150 MHz

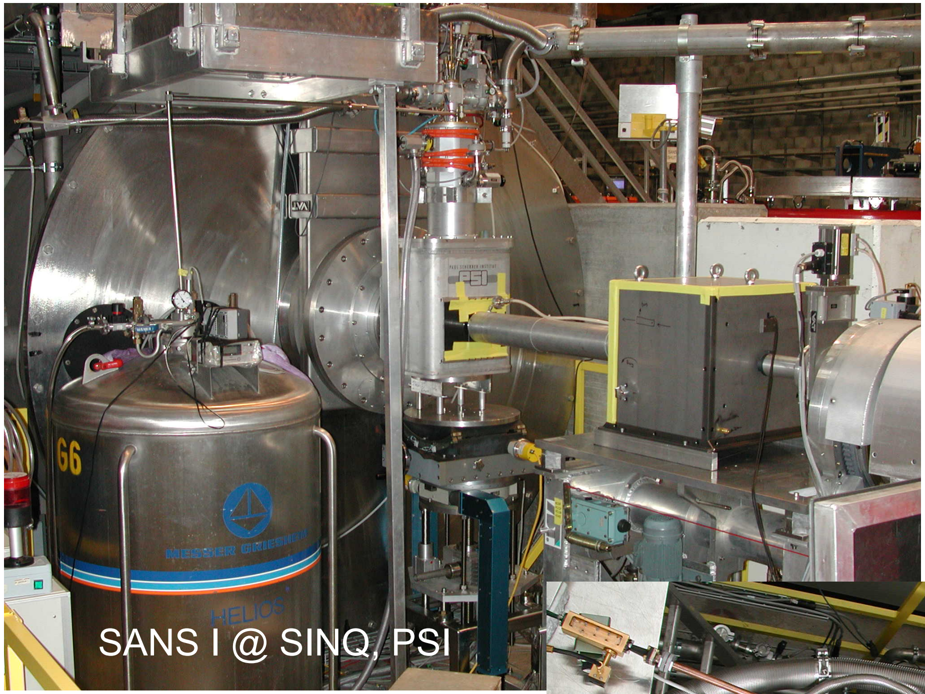
f_{μ} ~ 70 - 140 GHz

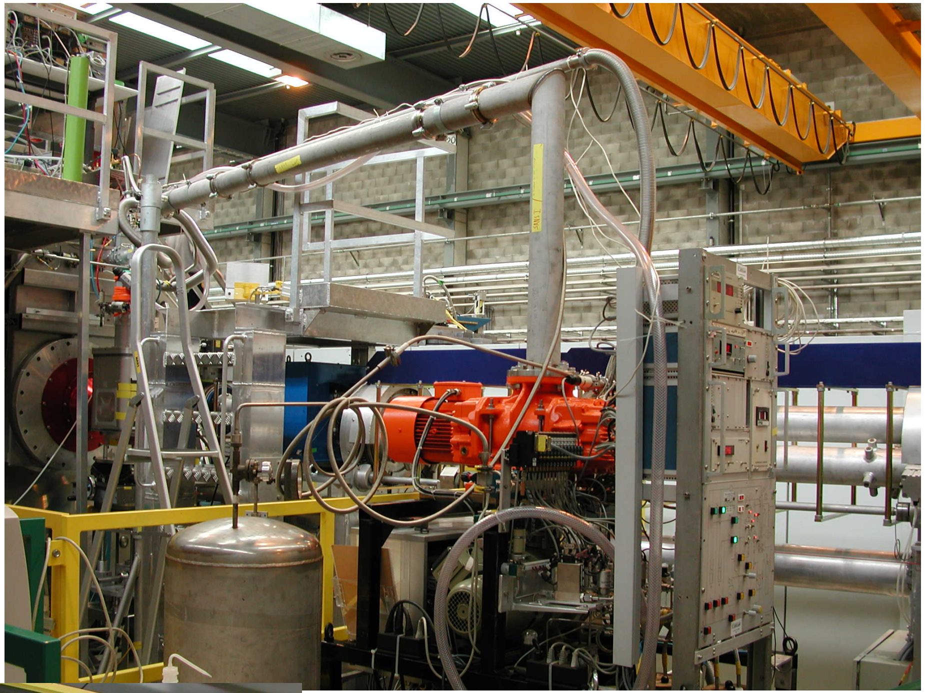
T ~ 0.1 – 1 K

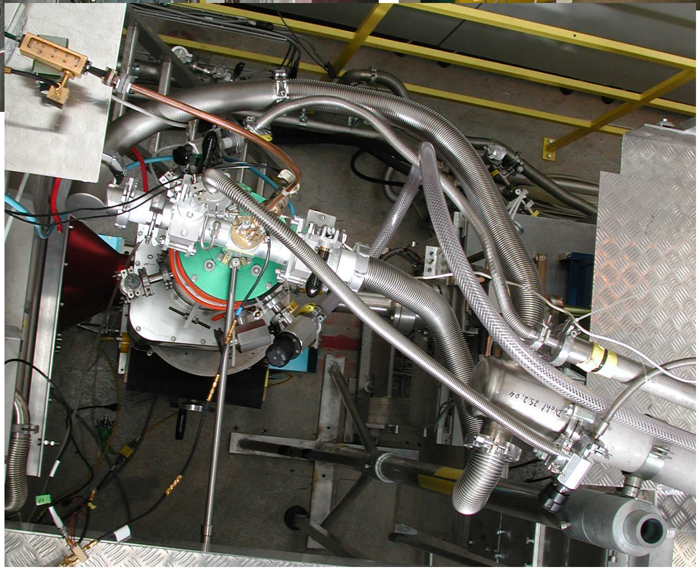
H = 2.5 – 5 T

alcohol, plastic, chemically doped with free radicals or paramagnetic complexes

Classical DNP system







This is a compact system !!

1 K ^4He cryostat
(~ 50 l LHe per day)

1000 m^3/ h + 250 m^3/ h
roots blower pumping system

2.5 / 3.5 T magnet system

“classical” DNP: electron polarization created thermally

- **low temperature** (typically around 1 K)
- **high magnetic field** (typically 2.5 – 5 Tesla)

an “exotic” DNP mechanism …

Volume 165, number 1 CHEMICAL PHYSICS LETTERS 5 January 1990

HIGH DYNAMIC NUCLEAR POLARIZATION AT ROOM TEMPERATURE

A. HENSTRA, T.-S. LIN [1], J. SCHMIDT and W.Th. WENCKEBACH

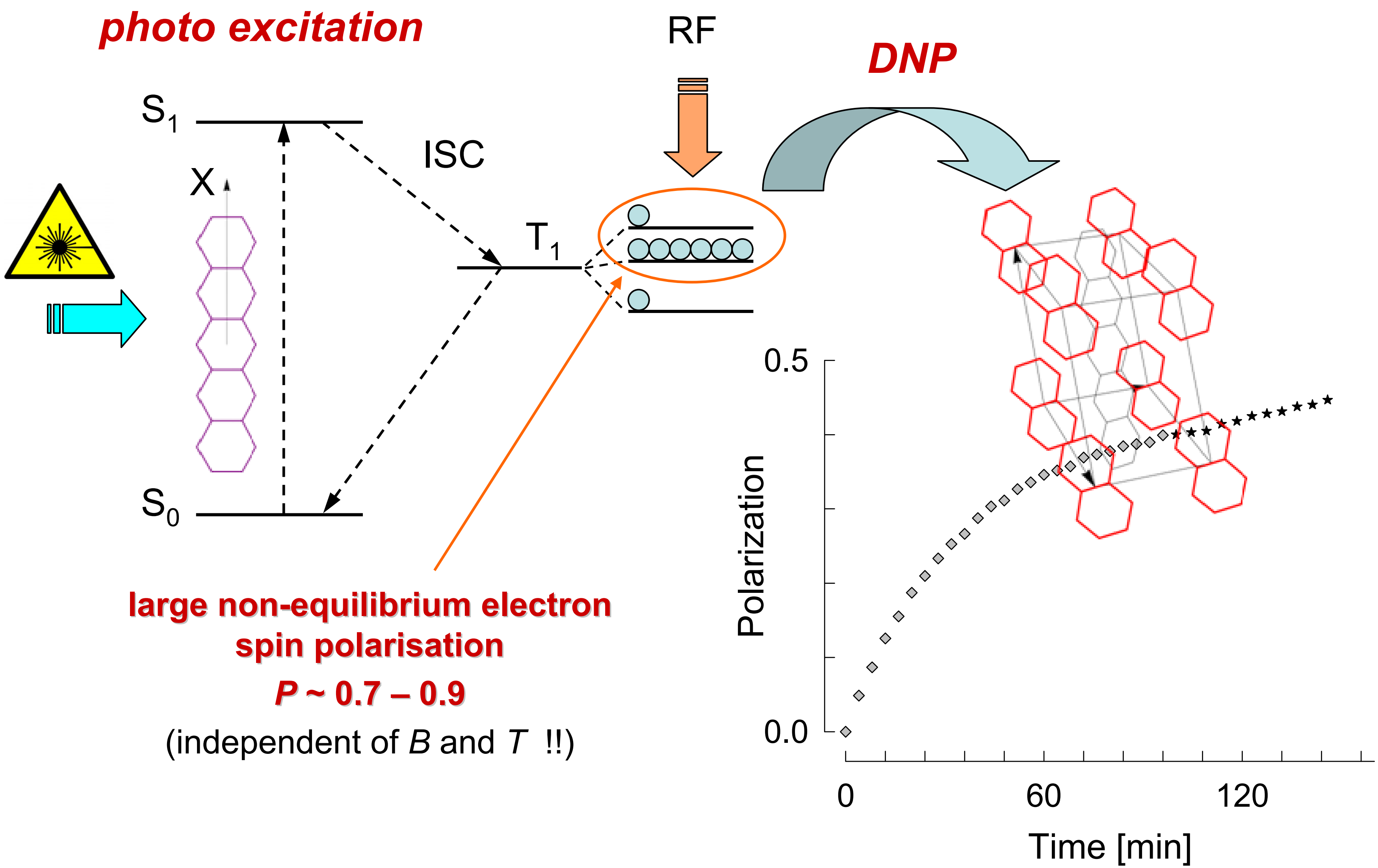
Kamerlingh Onnes and Huygens Laboratories, University of Leiden, P.O. Box 9504, 2300 RA Leiden, The Netherlands

Received 10 November 1989

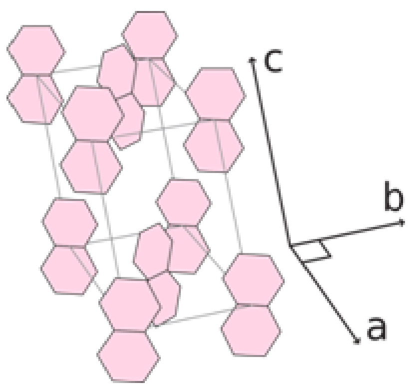
The highly polarized photo-excited triplet state of pentacene in a naphthalene crystal is used for pulsed dynamic nuclear polarization at room temperature. Thus far an enhancement of 5500 of the naphthalene proton polarization has been reached. For this purpose, a newly developed technique, the integrated solid effect, performed while obeying the Hartmann–Hahn condition, is used to transfer the triplet polarization efficiently to the nuclear spin system.

create electron polarization with laser light which can be used for DNP

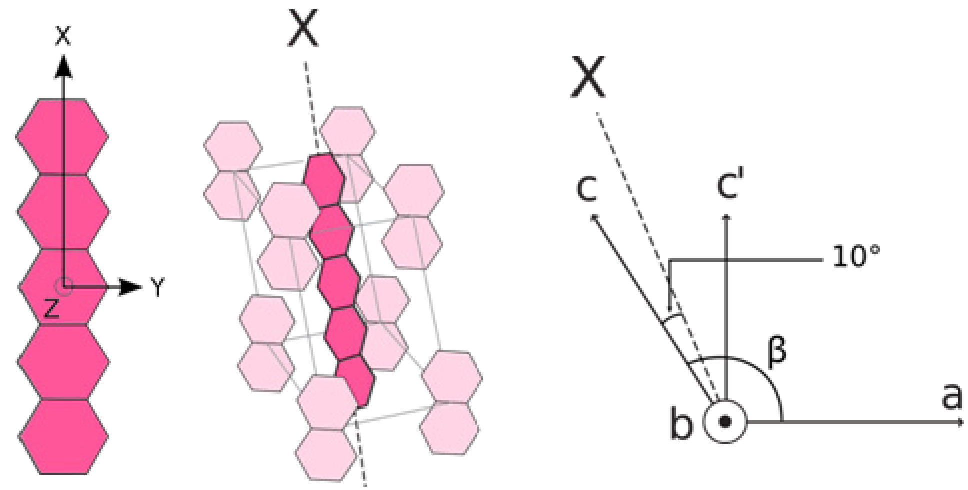
DNP using triplet states (pentacene in naphthalene)



Naphthalene – Pentacene Crystals



unit cell of pure naphthalene ($C_{22}H_{14}$)



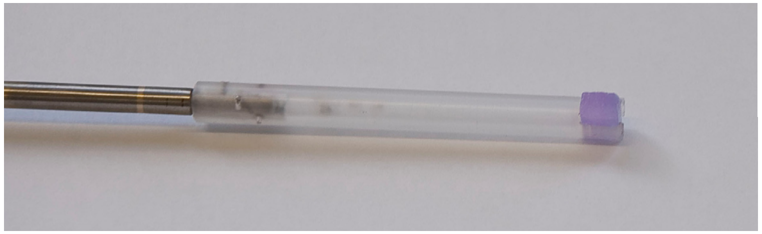
unit cell when a pentacene molecule ($C_{22}H_{14}$) has taken the position of two naphthalene molecules

large single crystal grown using a selective self-seeding Bridgman technique

Pentacene conc ~ 10^{-5} mol/mol

sample size: 5 ´ 5 ´ 5 mm^3

mounted on KelF holder



Harware for triplet state DNP & neutron experiments

^{4}He flow cryostat operates from 4 K < T < RT

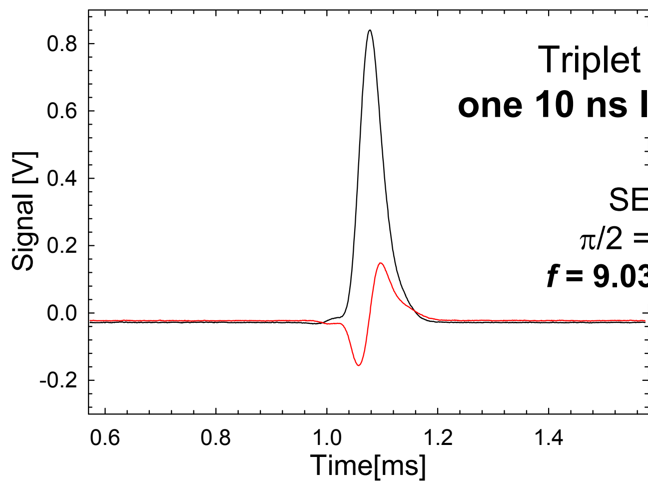
typically operated at T = 100 K, ~ 100 l LHe / week

90% transparency for neutrons

Pulse X-band ESR spectrometer

combines pulse EPR with pulse DNP capabilities

(home built, design by J.J. van der Klink, EPFL)

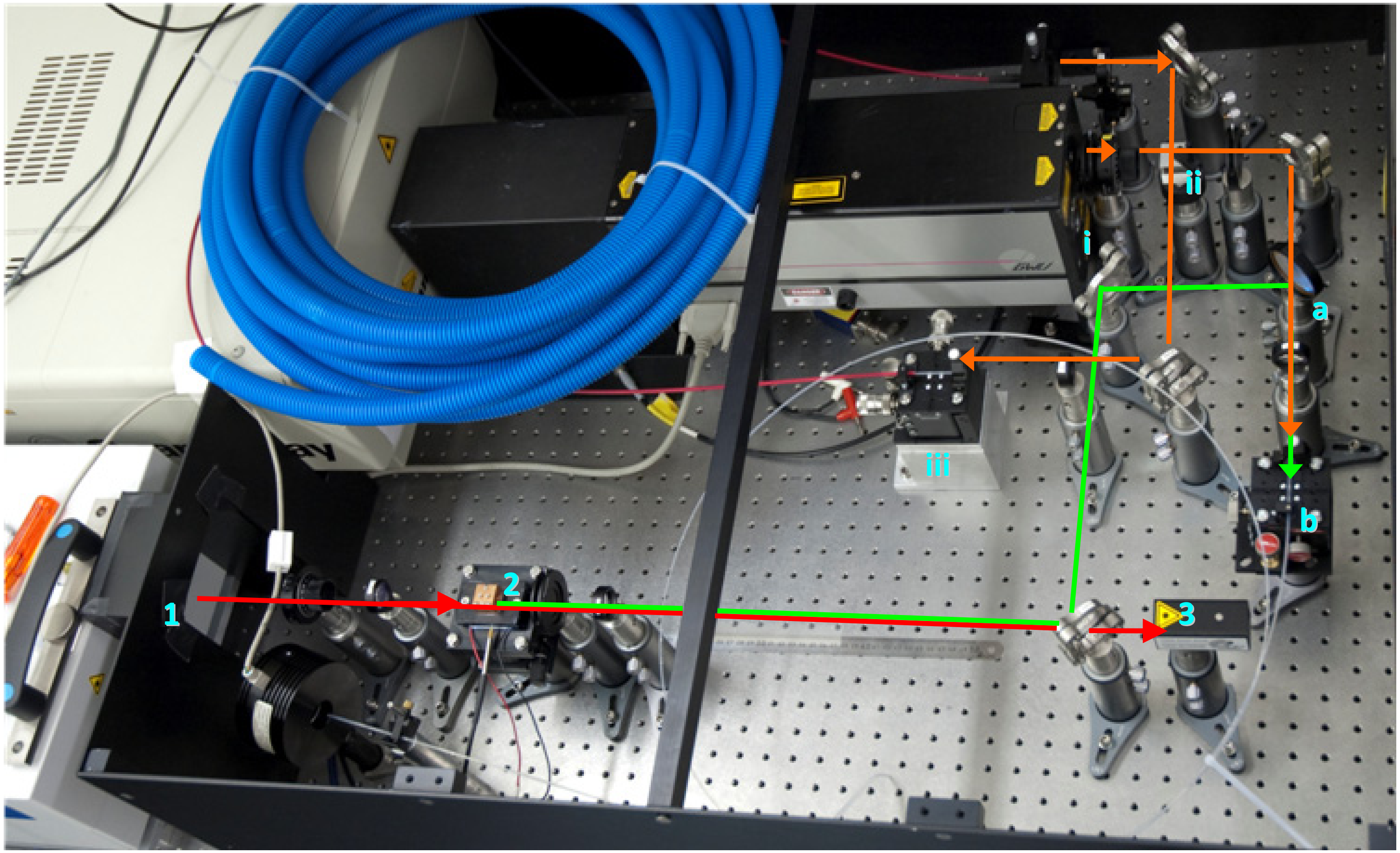


Triplet ESR Signal after
one 10 ns laser shot @ 600 nm

SESE: $\pi/2$ - $\pi/2$ - $\pi/2$
$\pi/2$ = 35 ns, T = τ = 1 ms
***f* = 9.0311 GHz**, B = 0.3015 T

Laser system + optical fiber YAG/OPO (600 nm) / disk laser (515 nm)

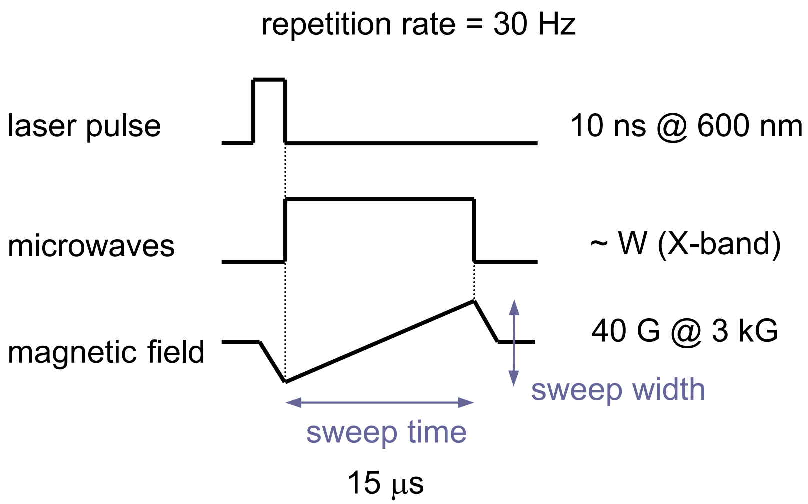
Multipurpose laser setup: YAG/OPO + *Disk Laser*

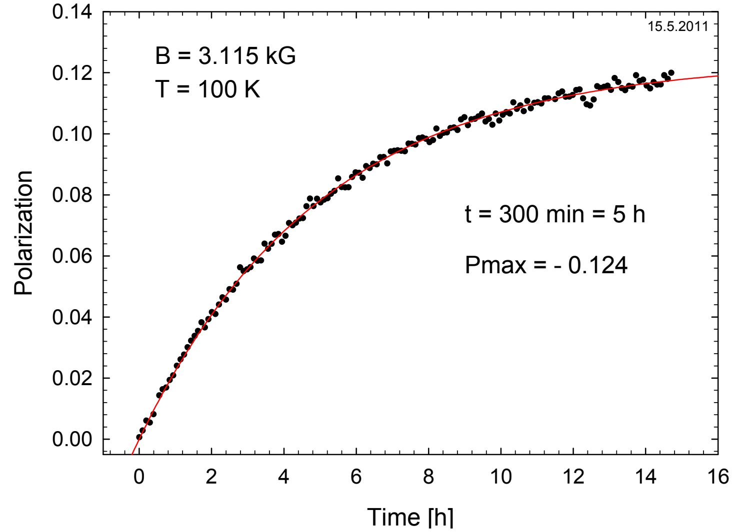


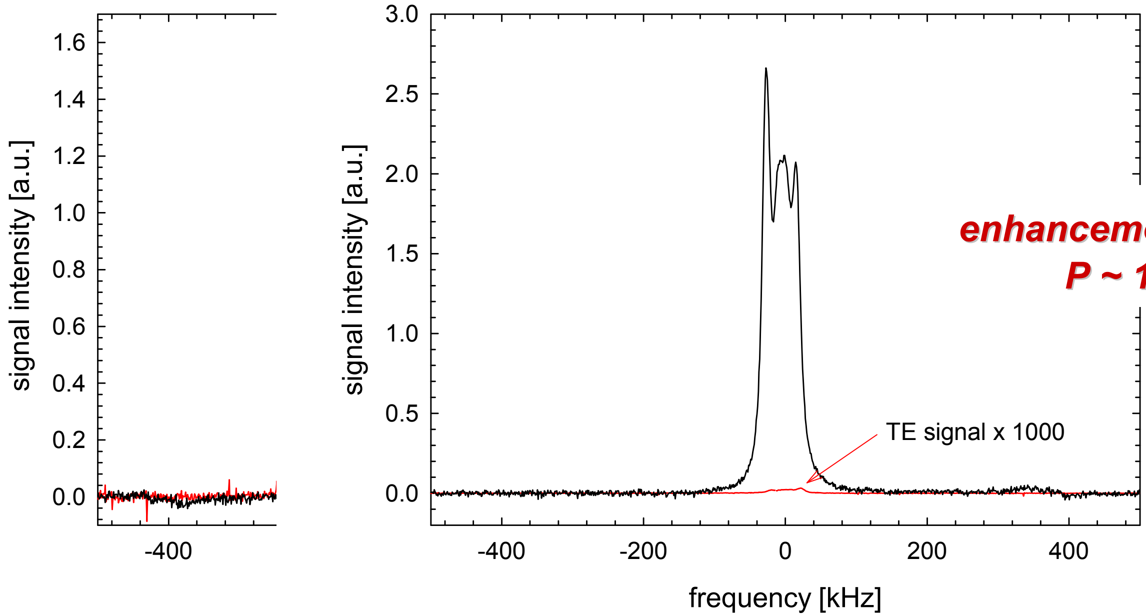
1: **IR disk laser** (1030 nm, 50 W, < 10 kHz, < 500 ns pulses),

2: **LBO** crystal for **SHG** to 515 nm with > 20 % efficiency => 1.2 mJ at end of fiber

DNP via ISE







enhancement ~ 35500

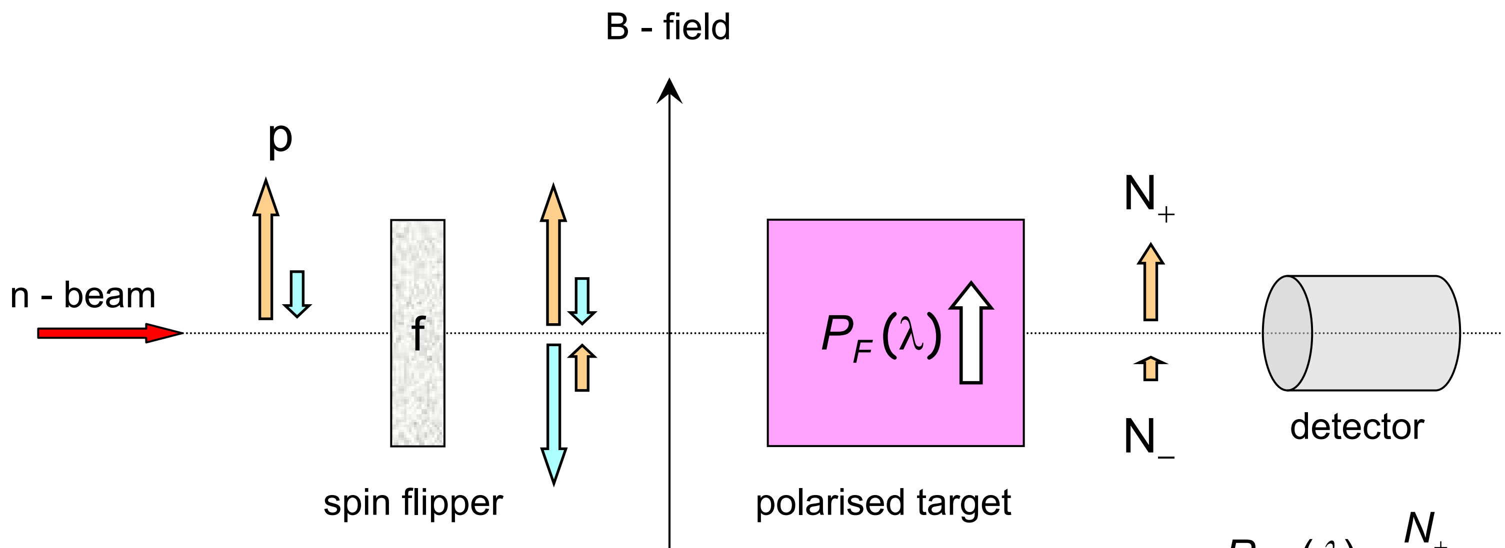
P ~ 11.3 %

TE polarization

P_{TE} = 3.1826 ´ 10^{-6}

[summer 2011]

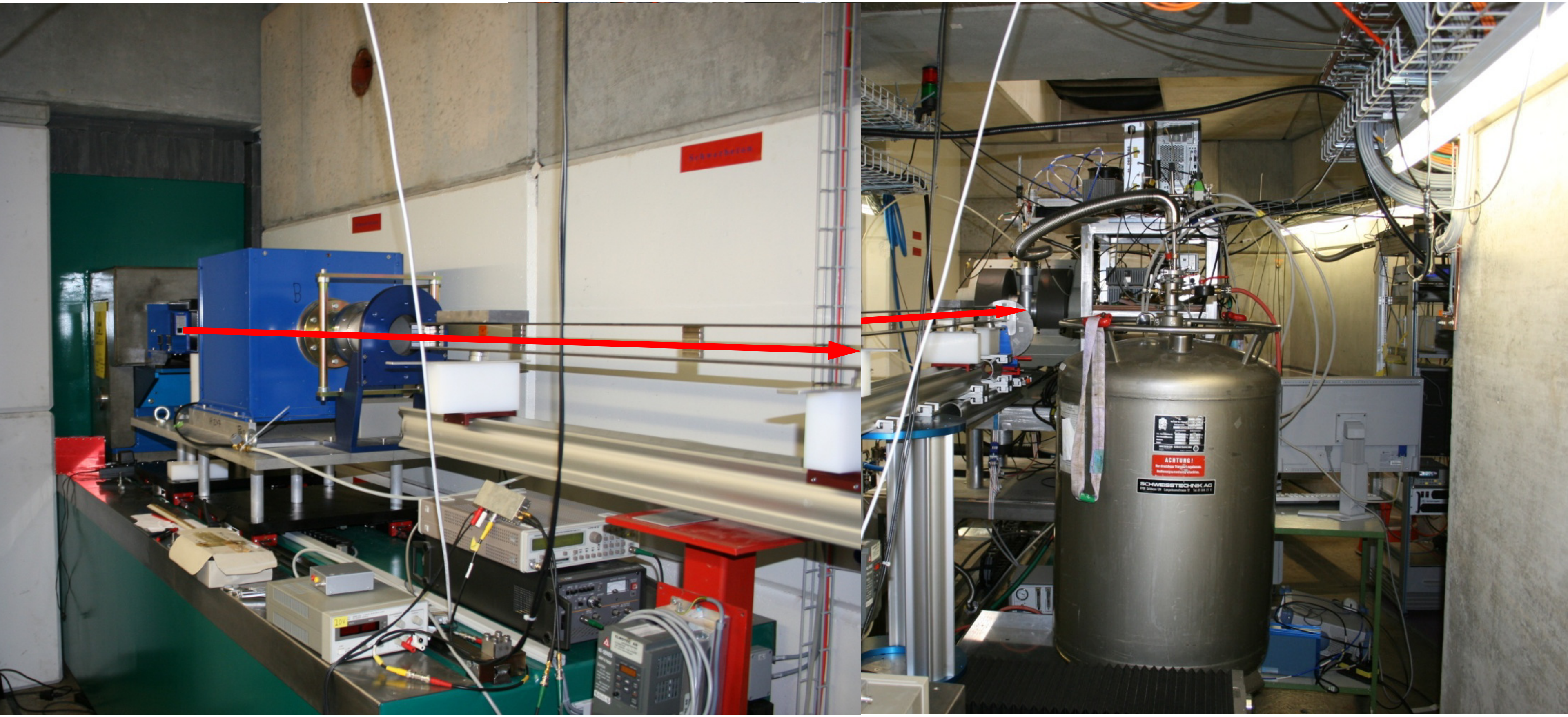
Test of Principle - Experimental Scheme



$$R_{p,F}(\lambda) = \frac{N_+}{N_-} = \frac{1 + p P_F}{1 - f\, p P_F}$$

- perform a ***test of principle*** for a triplet spin filter
- prove the spin filter effect
- measure the ***polarization cross section***
- use the neutrons to ***characterize the target performance*** (DNP, relaxation etc..)
- long term stability of system under adverse conditions

Set up on neutron beamline I



BOA beamline @ SINQ (PSI), flux ~ 2 ´ 10^7 /cm^2 s

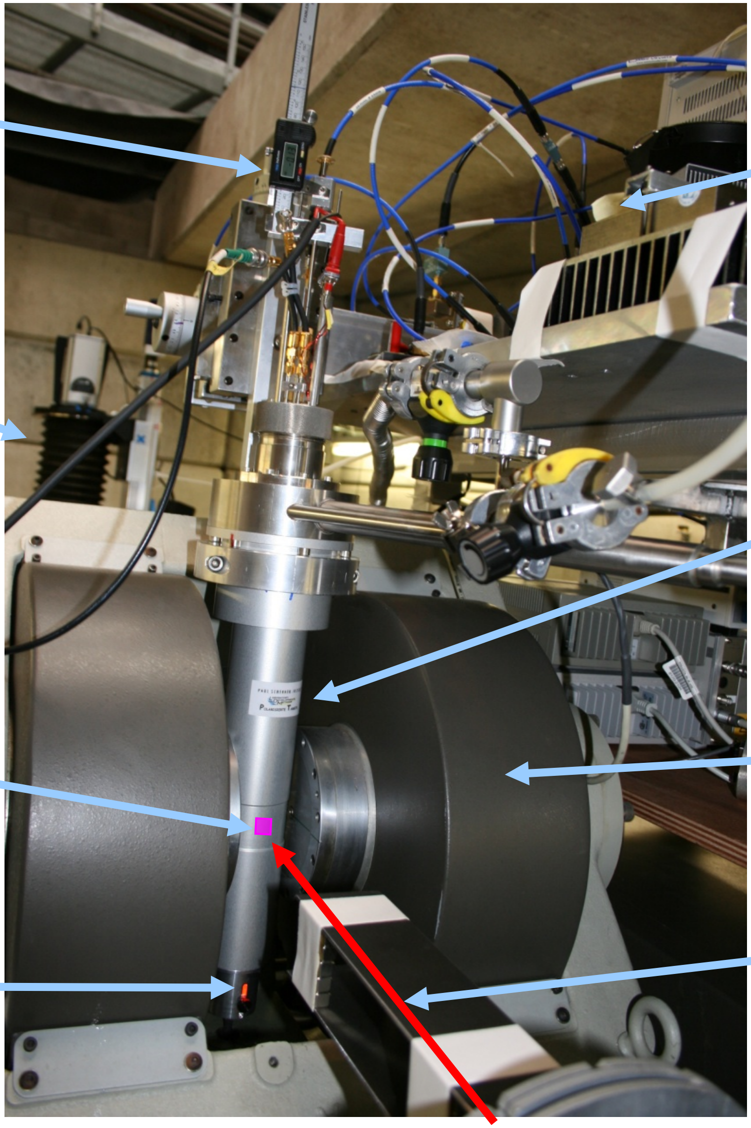
Set up on neutron beamline II

Caliper for sample positioning

Neutron detector

Target Crystal

Fiber coupled laser light @ 600 nm



pulse ESR / DNP system 9 GHz

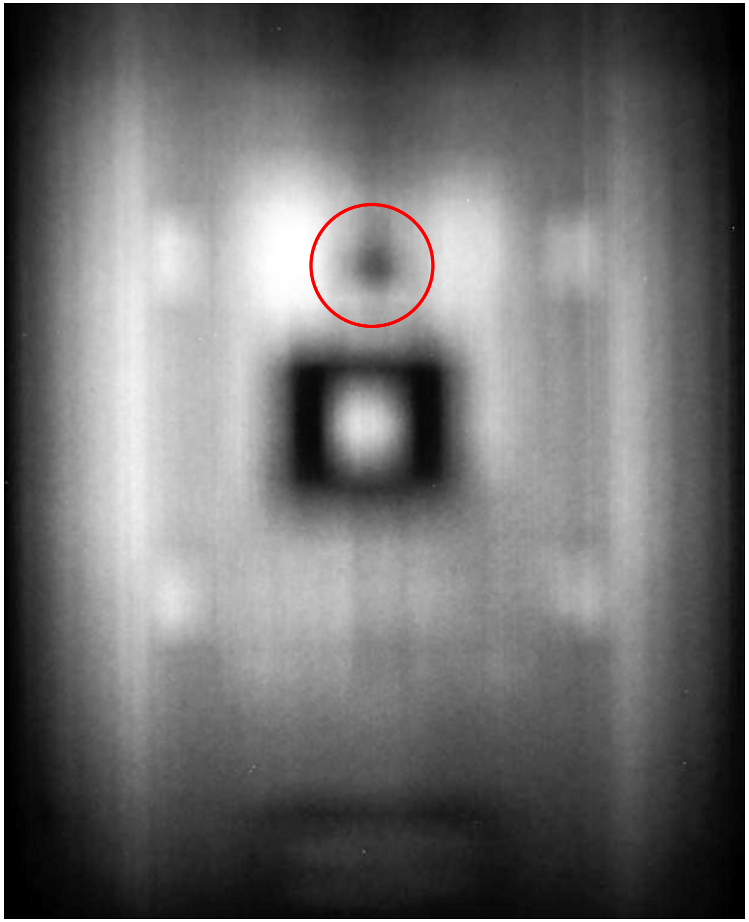
Pulse NMR system

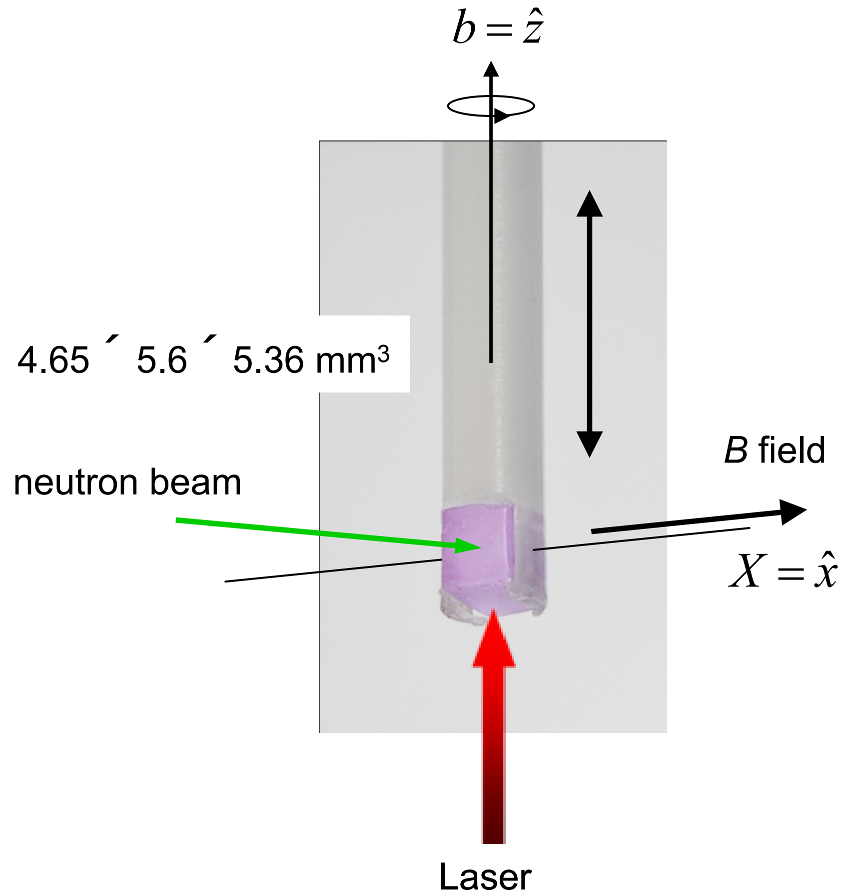
Cryostat T ~ 100 K

Magnet, 0.3 T (max 0.6 T)

Neutron beam

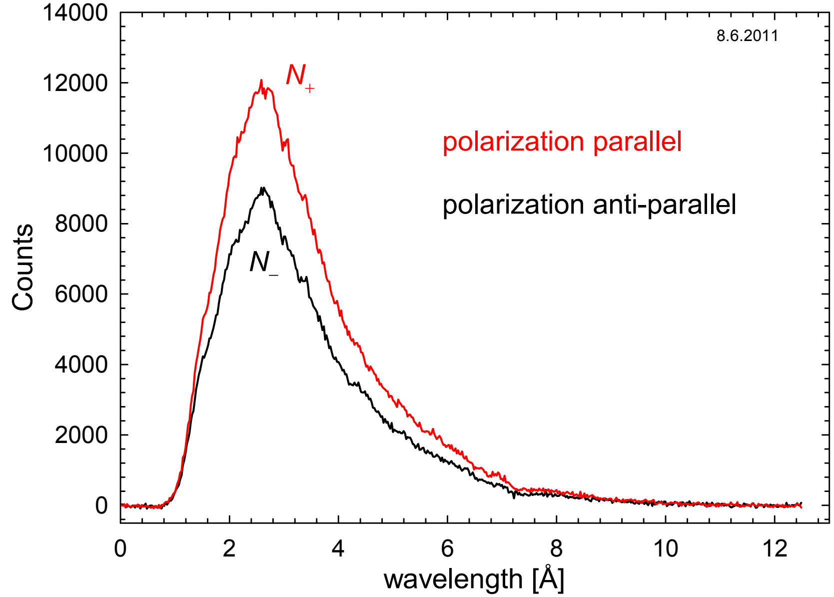
Sample positioning





Pentacene conc. = 2.0 ± 0.1 ´ 10^{-5} mol/mol

Neutron measurements – polarization cross section



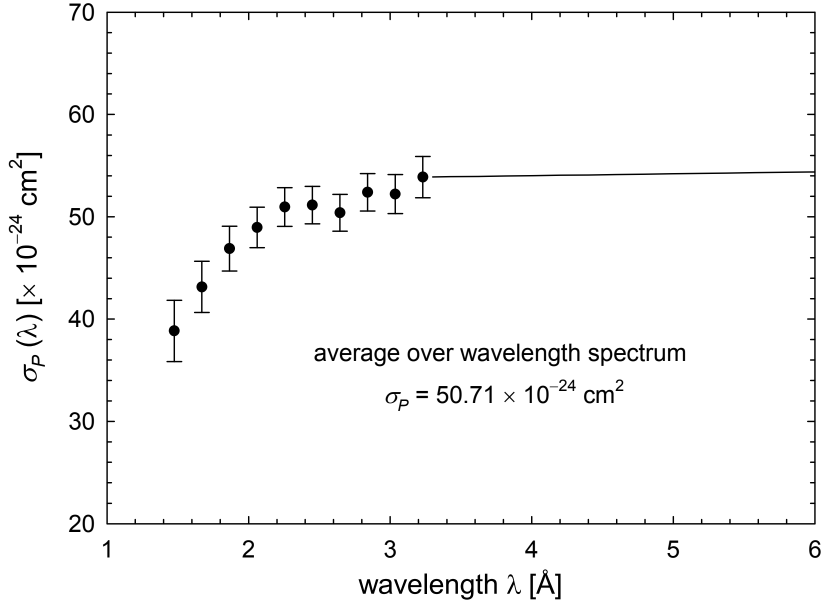
wavelength resolved measurements

time of flight system

flipping ratio $$R_{p,F}(\lambda) = \frac{N_+}{N_-} = \frac{1 + p P_F(\lambda)}{1 - f\, p P_F(\lambda)}$$

polarization cross section normalized

$$\sigma_{-1,P}(\lambda) = -\frac{\sigma_P(\lambda)}{P}$$



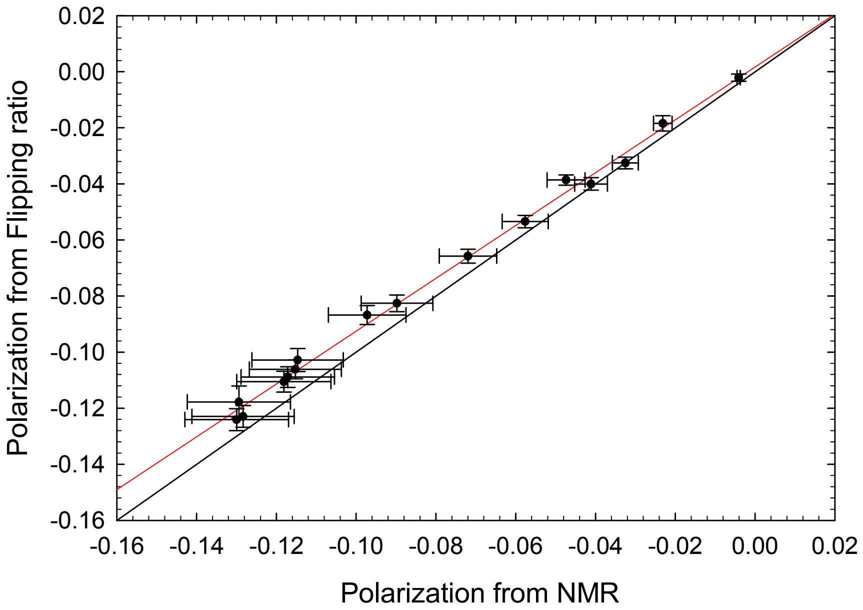
Neutrons as local polarization probe

measure ***two neutron count rates***

2 ´ ~ 60 s

$$P = \frac{1}{\overline{\sigma}_{-1,P}} \cdot \frac{1}{N d} \operatorname{artanh}\left(\frac{R_{p,F} - 1}{R_{p,F} \cdot f + 1} \cdot \frac{1}{\overline{p}} \right)$$

accurate polarization value of the sample



Triplet spin filter – proof of principle

Nuclear Instruments and Methods in Physics Research A 678 (2012) 91–97

ELSEVIER

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Spin filtering neutrons with a proton target dynamically polarized using photo-excited triplet states

M. Haag [a], B. van den Brandt [a], T.R. Eichhorn [a,b], P. Hautle [a,*], W.Th. Wenckebach [a,1]

[a] *Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*
[b] *Laboratory for Functional and Metabolic Imaging, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*

ARTICLE INFO

Article history:
Received 12 December 2011
Received in revised form
24 February 2012
Accepted 4 March 2012
Available online 22 March 2012

Keywords:
Neutron polarization analysis
Spin filters
Dynamic nuclear polarization
Photo-excited triplet states

ABSTRACT

In a test of principle a neutron spin filter has been built, which is based on dynamic nuclear polarization (DNP) using photo-excited triplet states. This DNP method has advantages over classical concepts as the requirements for cryogenic equipment and magnets are much relaxed: the spin filter is operated in a field of 0.3 T at a temperature of about 100 K and has performed reliably over periods of several weeks.

The neutron beam was also used to analyze the polarization of the target employed as a spin filter. We obtained an independent measurement of the proton spin polarization of ~ 0.13 in good agreement with the value determined with NMR. Moreover, the neutron beam was used to measure the proton spin polarization as a function of position in the naphthalene sample. The polarization was found to be homogeneous, even at low laser power, in contradiction to existing models describing the photo-excitation process.

Martin Haag, PhD Thesis, ETHZ December 2012

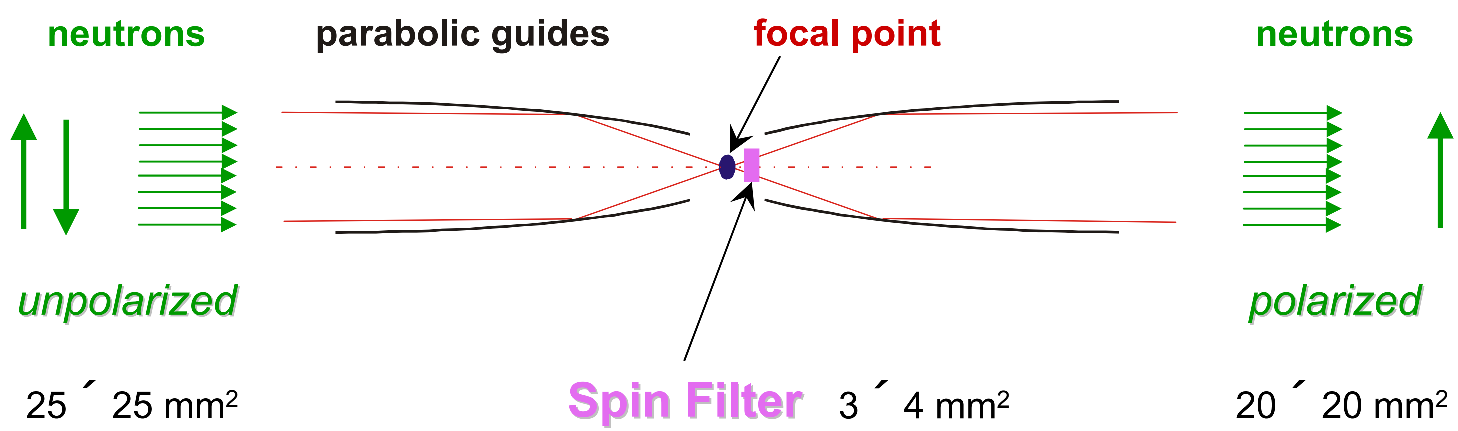
Triplet spin filter + neutron optics

- development of *focusing neutron guides* (elliptic, parabolic)

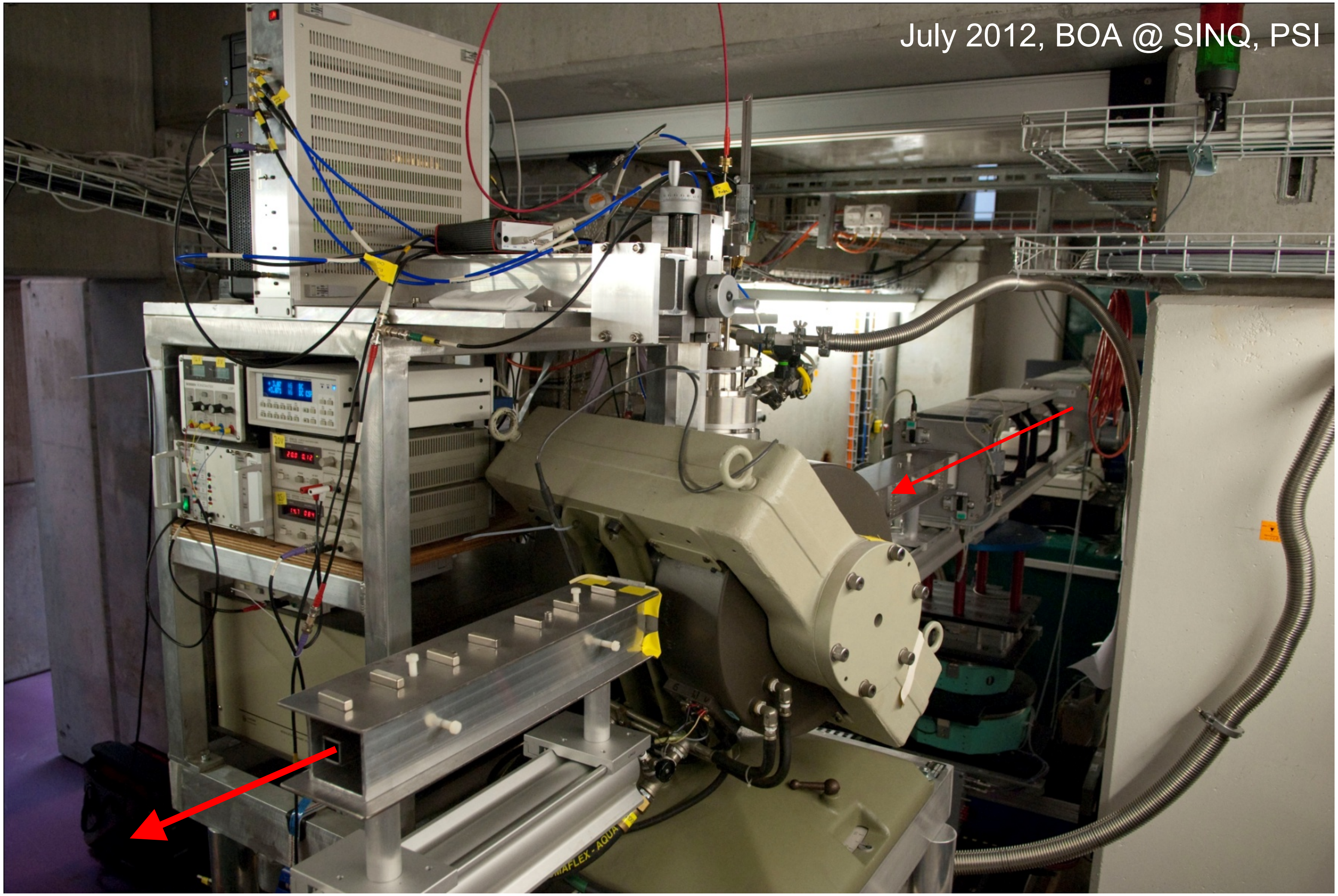
  large gains in neutron flux possible

- integration of small triplet spin filter
  into a focusing guide system close to focus

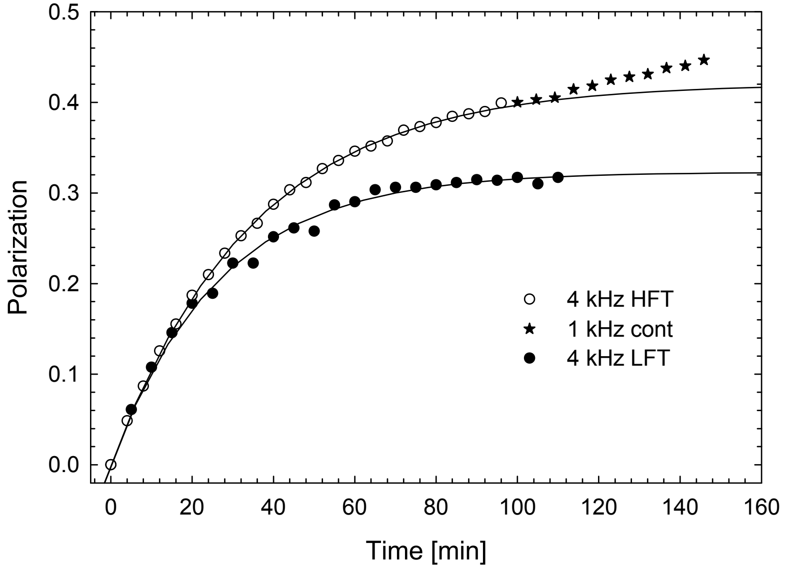
<u>Primary polarizer set up</u>*: *proof of principle experiment at PSI / BOA July 2012



Triplet spin filter + neutron optics



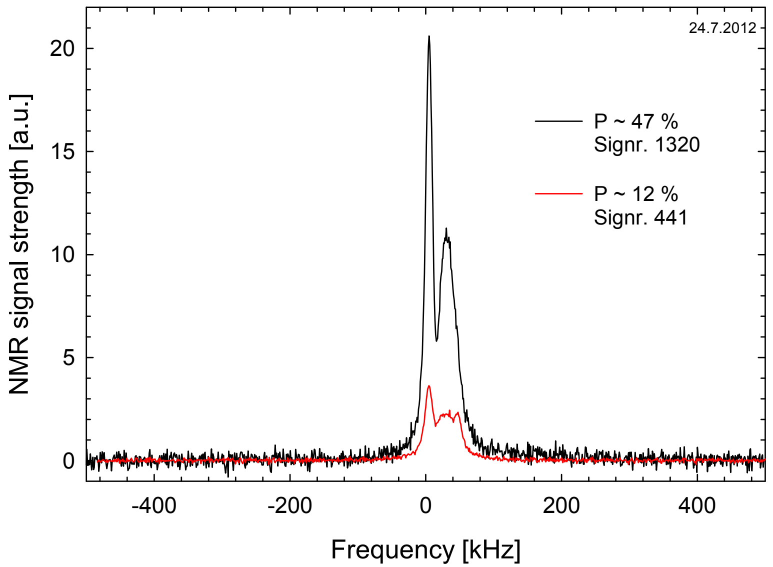
DNP results with disk laser & pentacene-d_{14}



Initial polarization build-up proportional to ISE repetition rate

ISE @ 4 kHz : dP/dt ~ 1.2 % / min

Limit is given by cooling power of the cryostat



Polarization measured with NMR and neutron transmission

P ~ 50 %

enhancement ~ 160'000

TE polarization

$P_{TE} = 3.1826 \times 10^{-6}$

[T.R. Eichhorn et al., Chem Phys Lett 555 (2013) 296]

Output of BOA beam times

3 weeks in June 2011

- proof of principle of triplet spin filter
- M. Haag et al, NIM A 678 (2012) 91
- PhD Thesis Martin Haag

3 weeks in July 2012

- first experiment of focus / defocus concept with spin filter
- E. Rantsiou et al., NIM A, in preparation for NOP-2013
- so far highest polarization P ~ 0.5 achieved with triplet DNP system
- T.R. Eichhorn et al, Chem Phys Lett 555 (2013) 296

Outlook & Planning for SF itself

improve filter performance

- higher polarization
  => better cooling, new liquid Argon cryostat
- increase sample lenght (~10 mm)

simplify spin filter

- reduce the technical complexity and „footprint“
  => permanent magnet, simple cryostat
- study optimum integration into focusing optics

fundamental investigations

- optimize ISE polarization process
- fully understand optical excitation

Experiment planning / requirements

Goal of next Spin Filter experiments

1. provide conclusive proof of the focus / defocus principle
2. demonstrate an efficient primary polarizer for large beam area
3. demonstrate polarization analysis with reference scatterer
   SANS on CuNiFe alloy

Prerequisites

1. spin flipper
   housing has been rebuildt, needs to be tuned and integrated
2. saphire cyrstals for background reduction
3. 2D detector for SANS
4. LN cooled Be filter for SANS
   cut spectrum below 4 A, checked at SANS I

Beam time request

1. spin filter experiment : 1 full block (= 4 weeks)
2. general preparation : 1 week at start of SINQ (Flipper, SANS test)

Team

T. R. Eichhorn (PhD, SNF funded until April 2014, PSI – EPFL)
M. Haag (PhD, FOKO funded, finished December 2012)
X.X (PhD, SNF funded, PSI)
B. van den Brandt
P. Hautle
W. Th. Wenckebach (academic guest)

Paul Scherrer Institut

EPFL Lausanne

http://ltf.web.psi.ch
patrick.hautle@psi.ch