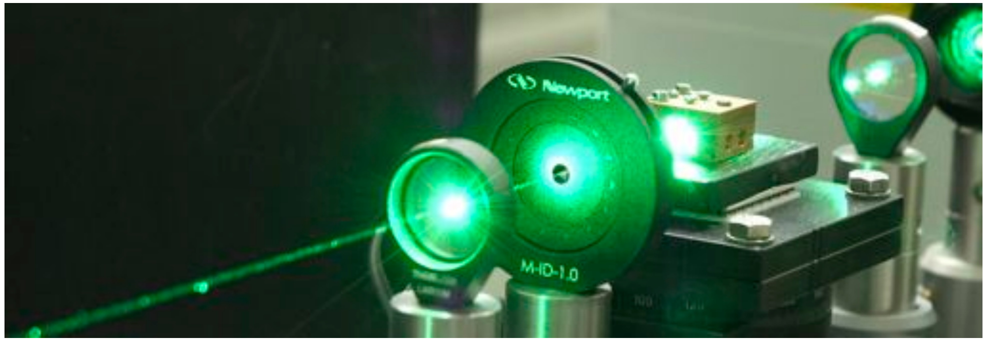
The 2013 International Workshop on Polarized Sources, Targets & Polarimetry

Neutron spin filter based on dynamically polarized protons using photo-excited triplet states



Tim Eichhorn[a,b], Ben van den Brandt[a], Martin Haag[a], Patrick Hautle[a], Tom Wenckebach[a]

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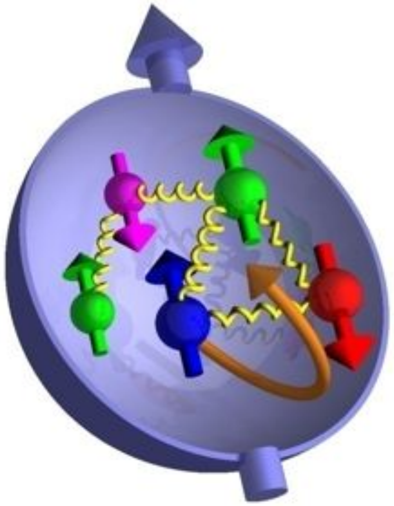
cw Proton accelerator (590 MeV, 2.2 mA)

Outline



WHY

Neutron spin filtering with polarized protons

HOW to polarize protons :

DNP using photo-excited triplet states

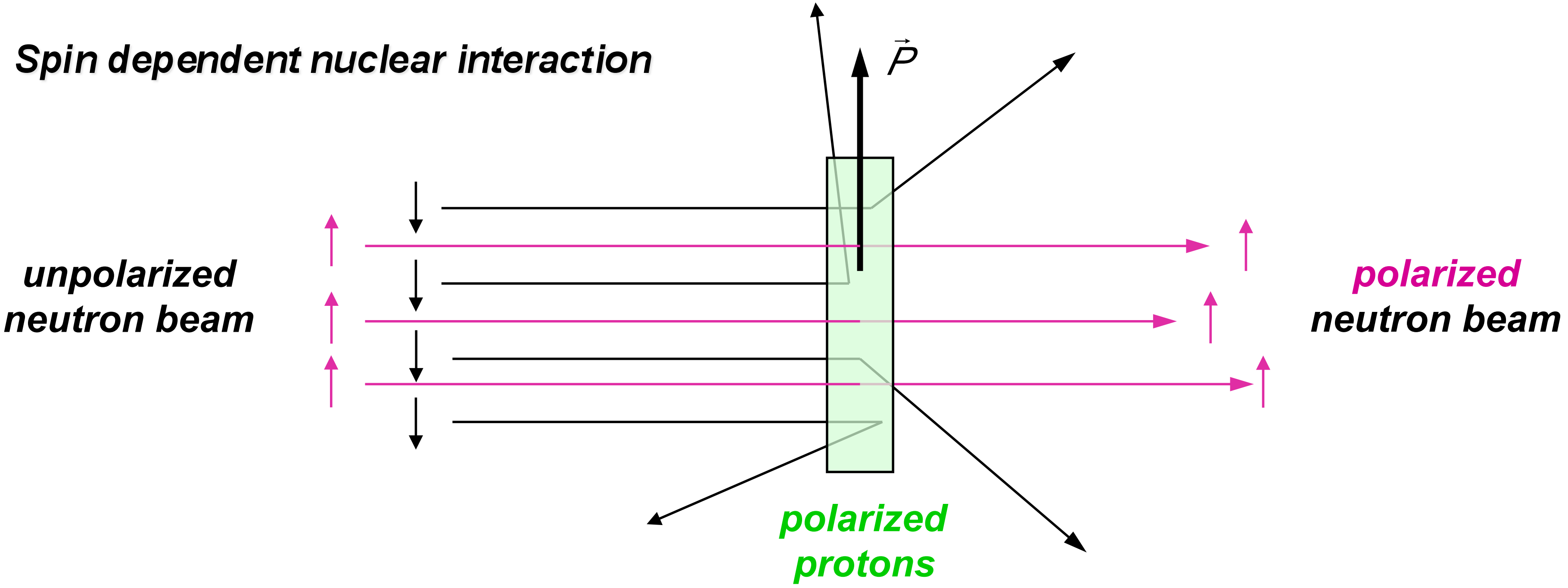
RESULTS / PRESENT STATUS

Experiments @ BOA / PSI

Polarized proton target as a neutron spin filter

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

Spin dependent nuclear interaction



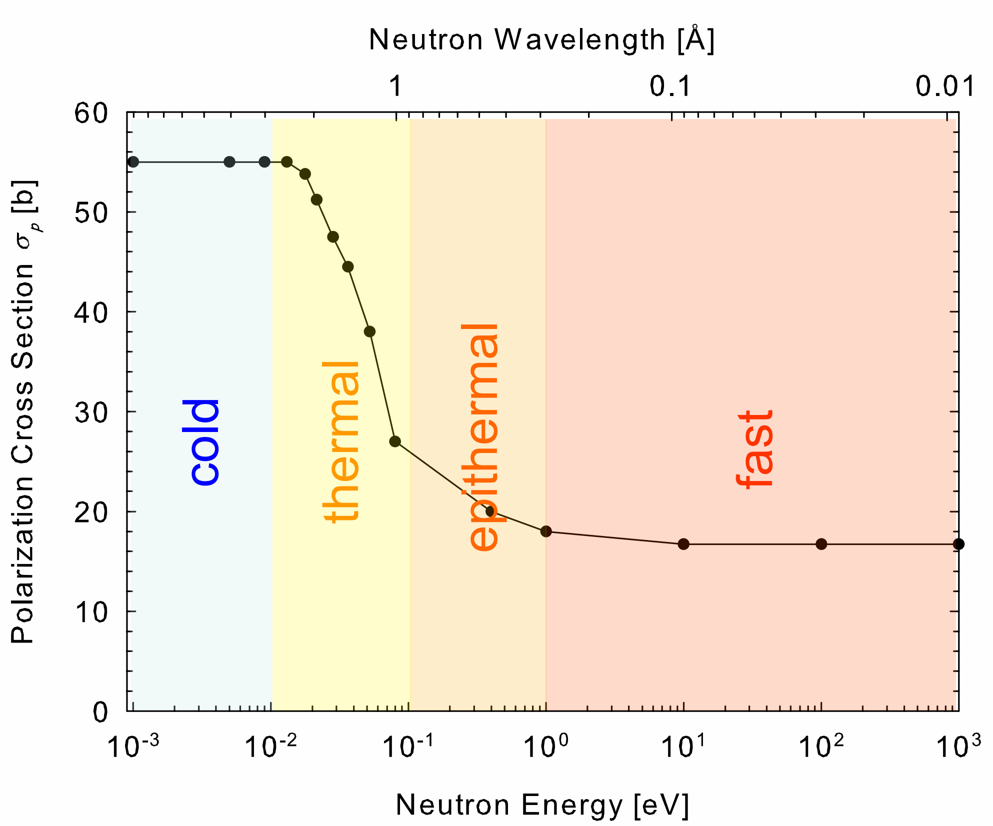
polarized protons : **spin dependent scattering**

difference between triplet & singlet scattering

effective cross section :

$$\boxed{\sigma_{\pm} = \sigma_0(\lambda) \pm \sigma_P(\lambda) \cdot P}$$

Neutron scattering on condensed hydrogenous material



E < E_{lim} for Bragg scattering :

elastic incoherent scattering
+ absorption on bound nuclei

transition region

inelastic scattering
interference…

isolated free nuclei

➡ **Polarized protons are the ultimate broad-band spin filter**

Characteristics of opaque spin filters

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

Effective cross section

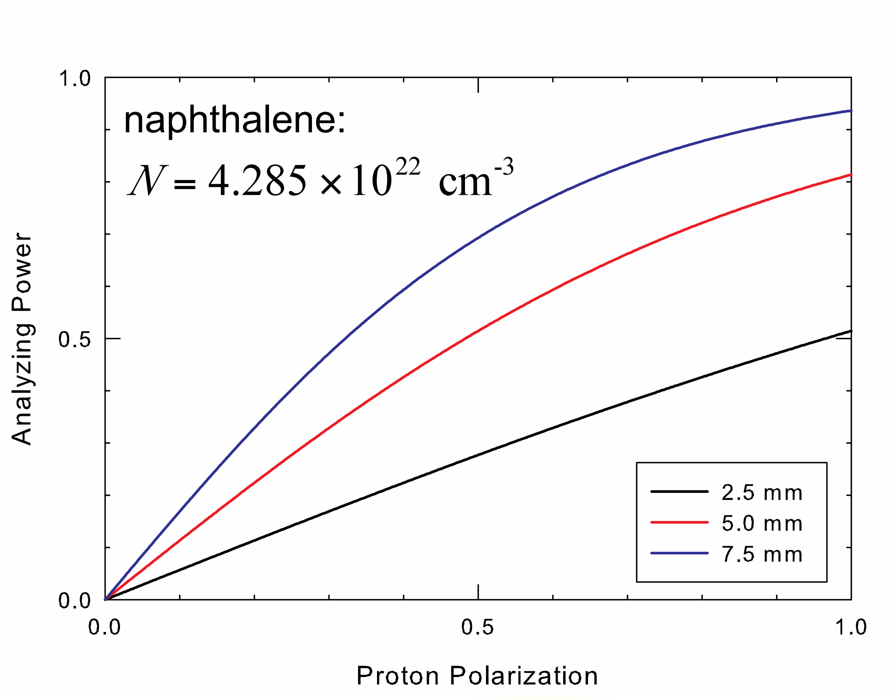
$$\sigma_{\pm} = \sigma_0(\lambda) \pm \sigma_P(\lambda) \cdot P$$

Intensity of beams behind the spin filter

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

Analyzing power (neutron polarization after filter)

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



→ **high opacity for small samples**

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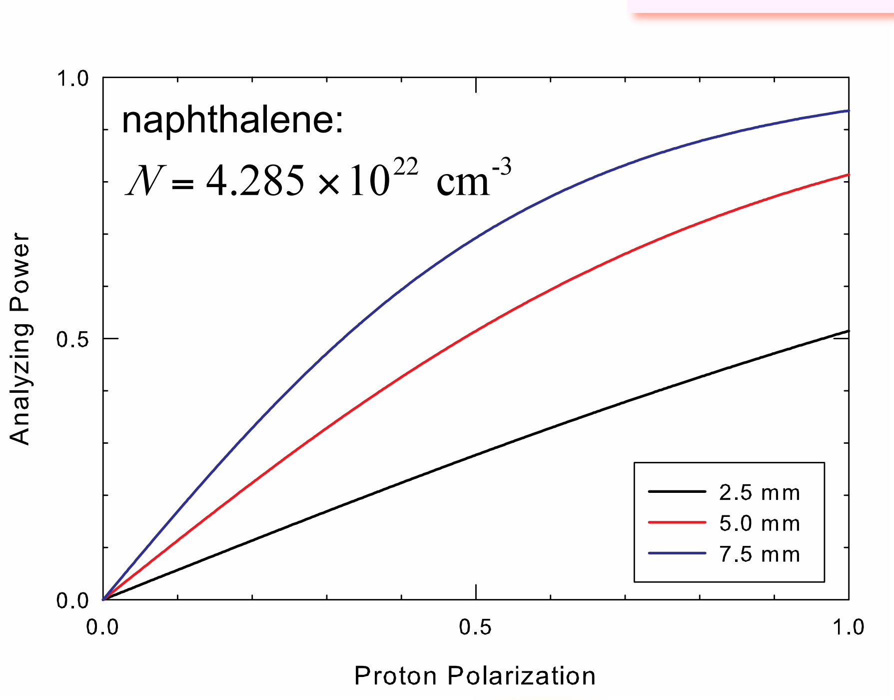
Characteristics of opaque spin filters

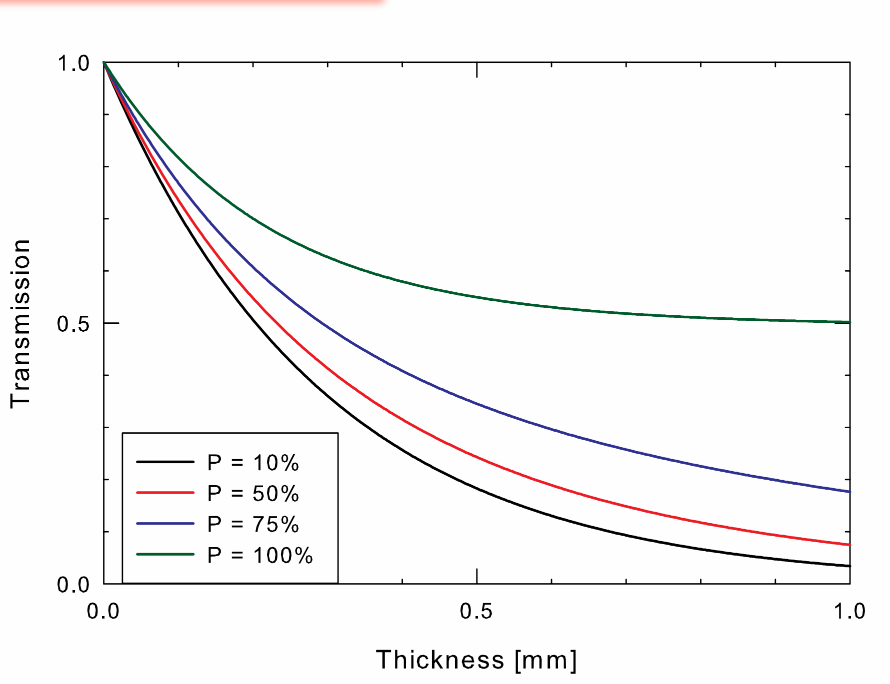
[Zimmer, Müller, Hautle, Heil, Humblot, Phys. Lett. B 455 (1999) 62]

Effective cross section $$\sigma_{\pm} = \sigma_0(\lambda) \pm \sigma_P(\lambda) \cdot P$$

Intensity of beams behind the spin filter $$N_{\pm} = \frac{I_0}{2} \exp[-(\sigma_0 \pm \sigma_P P)Nd]$$

Neutron Transmission $$T = \frac{N_+ + N_-}{I_0} = \exp(-\sigma_0 Nd)\cosh(-\sigma_P PNd)$$





Characteristics of opaque spin filters

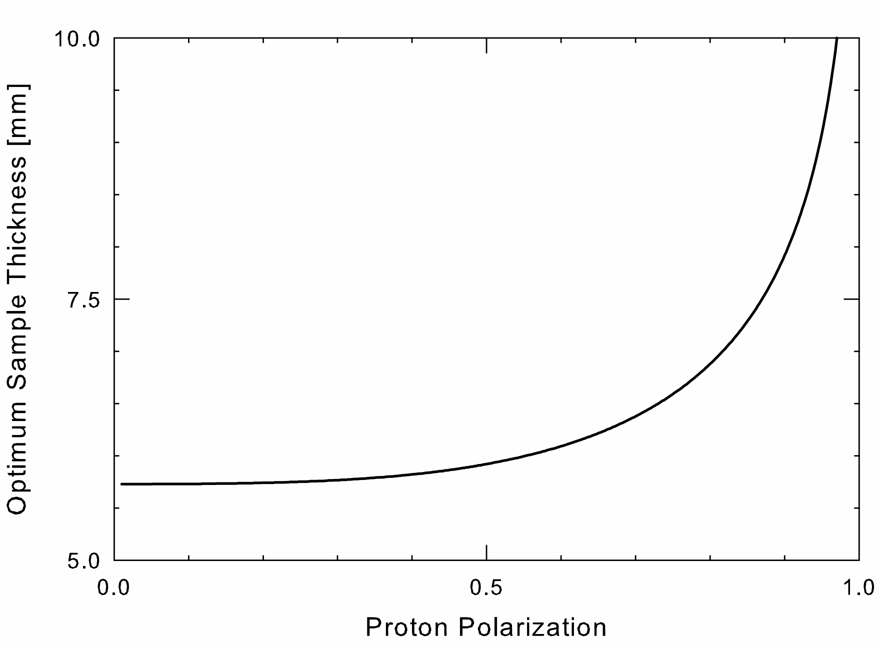
[Zimmer, Müller, Hautle, Heil, Humblot, Phys. Lett. B 455 (1999) 62]

Effective cross section $\sigma_{\pm} = \sigma_0(\lambda) \pm \sigma_P(\lambda) \cdot P$

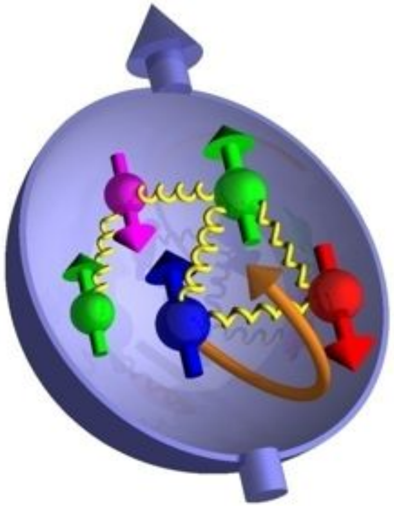
Intensity of beams behind the spin filter $N_{\pm} = \frac{I_0}{2} \exp[-(\sigma_0 \pm \sigma_P P)Nd]$

optimization of analyzing power and transmission given by Figure of merit $M = A^2 \cdot T$

➡



Outline



WHY

Neutron spin filtering with polarized protons

HOW to polarize protons :

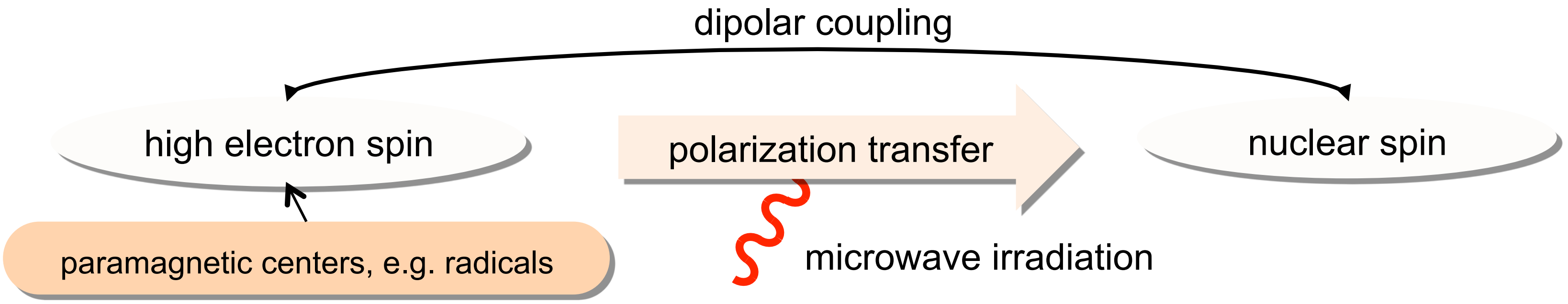
DNP using photo-excited triplet states

RESULTS / PRESENT STATUS

Experiments @ BOA / PSI

Dynamic Nuclear Polarization (DNP)

polarization transfer from electron spins to surrounding nuclei


dipolar coupling

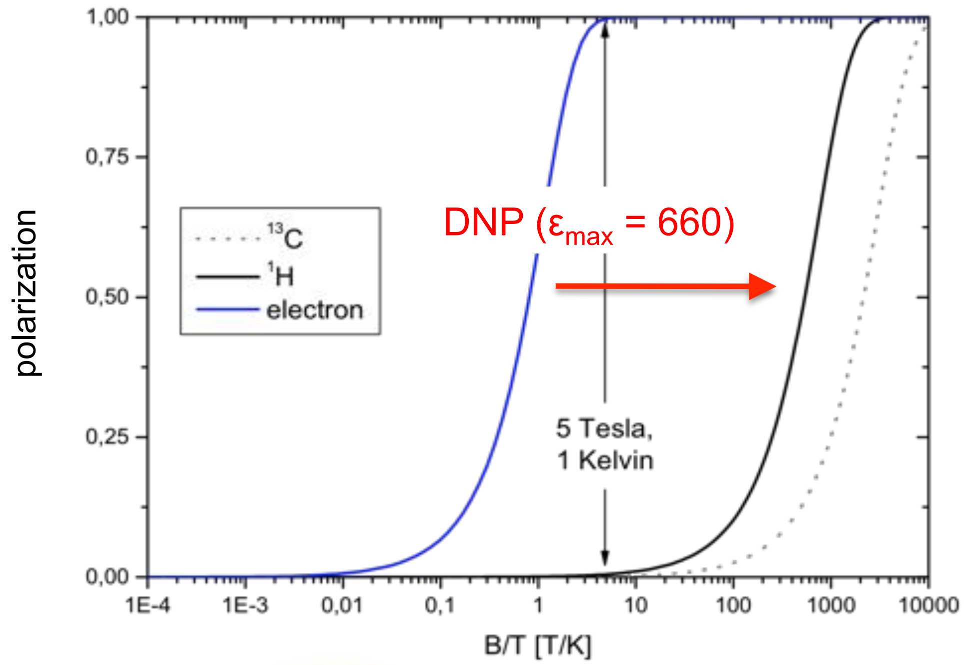
high electron spin

polarization transfer

nuclear spin

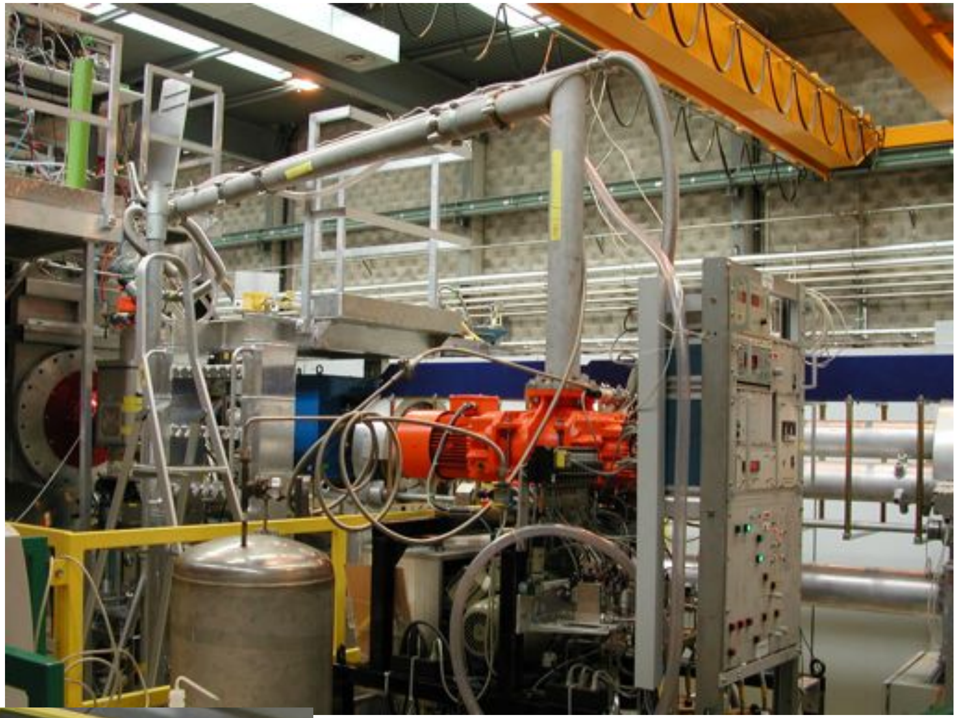
paramagnetic centers, e.g. radicals

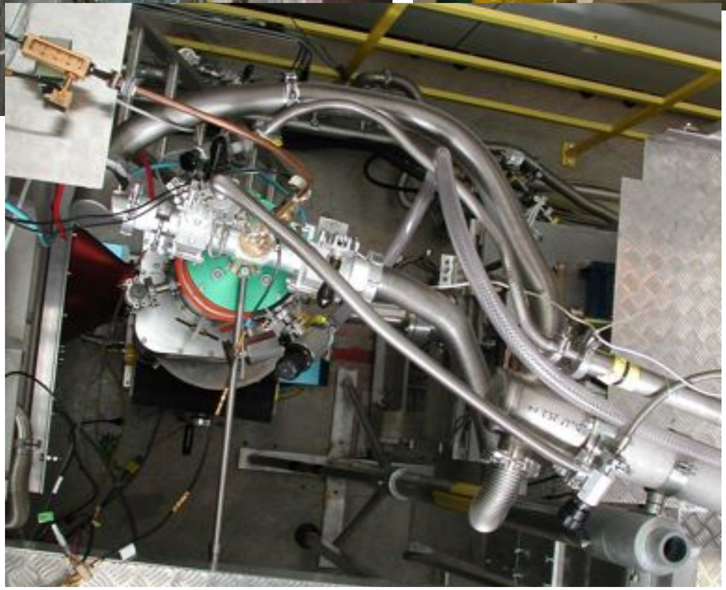
microwave irradiation


DNP (ε_{max} = 660)

- thermal electron polarization
- high magnetic field (2.5 – 5 T)
- low temperature (ca. 1 K)

Classical DNP system





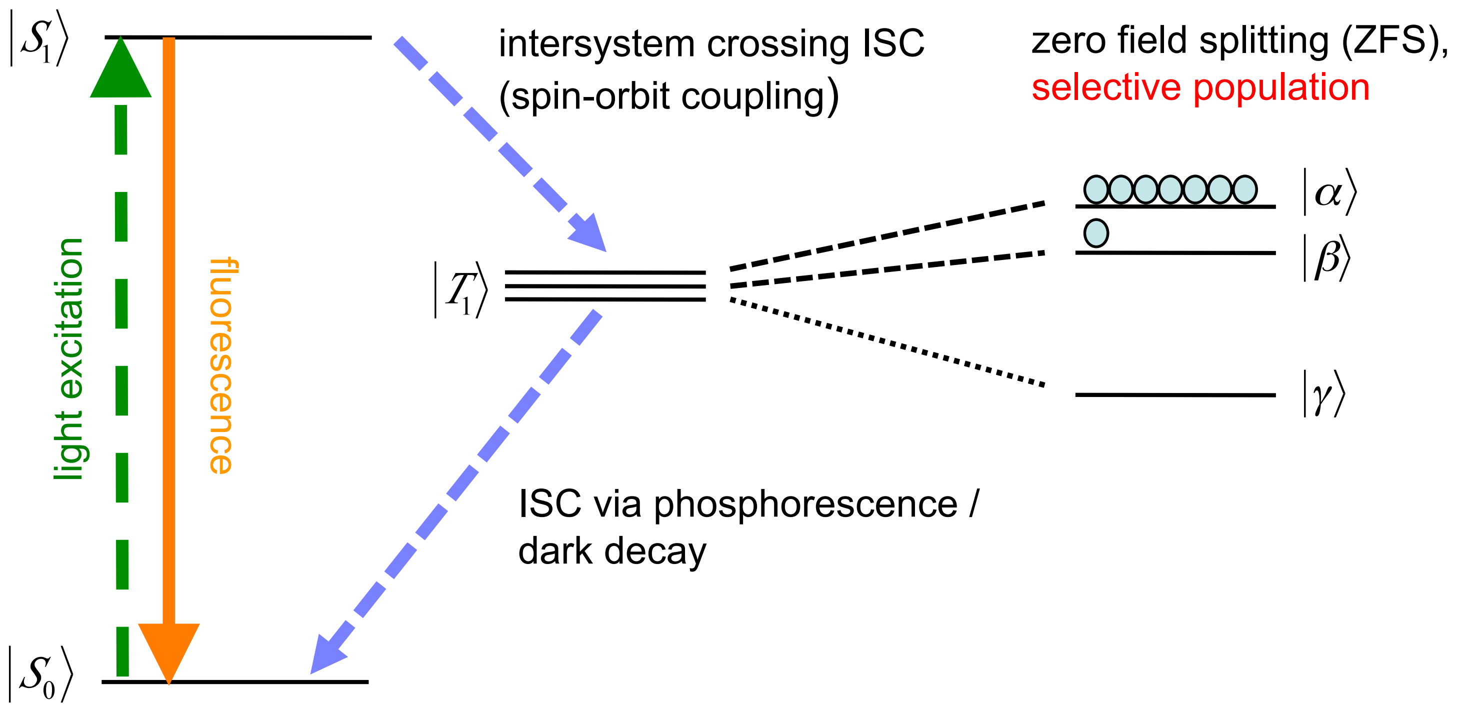
This is a compact system !!

1 K ^{4}He 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

The photo-excited triplet state as source of paramagnetism



electronic singlet states (S = 0)

electronic triplet states (S = 1) (paramagnetic)

$|S_1\rangle$

$|S_0\rangle$

light excitation

fluorescence

intersystem crossing ISC
(spin-orbit coupling)

$|T_1\rangle$

ISC via phosphorescence /
dark decay

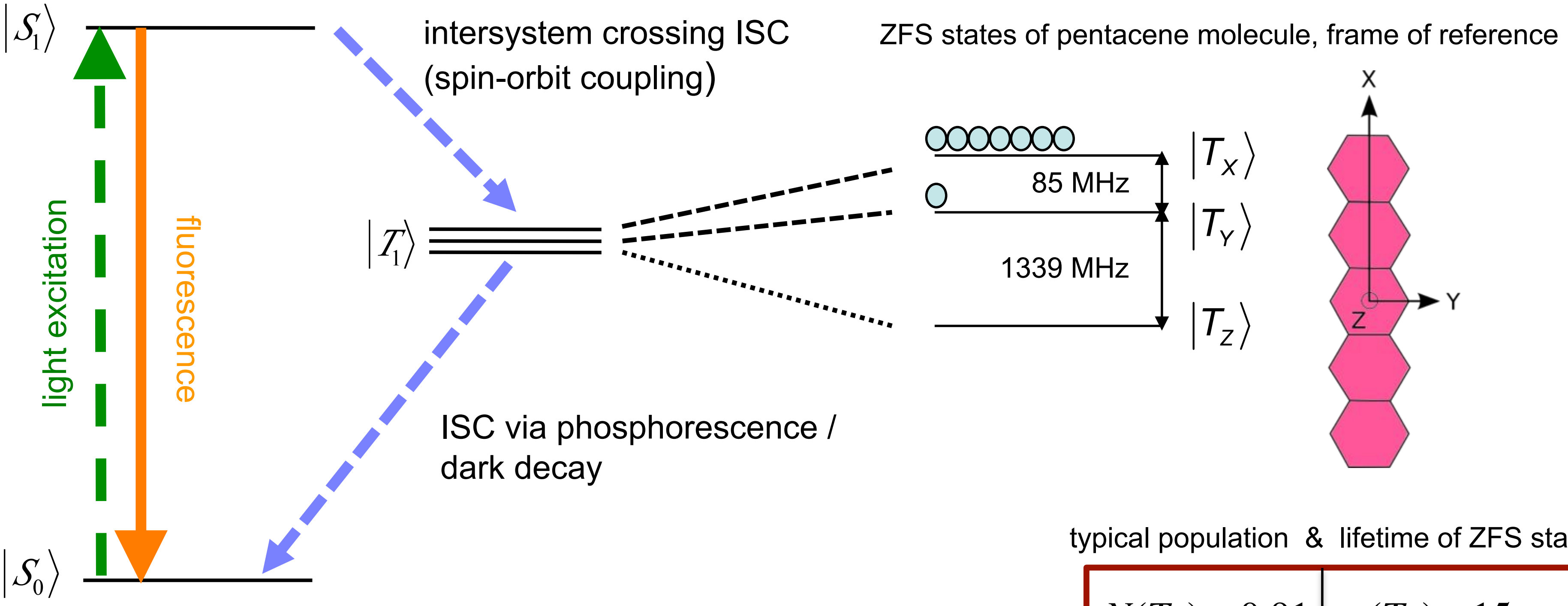
zero field splitting (ZFS),
selective population

$|\alpha\rangle$

$|\beta\rangle$

$|\gamma\rangle$

The photo-excited triplet state of pentacene



electronic singlet states (S = 0)

electronic triplet states (S = 1) (paramagnetic)

intersystem crossing ISC
(spin-orbit coupling)

ZFS states of pentacene molecule, frame of reference

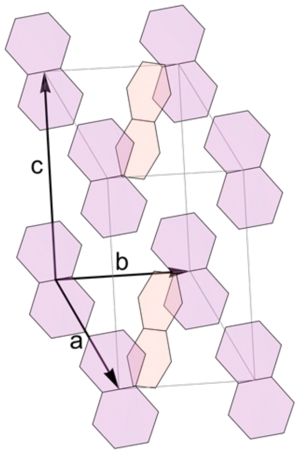
ISC via phosphorescence /
dark decay

typical population & lifetime of ZFS states

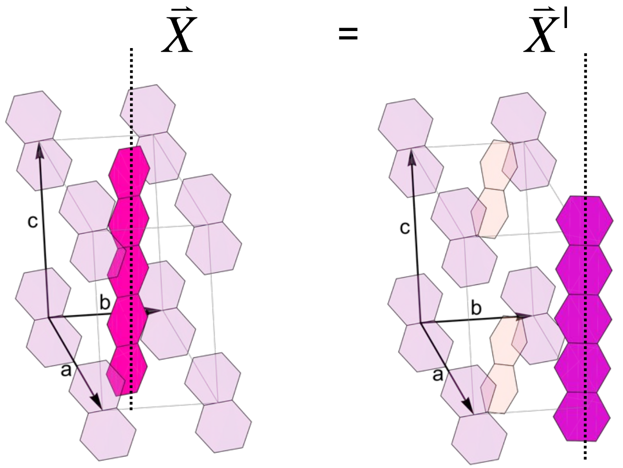
$N(T_X) = 0.91$	$\tau(T_X) = 15\mu s$
$N(T_Y) = 0.09$	$\tau(T_Y) = 35\mu s$
$N(T_Z) \approx 0$	$\tau(T_Z) > 200\mu s$

- short-lived high electron spin order within ZFS substates independent on magnetic field and temperature
- no paramagnetic relaxation without light
- ZFS triplet states linked to molecular frame, mixed in external magnetic field; orientational dependence of spin order

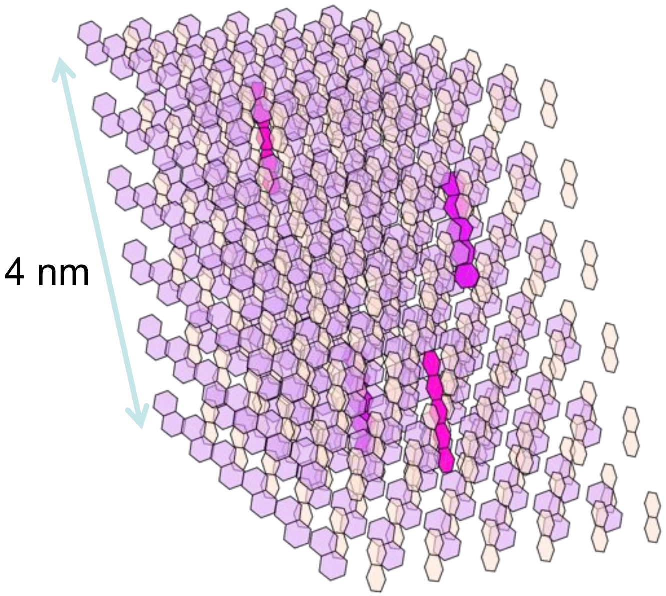
Pentacene:naphthalene crystals

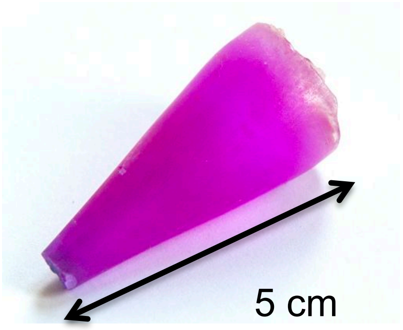


unit cell of naphthalene crystal



pentacene:naphthalene (two dopant sites)





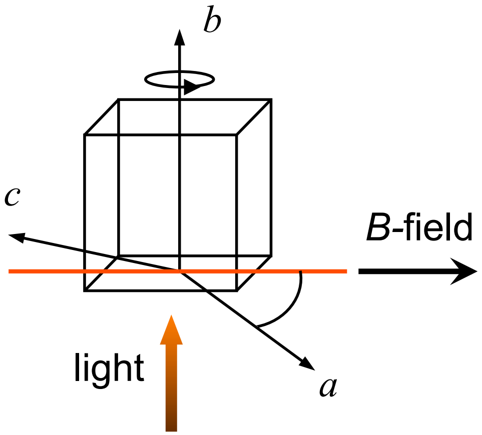
mixed single crystal
pentacene:naphthalene (5 x 10^{-5})

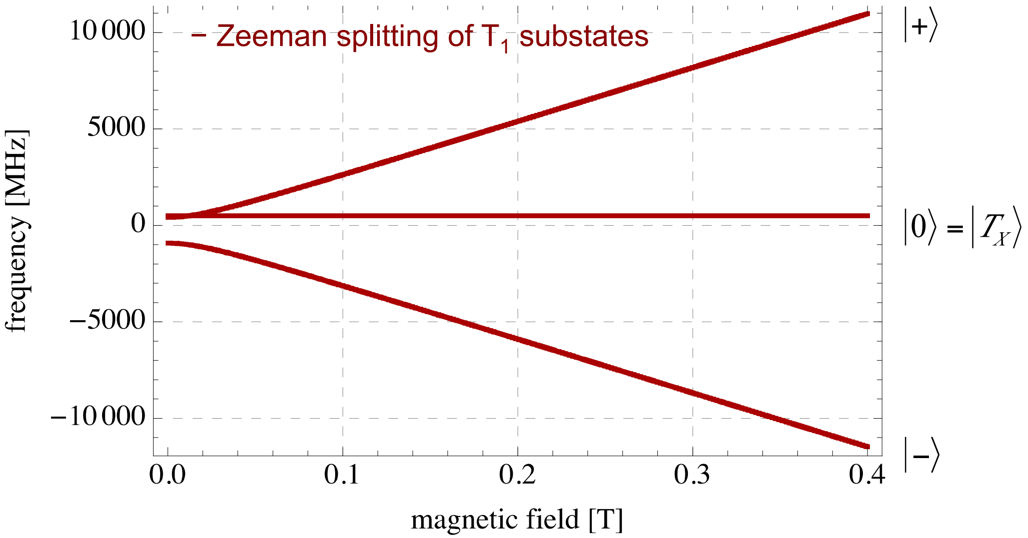
pentacene:naphthalene mixed single crystals

- magnetic field || X-axis as preferred / conventional alignment
- exceptional candidate for high bulk spin polarization in sizable crystals at moderate magnetic fields and temperature
- fast, repetitive DNP transfer scheme required: integrated solid effect (ISE) ➔ A. Henstra et al., Phys. Lett. A, 134 (1988)

Alignment of pentacene:naphthalene crystal

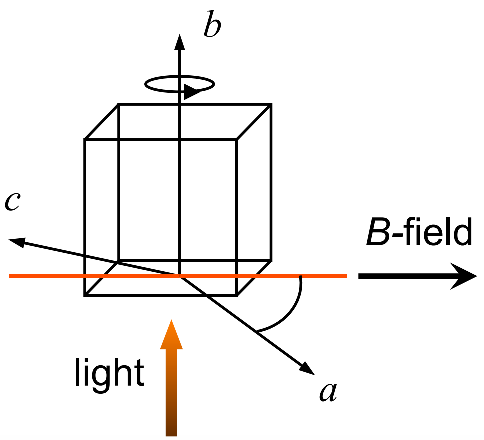
magnetic field || long molecular (X-) axis of pentacene

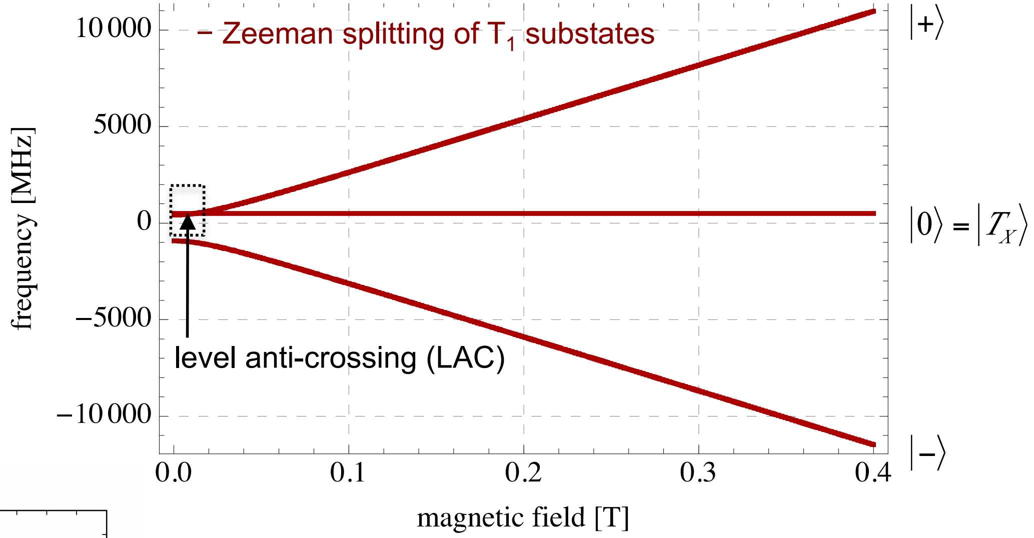


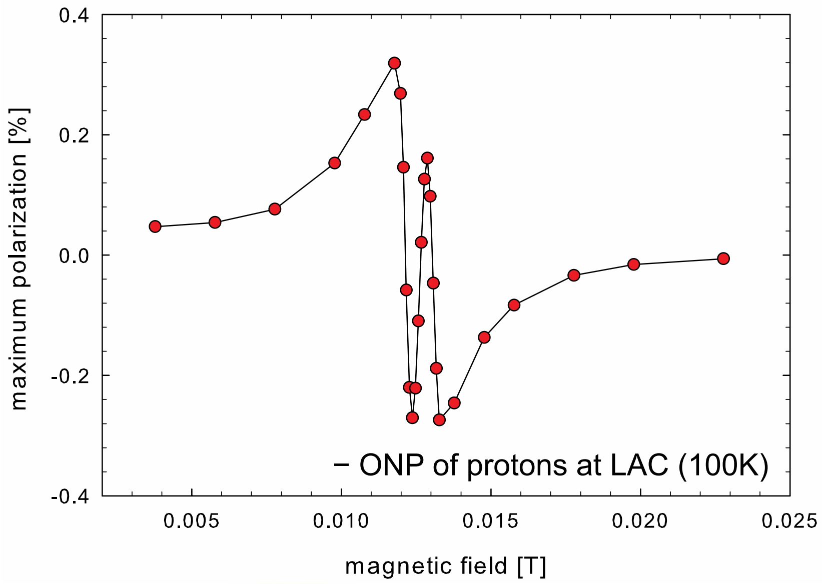


Alignment of pentacene:naphthalene crystal

magnetic field || long molecular (X-) axis of pentacene







optical nuclear polarization (ONP):

- “easy” to acquire high enhancements (> 4 orders) in short time (seconds to minutes)
- final polarization limited by fast relaxation at very low magnetic fields (order of hyperfine interaction)

Alignment of pentacene:naphthalene crystal

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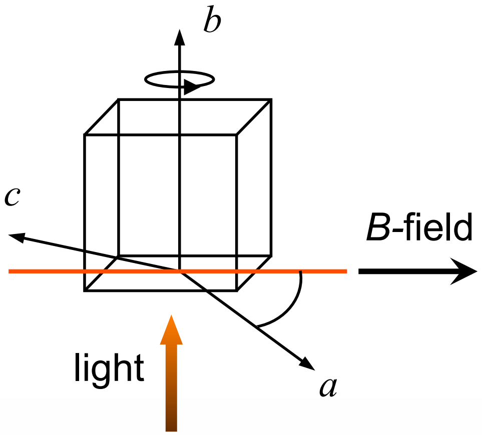
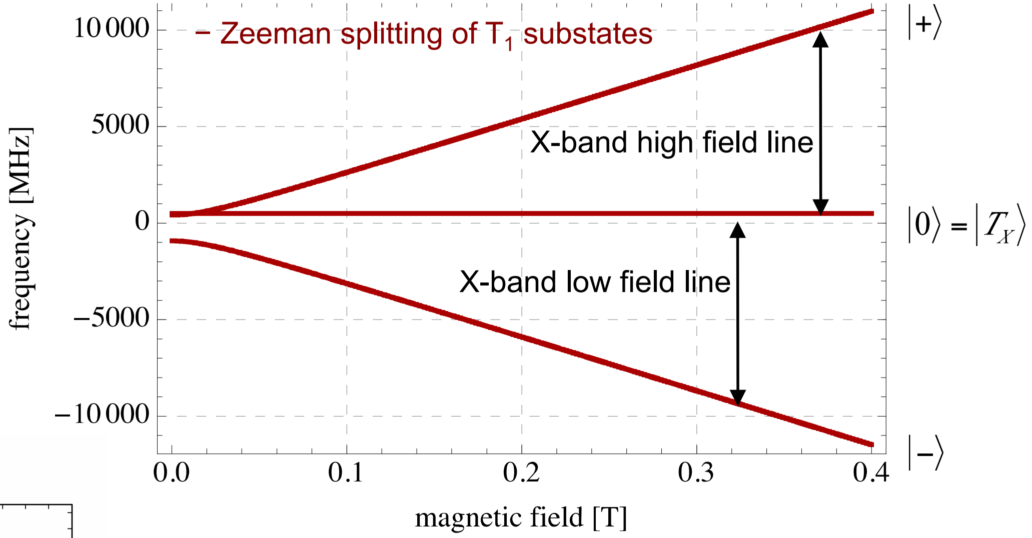
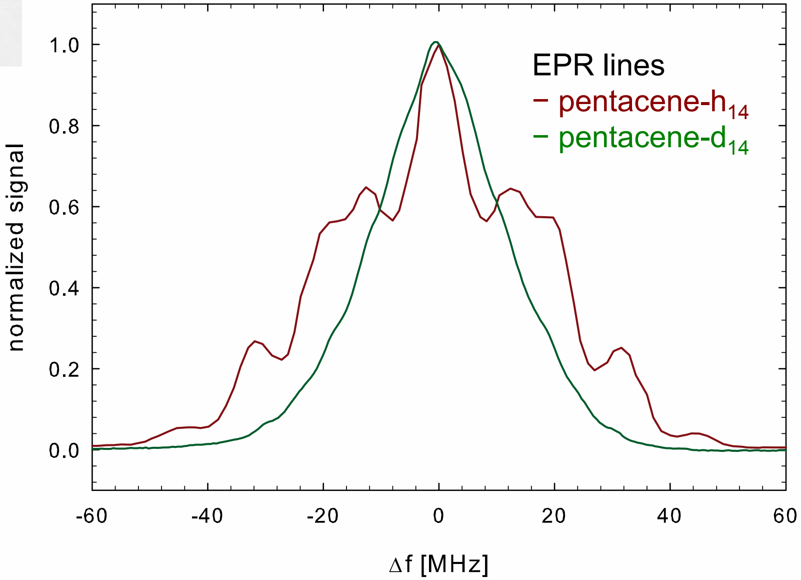




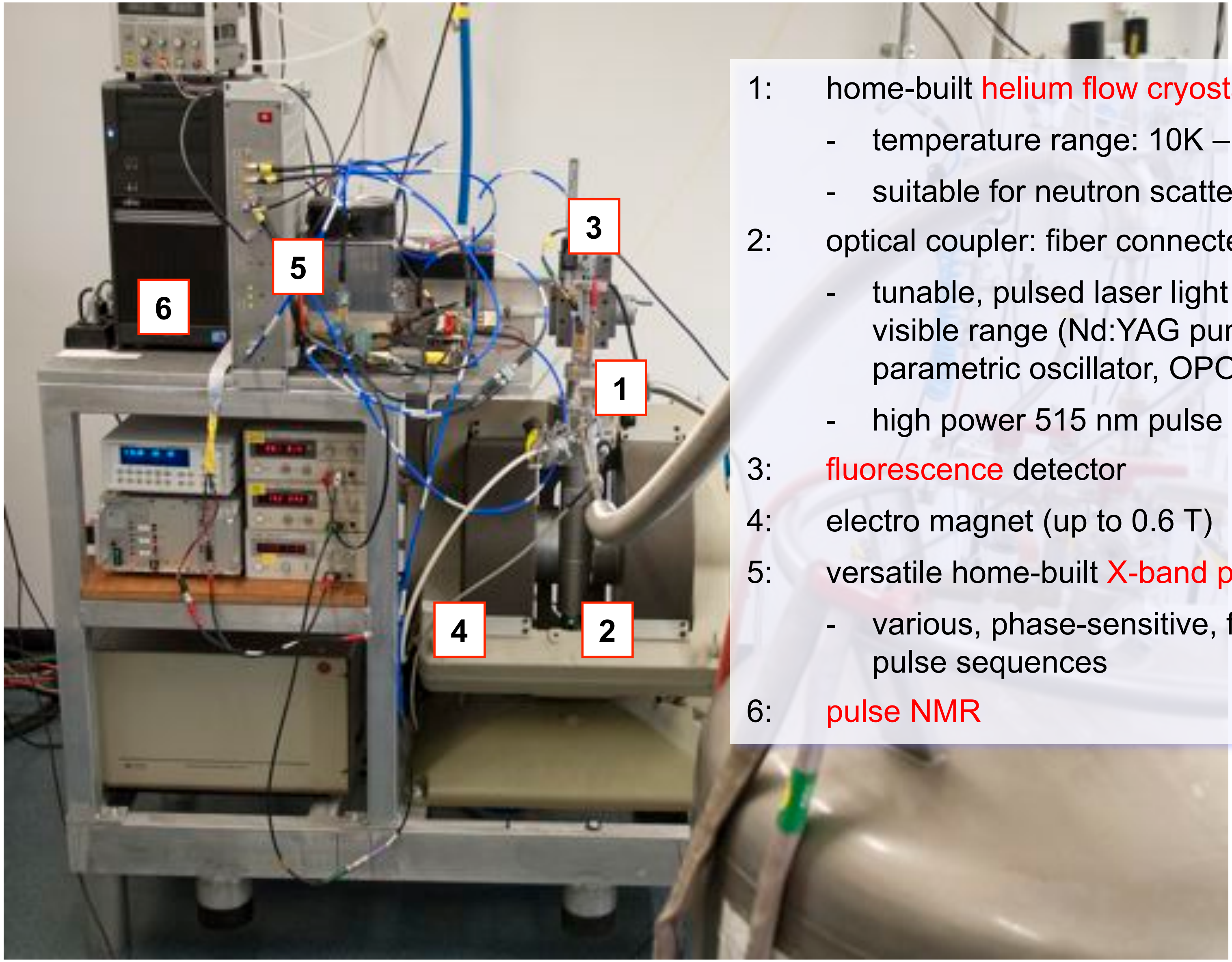


crystal alignment quality inspected by EPR:

- very symmetric, single lines
- hyperfine structure resolved
- maximum splitting between low field and high field line

Compact DNP setup

[T.R. Eichhorn et al., J. Magn. Res. 234, 58 – 66 (2013)]



1: home-built helium flow cryostat
   - temperature range: 10K – 300K
   - suitable for neutron scattering experiments

2: optical coupler: fiber connected to separate laser lab
   - tunable, pulsed laser light source in the visible range (Nd:YAG pumped optical parametric oscillator, OPO)
   - high power 515 nm pulse laser, optimized for DNP

3: fluorescence detector

4: electro magnet (up to 0.6 T)

5: versatile home-built X-band pulse ESR spectrometer
   - various, phase-sensitive, fast (ns) DNP pulse sequences

6: pulse NMR

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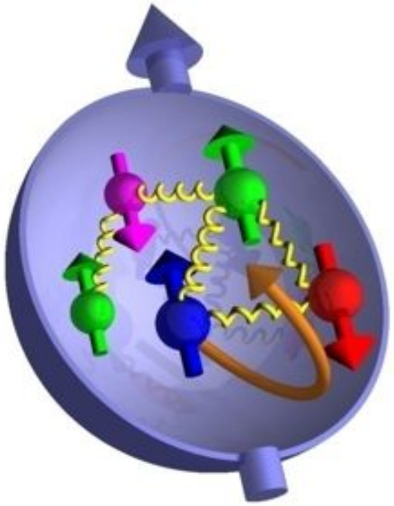
Outline



WHY

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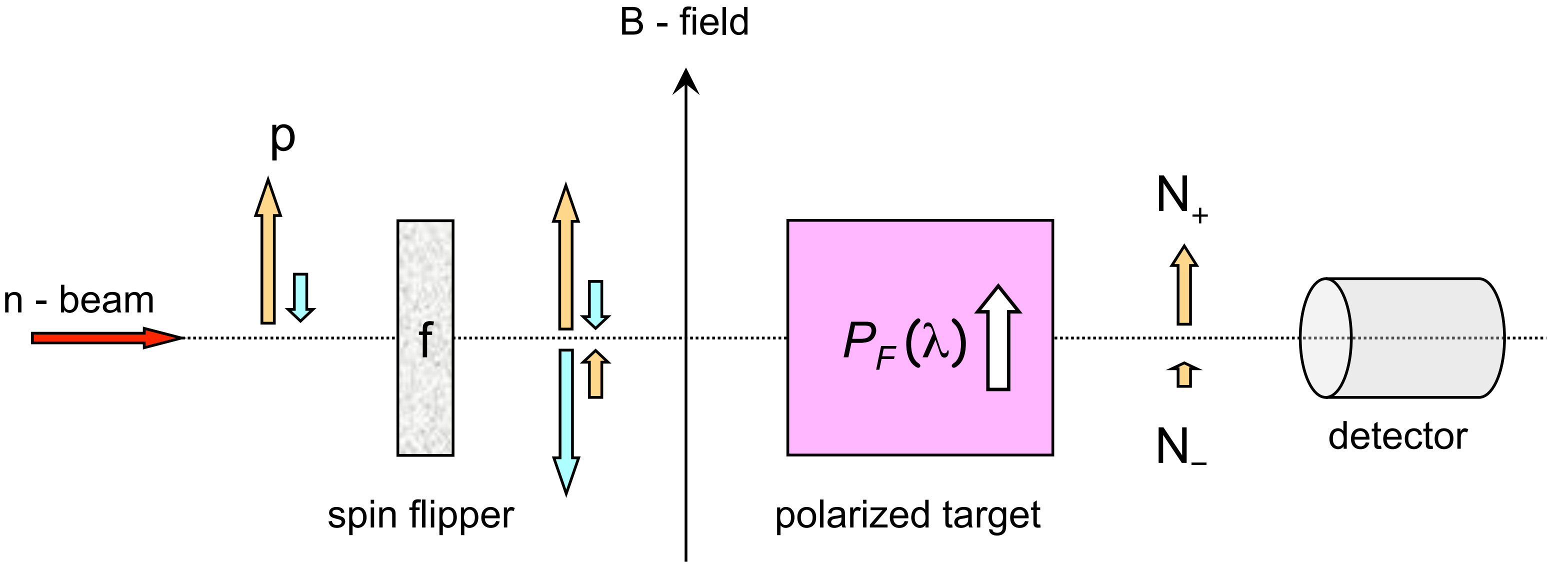
RESULTS / PRESENT STATUS

Experiments @ BOA / PSI

Spin Filter, Test of Principle - Experimental Scheme

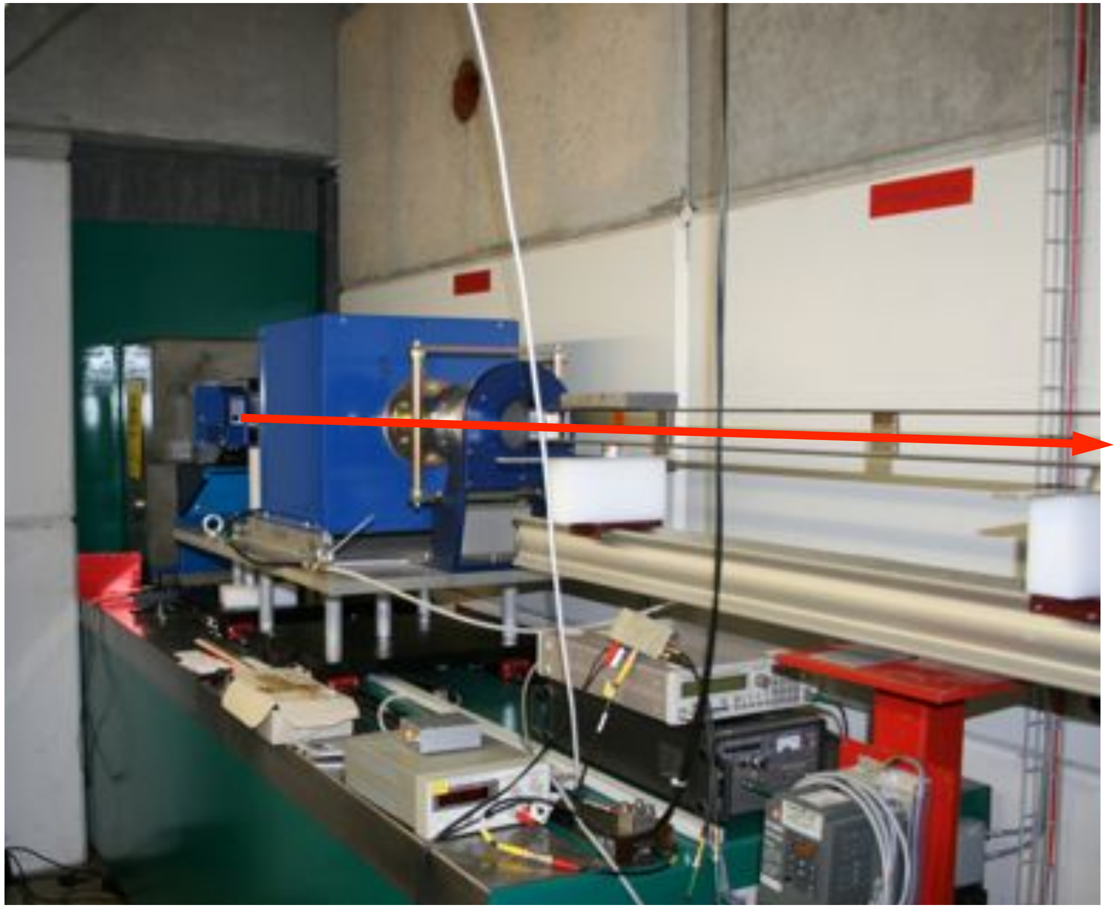


flipping ratio $R(\lambda) = \frac{N_+}{N_-} = \frac{1 + pP_F(\lambda)}{1 - fpP_F(\lambda)}$

- perform a ***test of principle*** for a triplet spin filter
- measure the ***polarization cross section*** as function of λ
- use the neutrons to ***characterize the target performance*** (DNP, relaxation etc..)

1st setup on neutron beamline BOA

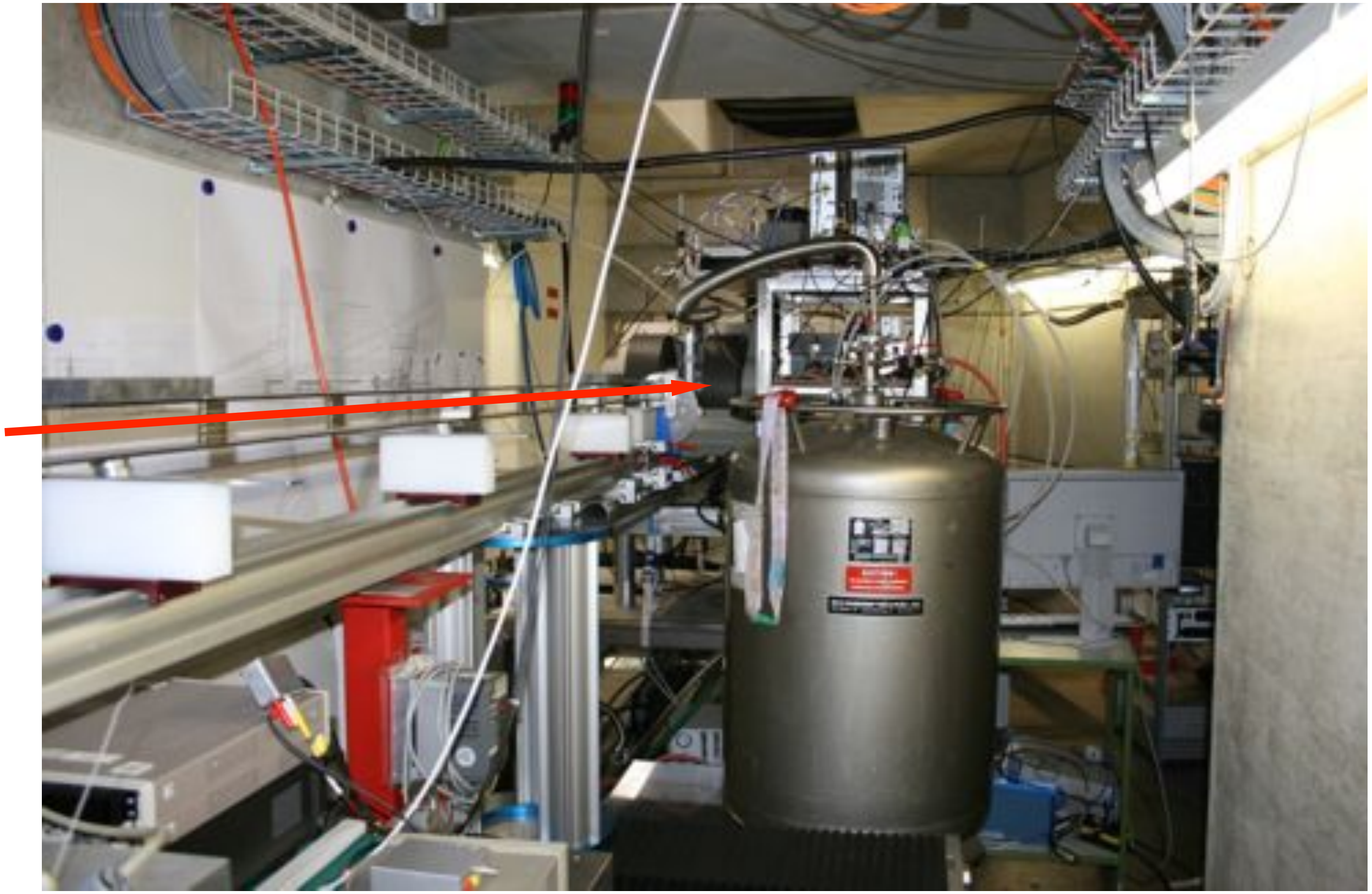
[M. Haag et al., Nucl. Instr. and Meth. A 678 (2012)]



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

1st setup on neutron beamline BOA

[M. Haag et al., Nucl. Instr. and Meth. A 678 (2012)]



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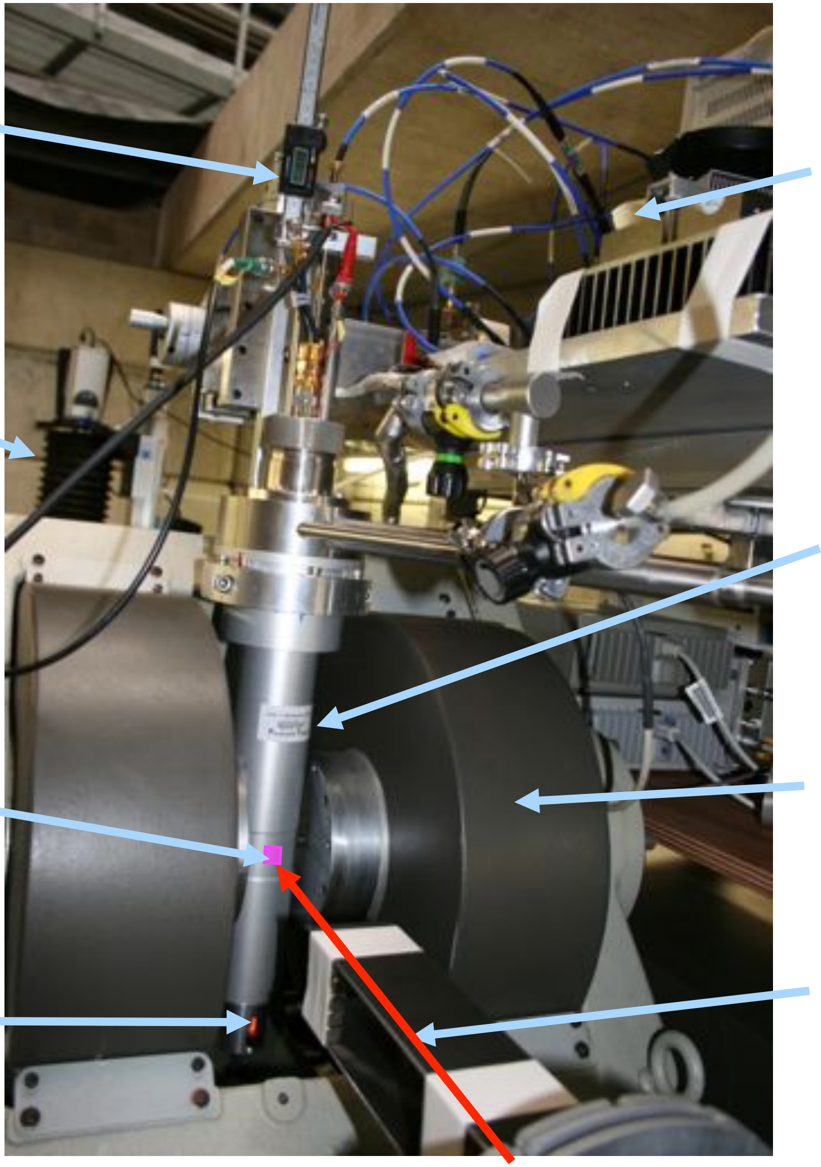


Caliper for sample positioning

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)

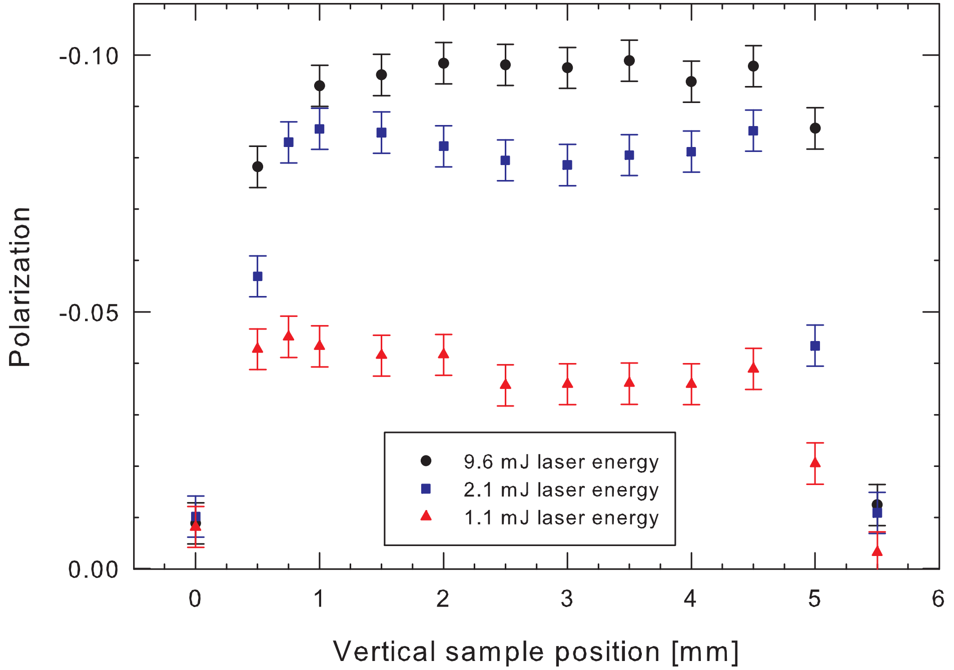
Neutron beam

Tim R. Eichhorn, PSTP workshop 2013
Tim.Eichhorn@psi.ch

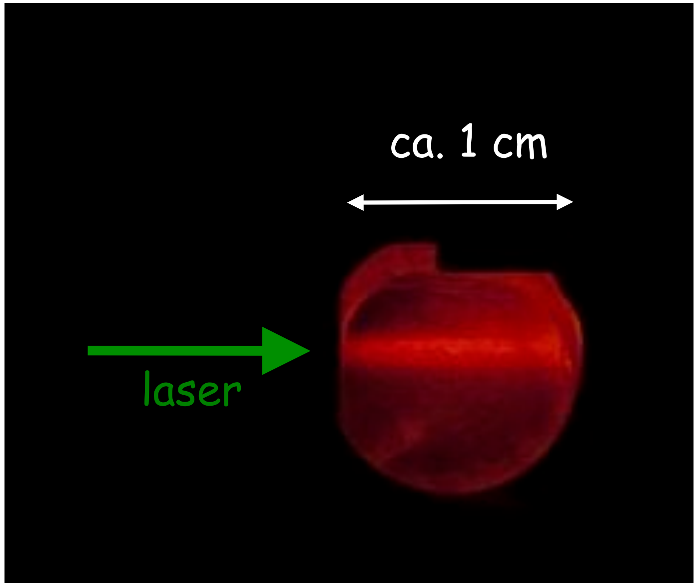
Polarization homogeneity

[M. Haag et al., Nucl. Instr. and Meth. A 678 (2012)]

homogeneous polarization in sizable crystals



light penetration studied by fluorescence



- pentacene concentration < 10^4 mol/mol absorption length of visible light > several mm (regime of linear absorption)
- homogeneous polarization enhancement (after complete buildup) for any laser intensity

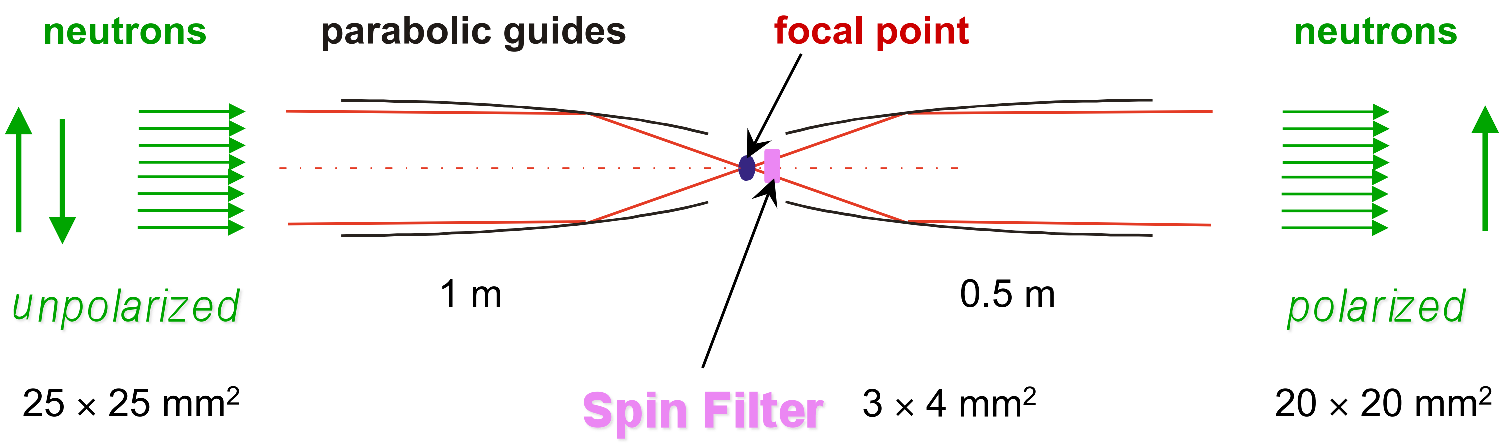
2nd generation: triplet spin filter + neutron optics

- 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

***Primary polarizer setup**: first test of principle experiment at PSI / BOA 2012*



neutrons

parabolic guides

focal point

neutrons

unpolarized

1 m

0.5 m

polarized

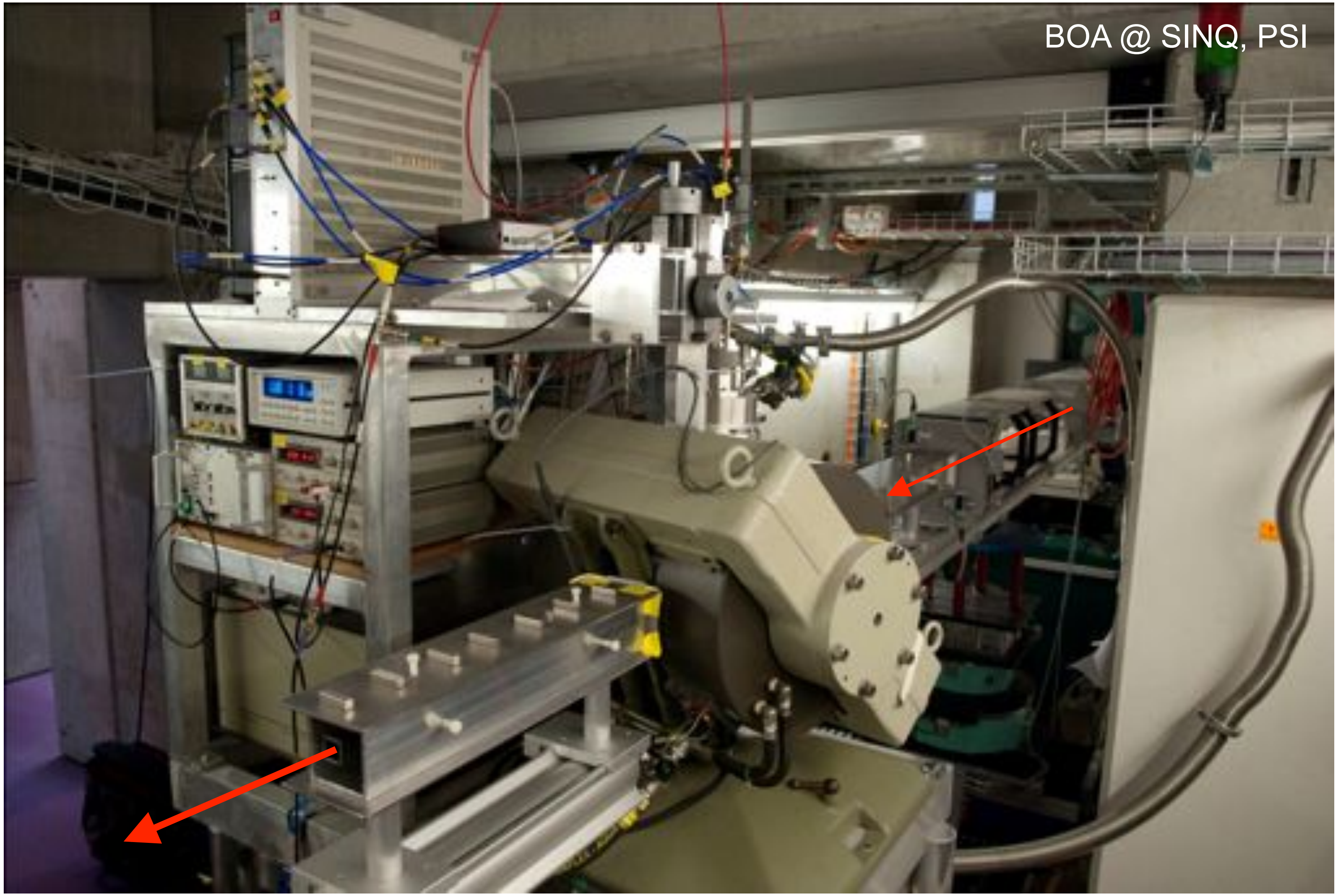
25 × 25 mm^2

Spin Filter

3 × 4 mm^2

20 × 20 mm^2

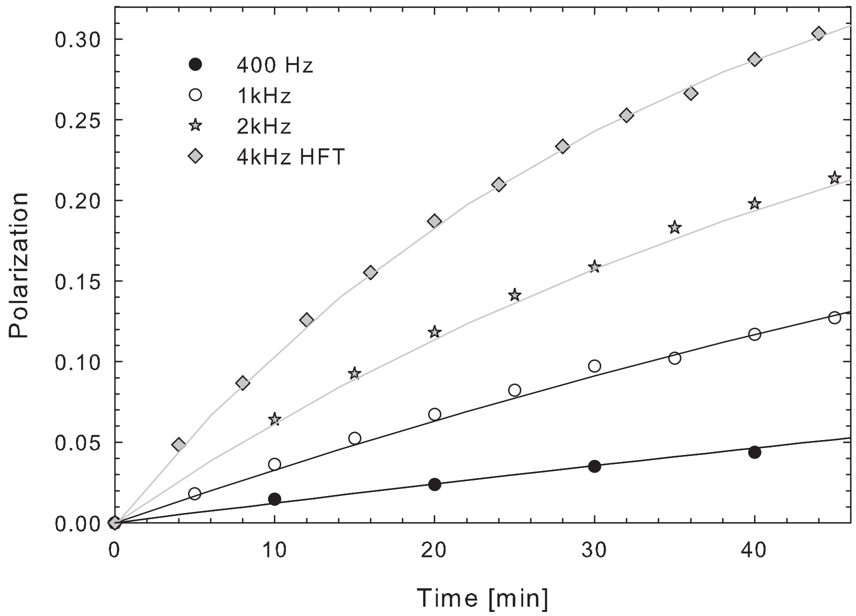
Triplet spin filter + neutron optics



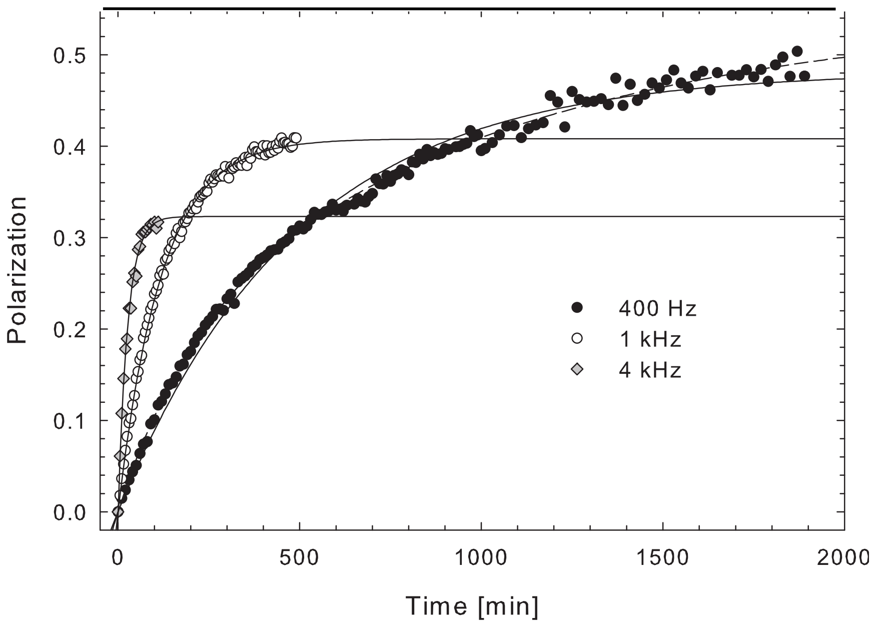
Improved DNP performance

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

^{1}H buildup, optimized ISE



complete buildup curves, ISE on low field line



experimental parameters / results:

- buildup efficiency: > 1% ^{1}H bulk polarization per minute possible
- up to 50% ^{1}H bulk spin polarization (enhancement of 5 orders) at 40 - 100K and 0.35T
- analyzing power of 0.5 for sample with d = 5 mm (up to 0.8 expected for sample with d = 1 cm)
- proton spin relaxation time up to 30h at 100K

Acknowledgements

Thank you for your attention!

M. Haag
P. Hautle
B. van den Brandt
W.Th. Wenckebach

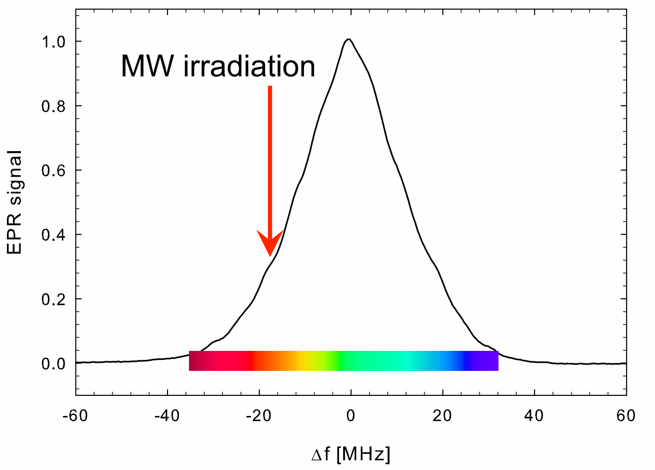
Paul Scherrer Institute

S. Jannin
A. Comment
J.J. van der Klink

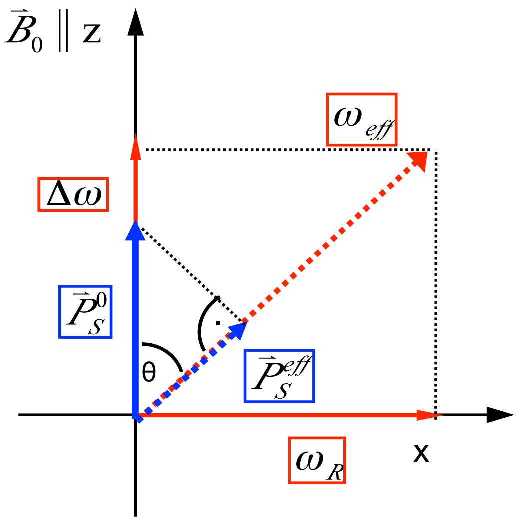
EPFL Lausanne

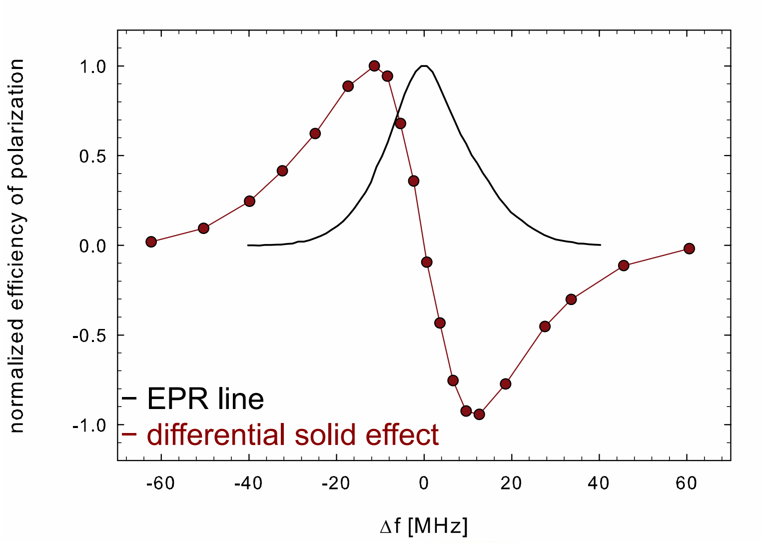
Solid effect

irradiation with microwaves (MW) during triplet state lifetime, microwave amplitude ω_R , offset $\Delta\omega$



concept in rotating frame, polarization $\vec{P}_S$





drawbacks / low efficiency of solid effect:

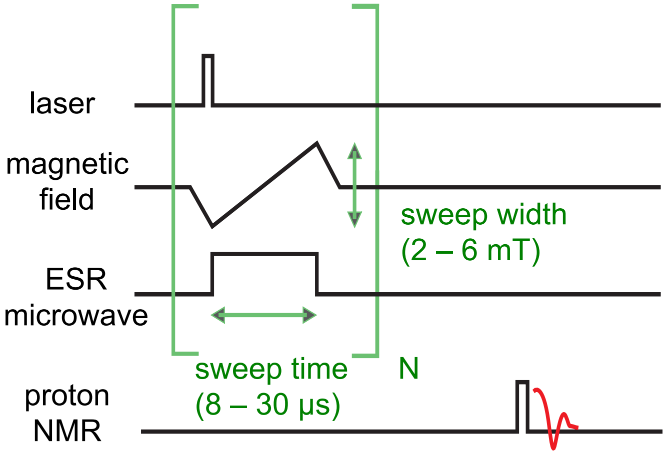
- EPR line is inhomogeneously broadened: only few spin packets participate in polarization transfer
- ΔEPR > f_{NMR} at X-band: differential solid effect
- effective polarization vector reduced by cos(θ)
- short lifetime of triplet states (small “duty cycle”)

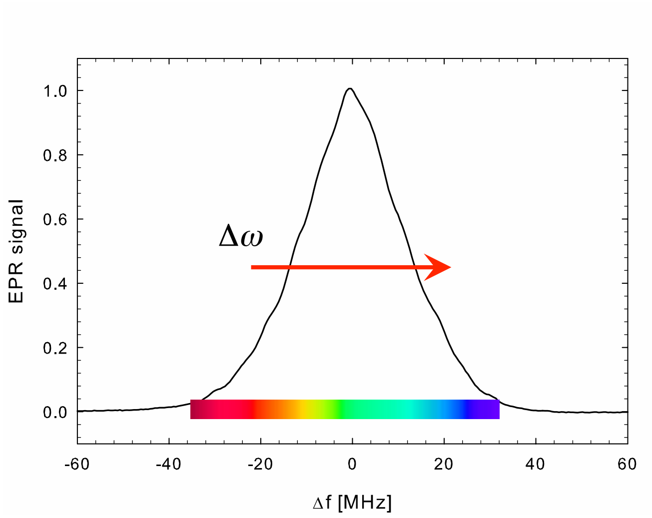
Integrated solid effect, ISE

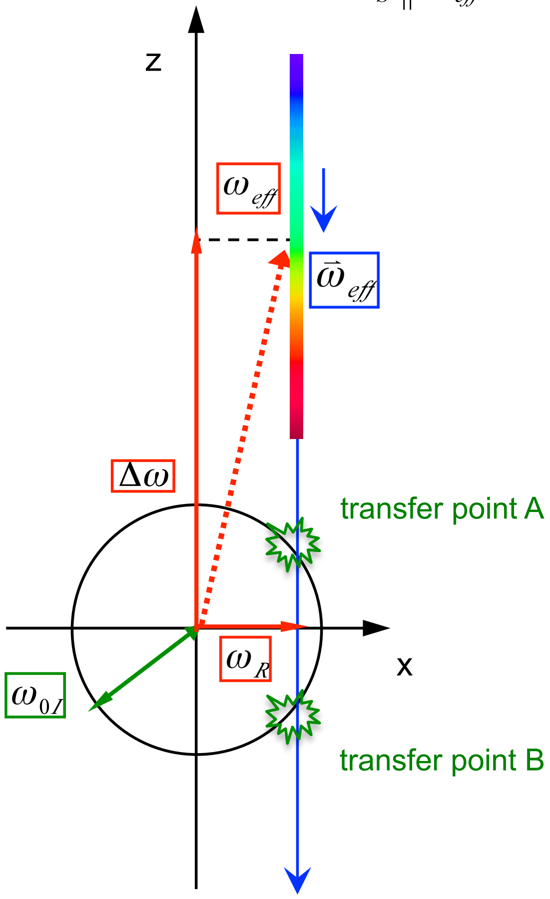
[A. Henstra et al., Phys. Lett. A, 134 (1988)]

irradiation with microwaves (MW) during triplet state lifetime,
microwave amplitude ω_{1S} on resonance,
adiabatic magnetic field sweep through EPR line: $\Delta\omega$
Hartmann-Hahn type resonance condition $\omega_{0I} = \omega_{eff}$

concept in rotating frame, polarization $\vec{P}_S \parallel \vec{\omega}_{eff}$



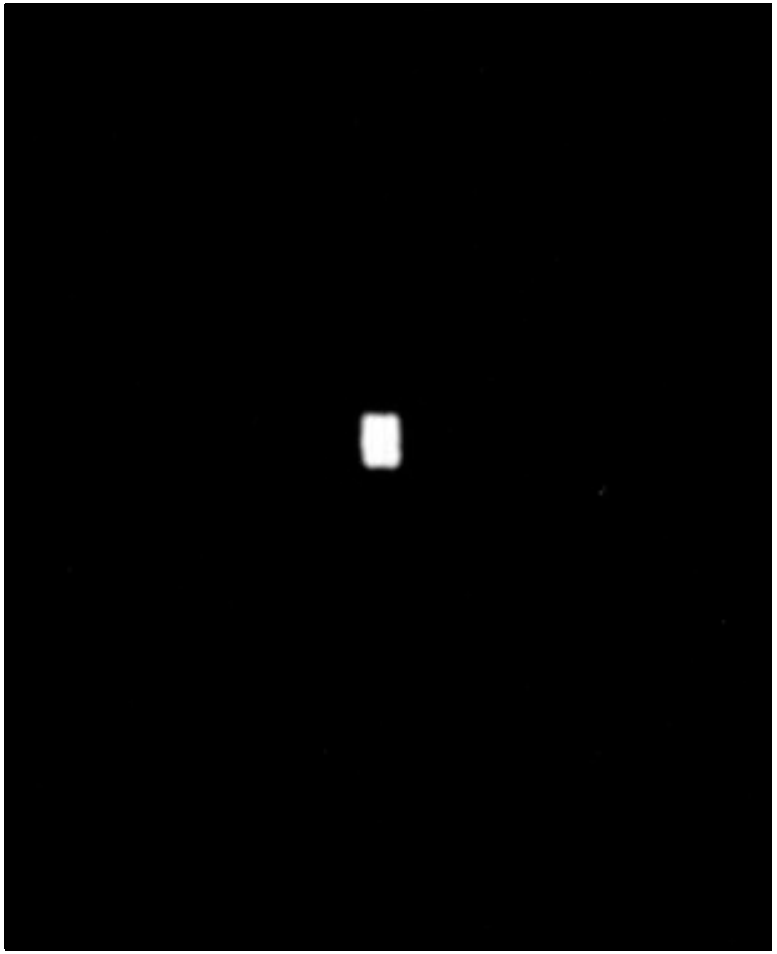


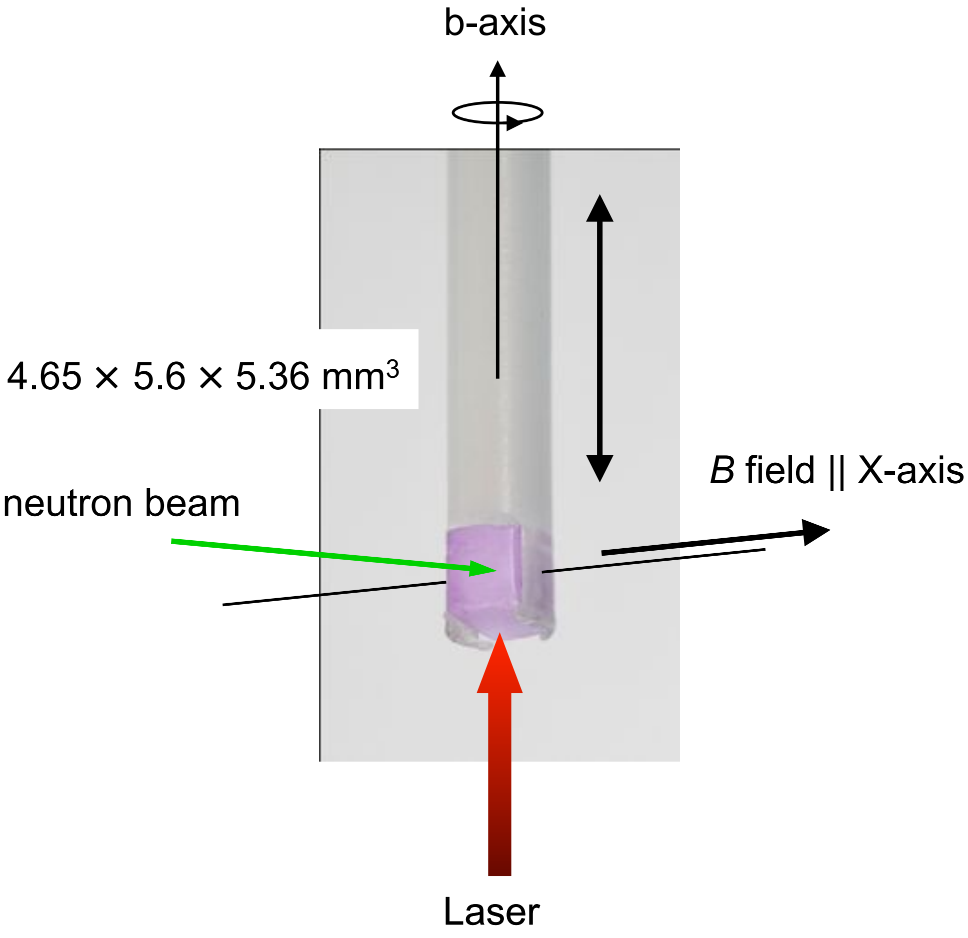


advantages of the integrated solid effect (ISE):

- all spin packets of the EPR line participate in the polarization transfer
- no differential effects
- polarization is not reduced in rotating frame
- fast and efficient polarization transfer

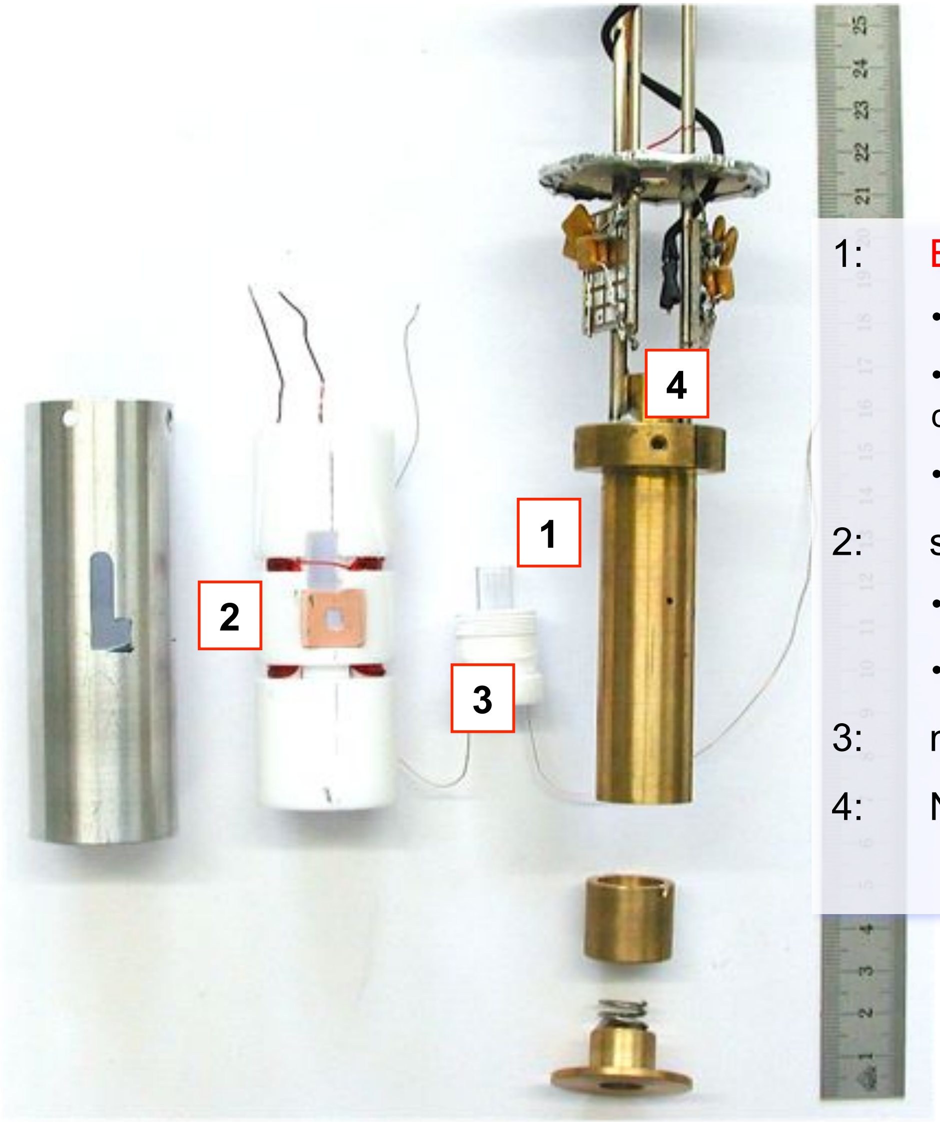
Sample positioning





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

The cryostat insert



1: ESR dielectric ring resonator with TE_{011} brass cavity

- sample size: 14 mm height, 7 mm inner diameter (< 100 mm^3)
- dielectric material: sapphire (ε ca.10): magnetic field is concentrated (filling factor 0.76)
- Q value up to 2000, B_1 field amplitude up to 0.6 mT

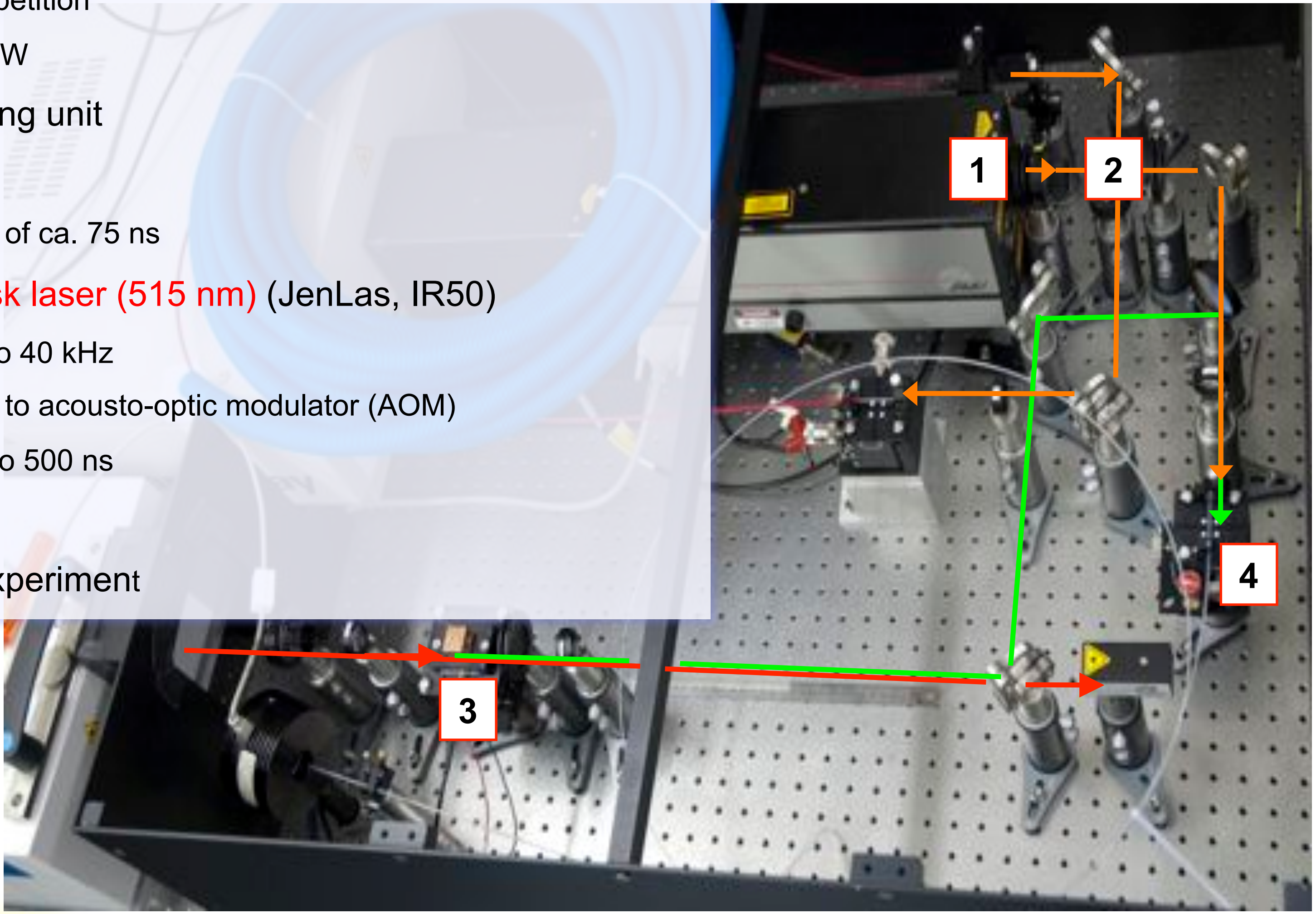
2: saddle coil for magnetic field sweep

- driven by a linear power operational amplifier (Servowatt DCP390)
- up to 0.6 mT/us sweep speed

3: monitoring NMR coil (below sample)

4: NMR coil for TE calibration (sample shuttle)

Multipurpose laser setup



1: Nd:YAG (3rd harmonic) pumped optical parametric oscillator (OPO) (GWU, premiScan)

- tunable wavelength: 480 nm – IR
- 10 ns pulselength, 30 Hz repetition
- average power typically 0.5 W

2: optional pulse train forming unit

- beam splitter / combiner
- 20 m fiber, pulse separation of ca. 75 ns

3: frequency doubled IR disk laser (515 nm) (JenLas, IR50)

- base repetition from 8 kHz to 40 kHz
- pulse selection via interface to acousto-optic modulator (AOM)
- pulse length range: 150 ns to 500 ns
- average power: > 10 W

4: fiber coupling stage to experiment