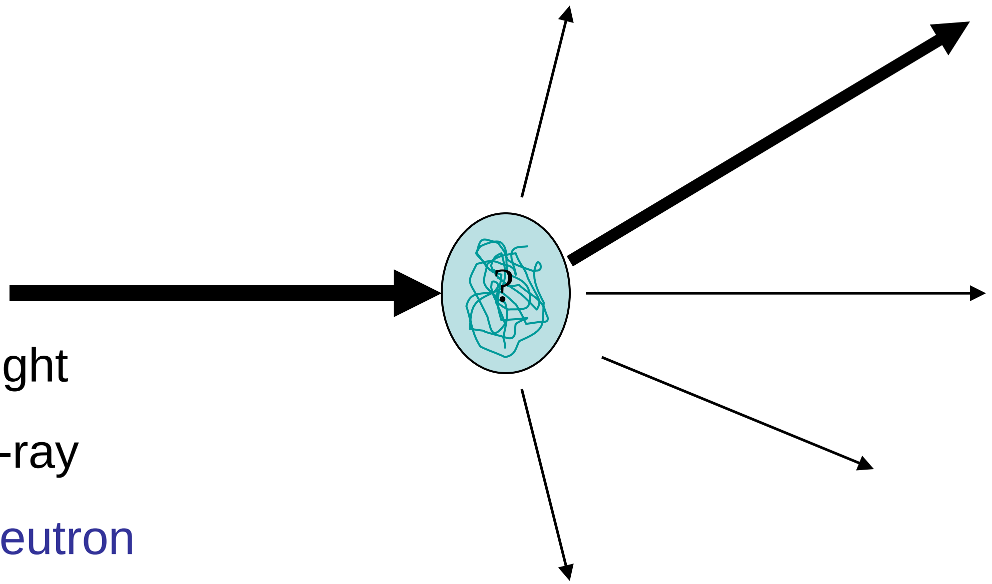
Neutron Spin Echo: The Instrumentation and the Science

Oak Ridge 2010

Michael Ohl

Scattering Experiment



$$k = \frac{2\pi}{\lambda}$$

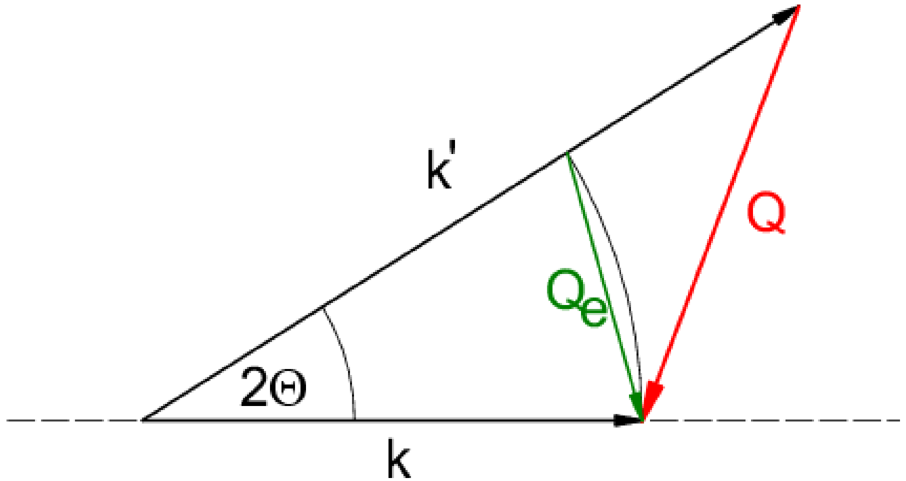
Light

X-ray

Neutron

……

Reveals structure and eventual Dynamics



Relations for neutrons

- $k = 2\pi / \lambda$ wavenumber → wavelength
- $k = 2\pi\, mv/h$ wavenumber → momentum
- $\lambda = h / mv$ wavelength → velocity
- $E = ½\, mv^2$ energy → velocity
- $E = ½\, h^2/m^2\lambda^2$ energy → wavelength
- ......

momentum transfer

$\underline{Q} = \underline{k}_i - \underline{k}_f$

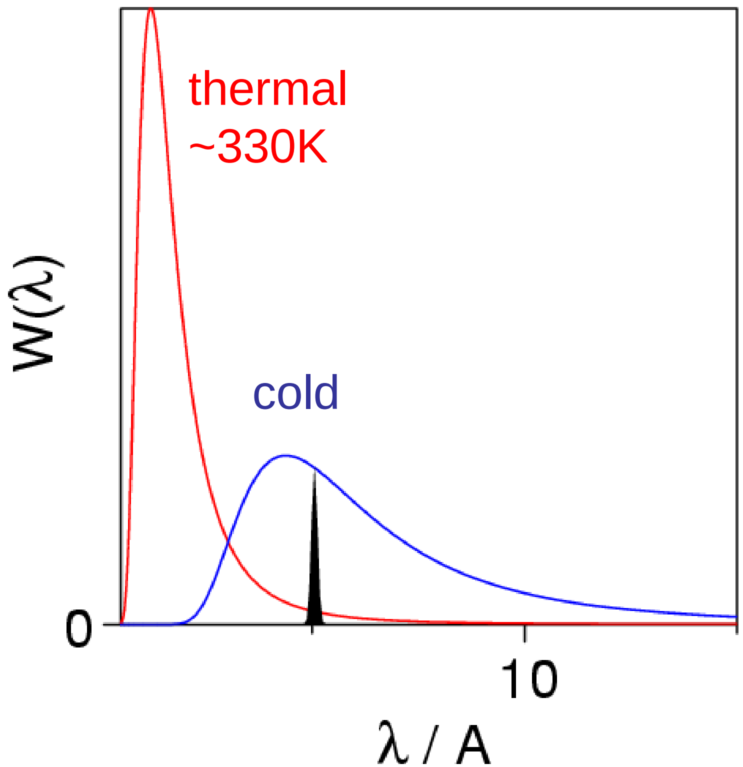
energy transfer

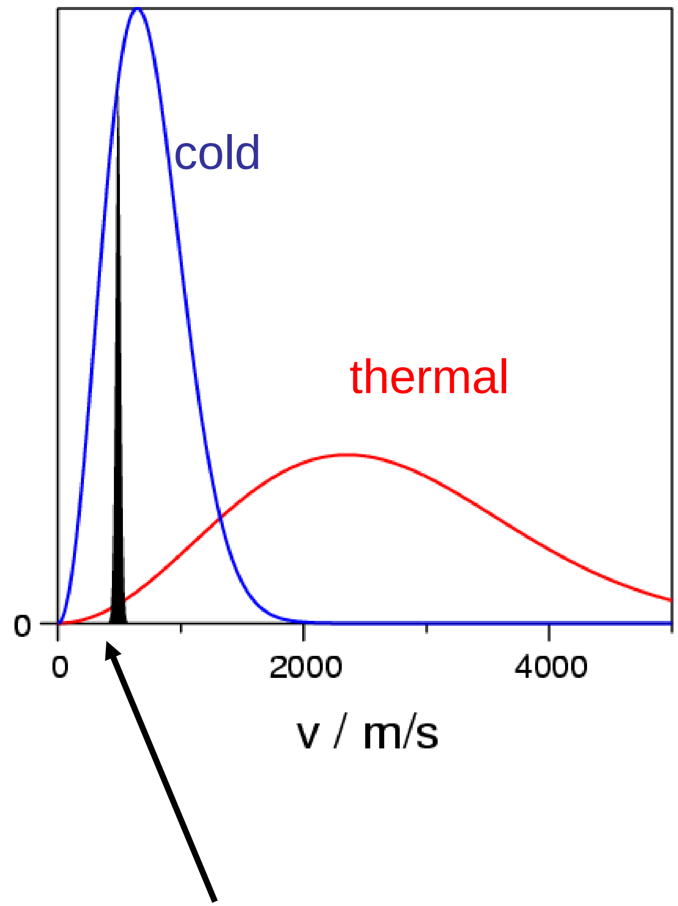
$\Delta E = E_i - E_f = \hbar\omega$

$= ½\, h^2/[m^2\ (\lambda^2_i - \lambda^2_f)]$

$= ½\, m\ (v^2_i - v^2_f)$

Neutron sources yield Maxwellian type spectra



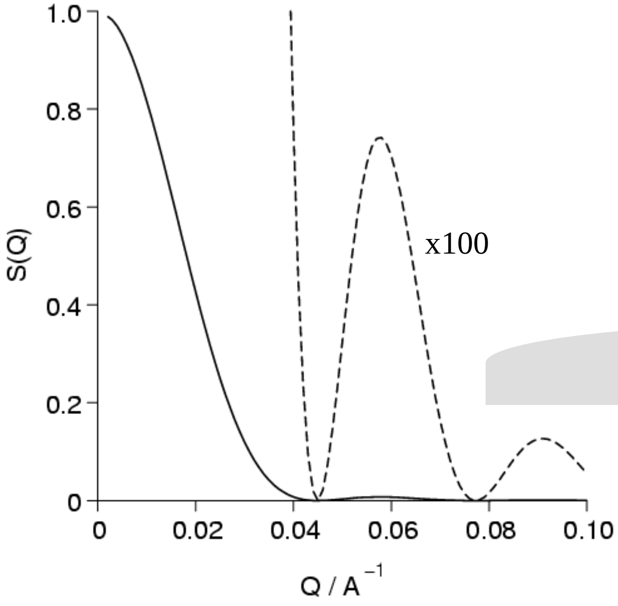


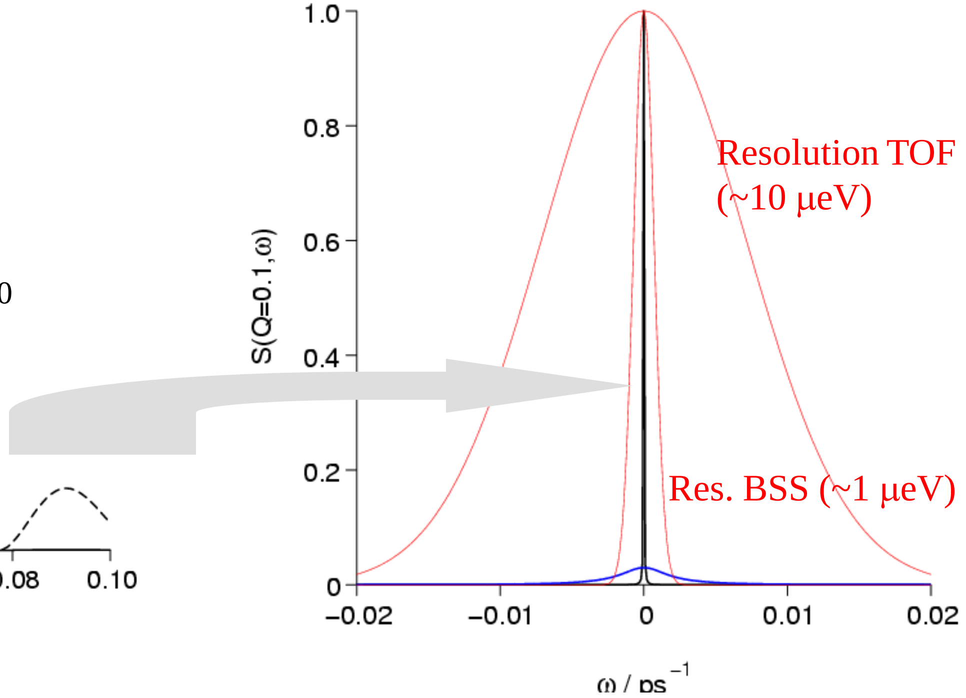
M(λ,T) = 2 a2 λ^(-5) exp(-a/λ^(2))
is the normalized Maxwellian spectrum, i.e. its integral over wavelength λ between 0 and ∞ equals 1. λ is measured in units of Å, **T is the effective spectral temperature in K, and**
a = 949 / T.

E=1.3 meV → 2 x 10^{12} s^{-1}

….in mesoscopia….

Sphere R=100Å ………………..diffusing in “water”



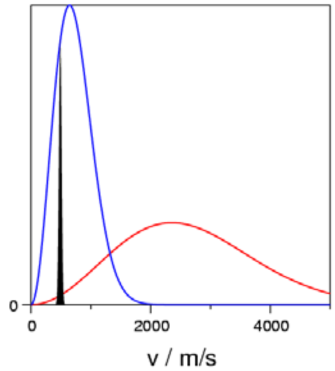


SANS

$$\omega \sim \Delta v$$

$$\Delta v/v < 10^{-4}$$

Need to detect Δv smaller than 10^{-4} v



direct filter-filter

NO intensity !

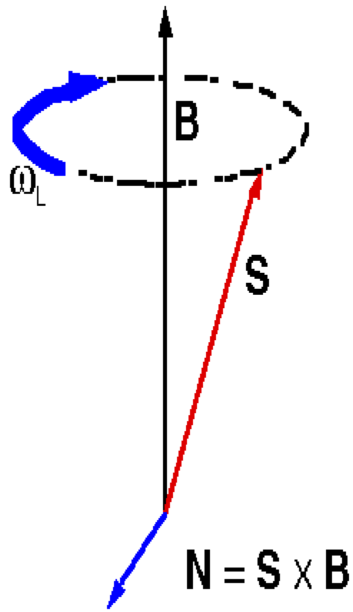
Spin-echo trick:

use precessing neutron spin as watch !

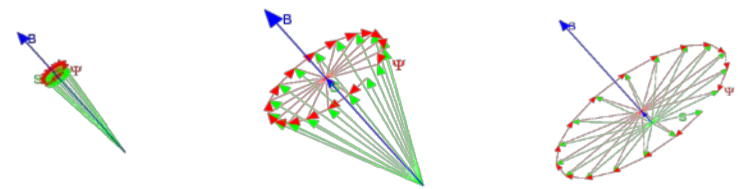
Neutrons in magnetic fields: Precession

$$\frac{d\underline{s}}{dt} = \gamma\, \underline{s} \times \underline{B}$$

- The neutron will experience a torque from a magnetic field perpendicular to its spin direction:
  - Precesses with the Larmor frequency $\omega_L = \gamma B$.
  - Only the strength of the field determines the precession rate.



$$\frac{d\mathbf{S}}{dt} = \gamma \mathbf{S} \times \mathbf{B} = \mathbf{S} \times \omega_L$$



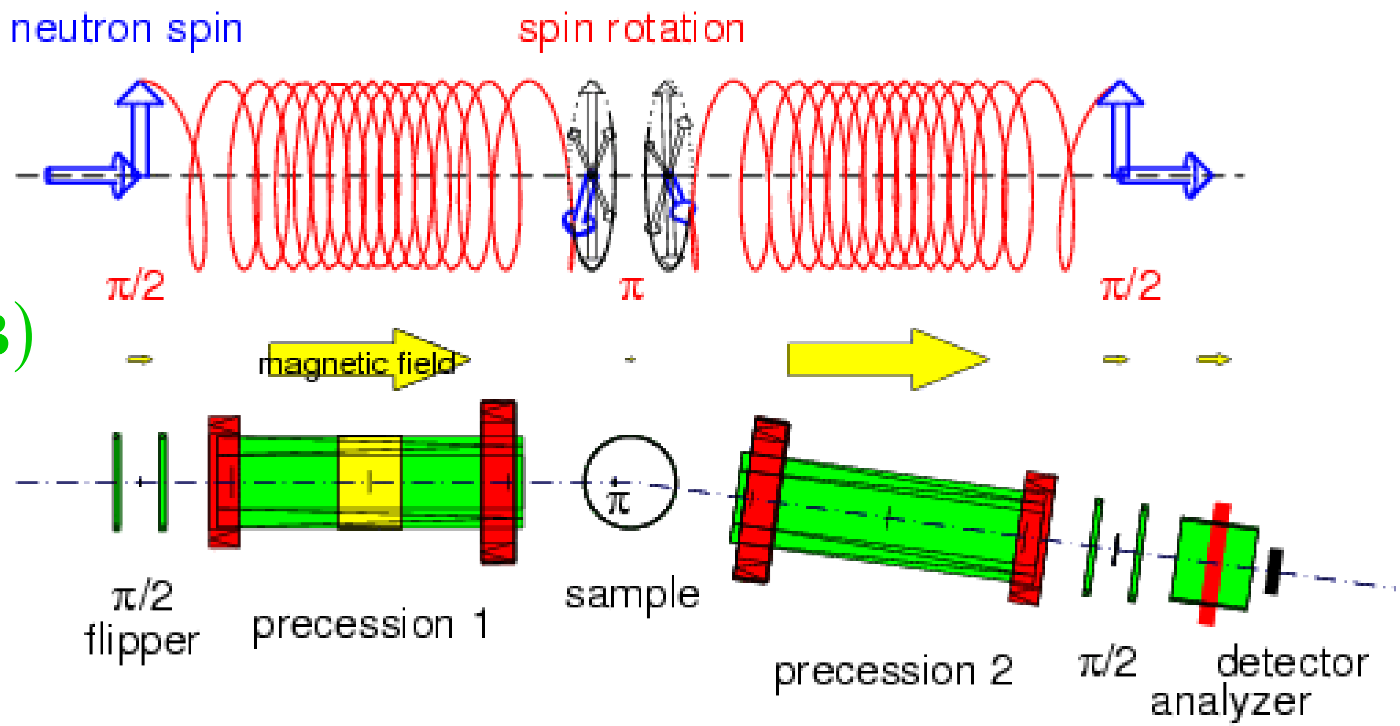
$$\Omega = \gamma\, B$$

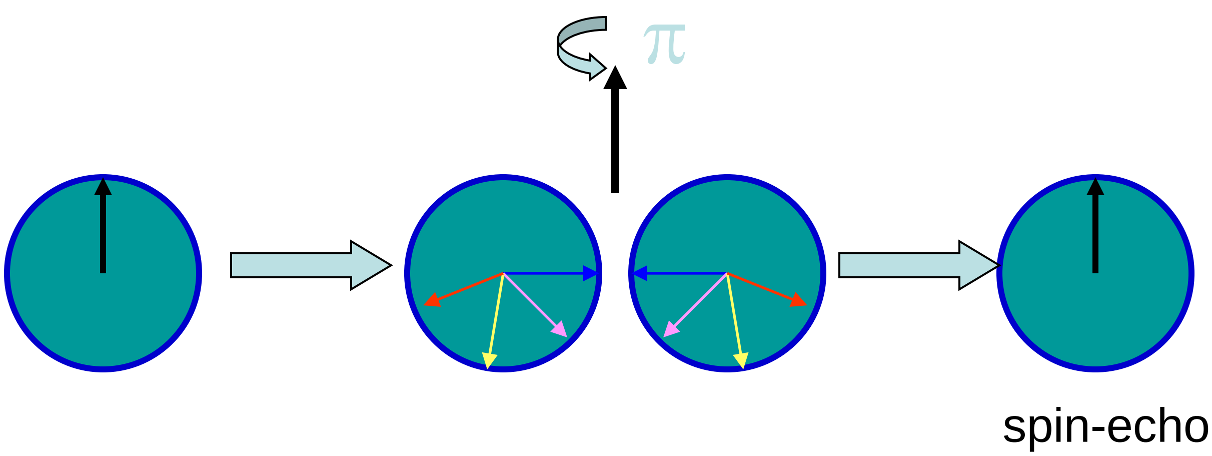
~3000 Hz at 1 Gauss

NSE spectrometer: elastic

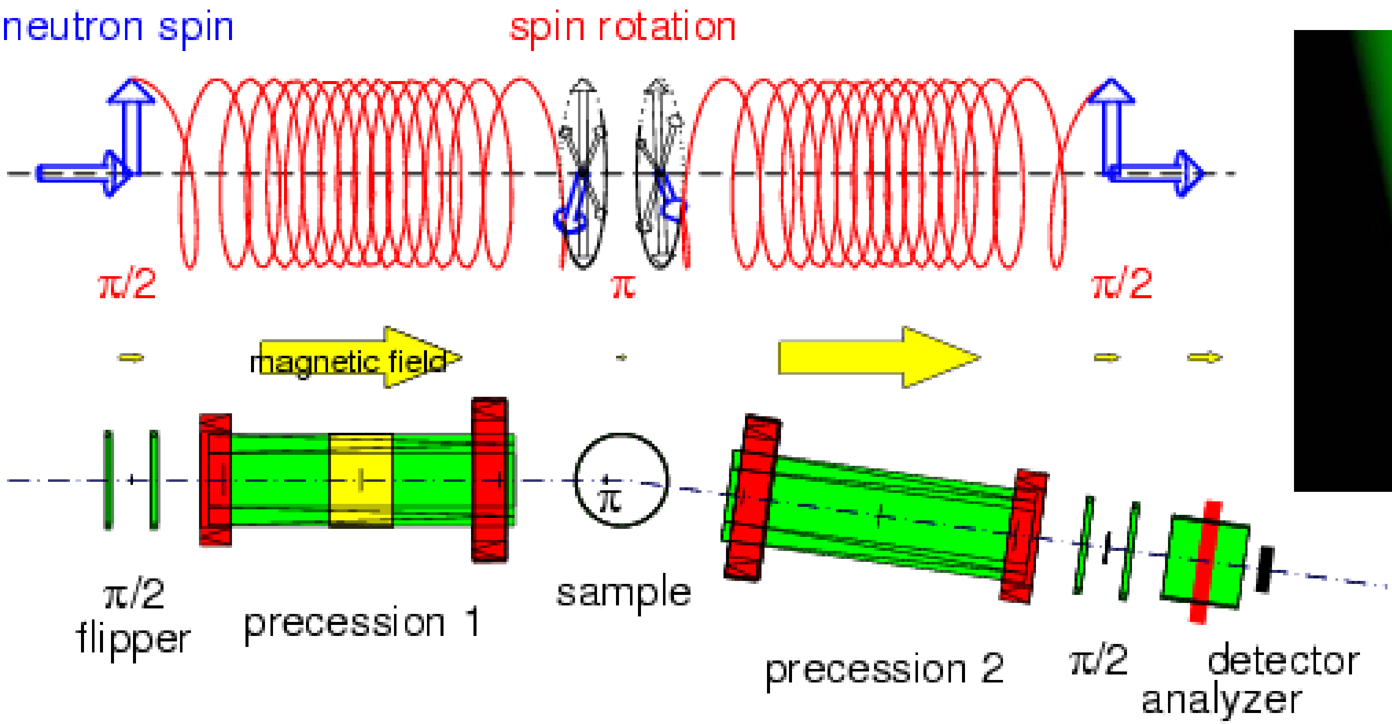
Neutron spin in magnetic field described by Bloch equation:

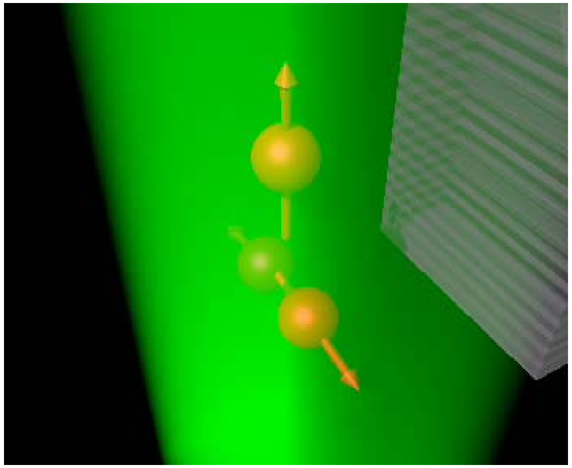
$$\frac{d\mathbf{P}}{dt} = \frac{g_n \mu_N}{\hbar}(\mathbf{P} \times \mathbf{B})$$

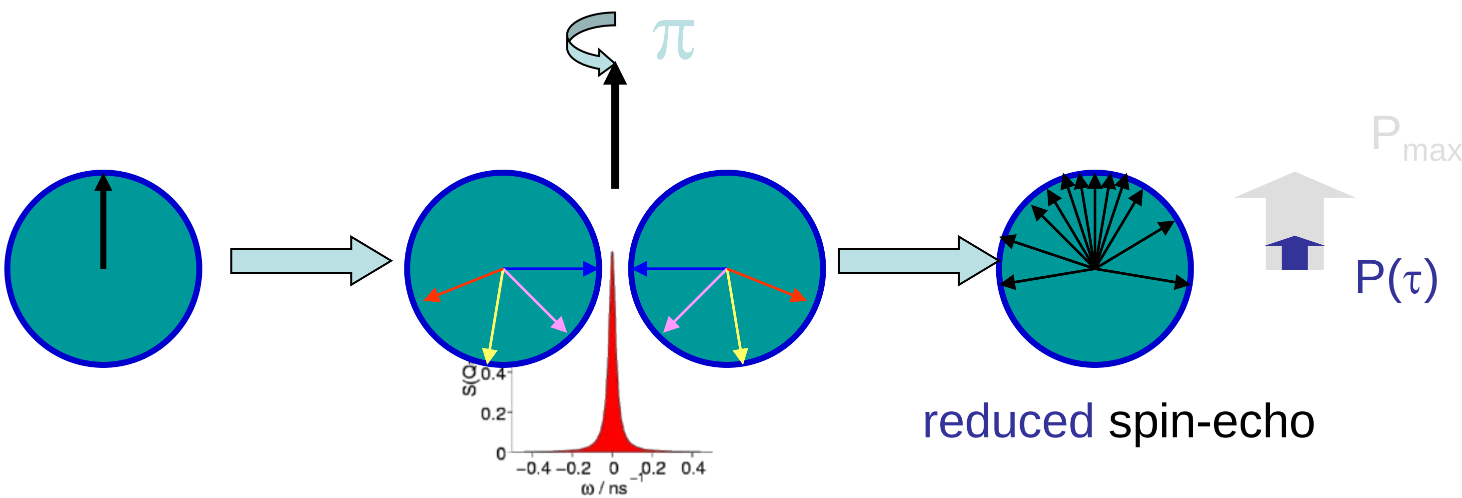




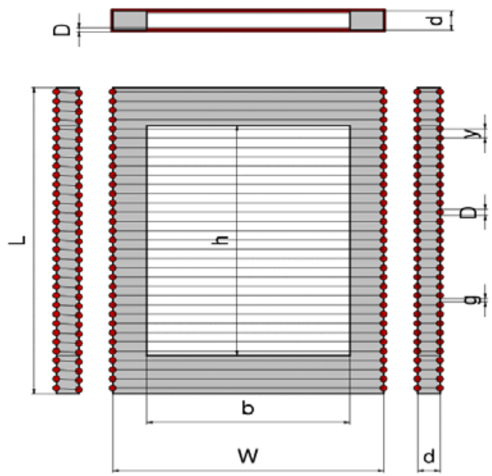
NSE spectrometer: (quasielastic scattering)



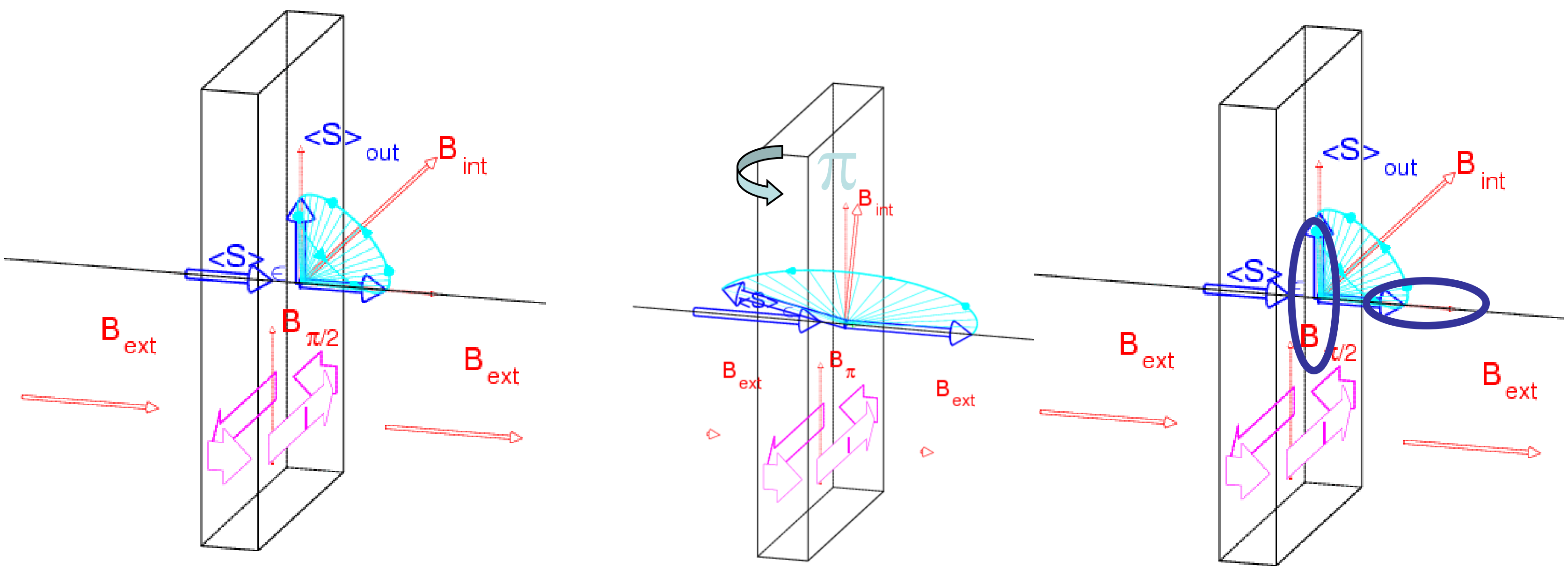




NSE spectrometer: technical aspects

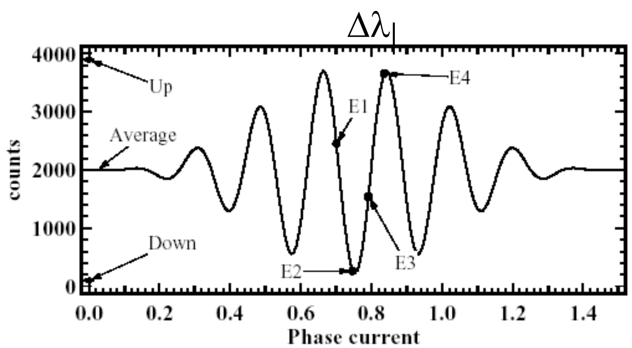


WNDING: material	Al, no insulation
cross section	circular 1 mm diameter
type	Mezei
length	33 m
weight	0.07 kg
resistance at 20°C [50°C]	1.03 □ [1.17 □]



TOF operation, echo evaluation and „ramped" flippers

Four points are sufficient



Envelope= FT(Φ(λ))

Amplitude =>S(Q,τ)

$$\Delta\varphi = 0.5817\,\lambda\, N\,\Delta I$$

1) Spin up and spin down => echo amplitude
2) Periodicity in terms of phase current

3) $\pi/2$ steps around the centre: E1 .. E4

$$E1 = Aver + E_{Ampl}\sin(\varphi)$$

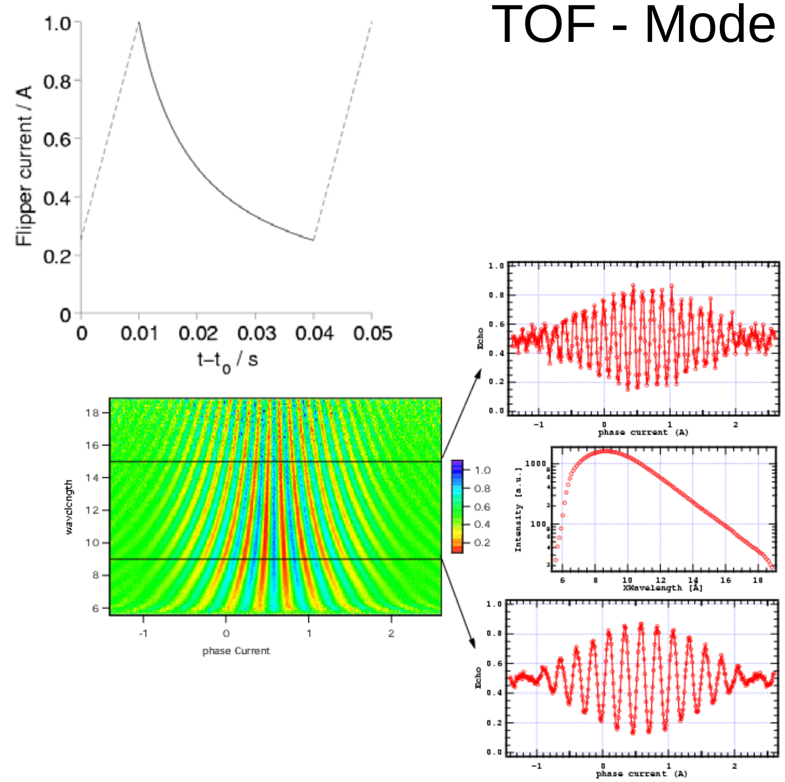
$$E2 = Aver - E_{Ampl}\cos(\varphi)$$

$$E3 = Aver - E_{Ampl}\sin(\varphi)$$

$$E4 = Aver + E_{Ampl}\cos(\varphi)$$

$$=> Aver, E_{Ampl}, \varphi => S(Q,\tau)$$

TOF - Mode



$$E1 = Aver + E_{Ampl}\cos(\varphi + n\varphi_{step}(\lambda))Envelop(n,\lambda)$$

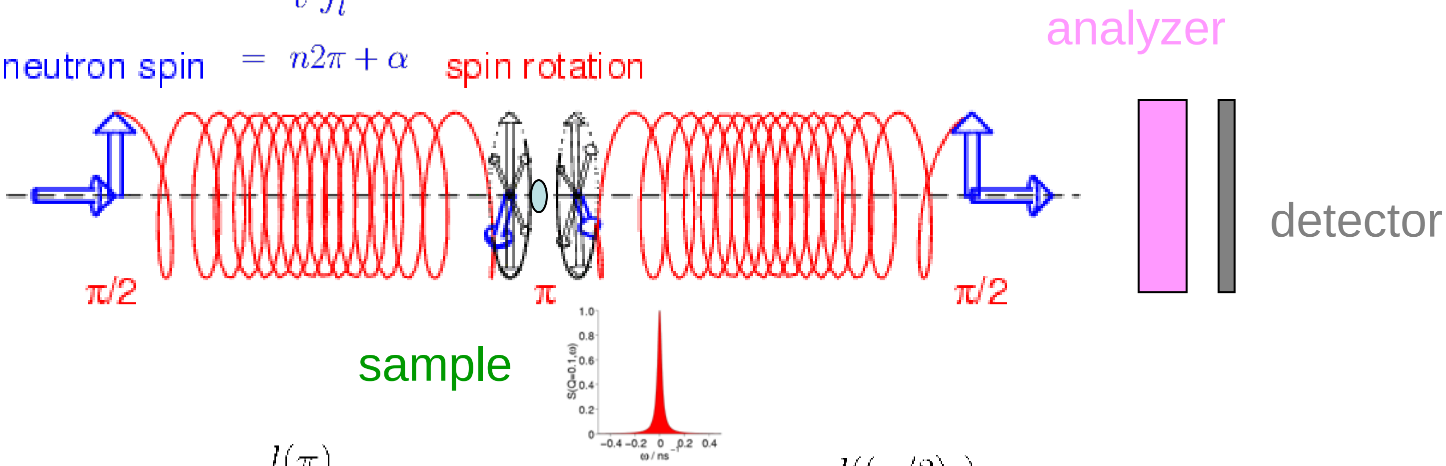
Measure echo group *Envelop(n,* λ*)* and the echo step size $\varphi_{step}(\lambda) \sim \Delta I_{step}\,\lambda$

Feasibility shown by B. Farago, A. Wischnewski and D. Richter

Phase calculation

$$\Psi = \frac{\gamma}{v}\int_l |B|dl$$

neutron spin $= n2\pi + \alpha$ spin rotation



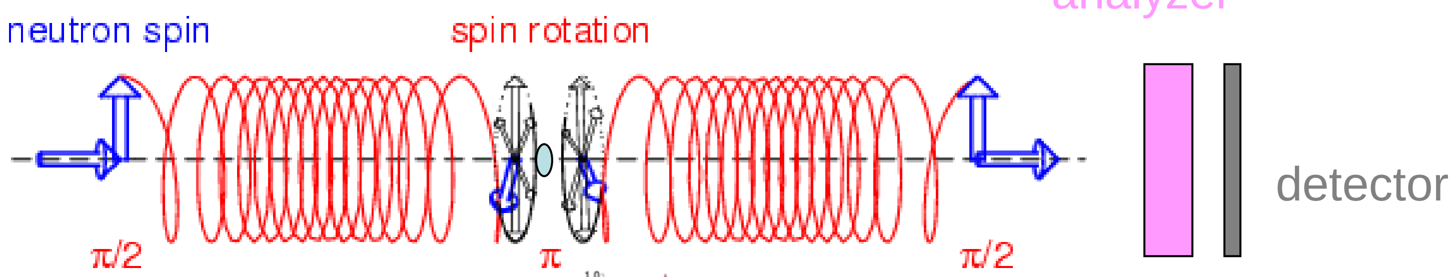
$$J_1 = \int_{l((\pi/2)_1)}^{l(\pi)} |B|dl \qquad J_2 = \int_{l(\pi)}^{l((\pi/2)_2)} |B|dl$$

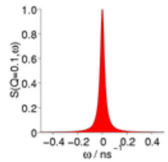
$$\Psi_{1,2} = -\frac{\gamma J_1}{v} + \frac{\gamma J_2}{v+\Delta v} + 4n\pi$$

$$T_a = \frac{1}{2}\left[1 + \cos(\underbrace{-\frac{\gamma J_1}{v} + \frac{\gamma J_2}{v+\Delta v}}_{\Delta\Psi})\right]$$

$$I = \eta S(Q) \iint \frac{1}{2}\left[1 \pm \cos(-\frac{\gamma J_1}{v} + \frac{\gamma J_2}{v+\Delta v})\right] w_\omega(\Delta v) w_\lambda(v)\, d\Delta v\, dv$$

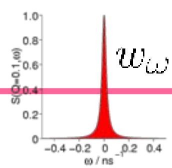
NSE what do we measure ?





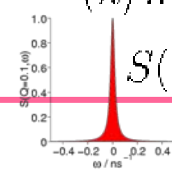
$$I = \eta S(Q)$$

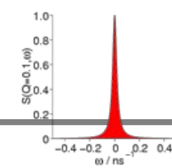
$$\iint \frac{1}{2}\left[1 \pm \cos\left(-\frac{\gamma J_1}{v} + \frac{\gamma J_2}{v+\Delta v}\right)\right] w_\omega(\Delta v) w_\lambda(v)\, d\Delta v\, dv$$



$$I \top \eta/2$$

$$\iint \left[1 \pm \cos\left(-\frac{\gamma J_1}{(h/m_n)\lambda^{-1}} + \frac{\gamma J_2}{(h/m_n)\lambda^{-1} + \lambda\omega/2\pi}\right)\right] S(Q,\omega) w_\lambda(\lambda)\, d\omega\, d\lambda$$





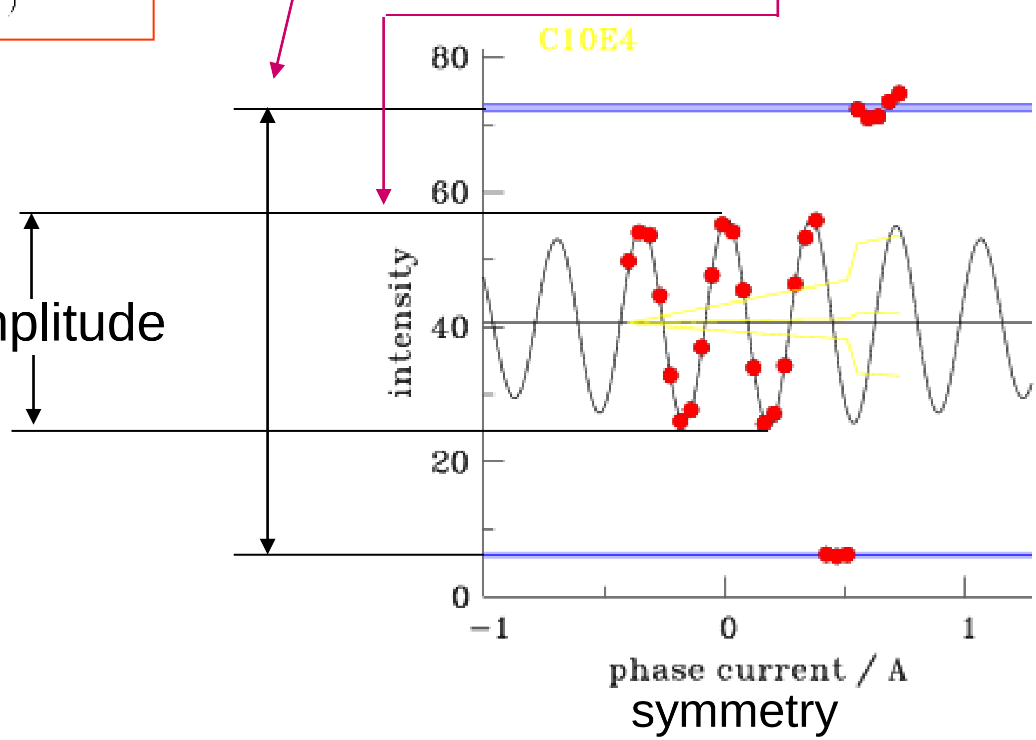
$$I = \eta \frac{1}{2}\left[S(Q) + \underbrace{\int \cos(\underbrace{\gamma J \frac{m_n^2}{h^2 2\pi}\lambda^3}_{t}\ \omega) S(Q,\omega) d\omega}_{S(Q,t)}\right]$$

The NSE signal, where is the information?

$$I = \eta\frac{1}{2}\Bigg[S(Q) + \underbrace{\int \cos(\underbrace{\gamma J \frac{m_n^2}{h^2 2\pi}\lambda^3}_{t}\ \omega) S(Q,\omega)\, d\omega}_{S(Q,t)}\Bigg]$$

$t \sim \lambda^3$!

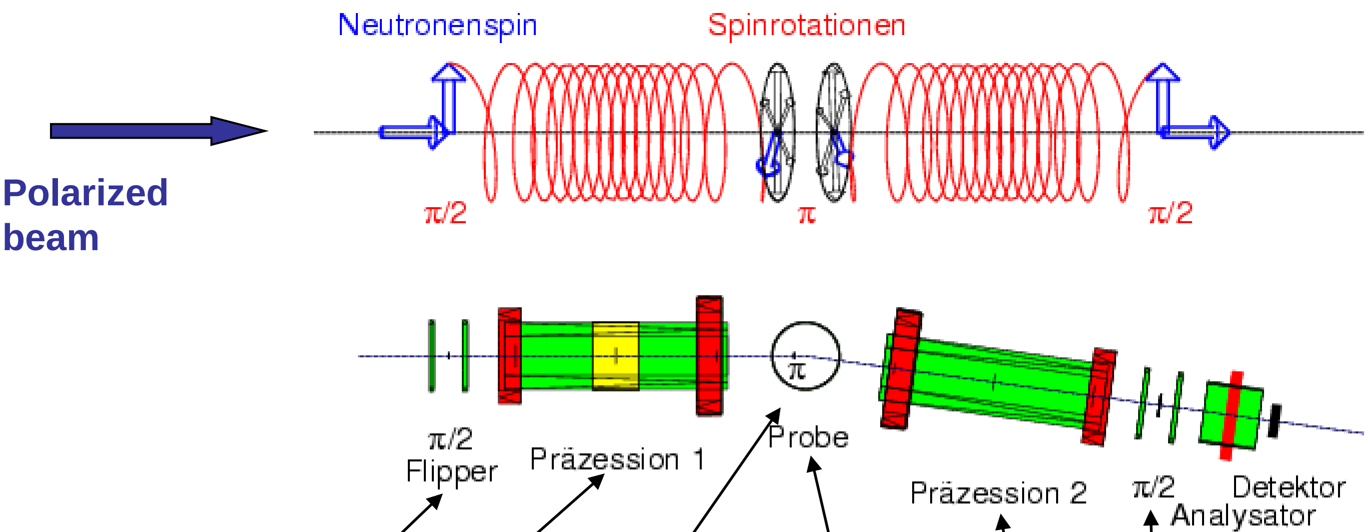
$$J_1 = \int_{l((\pi/2)_1)}^{l(\pi)} |B|\, dl$$



$$\frac{S(Q,t)}{S(Q)}$$

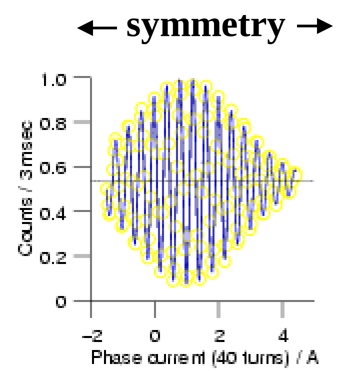
finally

Principle of NSE : Summary

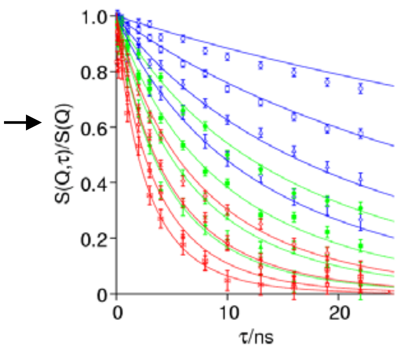


Start---run---timereversal---sample---run---Stop-readout

States of the indivdual spin-clocks



Echoform



$$\tau \sim \lambda^3 BL$$

Neutron spin Echo at the SNS

Note that for the SNS - NSE:

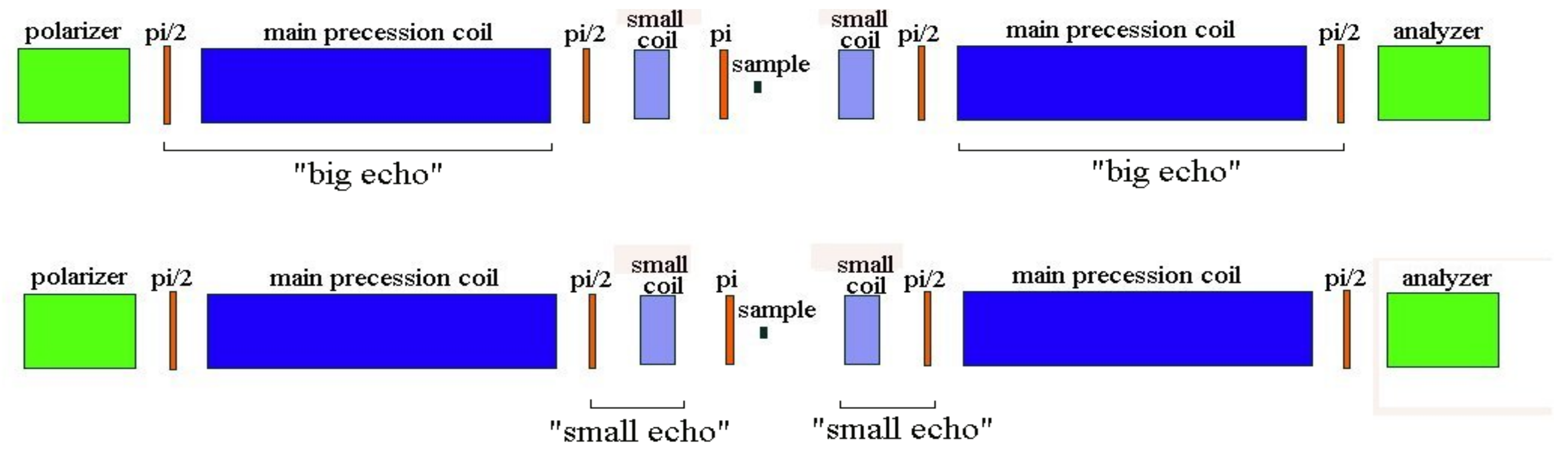
J ≅ 1T acting along 1.2m

≅ 150000 Precessions

for 200m/s (λ=2.0nm).

Accuracy: 0.1*2π

Neutron spin Echo how to get the dynamic range



Homogeneity of the field integral 10^{-3} without Fresnel coils & 10^{-6} with Fresnel coils required

- Ratio between τ_{min} / τ_{max} maybe of about the 500 for the „big echo mode“

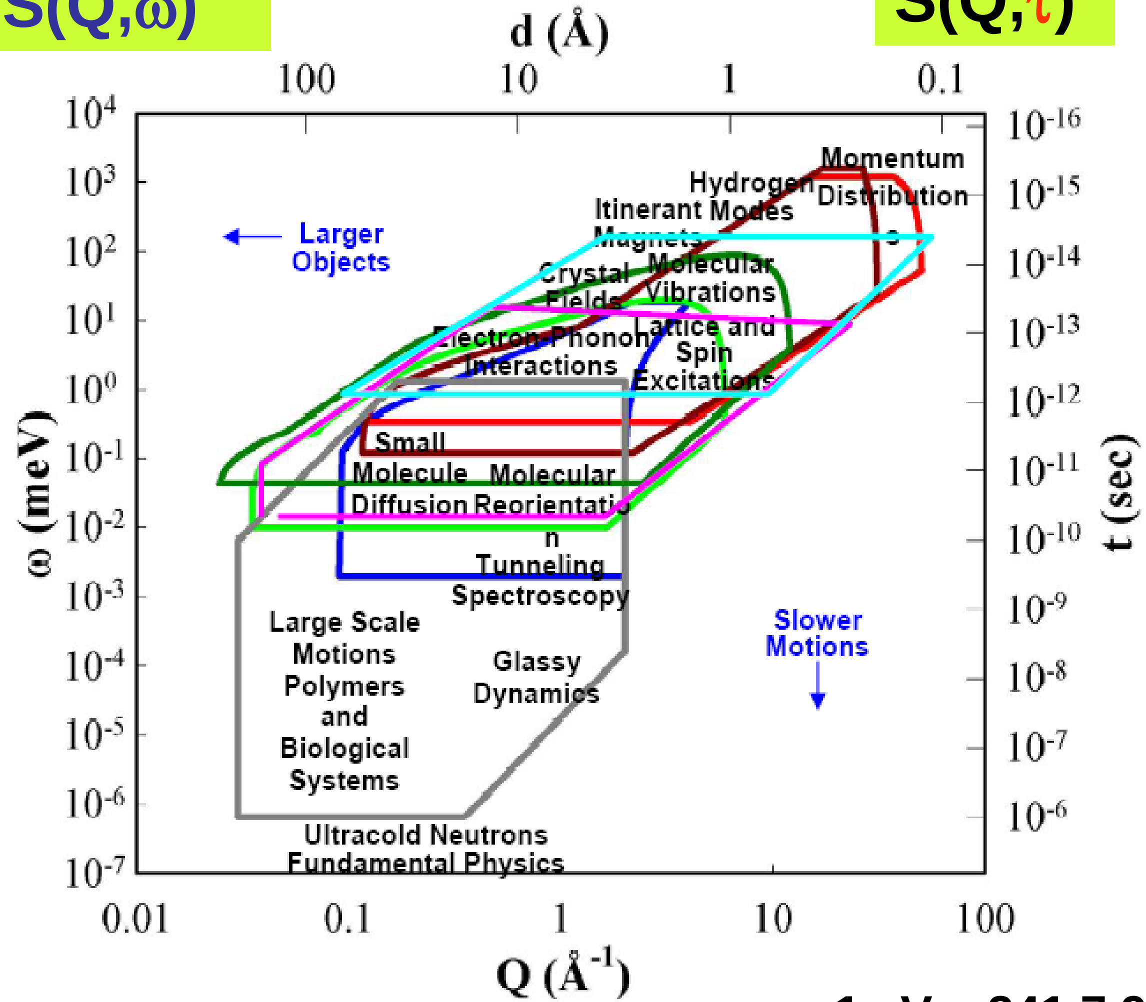
- Ratio between τ_{min} / τ_{max} maybe of about the 10 for the „small echo mode“

-**Wavelength band from $0.3 < \lambda/nm < 2.0$ => 300**

e.g. NSE @ SNS domain … 1ps to 1μs

S(Q,ω)

S(Q,τ)



- ARCS Fermi Chopper
- SEQUOIA Fermi Chopper
- HYSPEC
- Cold Neutron Chopper Spectrometer
- Backscattering
- Neutron Spin Echo
- Cold triple Axis
- Thermal Triple Axis

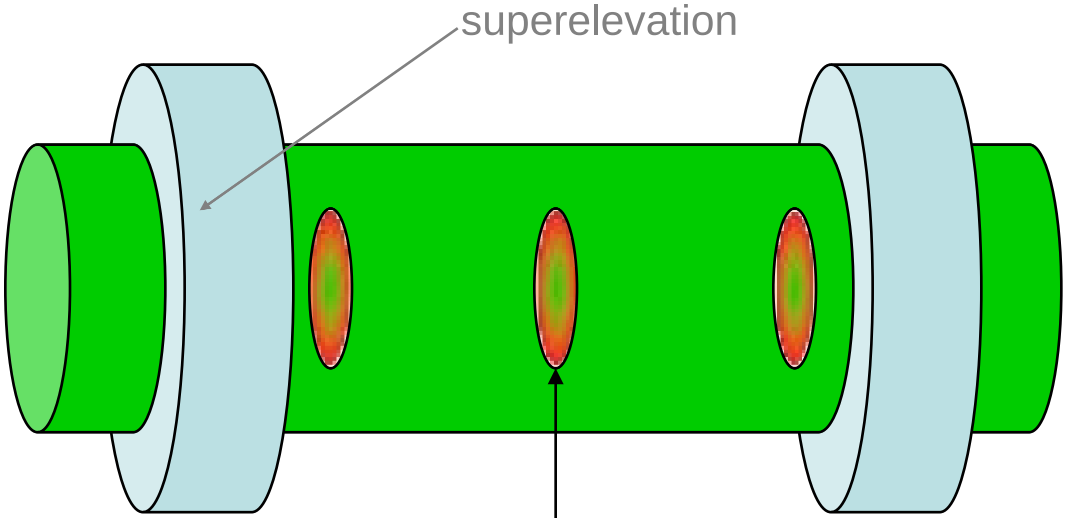
adapted from "Neutron Scattering Instrumentation for a High-Powered Spallation Source" R. Hjelm, et al.,

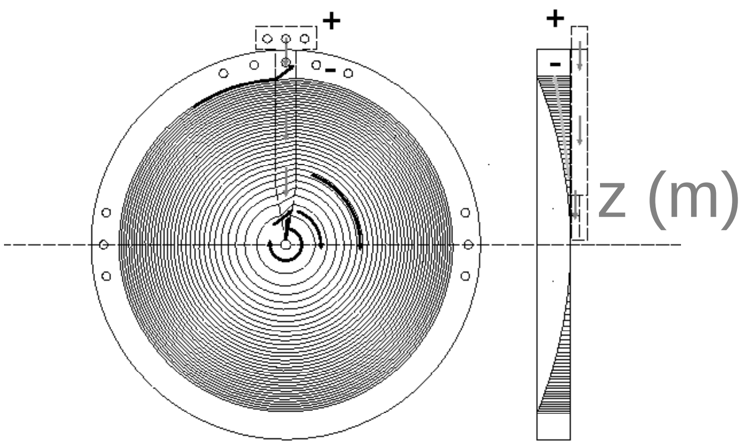
1 eV = 241.7 988 36 (72) THz

Corrections of Field Integral

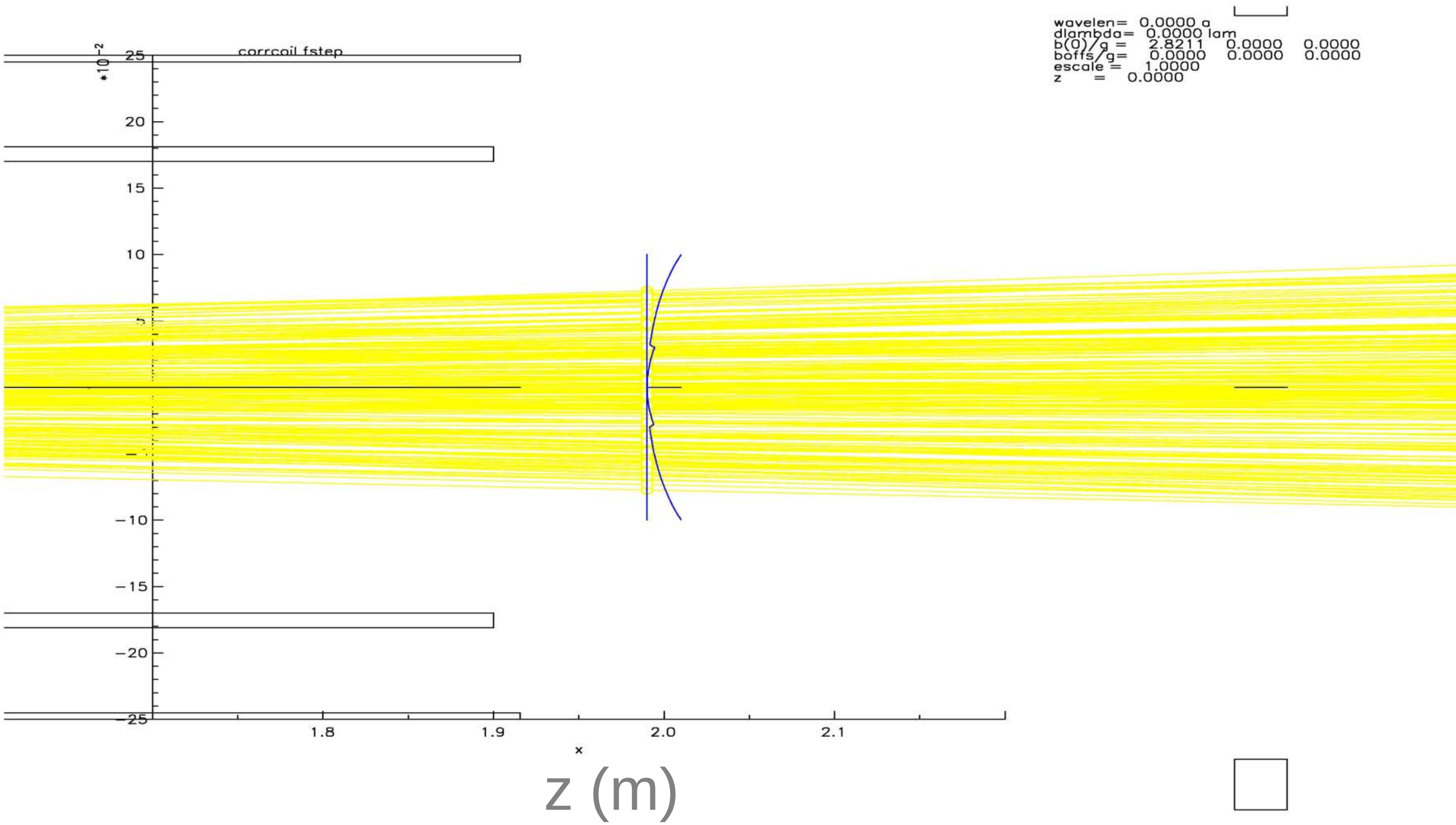
Bl – or rather $\int |\mathbf{B}|\, \mathrm{d}l$ – is not constant for all neutron paths!

⇒ correction coils:





Corrections of Field Integral



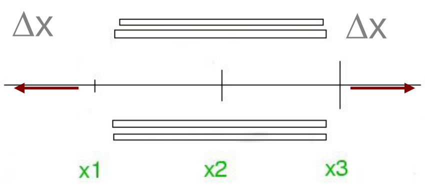
Useful criterion: Average relative spread (ARS)

$$\delta = \sqrt{\sum_i \left(J_i - J_0\right)^2} / J_0 < 10^{-6}$$

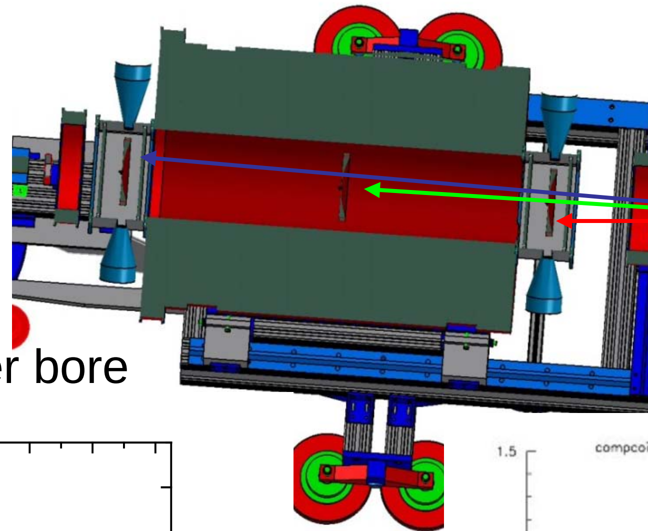
Simulation $\delta = 8*10^{-7}$ **OK!**

Corrections of Field Integral

CC 2 should be negligible !

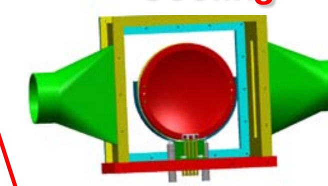


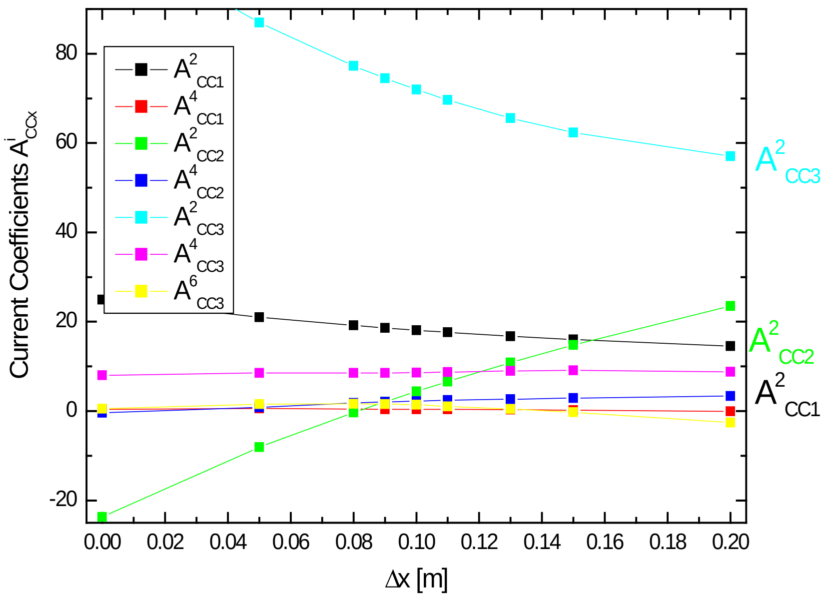
shift cc1 and cc3 outside inner bore

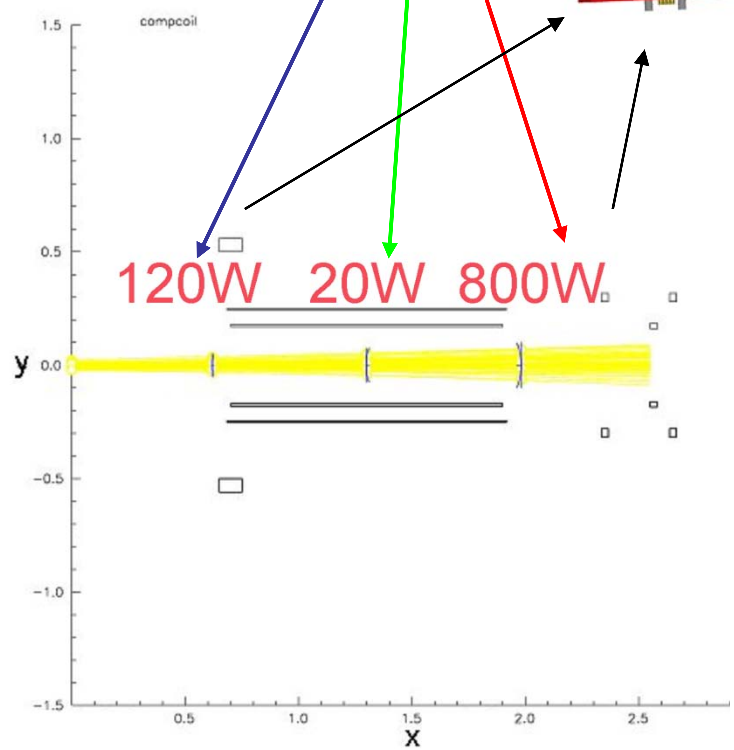


Fresnel coils

Cooling







$$\mu_0 j_{CCx}(r) = 2A^2_{CCx} r^1 + 4A^4_{CCx} r^3 + 6A^6_{CCx} r^5$$

Requirements to magnetic environment

Phasenstability better 10^0 !

Means with λ=15Å (300m/s)

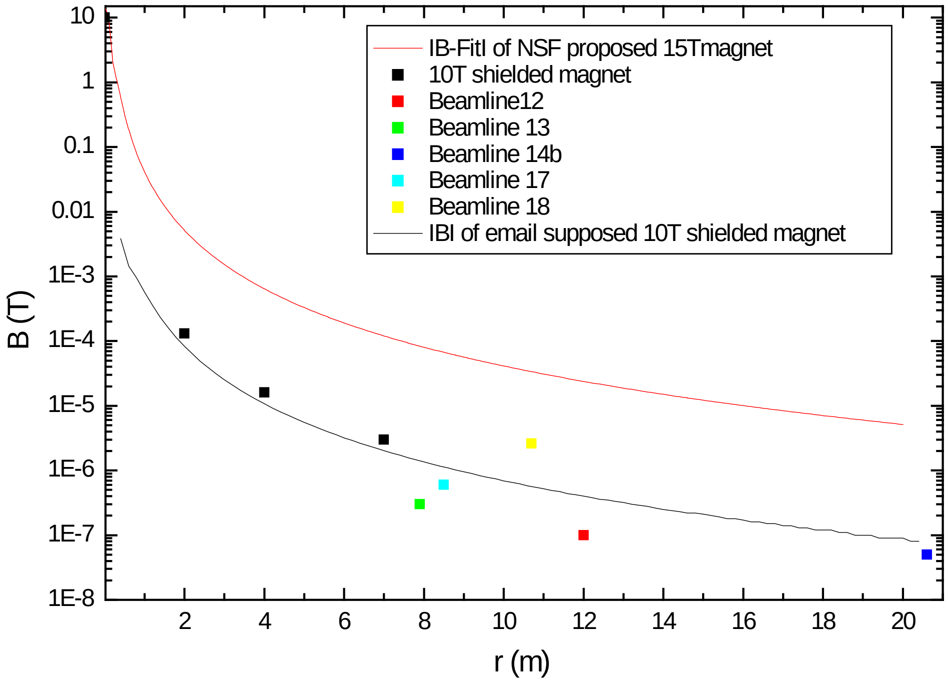
$\Delta\phi = 360^0 \times$ 3000 Hz/Gauss $\times$ 3m / 300m/s = 10800^0 / Gauss

→ ΔB < 10/10800 G ~ 1 mG !

Magnetic stray field sources

Parameterization of SNS delivered magnets !!! More than 1mG ?

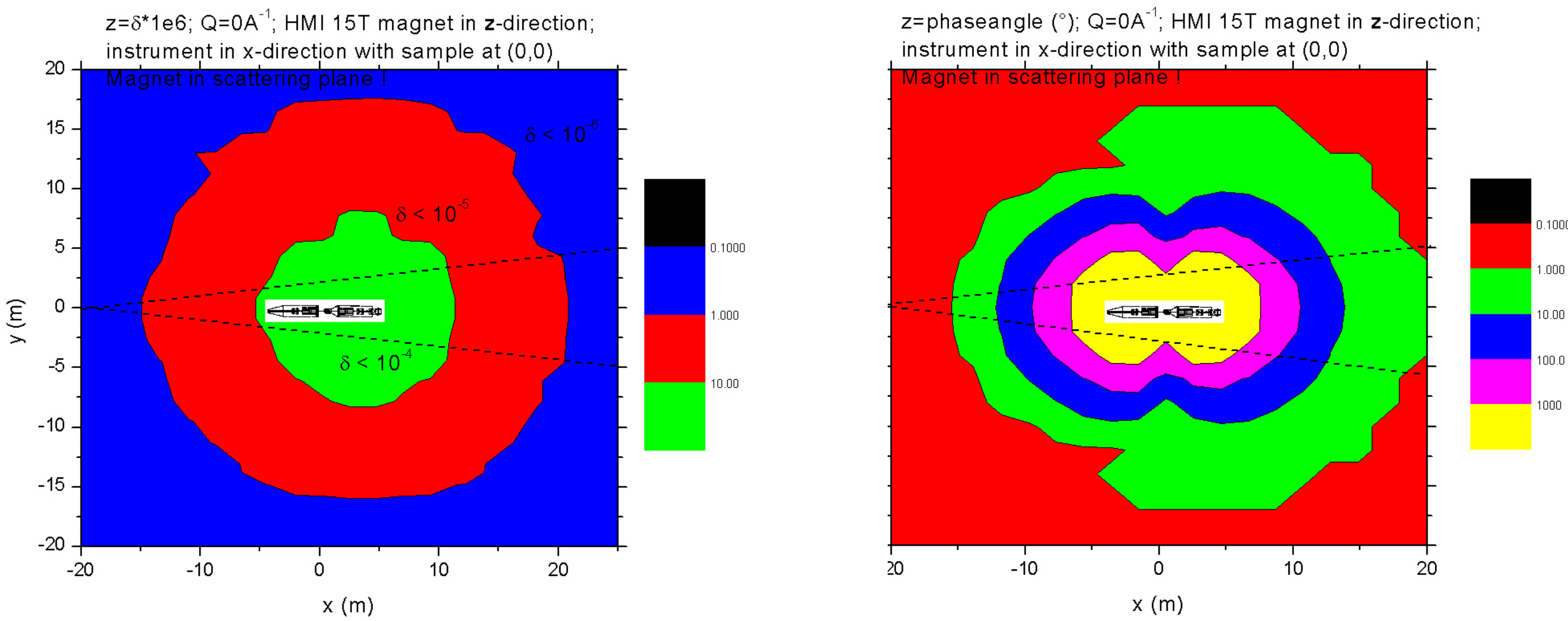
15T Magnet winding parameters and self-shielded 10T magnet fitted at several beamlines near the sample stage of the NSE!



Parameter of the HMI 15T Magnet:red curve
Main coil:
0 0 -0.173 /x y z
0 0 0.173 /x y z
0.03776 0.248 /ri ra
0.5283e7 /nI
Shielding coil:
0 0 -0.205 /x y z
0 0 0.205 /x y z
0.331 0.363 /ri ra
-0.3018e5 /nI

Parameter of the 10T Magnet:black curve
main coil
0 0 -0.12
0 0 0.12
0.025 0.09
0.243e7
shielding coil 1
0 0 -0.15
0 0 -0.10
0.12 0.16
-0.2813e6
shielding coil 2
0 0 0.10
0 0 0.15
0.12 0.16
-0.2813e6

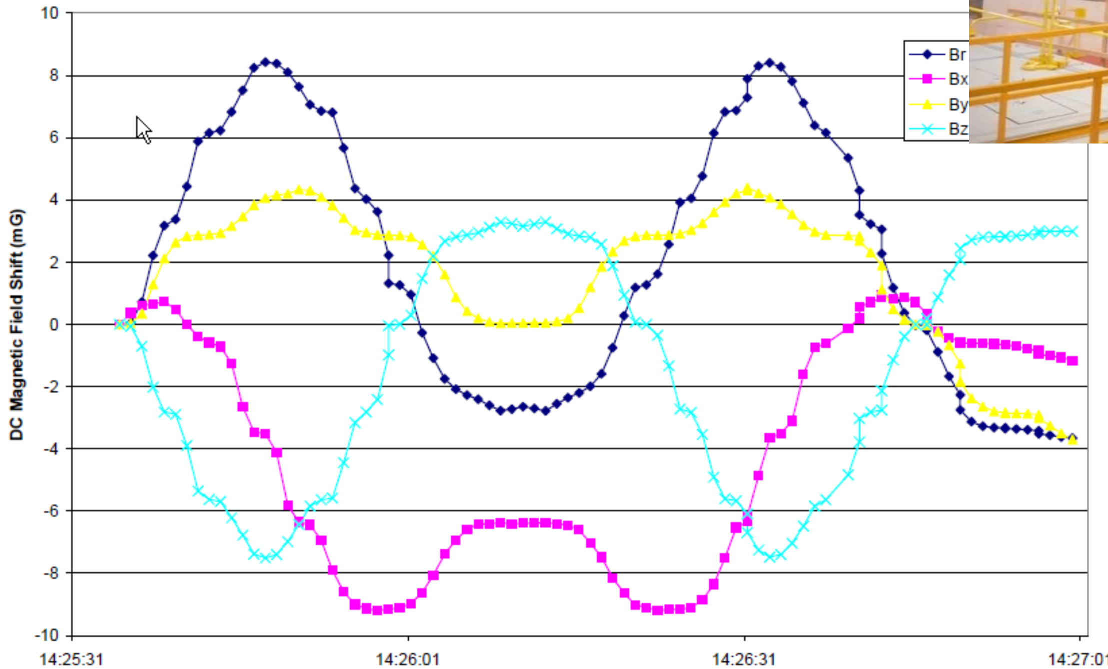
Sensitivity - two criteria: phase shift and homogeneity (15T magnet)



Already at distances < 19m the instrument will be disturbed !

Motivation to build a magnetic enclosure comprising the whole secondary spectrometer

High permeability magnetic field without shielding chamber

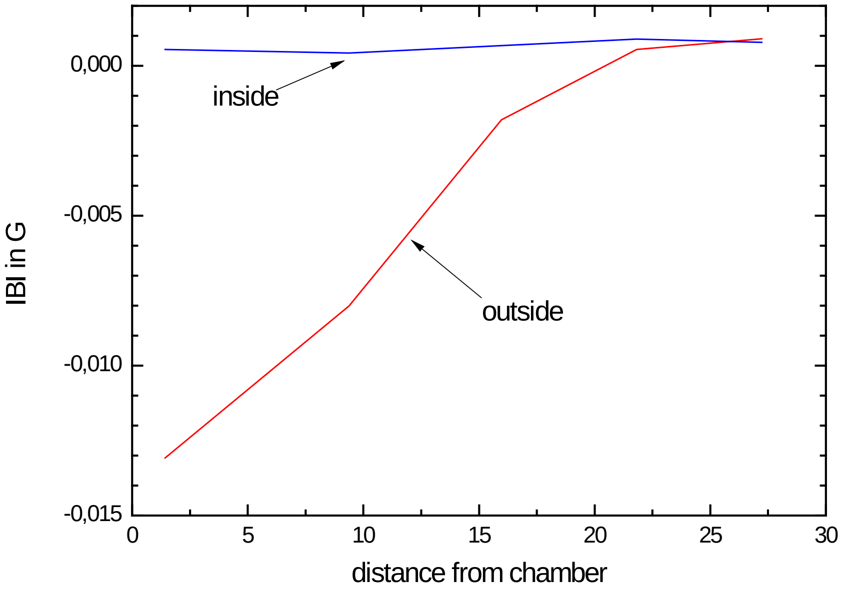


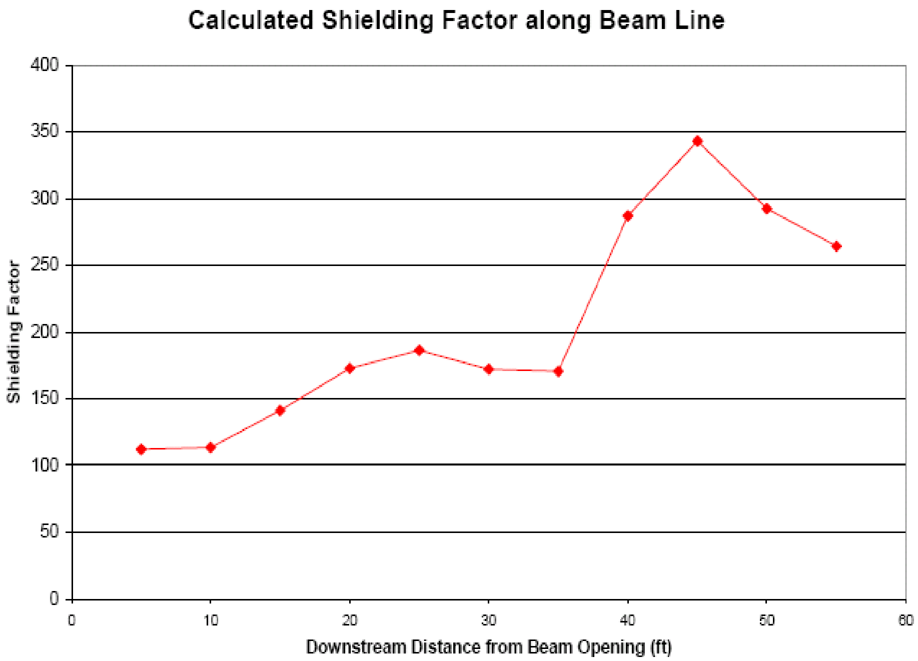
The magnetic shielding enclosure

Final acceptance test already took place in January 2008 !

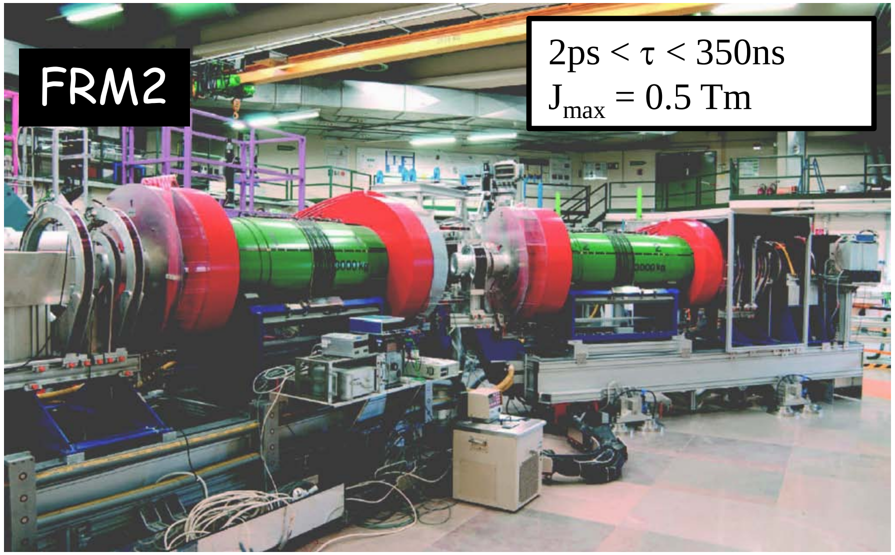
Shielding capabilities of the enclosure

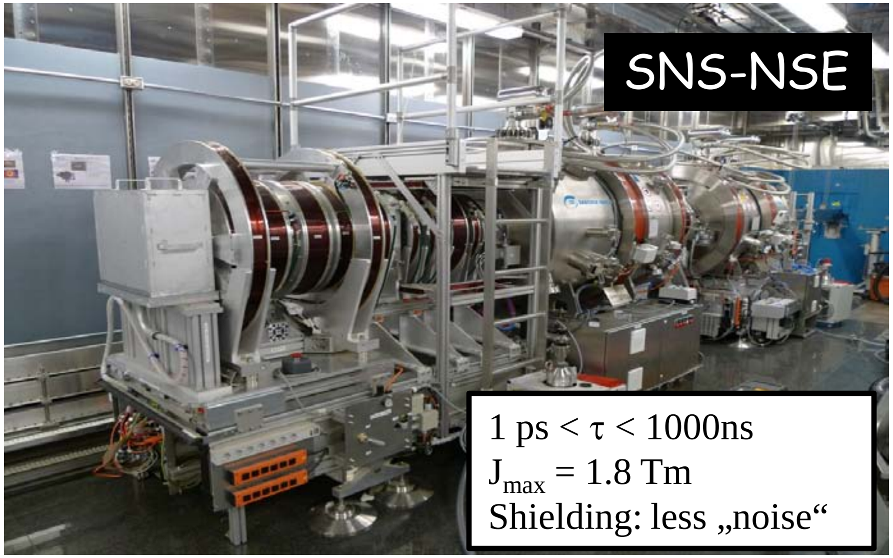
Crane moving over enclosure

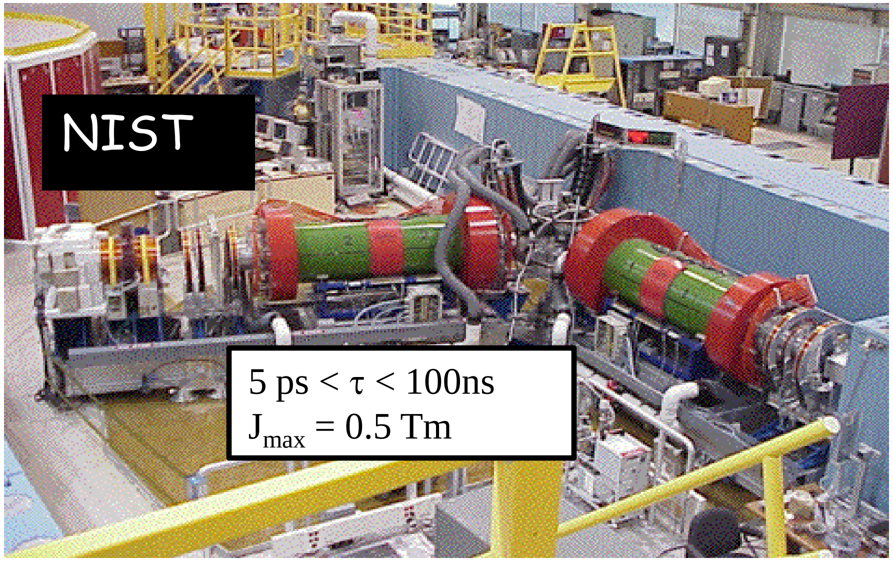


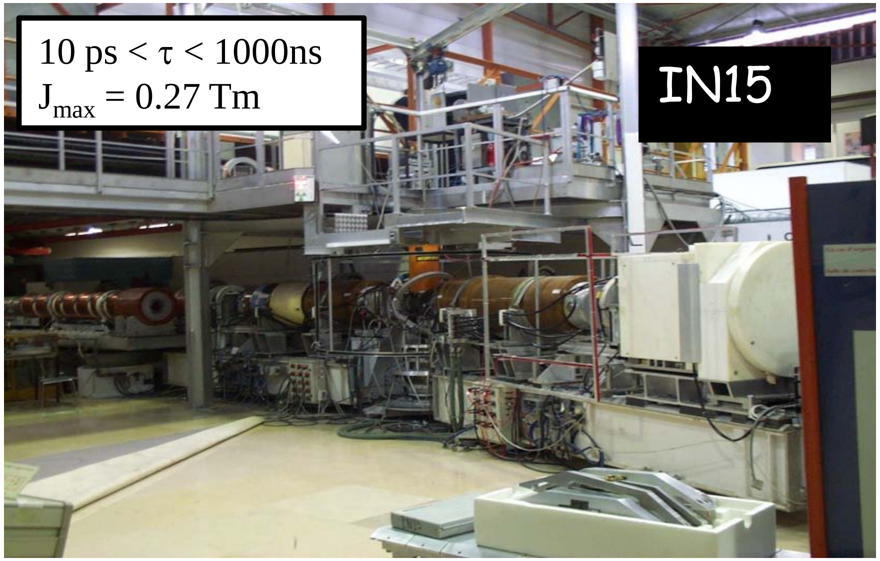


Comparison: The NSE @ SNS domain ... 1ps to 1μs

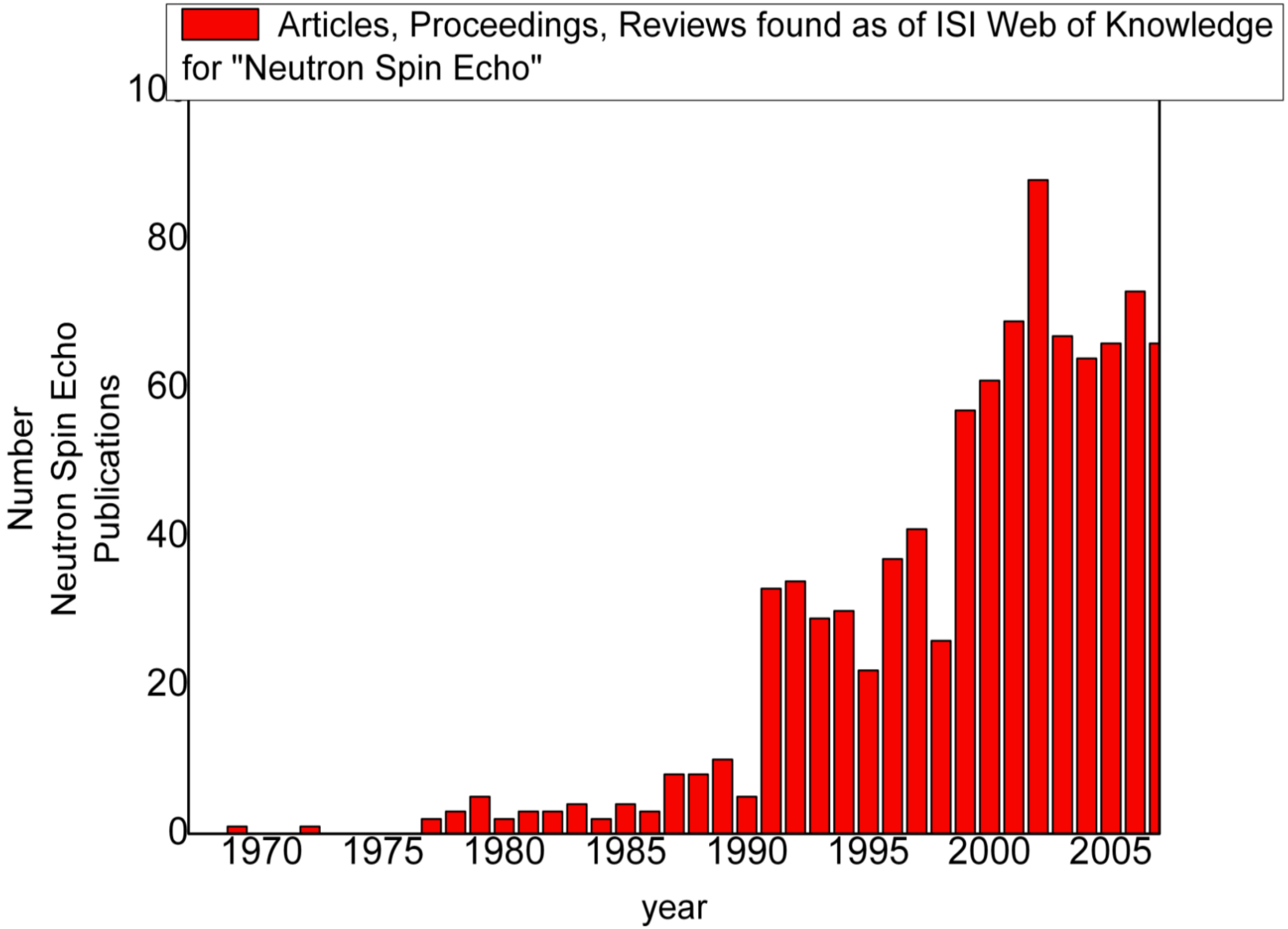






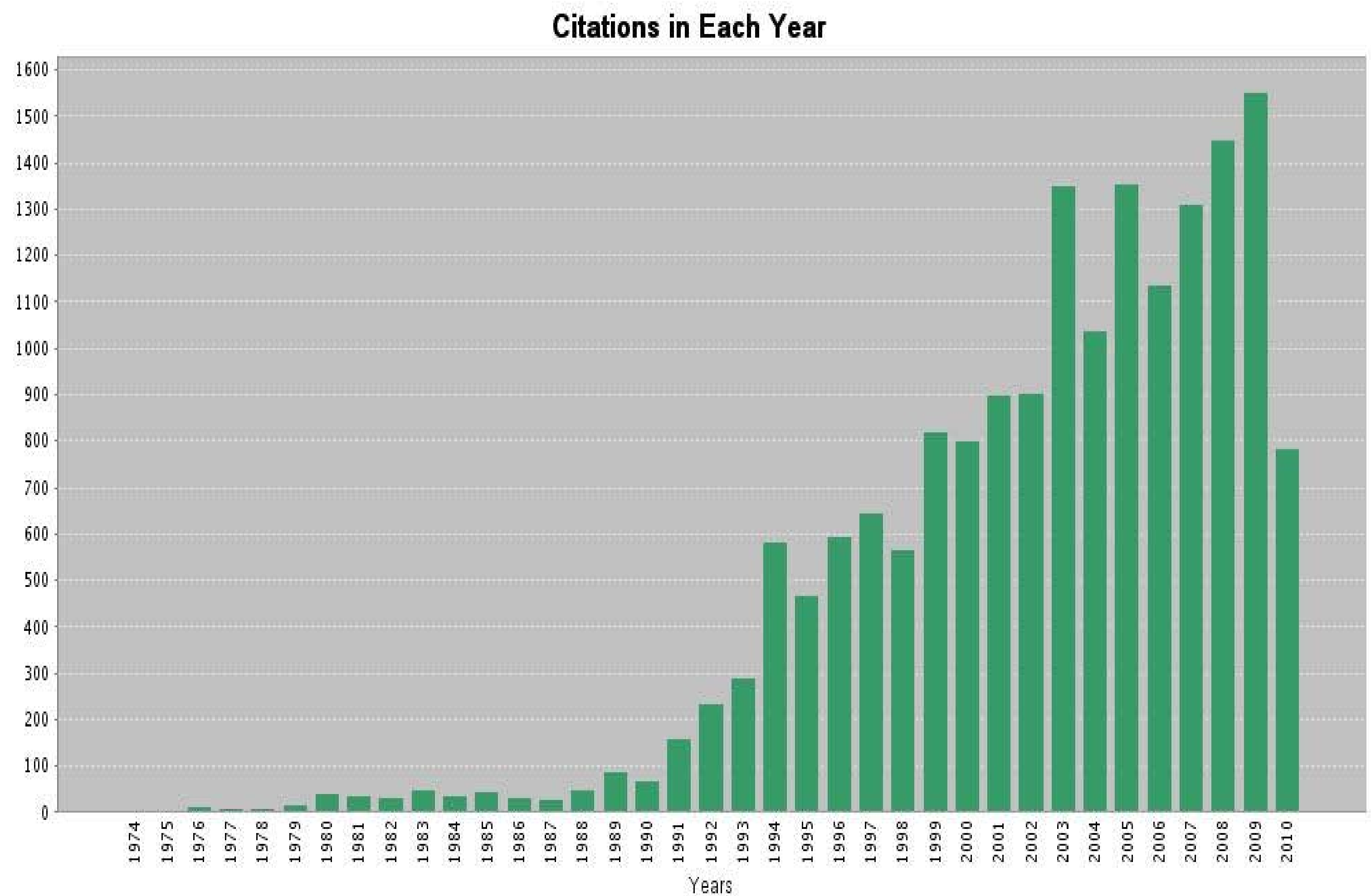


Motivation



Articles, Proceedings, Reviews found as of ISI Web of Knowledge for "Neutron Spin Echo"
Number
Neutron Spin Echo
Publications
0
20
40
60
80
1970
1975
1980
1985
1990
1995
2000
2005
year

Citations per year



What science is relevant for the NSE

Soft and Complex Condensed Matter

Polymers melts and molecular rheology
Microemulsions and worm-like micelles
Complex fluids
Rubbers and molecular networks
Gels and polyelectrolytes
Polymeric electrolytes

Biophysics
Protein dynamics
Membranes

Relaxations and the Glass Transition
Role of primary and secondary relaxations
Displacement patterns in space and temporal evolution

Science relevance: part 2

Nanostructured materials

Nanoparticles

Diffusion in suspension of nanoparticles

Ferro-fluids and magneto-fluids

Electrorheological fluids

Transport in porous media

Diffusion or migration in gels or granular media

science case: after some instrumental changes

Magnetism

Spin glasses

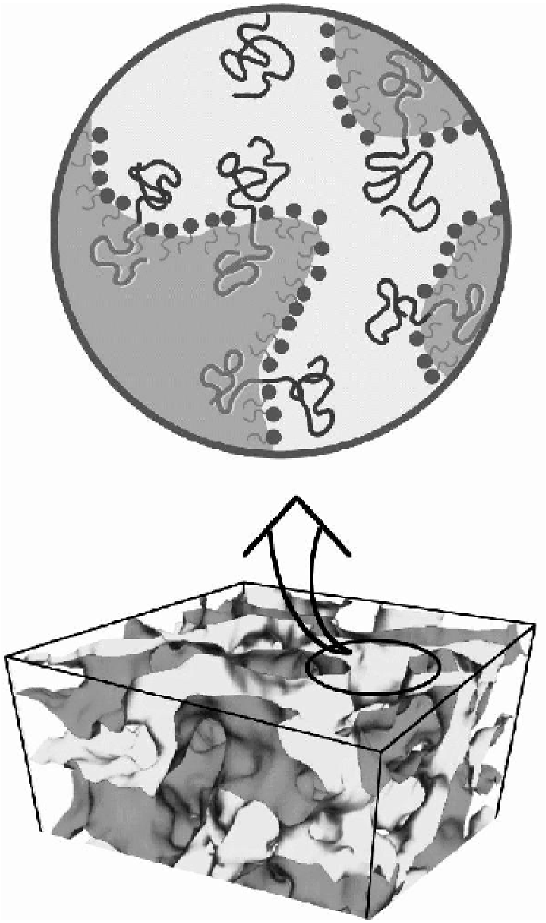
Superparamagnetic fluctuations in magnetic nanoparticles

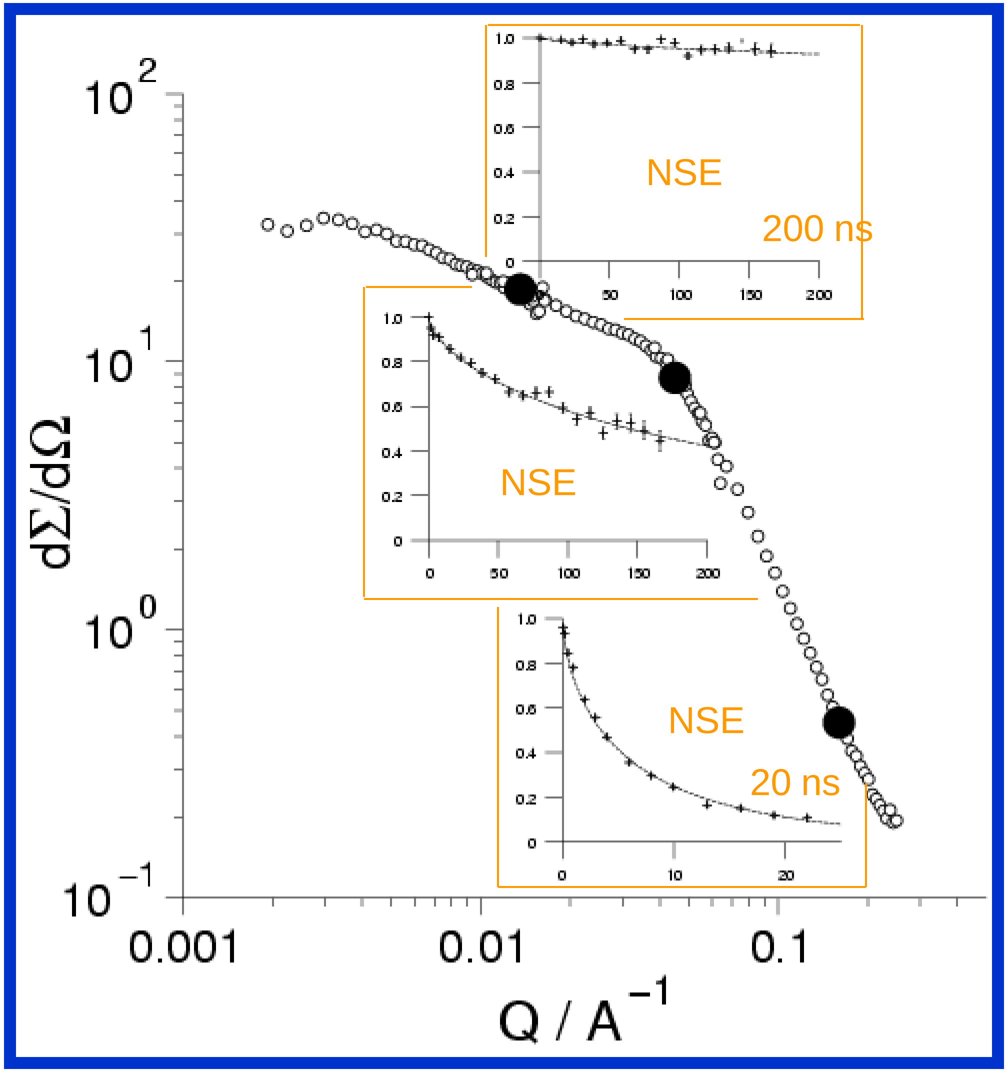
Low frequency excitations

Flux line motions in superconductors

NSE = SANS (t=0ns) + Dynamics

E.g.: Bicontinuous microemulsion
(water and oil)





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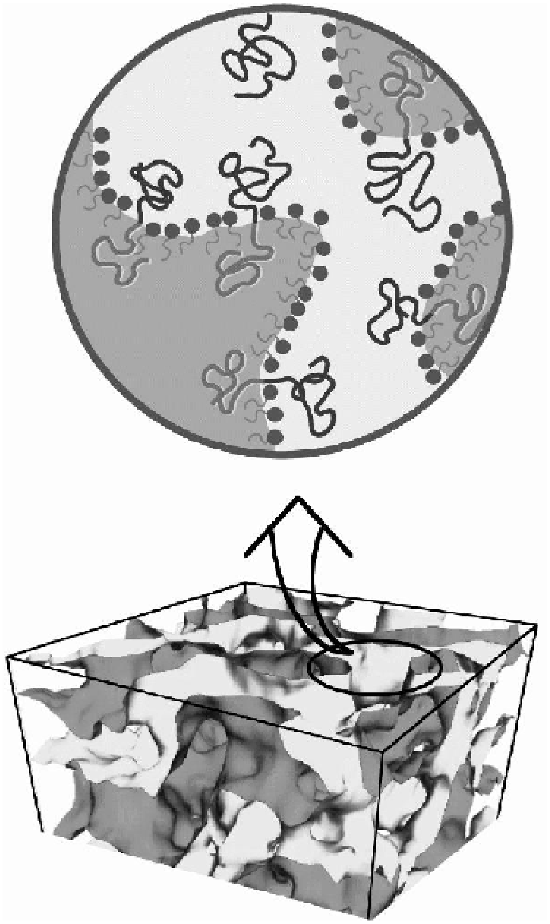
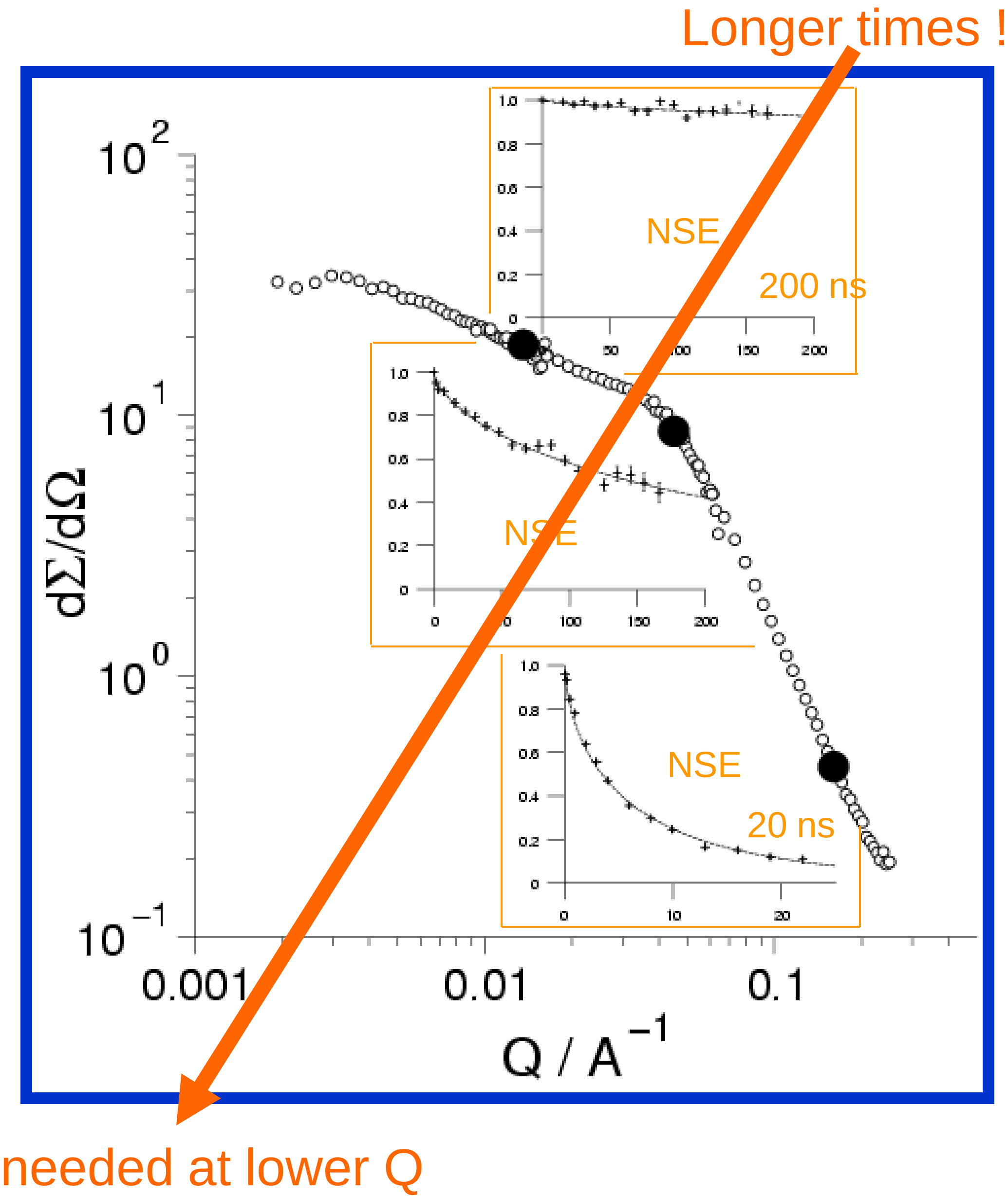
E.g.: Bicontinuous microemulsion



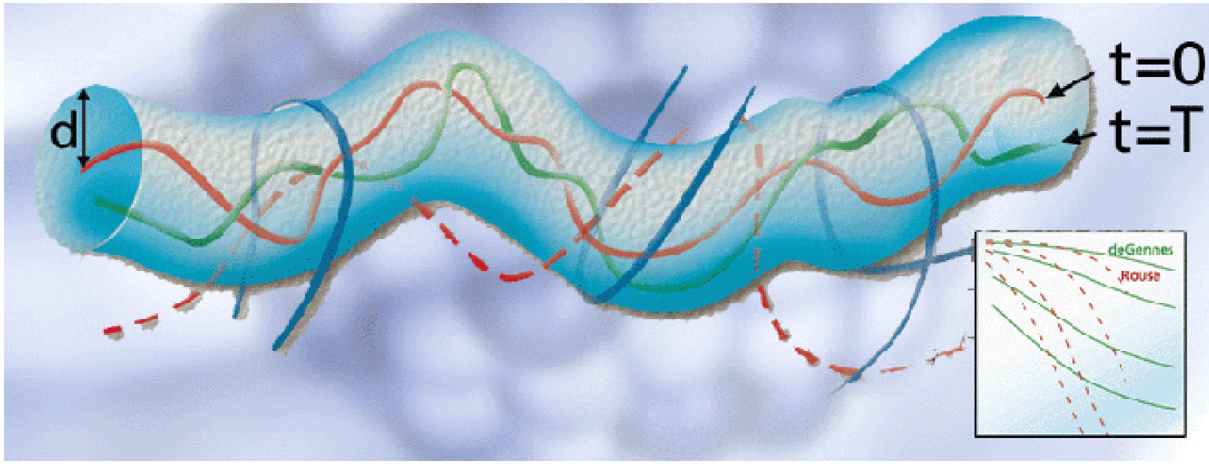


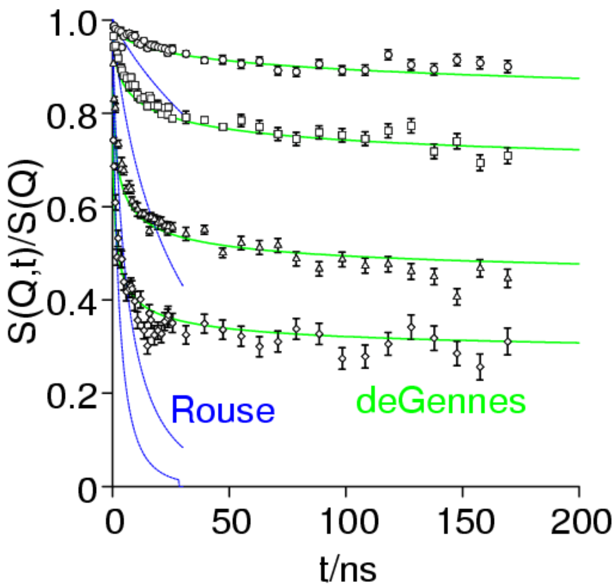
Reptation model (Edwards/deGennes)

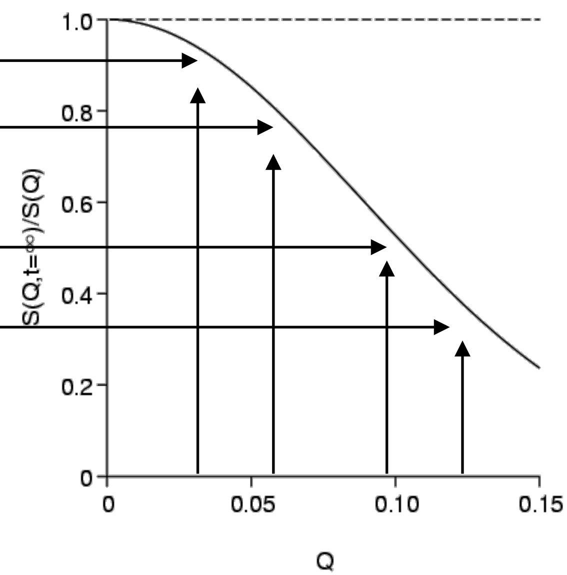
Confinement in a tube seen by NSE

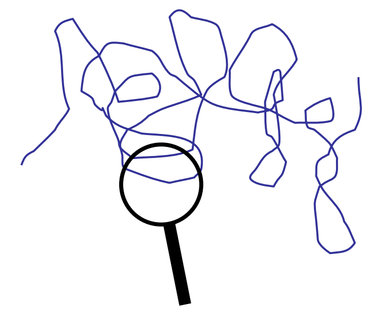
coherent scattering, labeled chain

melt of long chain linear polyethylene





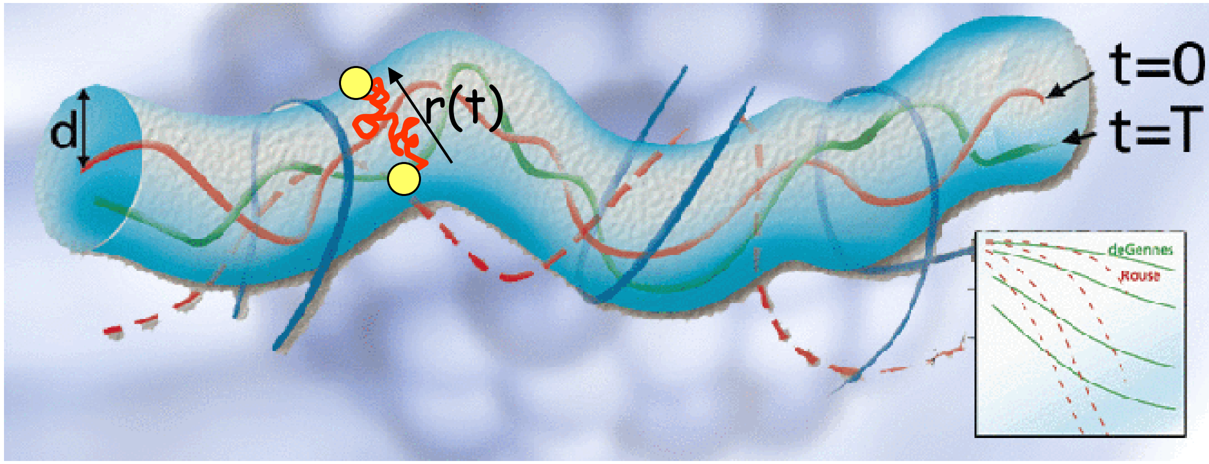


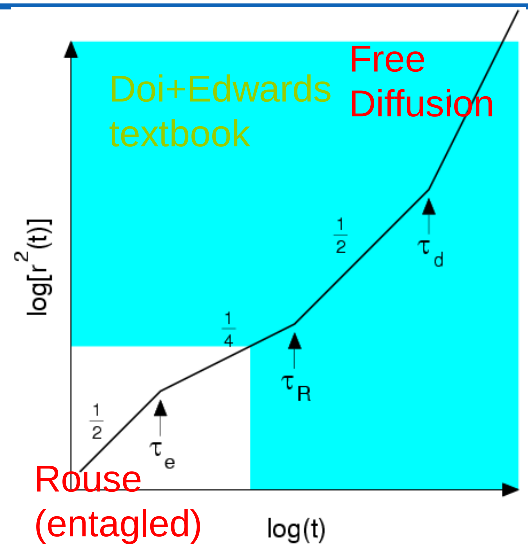


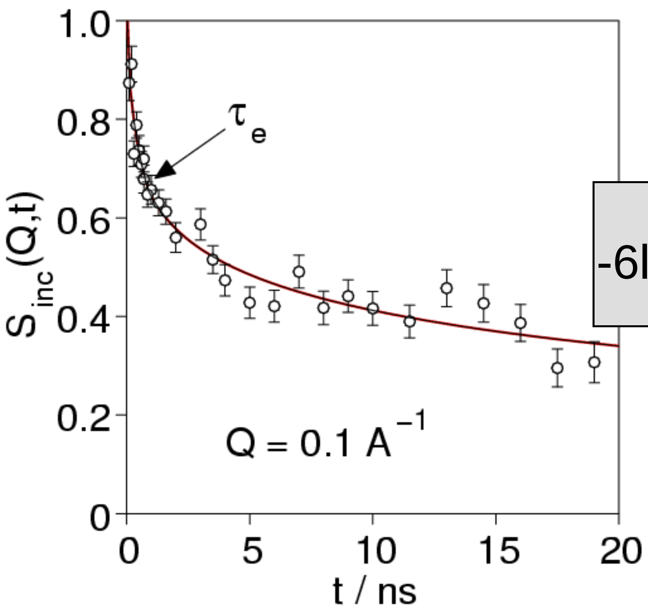
Rouse: mechanical rel. motivated, bead and spring model with friction

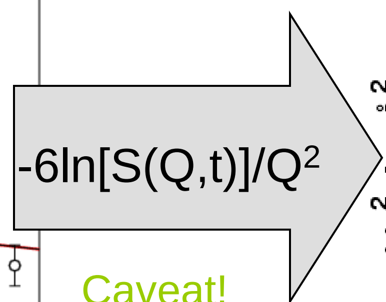
De Gennes: reptation

Segment displacement <r²(t)> from incoherent scattering

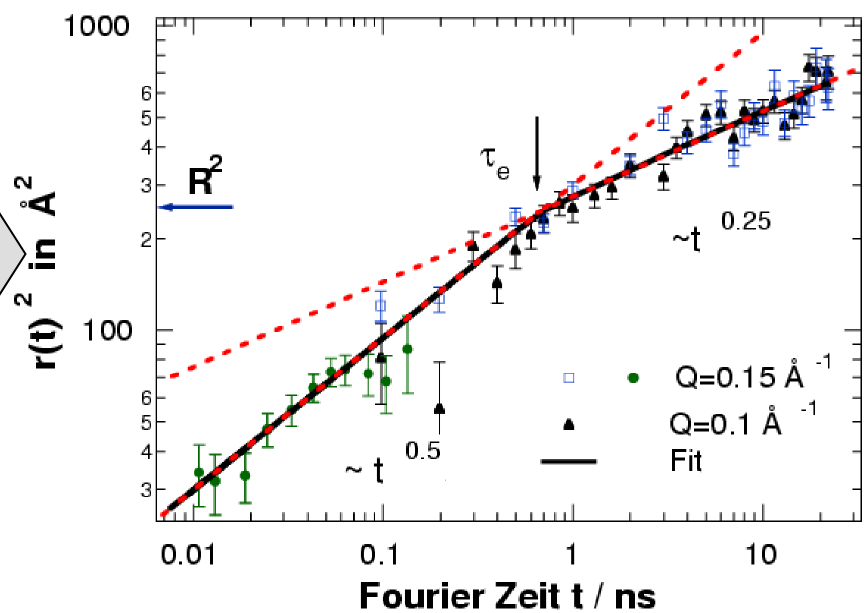








Caveat!



Neutrons & Soft Matter

Unique role:

- Suitability of length and time scales accessed, especially SANS and NSE
- Selectivity varying contrast: H ⟷ D → Decipher complex structures

..and...use dynamics to discriminate flexible from rigid structures ....

Molecular Rheology of topological Polymer Fluids

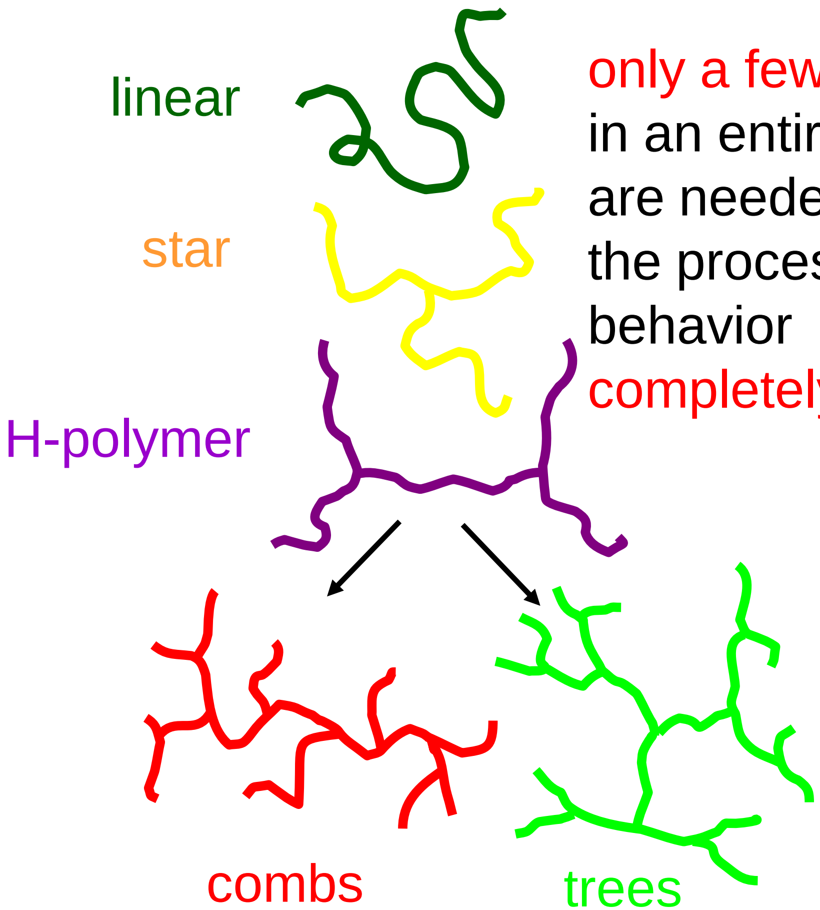
Industrial processing

↕

Macromolecular dynamics

↕

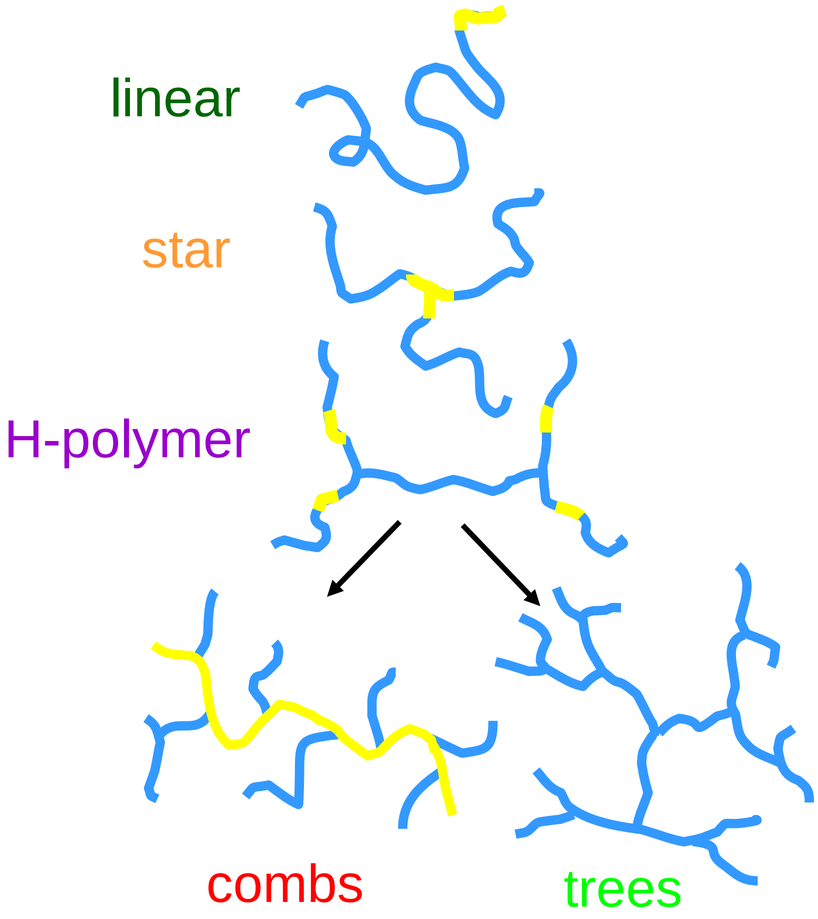
Topology of the molecules



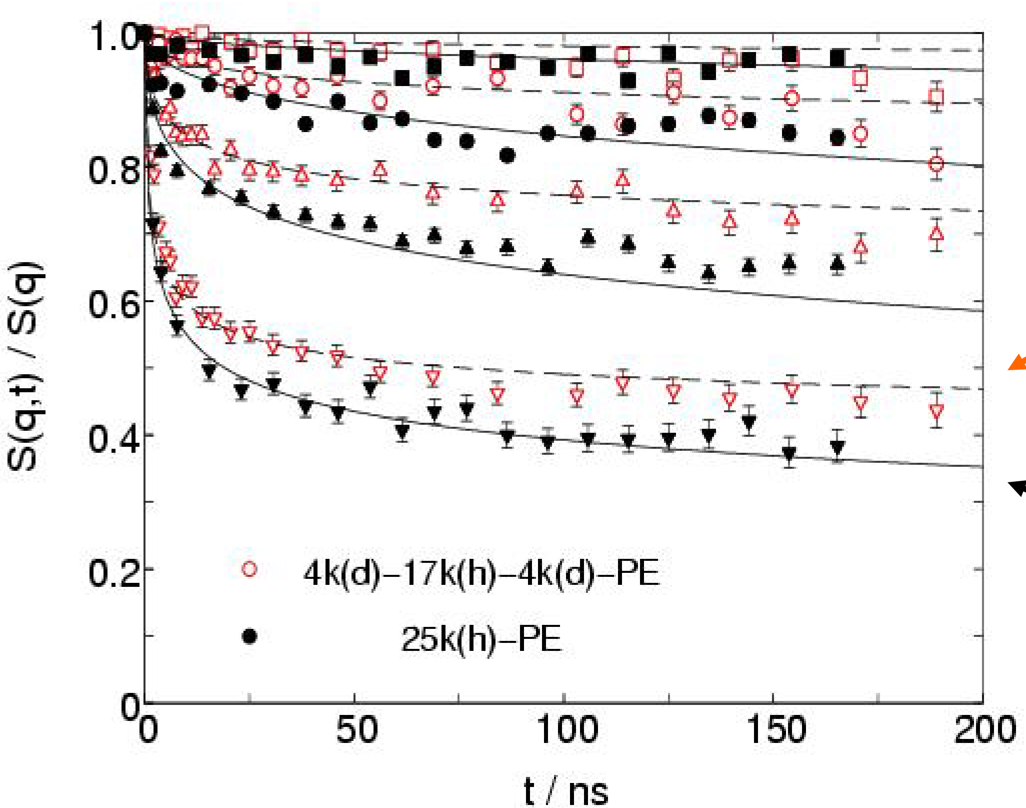
only a few branches in an entire molecule are needed to alter the processing behavior completely

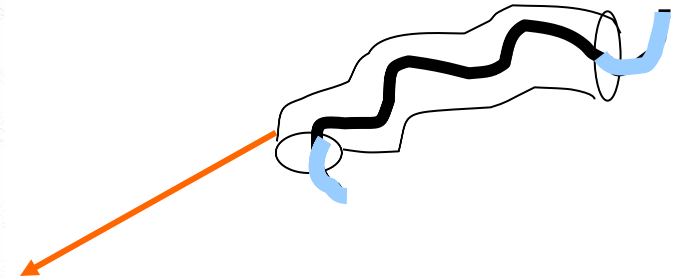
Next step: labelling !

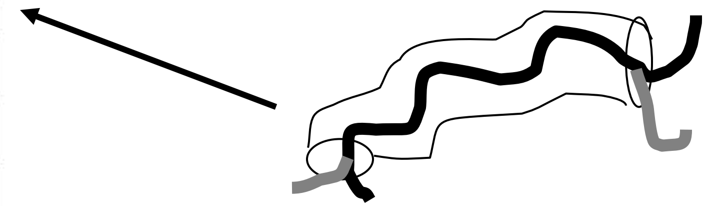
See how different sections of a molecule move !



A first step into selective labelling…….





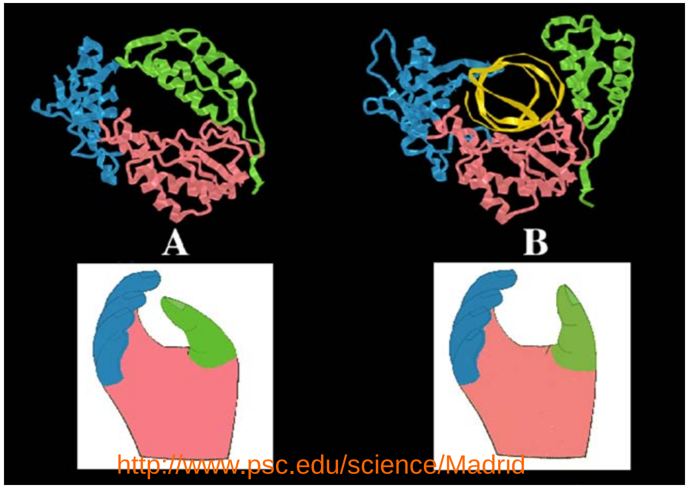


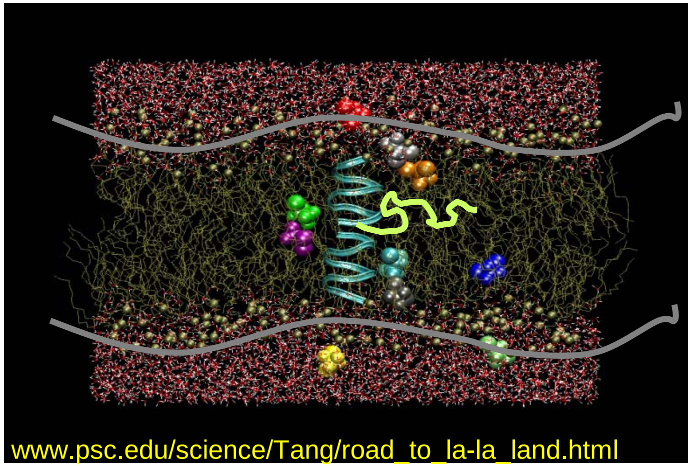
…….CLF explicitly shown !

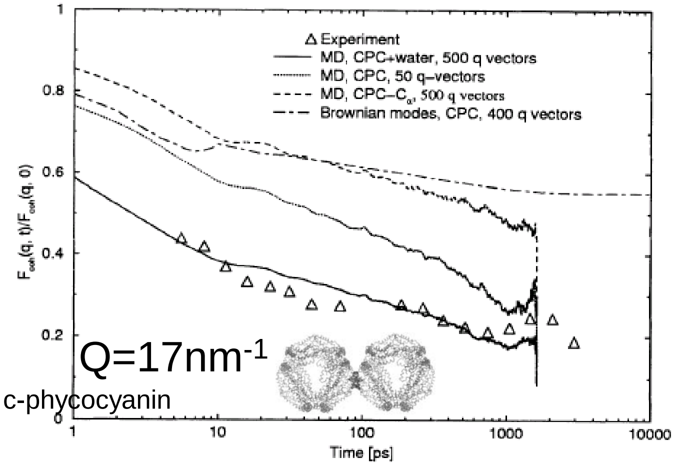
M. Zamponi et al.

e.g. applications in biology ?!

- fluctuations of biomolecules
- diffusion in membranes
- fluctuations of membranes
- diffusion in crowded solutions
- ......
- benchmark of MD of growing capabilities
- internal dynamics of modes







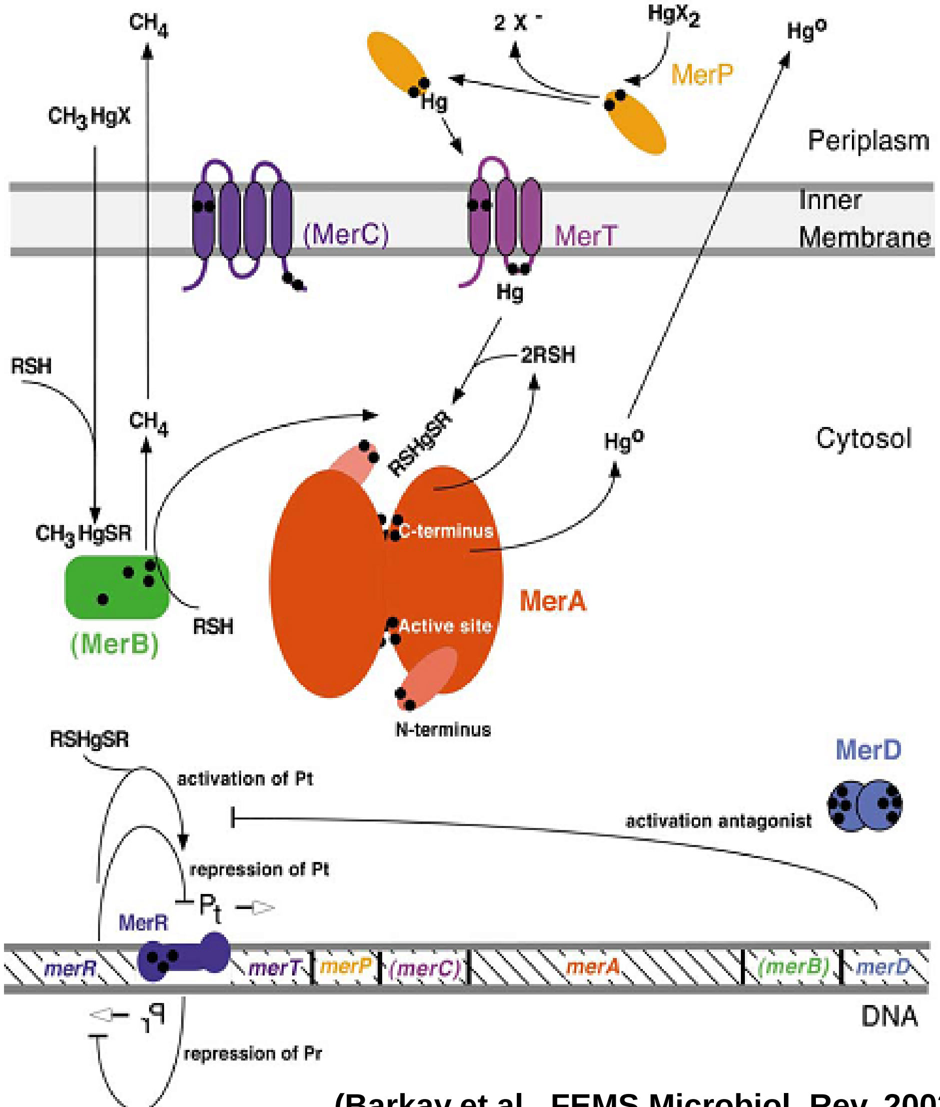
t

Q

Full potential of NSE will be explored within the next years….

K. Hinsen et al. J. Mol. Liq. 98-99,383(2002)

The mer system



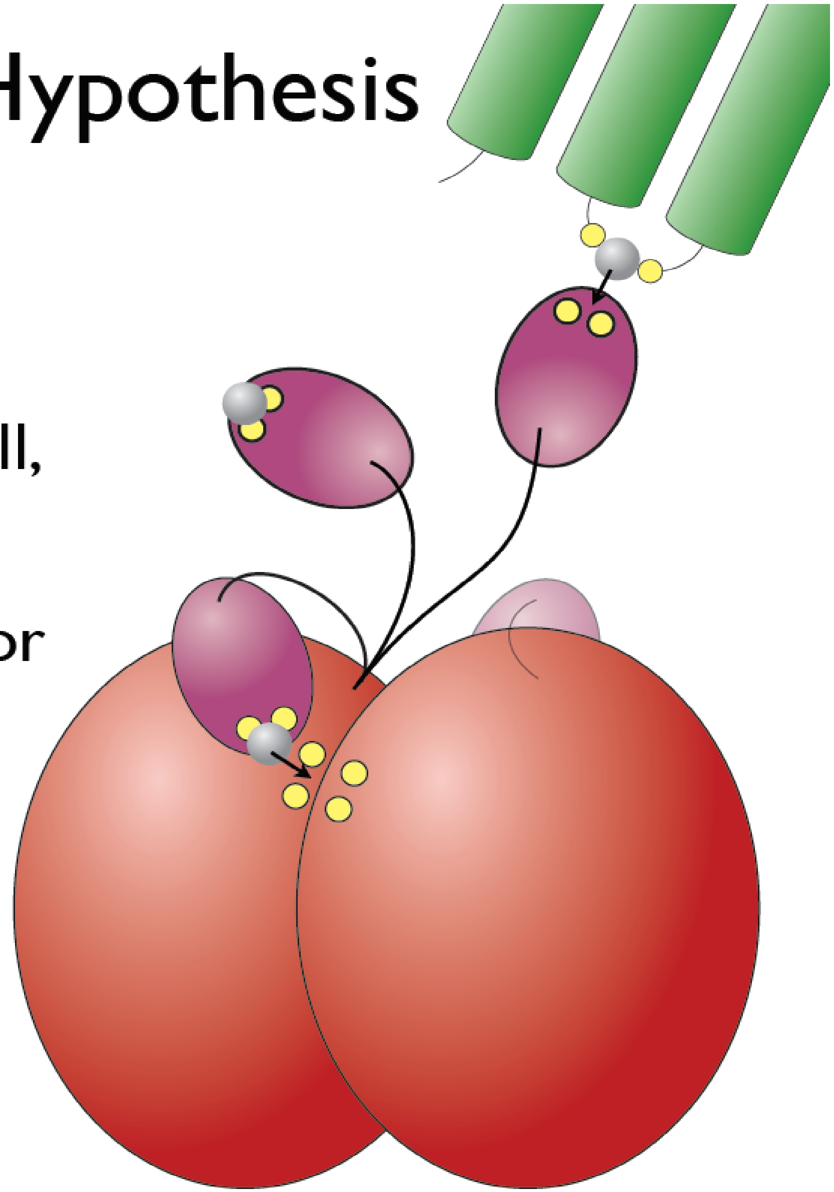
(Barkay et al., FEMS Microbiol. Rev. 2003)

- Regulation
  MerR, MerD
- Uptake & Transport
  MerC, MerT, MerP
  MerE, MerF, MerH
- Demethylation
  MerB
- Reduction
  MerA

The mer system : what it does !

N-A Hg^{2+} Handoff Hypothesis

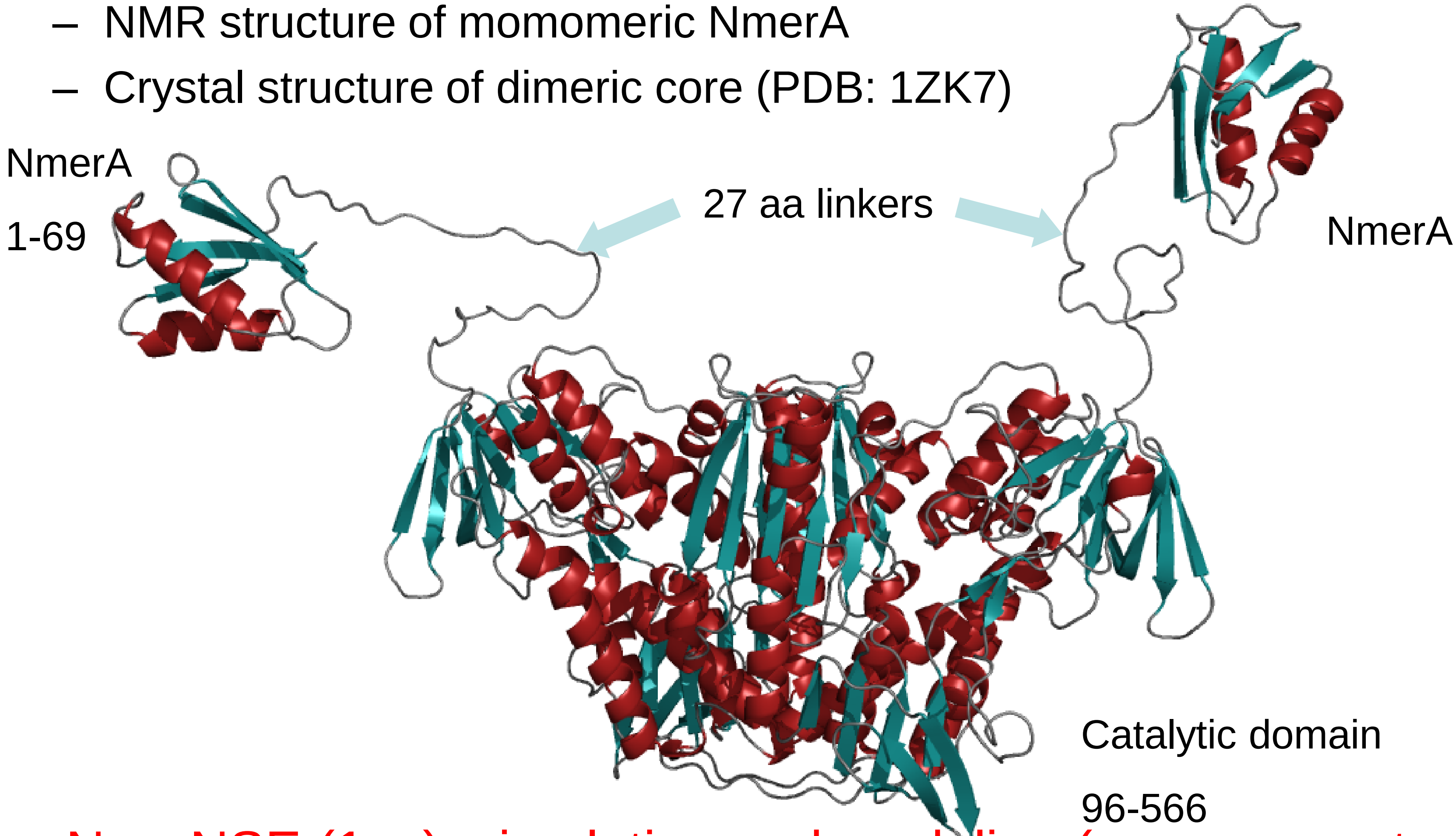
- Hg^{2+} handoff: T→N→A?
- N-terminal domain (N) hypothesized to protect cell, shuttle Hg^{2+} from T and A
- A previously solved; no N or full-length N-A structure



- Crystalize N-A and Hg^{2+} handoff intermediate: handling mechanism
- N→A model for T→N

Full-length MerA

- Model of full-length MerA with linkers
  - NMR structure of momomeric NmerA
  - Crystal structure of dimeric core (PDB: 1ZK7)



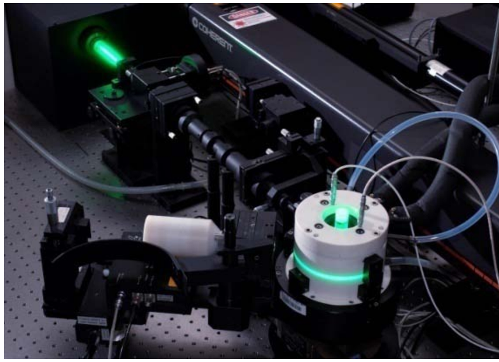
New NSE (1us), simulation and modeling (supercomputer) and sample preparation lab. in the ORNL => success !?!

Glass Methods

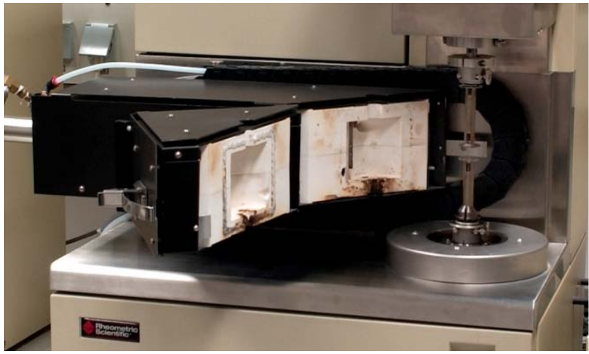
- Inelastic/Quasielastic Neutron Scattering
  - → combined information dynamics/structure
  - → isotopic labelling allows access to specific information on chemical groups/components

- Dielectric Spectroscopy
  - → large spectroscopic range (ns...s)

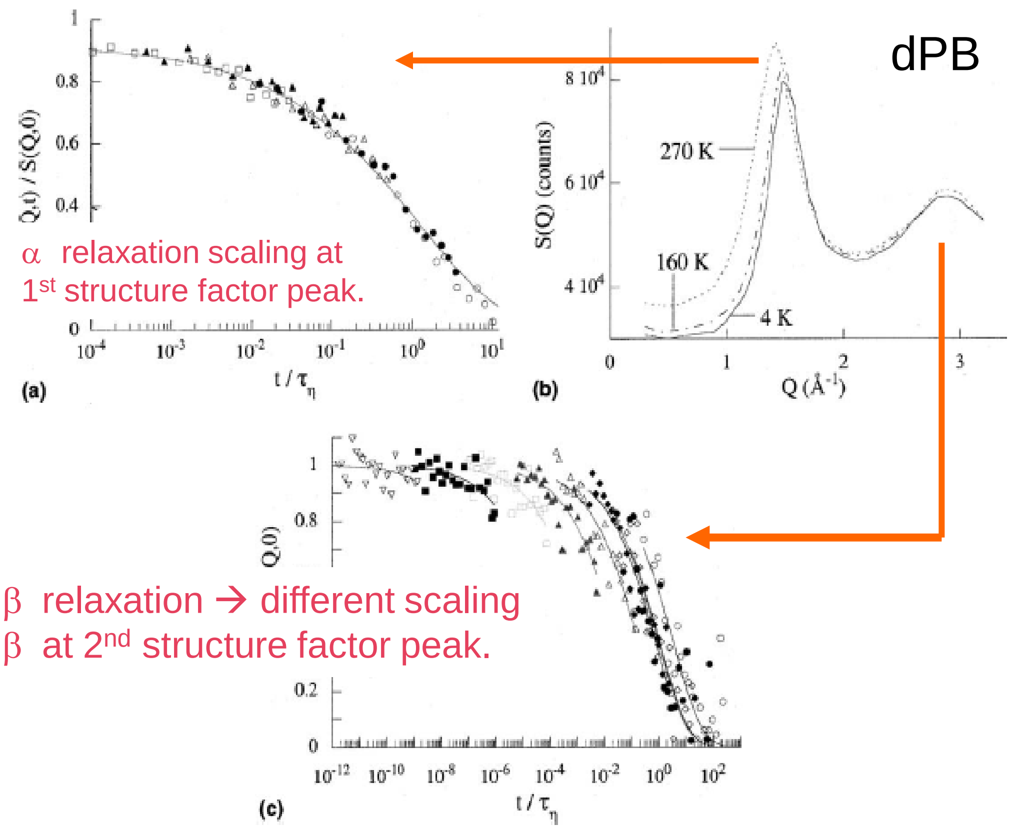
- Light Scattering
  - → large spectroscopic range (ps...s)
  - → large scale structure information



- Rheology
  - → macroscopic mechanical properties
- Calorimetry
  - → identification of glass transition
- Computer Simulation
  - → access to individual particle motion

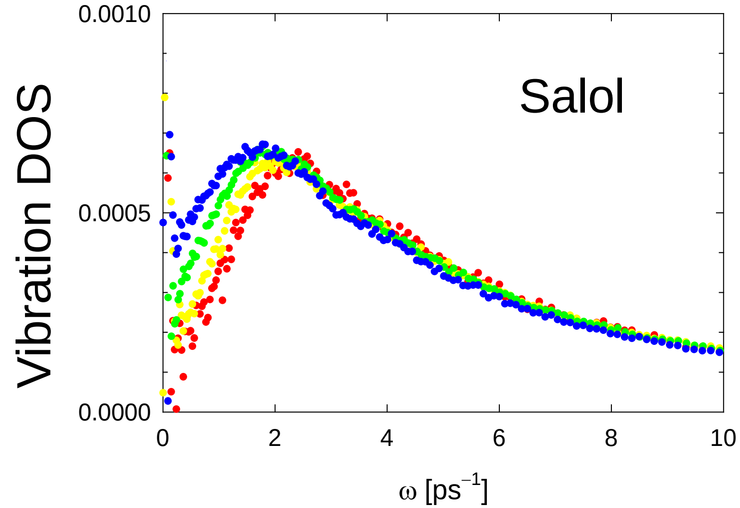


NS e.g. glass forming polymers: α- vs β-relaxation



α relaxation scaling at 1st structure factor peak.

β relaxation → different scaling
β at 2nd structure factor peak.

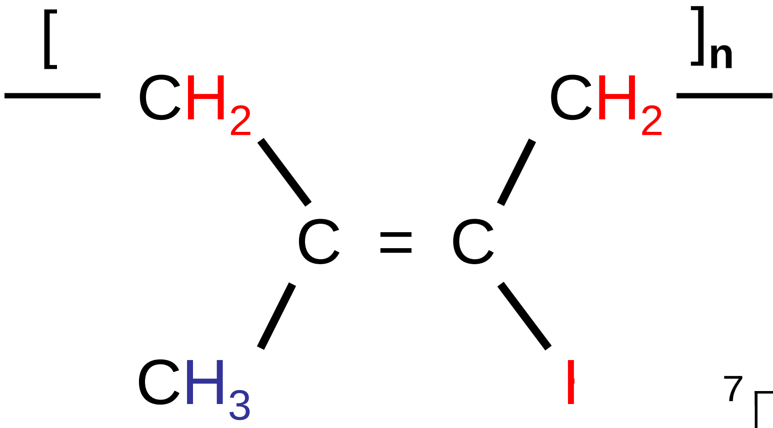


Cut-Off: lowest frequency vibrations suppressed in confinement – lowest resonator mode?

Time-temperature shift factors τ_η from macroscopic viscosity are applied

Challenges: dynamic length scales
nature of β-process and boson peak (PDOS ?)
α,β-merging

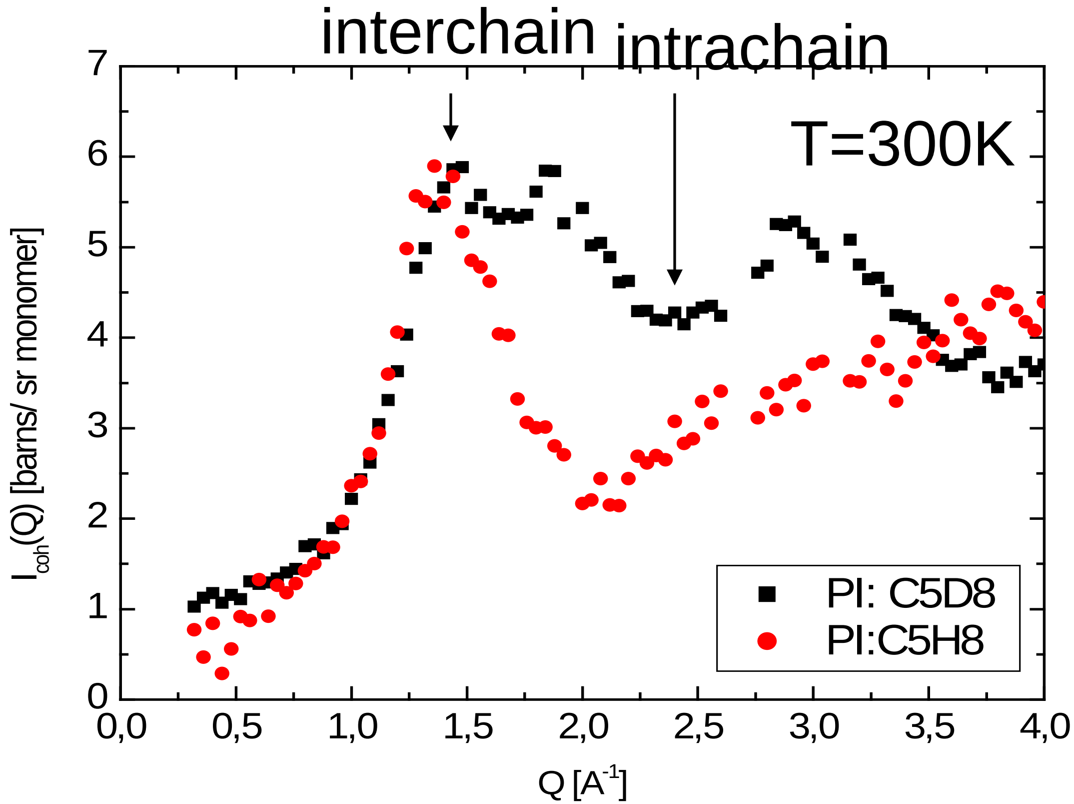
e.g. relaxation phenomena in polyisoprene



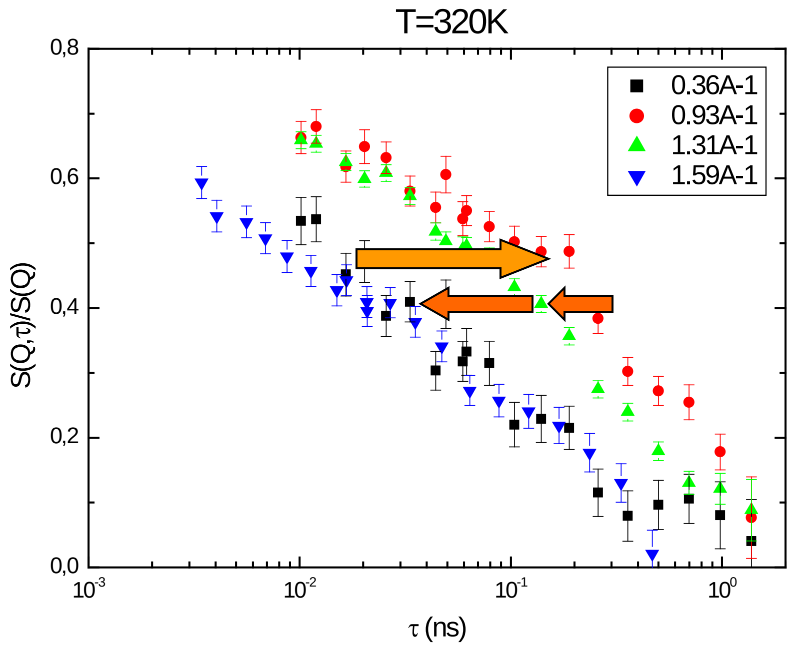
T_g=213K

M=10^5 g mol^{-1}

PI(D)/PI(D+H)>0.98

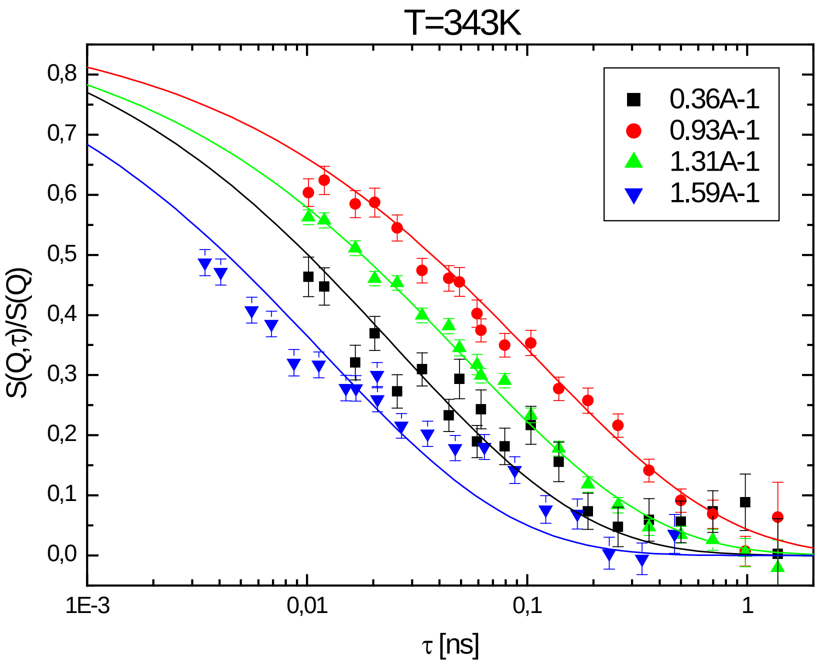


NSE: The Q - dependence (T<343K)

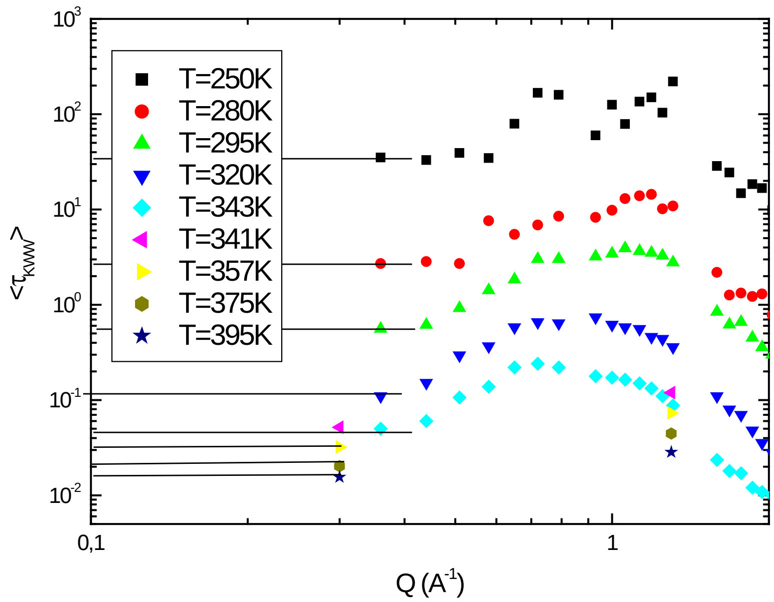


Kohlrausch-Williams-Watts

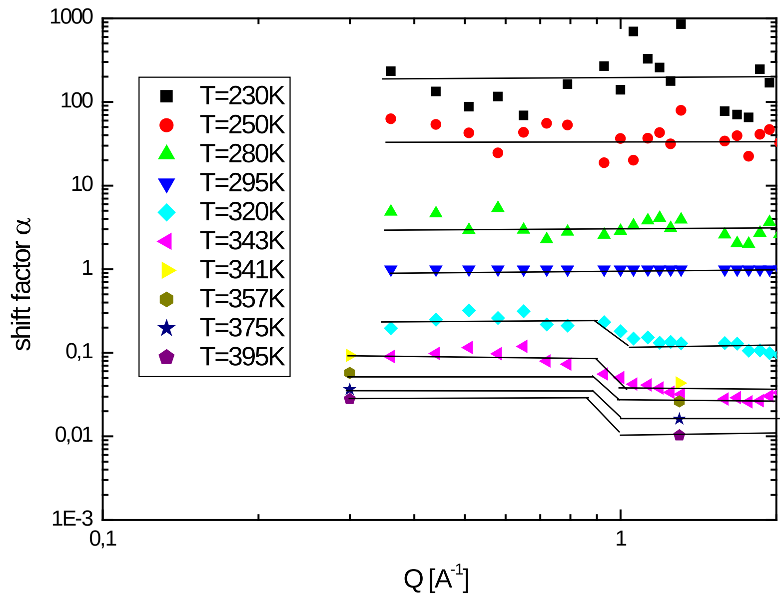
$$S(Q,\tau)/S(Q)=A\exp[-(\tau/\tau_o)^{\beta}]$$



Shift factor α_T and Q -dependence

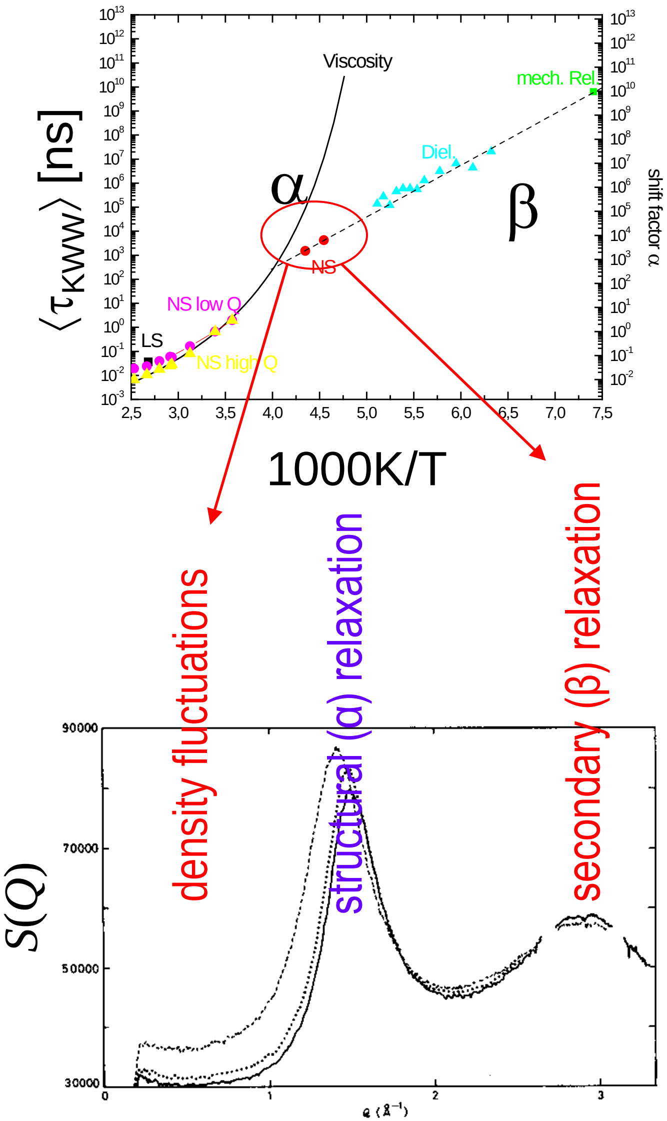


τ strongly Q dependent
... limited to Q < 1.9A^{-1} ...



For T > 320K two time scales dominate the Q - dependence

Intermediate scale collective dynamics



How do secondary relaxations couple to stress relaxations?

NSE Experiments at *low and high* Q for various polymers

Acitvation Energy
JG – process (τ_0=10^{-13}s/ 220K): ΔE= 3698K*k_b
Mech. relaxation* (τ_0=10^{-13}s): ΔE= 3883K*k_b

* K. Schmieder and K. Wolf, Kolloid Zeitschrift 134, 149 (1953)

Typical experiment

- sample size 3x3cm^2 , sample cells are Hellma Quartz or Aluminum cells
- transmission of about 60% for small angles and typical soft matter (polymers)
- 10^5n/(cm^2*s) on sample
- each tau and Q setting costs of about 30min to 6h (depending on sample)
- resolution measurement, graphite, elastic scatterer
- sometimes buffer like D2O
- typical experiment time 7 to 14days
- temperature from 10K to 600K

Summary

-NSE is the only neutron scattering method measuring the slow dynamics of materials

- The technique bases on encoding and decoding tiny velocity changes of the neutrons in the sample into neutron spin precession

- It is the highest energy resolution neutron scattering method and measures S(Q,T) instead of S(Q,w)

- With the invention of the technique in early 1970 a growing interest can be reported

Special thanks to … for preparation:

...
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R. Zorn (FZJ)