C. ALBA-SIMIONESCO

Laboratoire Léon Brillouin ,CNRS /CEA, Saclay France

National Neutron Scattering Facility

Outline QuasiElastic Neutron Scattering from an user point of view

Part I : why neutrons ?

context and experimental probes

Role of the experimentalist :user, facility scientist

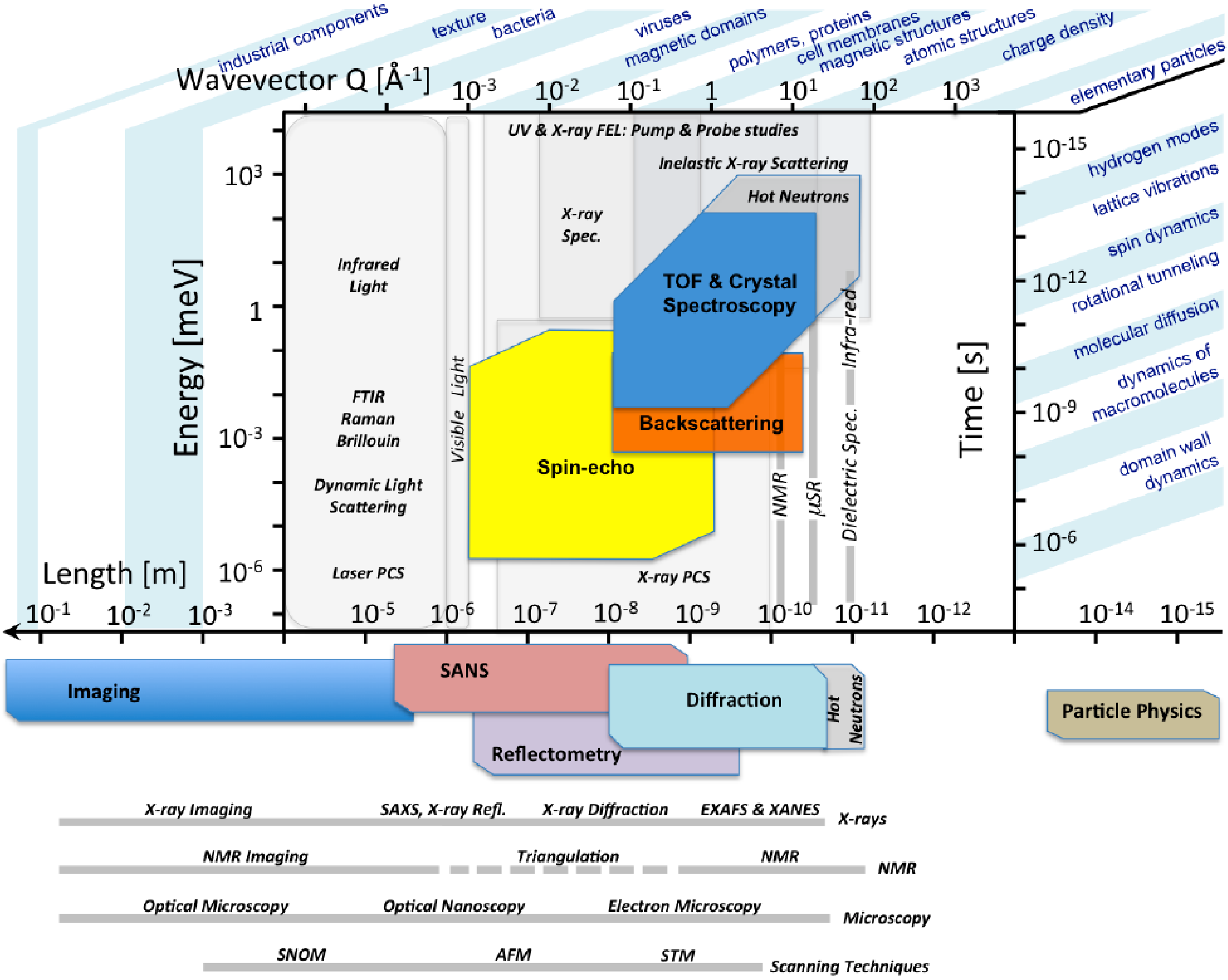
QENS spectrometers

Part II : what are the observables?

constraints on measurements, limitations

Models and theories

Sample environment : next challenges


industrial components
texture
bacteria
viruses
magnetic domains
polymers, proteins
cell membranes
magnetic structures
atomic structures
charge density
elementary particles
Wavevector Q [Å^{-1}]
10^{-3}
10^{-2}
10^{-1}
1
10^{1}
10^{2}
10^{3}
UV & X-ray FEL: Pump & Probe studies
Inelastic X-ray Scattering
Hot Neutrons
X-ray Spec.
Infrared Light
TOF & Crystal Spectroscopy
FTIR
Raman
Brillouin
Visible Light
Infra-red
Backscattering
Spin-echo
Dynamic Light Scattering
NMR
µSR
Dielectric Spec.
Laser PCS
X-ray PCS
Energy [meV]
10^{3}
1
10^{-3}
10^{-6}
Time [s]
10^{-15}
10^{-12}
10^{-9}
10^{-6}
hydrogen modes
lattice vibrations
spin dynamics
rotational tunneling
molecular diffusion
dynamics of macromolecules
domain wall dynamics
Length [m]
10^{-1}
10^{-2}
10^{-3}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-14}
10^{-15}
Imaging
SANS
Diffraction
Hot Neutrons
Reflectometry
Particle Physics
X-ray Imaging
SAXS, X-ray Refl.
X-ray Diffraction
EXAFS & XANES
X-rays
NMR Imaging
Triangulation
NMR
NMR
Optical Microscopy
Optical Nanoscopy
Electron Microscopy
Microscopy
SNOM
AFM
STM
Scanning Techniques

Laboratoire Léon Brillouin

good reasons to use neutrons

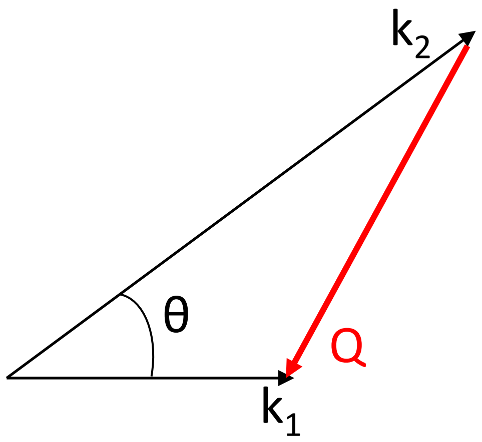
- **Cover large scales of time and space simultaneously**
- **A unique probe for the magnetism**
- **Complementary to the other techniques; give access directly in observable relevant and defined well (S(Q), S(Q,w), F (Q, t), dispersion curves)**
- **Neutr**

Their wavele
The interacti
The neutron
The neutron
The use of va

- Neutron scattering methods are pluridisciplinary
-it provides final answers to fundamental questions and
- a solid background to support any other techniques
(X rays, NMR, Numerical Simulations..)

thanks to well controlled and well known observables
providing absolute quantities.

Comparison with x rays and light



$\mathbf{Q} \equiv \mathbf{k_1} - \mathbf{k_2}$

$Q^2 = k_1^{\,2} + k_2^{\,2} - 2|\mathbf{k_1}||\mathbf{k_2}| \cos \theta$

$\hbar\omega \equiv E_1 - E_2$

$Q_{el} = 2|\mathbf{k_1}| \sin (\theta/2)$
$= (4\pi/\lambda) \sin (\theta/2)$

Neutrons:

$$E = \frac{h^2}{2m_n}\left(\frac{1}{\lambda}\right)^2$$

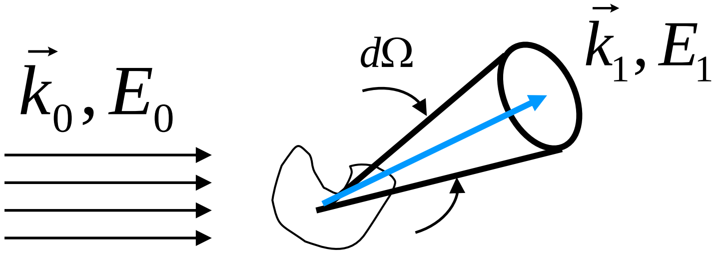
E (meV) = $81.81/ \lambda^2$

X Rays and Light scattering:

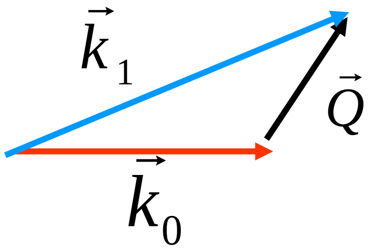
$$E = hc\left(\frac{1}{\lambda}\right)$$

E(keV) = $12.4 / \lambda$

(λ in Å)



$$\vec{Q} = \vec{k}_1 - \vec{k}_0$$
$$h\omega = E_f - E_i$$



$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} \approx N * \sigma_{Scat} * S(\vec{Q},\omega)$$

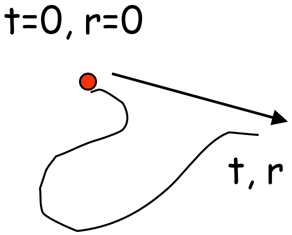
Measured Intensity

Scattering cross-section

Dynamical Structure Factor

<u>The “Physics” is here</u>

$S(\vec{Q},\omega)$ Is the FT **G(r,t)** (**<u>van Hove correlation function)</u>**



$$\boxed{S(\vec{Q},\omega) = \frac{1}{2\pi}\int G(\vec{r},t)e^{i(\vec{Q}\vec{r}-\omega t)}d\vec{r}dt}$$

<u>van Hove correlation function:</u> G(*r*,t) is the probability to find a particle at a distance *r*, at time t, provided it was at *r*=0, at t=0.

QUASI ELASTIC NEUTRON SCATTERING : QENS

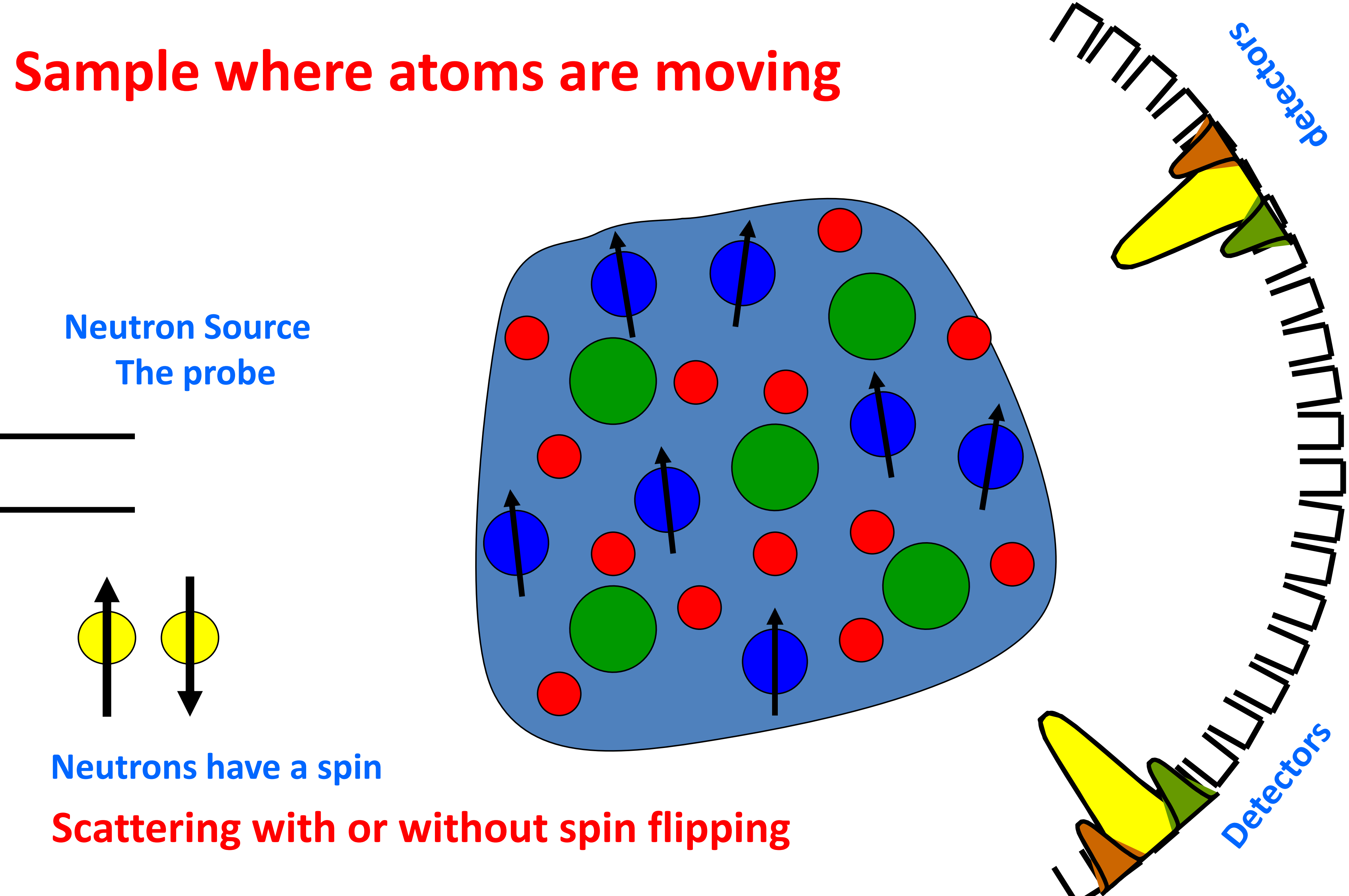
Measure of scattering processes involving small amounts of energy exchange, classical approximation ($|\hbar\omega| << \frac{1}{2} k_B T$)
i.e. in the low energy region of inelastic spectra close to 0

Dynamical phenomena at 10^{-13} to 10^{-7} s
Motions explored in space on legnthscales comparable with λ of the neutrons

Vibrational displacements, librations, jump distances, diffusion paths, corrlation lengths **(nano to micro)**

Observables : Dynamical structure factor, S(Q,w) or intermediate scattering function, mean square-displacements, self diffusion coefficient, relaxation time, Reptation Rouse modes, friction corfficient, Rotationl diffusion, EISF, VDOS, Cp

Sample where atoms are moving



Neutron Source
The probe

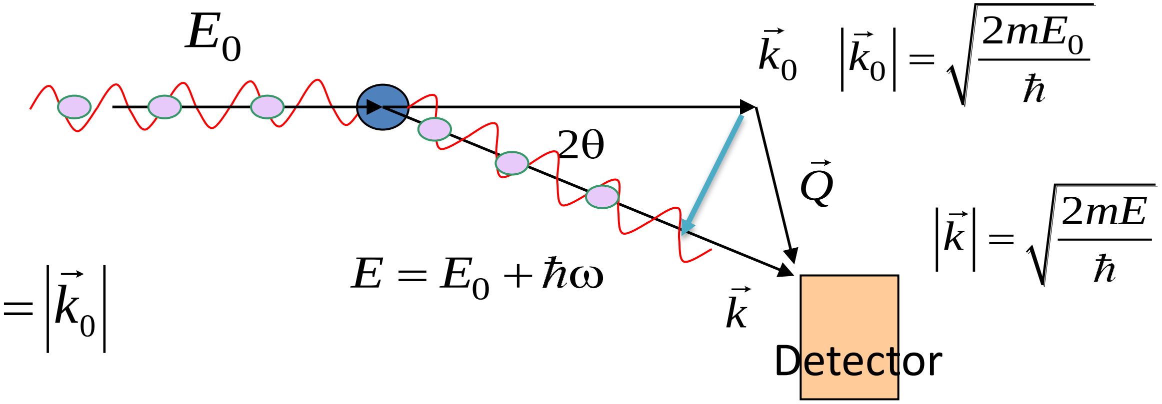
Neutrons have a spin

Scattering with or without spin flipping

Elastic quasielastic and inelastic scattering of neutrons

Incoming Neutron
λ : 2 to 10 Å
$E_{0:}$ 1 to 10 meV

$$Q_{elastic} = \frac{4\pi}{\lambda_0}\sin\theta \text{ and } |\vec{k}| = |\vec{k}_0|$$

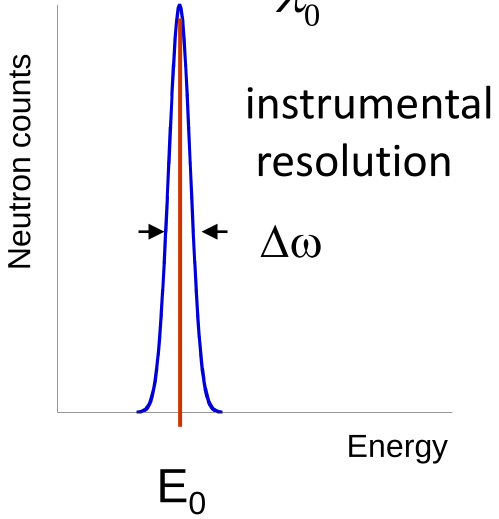


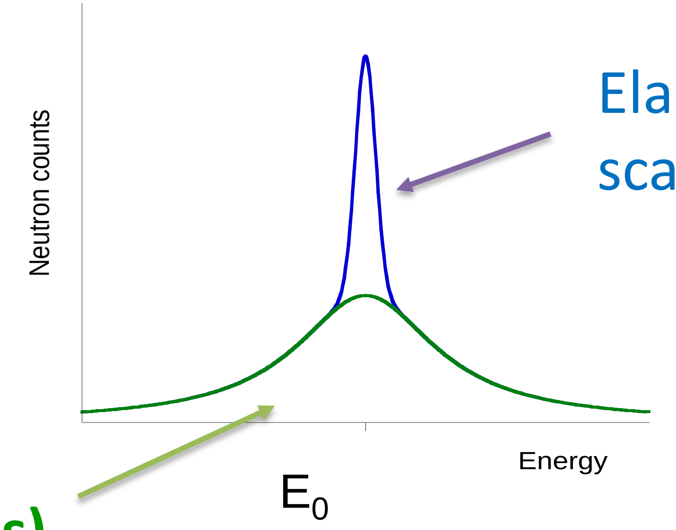
$$|\vec{k}_0| = \sqrt{\frac{2mE_0}{\hbar}}$$

$$|\vec{k}| = \sqrt{\frac{2mE}{\hbar}}$$

$$E = E_0 + \hbar\omega$$

Technically a TOF spectrum is accumulated at a fixed scattering angle for each detector

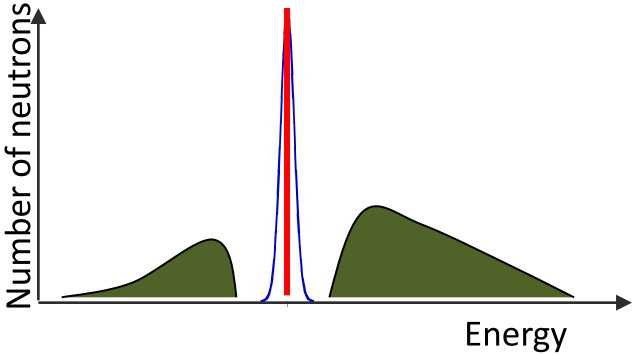




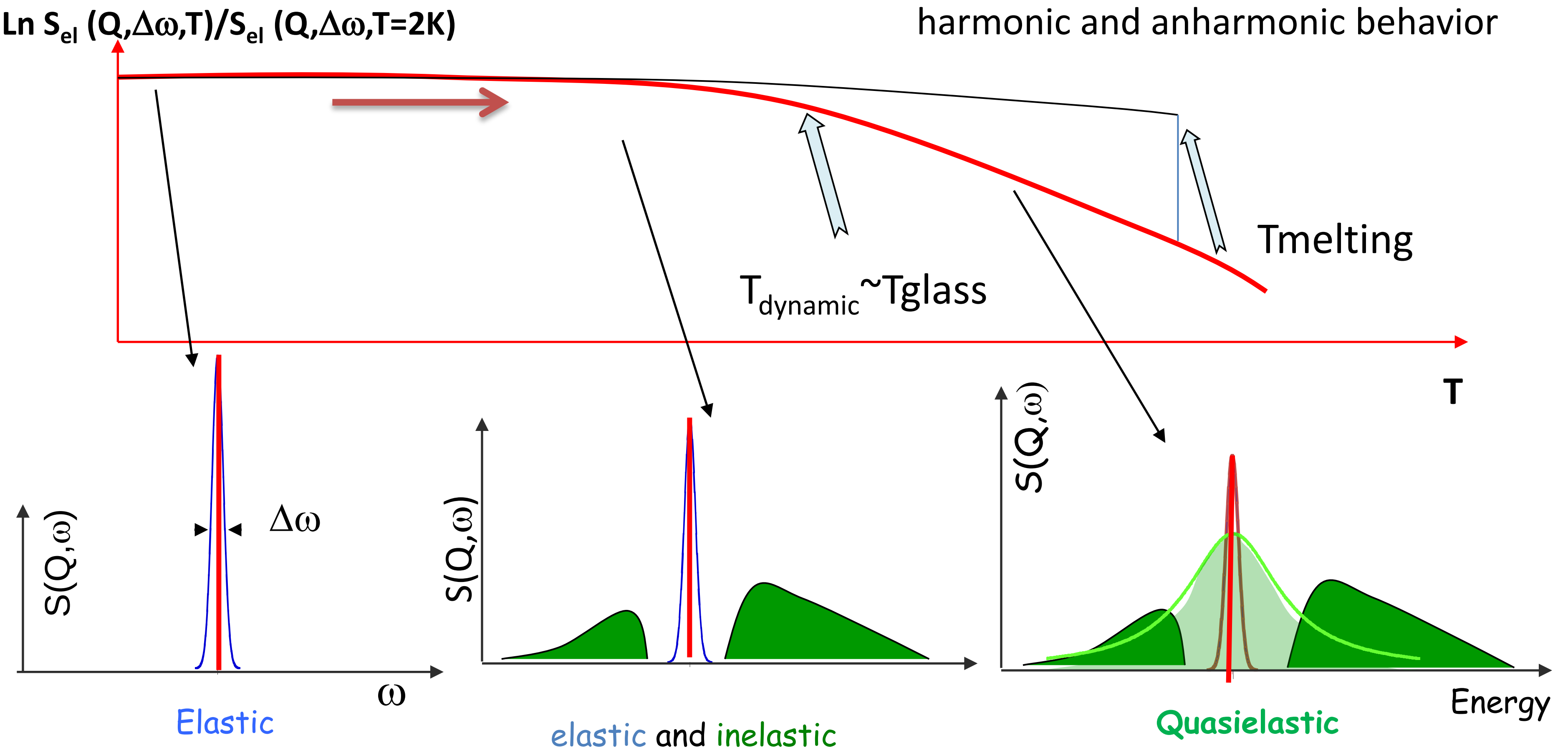
Elastic scattering

diffusif (10^{-9} – 10^{-12} s)
quasi elastic broadening

Elastic and inelastic



scenario of quasielastic scattering as T increases
Molecular liquids and polymers



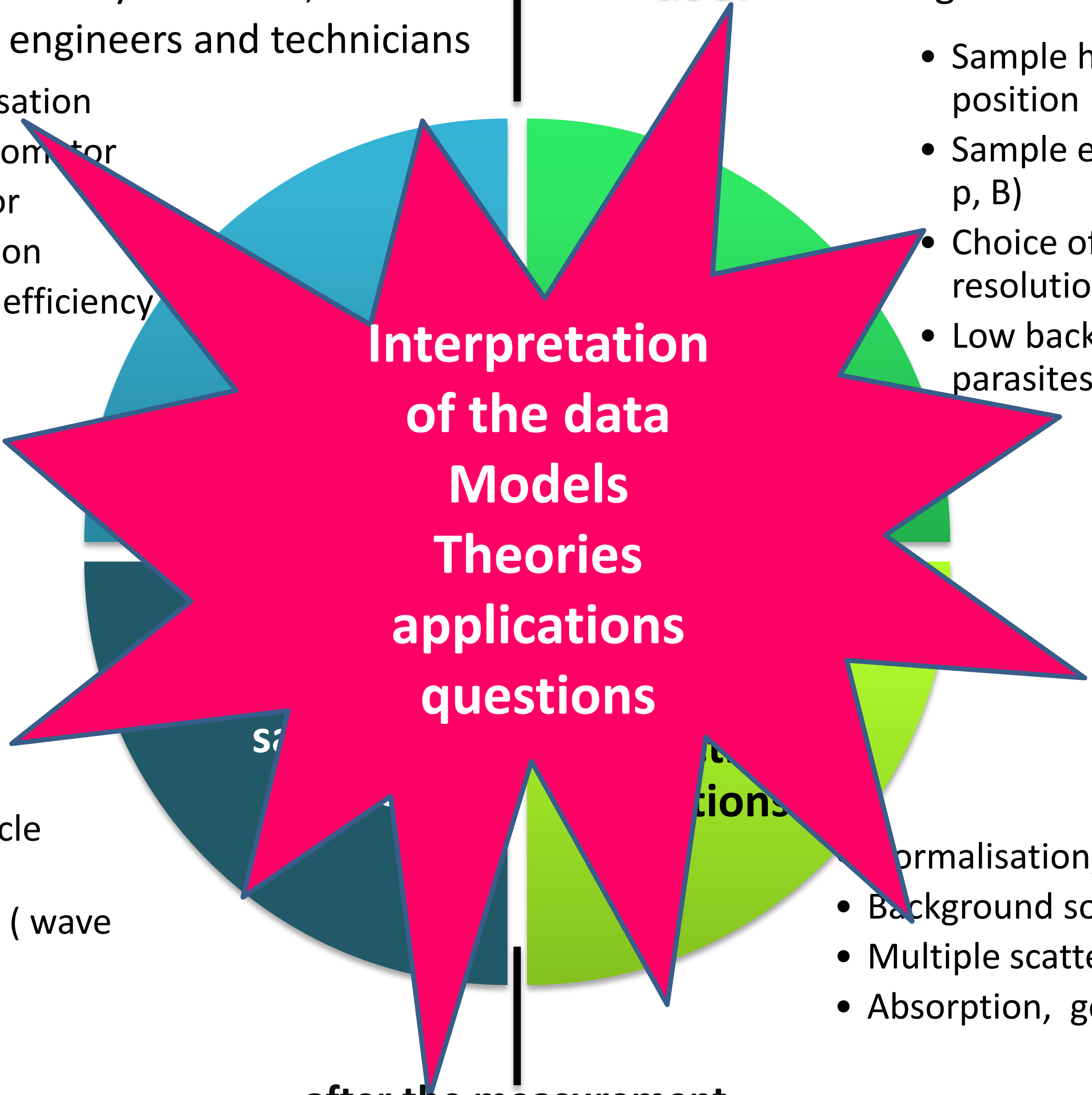
To cover a wide w range requires the combination of several instruments

Speed and wave length of the neutron

λ (A)	v (m/s)	k (A^{-1})
0,5	7 912	12,6
1,0	3 956	6,3
1,5	2 637	4,2
2,0	1 978	3,1
2,5	1 582	2,5
5,0	791	1,3
10,0	396	0,6
20,0	198	0,3

Sequences of steps for an experiment

Before the measurement



Facility scientists,
engineers and technicians

- Thermalisation
- Monochrom tor
- Collimator
- Polarisation
- Detector efficiency

user interacting at the facility

- Sample holder and position
- Sample environment (T, p, B)
- Choice of the E and Q resolution
- Low background , parasites

- Speed,(particle character)
- Wavelength (wave character)
- direction,
- polarisation

- ormalisation, Vanadium
- Background soustraction
- Multiple scattering
- Absorption, geometry

after the measurement

motions

$$\left\{\begin{array}{l}\textbf{0.1psec to 500nsec}\\ \textbf{.5meV à 20meV}\\ \textbf{1000Å à 1 Å}\end{array}\right\}$$

Several instruments

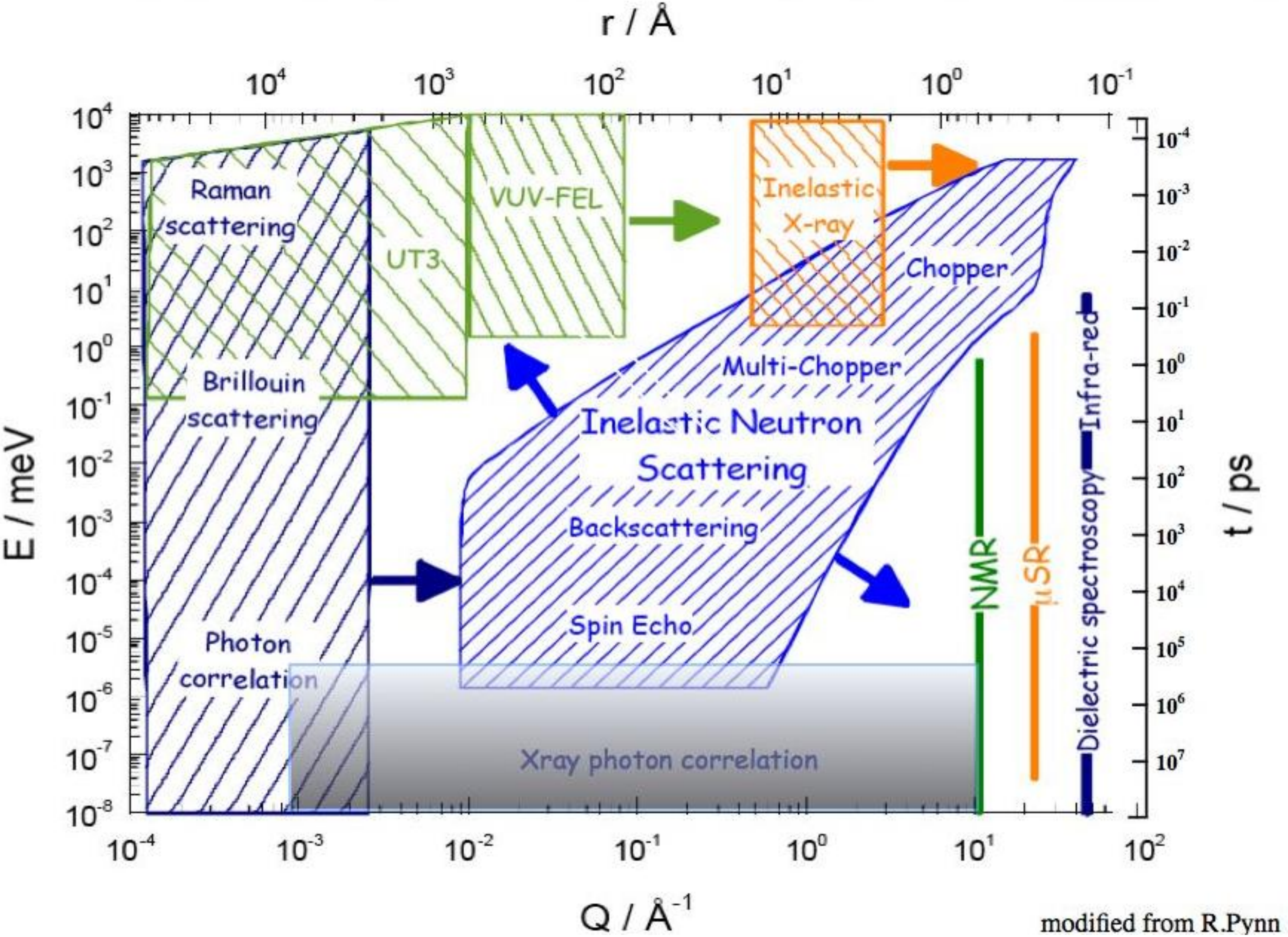
Triple Spectrometer

Time of Flight Spectrometer *(based on energy transfer analysis)*

Backscattering Spectrometer

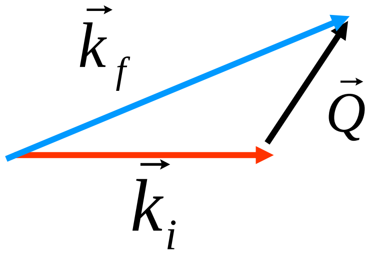
Neutron spin Echo Spectrometer (NSE and NRSE)

Based on the Fourier time analysis of the scattered intensity



modified from R.Pynn

Triple axis and Time of flight approaches



Create high continuous flux of monochromatic neutrons (ki fixed)
continuous detection of monochromatic neutrons (kf fixed)

Triple axis spectrometer

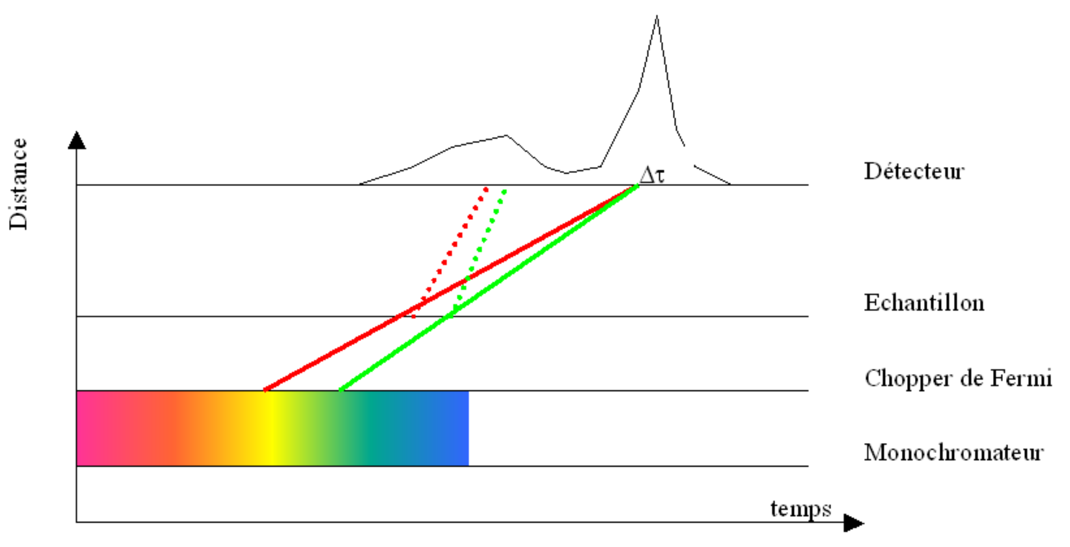
Create a high flux of pulsed monochromatic neutrons (ki fixed)
Detection as a function of time (kf variable)

Direct geometry Time of flight

Create high flux white neutron beam (ki variable)
Detection of monochromatic neutrons as function of time (kf fixed)

Indirect geometry Time of flight

$$\boxed{\left(\frac{d^2\sigma}{d\Omega d\omega}\right) = \frac{k_f}{k_i}\frac{\sigma_{sc}}{4\pi} N\, S(\vec{Q},\omega)}$$

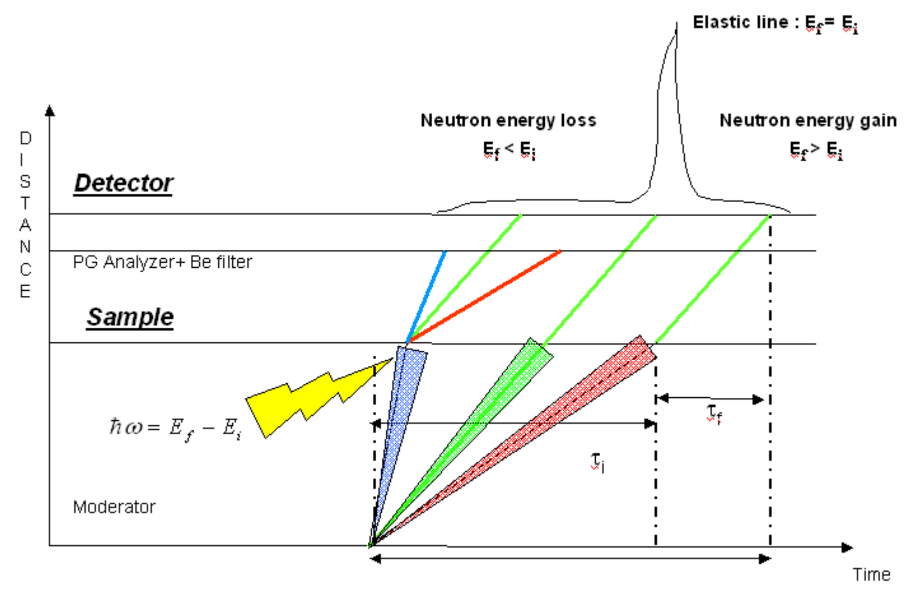


Time focusing
IN6 (ILL) – Focus (PSI)

Select a broad incident energy band
+
The « fastest » neutrons
Get to the detector at the same time that
the « slowest » ones.

Large flux but resolution not triangular

NB:
Can focus in the inelastic region



Inverted geometry
QENS (ANL/Intense Pulsed Neutron Source)
IRIS (ISIS)

Use a white beam
+ analyzer in front of detectors.

Concurent measurement of elastic (S(Q))
AND inelastic (S(Q,w))

On pulsed sources: measure the
neutron energy loss side: Bose factor not a limiting factor

Can probe far in the inelastic even at low temperature
Resolution± fixed
High background

FOCUS @ PSI

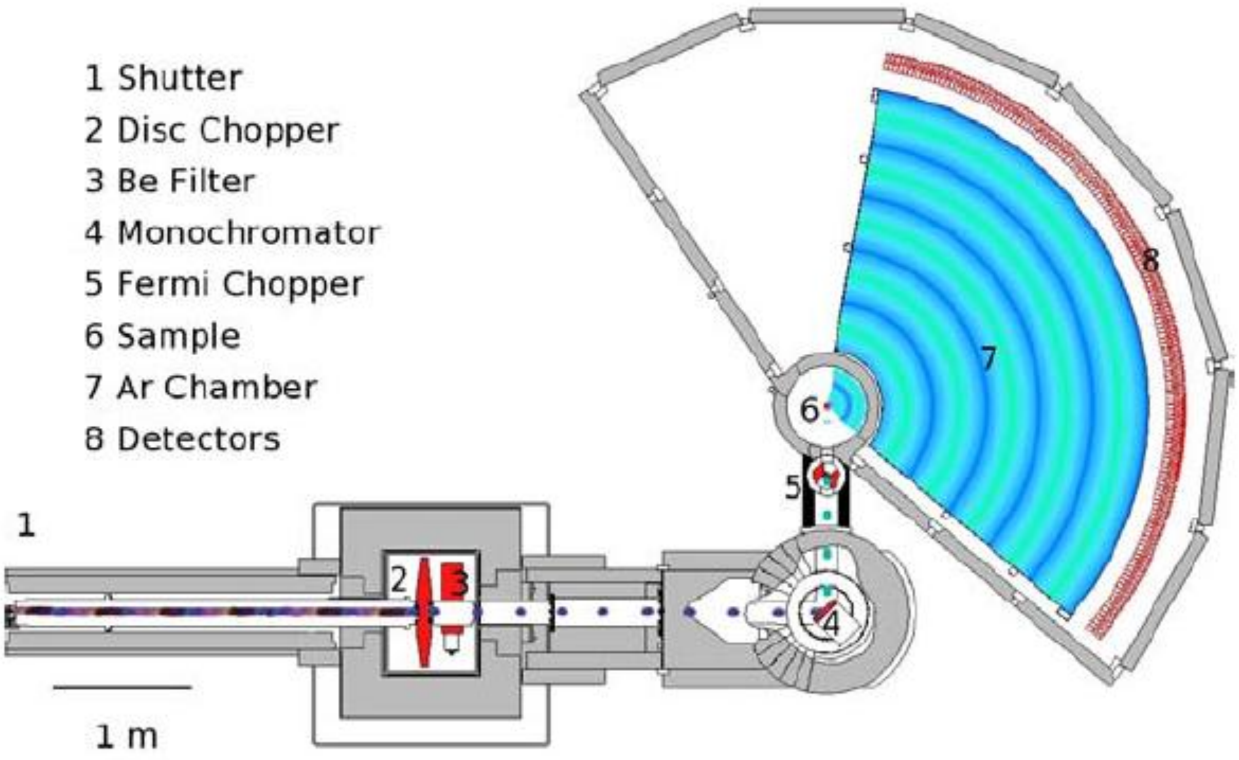


Fig. 3. FOCUS spectrometer at Paul-Scherrer Institut (PSI) [29]; FOCUS is a typical XTL-TOF spectrometer, i.e. a time-of-flight instrument with a Bragg monochromator and a time-of-flight analyzer. While the monochromator selects the incident neutron energy E_0, the energy of the scattered neutrons E is determined by measuring the neutron flight time.

The Fa# Project @ LLB
J-M Zanotti, S Rodriguez

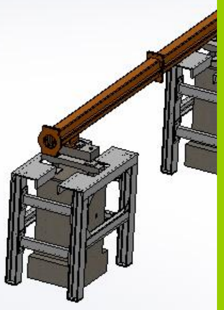
« Hybrid » Instrument :

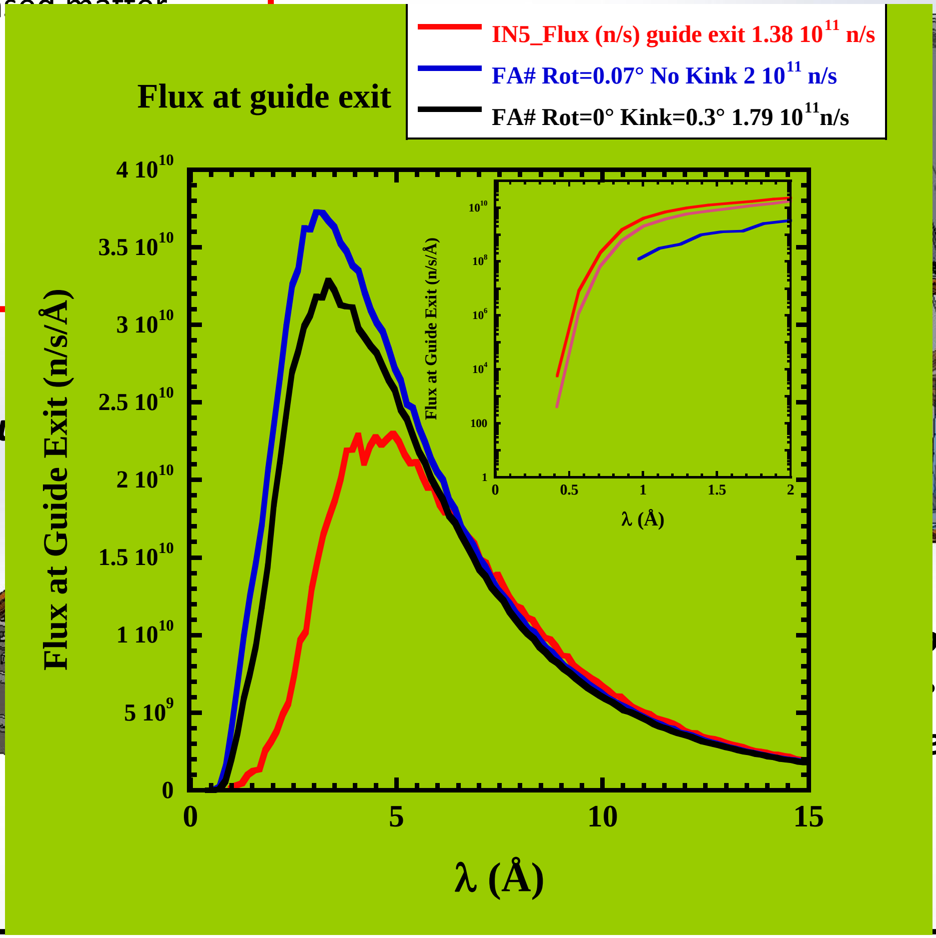
- **Time focusing (Soft matter / Biology)**
- **Energy focusing (Solid state physics)**

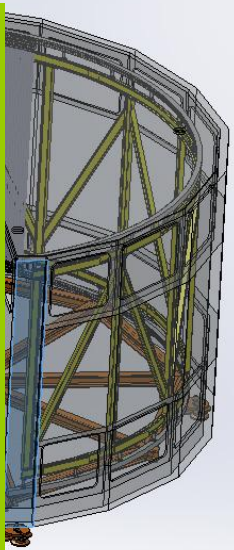
Scientific fields

- ○ Soft condensed matter
- ○ Materials
- ○ Magnetism
- ○ Soft Matter
- ○ Biology
- ○ Electrolites
- ○ …

Elliptic gu







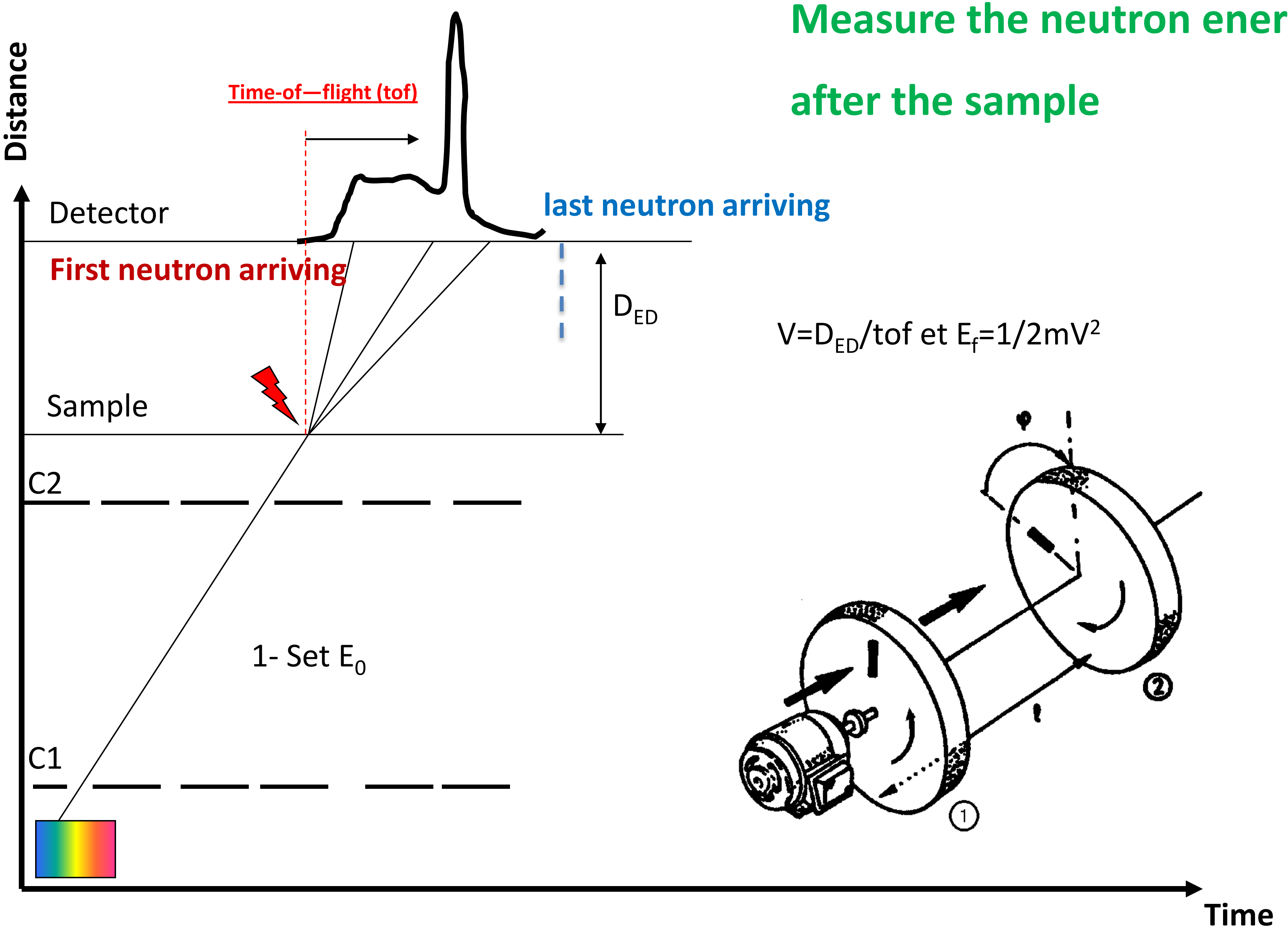
Detection
sition
nsitive
etectors (^{3}He)

P. Lavie, LLB

Disks choppers in cascade

Measure the neutron energy

after the sample



Distance

Time-of—flight (tof)

Detector

last neutron arriving

First neutron arriving

D_{ED}

Sample

C2

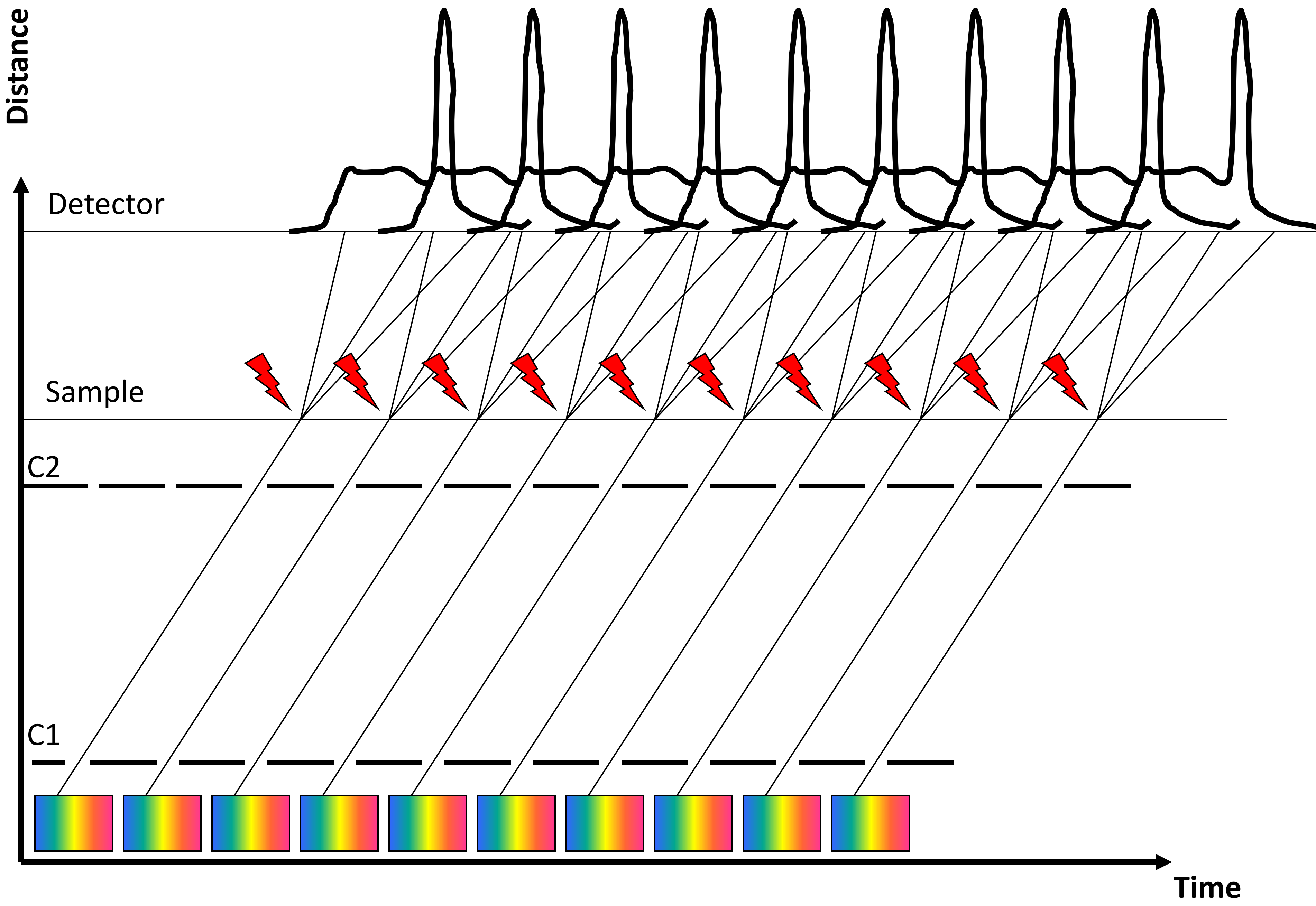
1- Set E_0

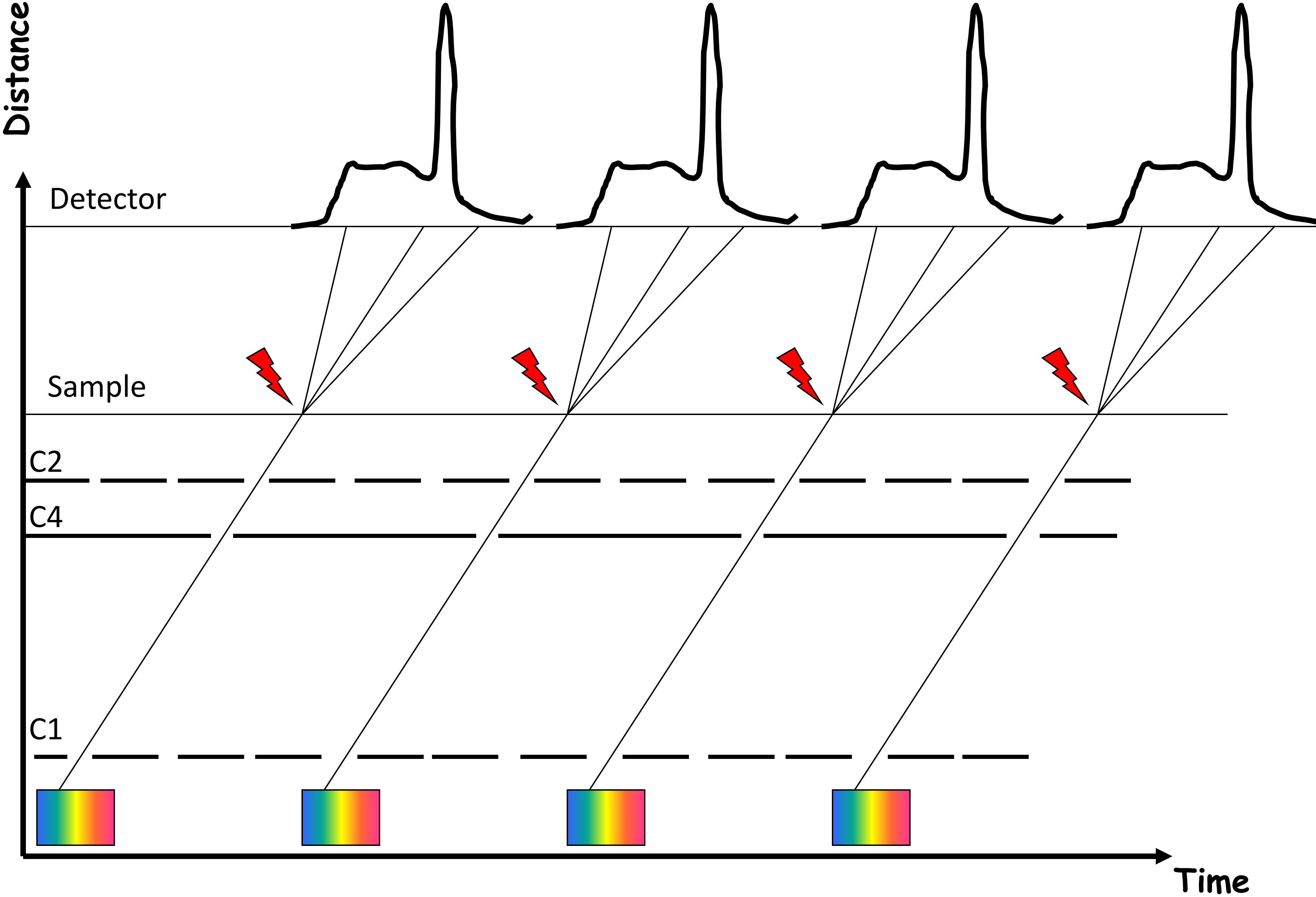
C1

Time

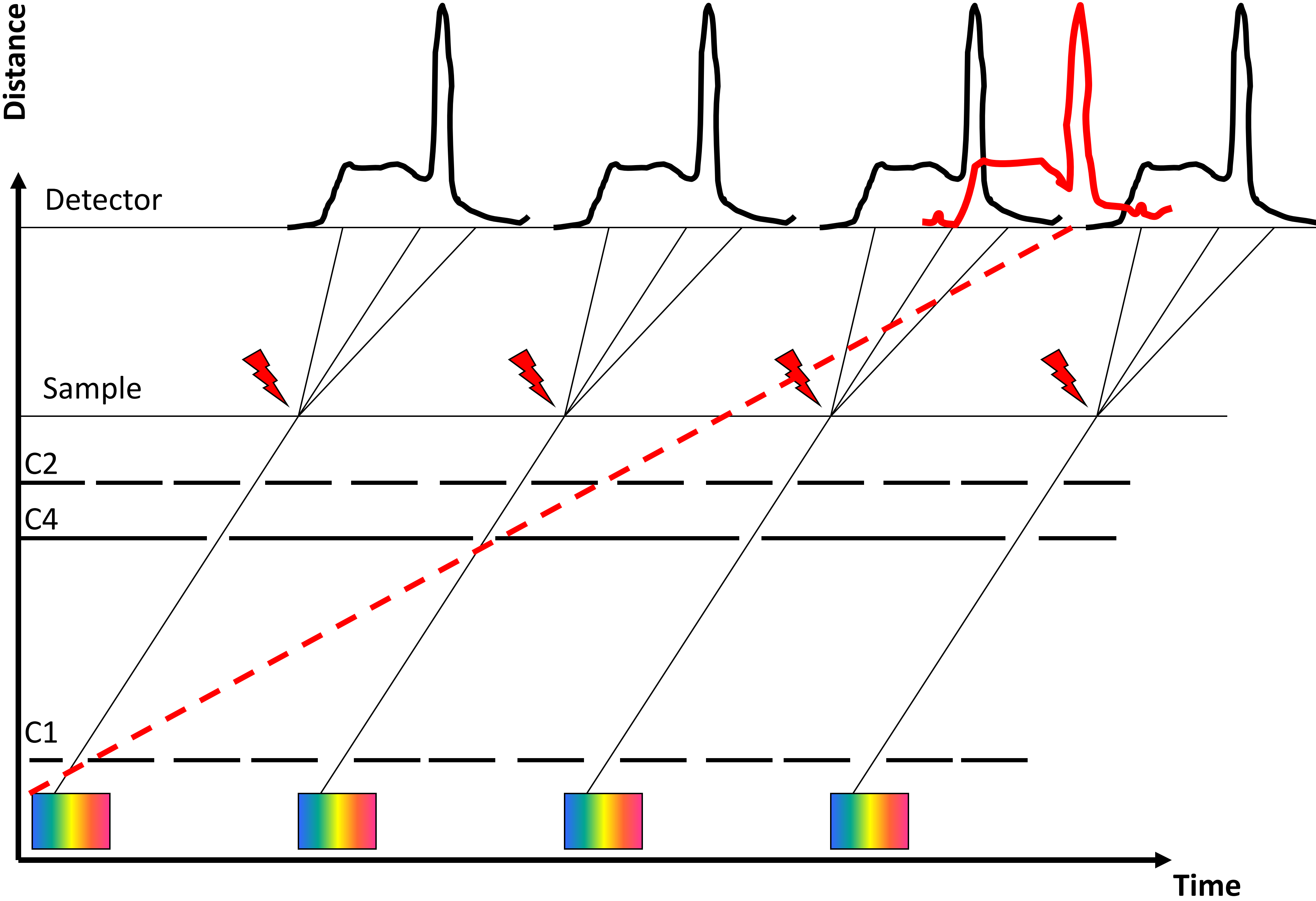
$V=D_{ED}/tof$ et $E_f=1/2mV^2$

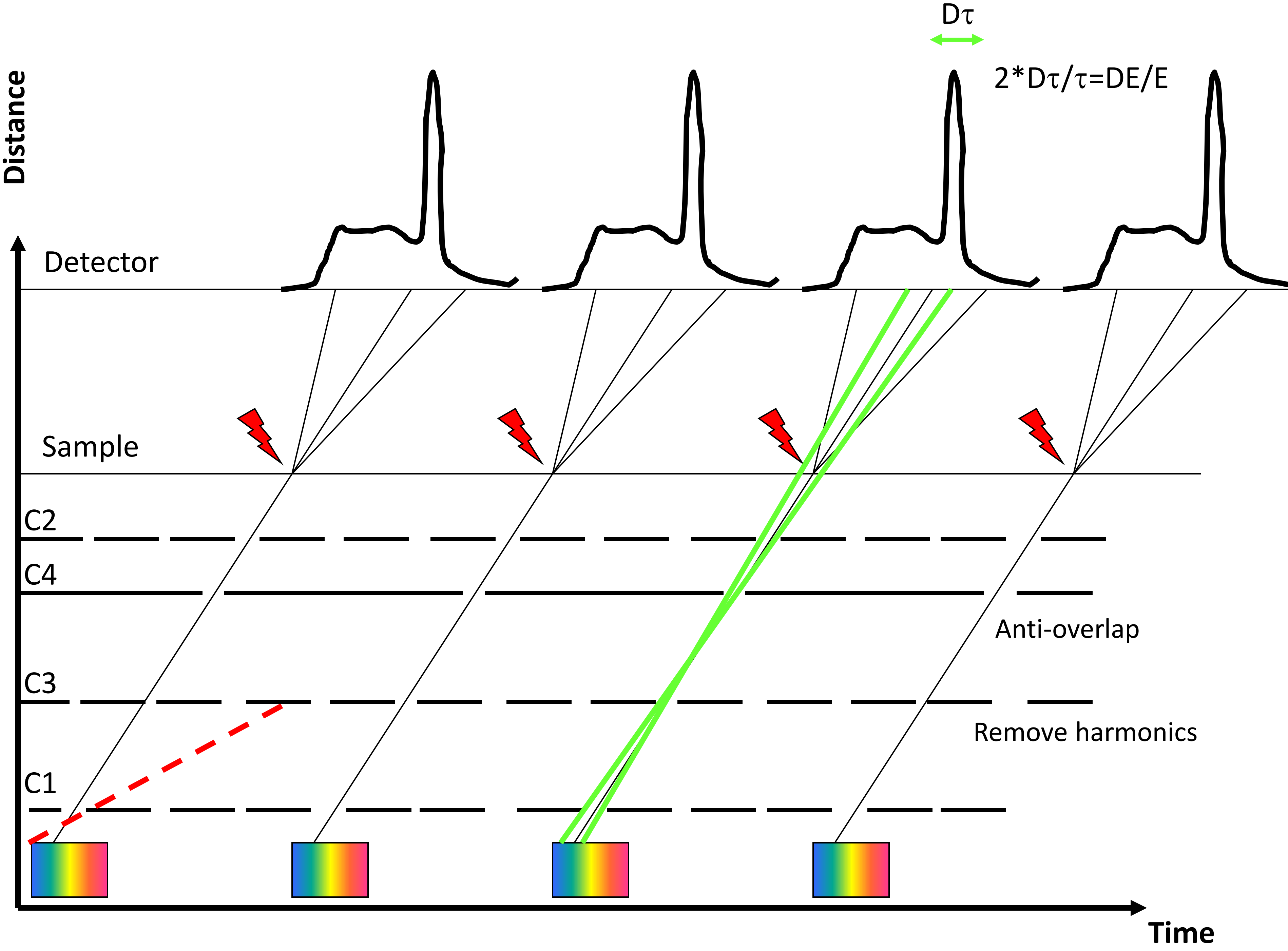
1- Avoid frame overlap



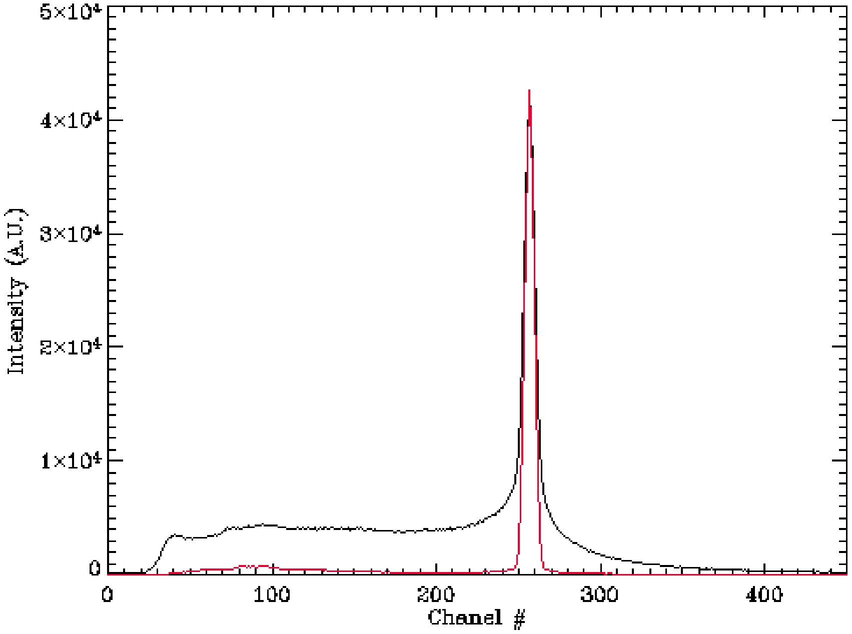

Distance
Detector
Sample
C2
C4
C1
Time

2- Avoid any harmonics

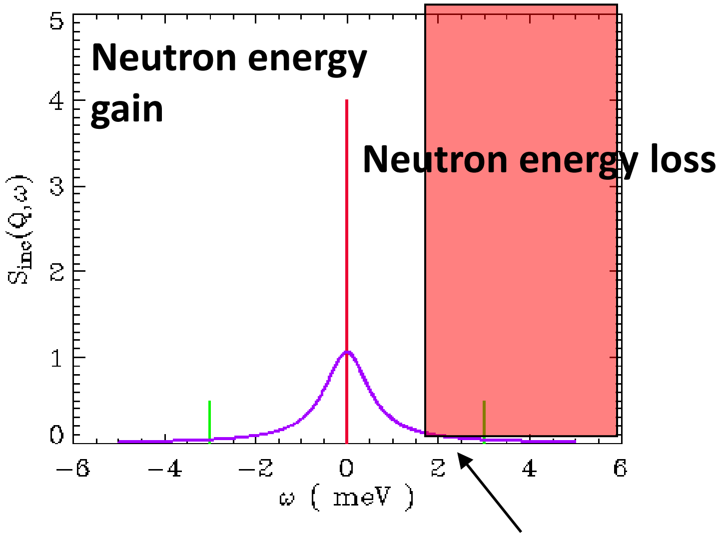



Dτ
2*Dτ/τ=DE/E
Distance
Detector
Sample
C2
C4
Anti-overlap
C3
Remove harmonics
C1
Time

Time-of-flight : Theory vs reality



$* \ 1/ \ t^4$



Neutron energy gain

Neutron energy loss

The maximum energy loss is E_0…

NB: At 298 K, $k_B T$= 25 meV

Conversion from $(2\theta, t_{time\ of\ flight})$ to (Q, ω)

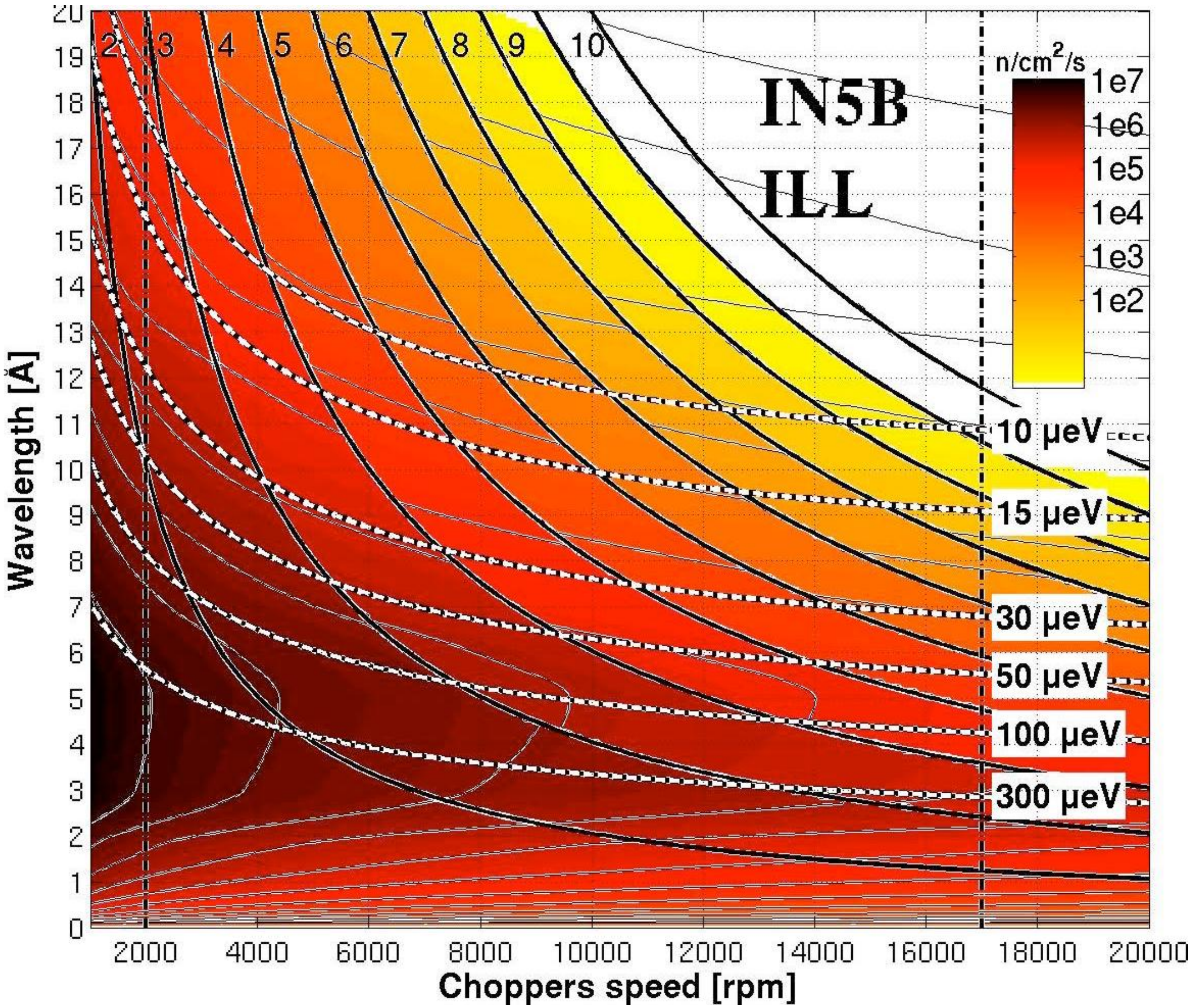
To measure long correlation times :

Increase the chopper speed and/or I_0 .

But flux drops !

$$S^{Mesuré}(Q,\omega) = S^{Théo}(Q,\omega) \otimes R(\omega)$$

Find a good compromise between flux, energy resolution and wavelength



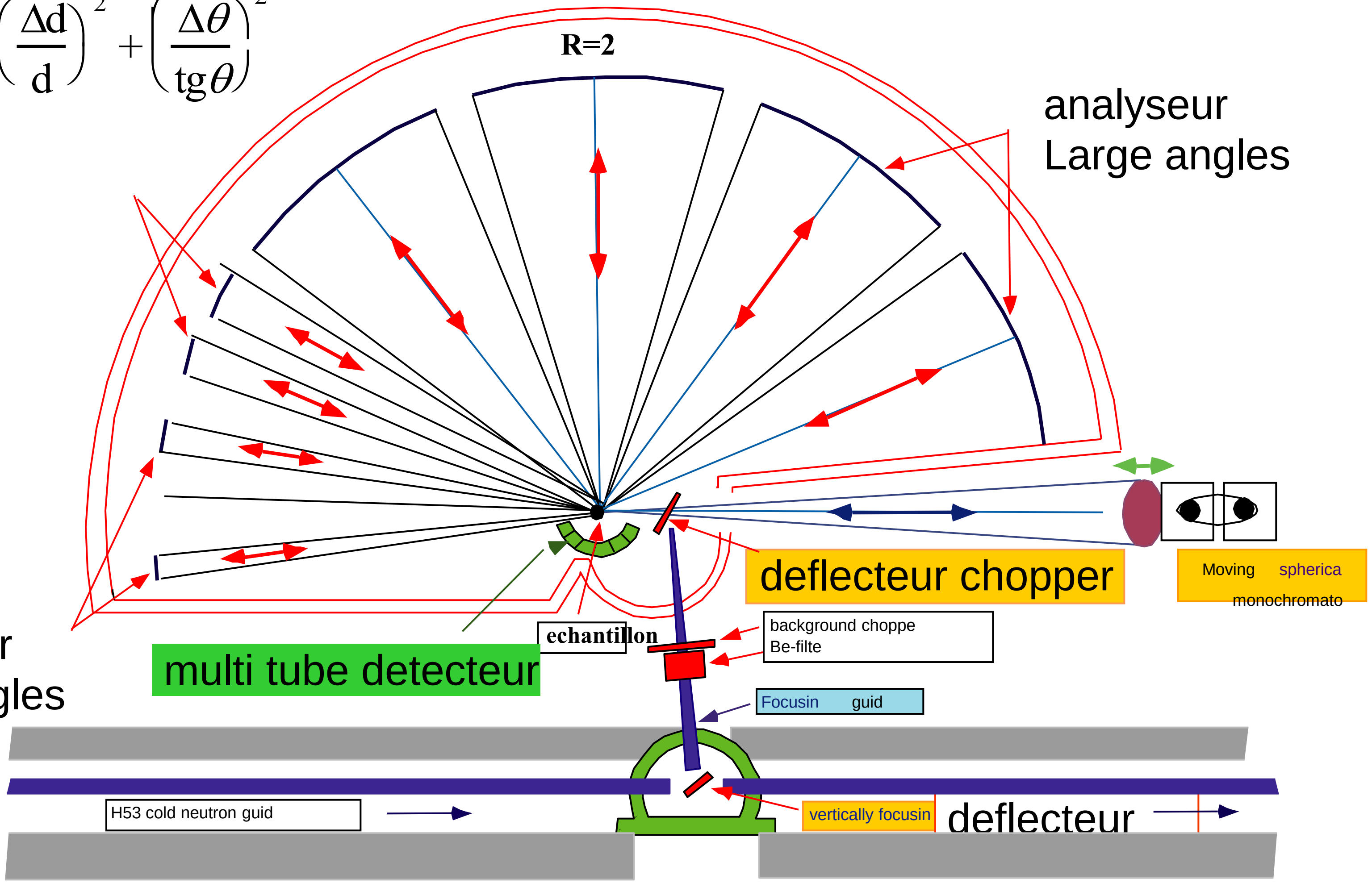
Backscattering Spectrometers

B. Frick, IN16, ILL

monochromator and analyseur : perfect crystals in backscattering (2q=180°)

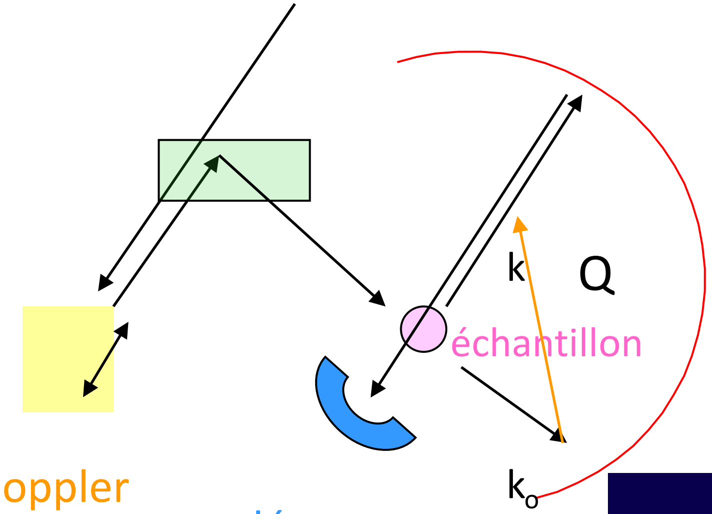
Bragg law differentiation

$$\left(\frac{\Delta\lambda}{\lambda}\right)^2 = \left(\frac{\Delta d}{d}\right)^2 + \left(\frac{\Delta\theta}{tg\theta}\right)^2$$



analyseur
small angles

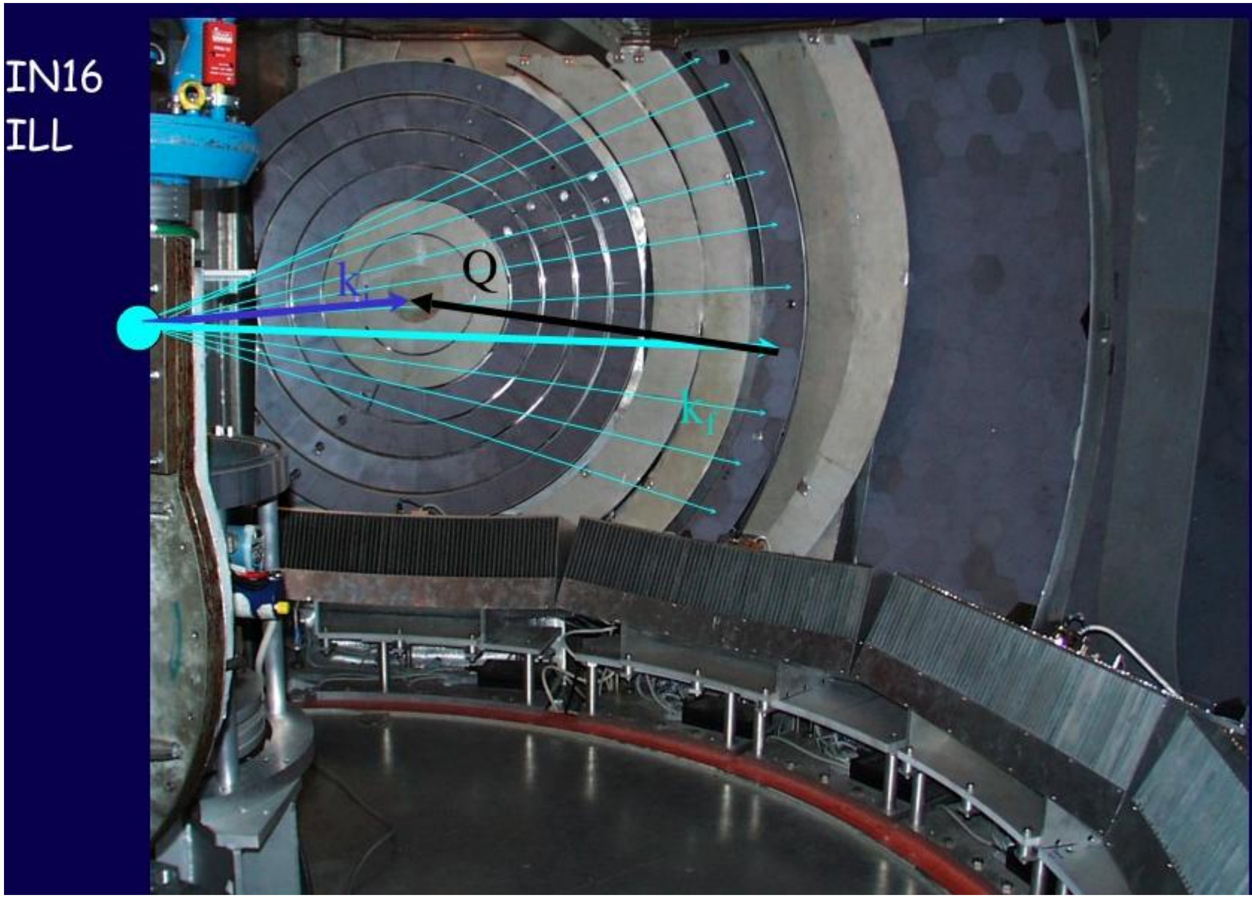
Backscattering Spectrometers



analyseurs

doppler

détecteurs



Q=Qélastique

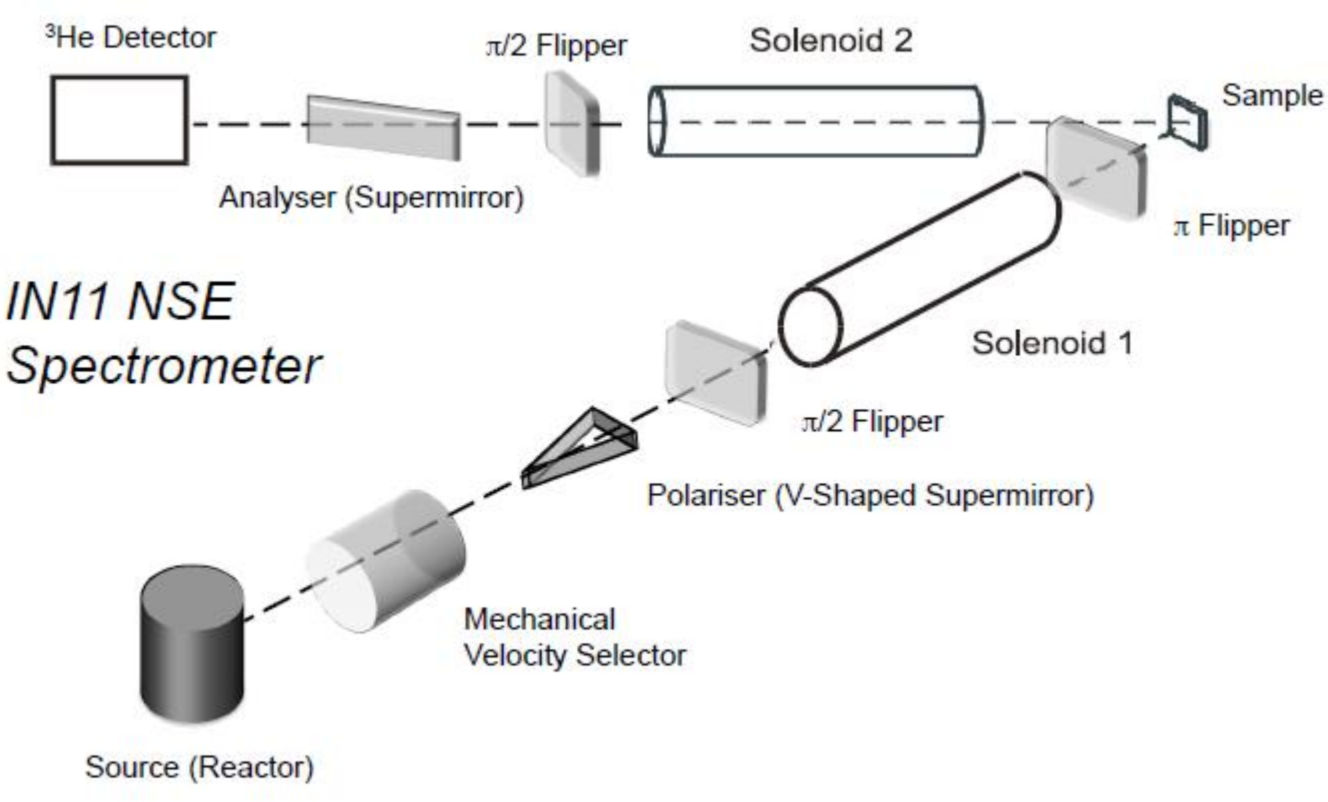
Résolution <1µeV
Gamme -15 à+15µeV

BASIS @SNS ± 300 µeV

Spin Echo Spectrometers

Mezei 1972 NSE, Gähler 1987 NRSE

NSE measures the sample dynamics in the time domain, via the determination of the intermediate scattering functions F(Q,t)

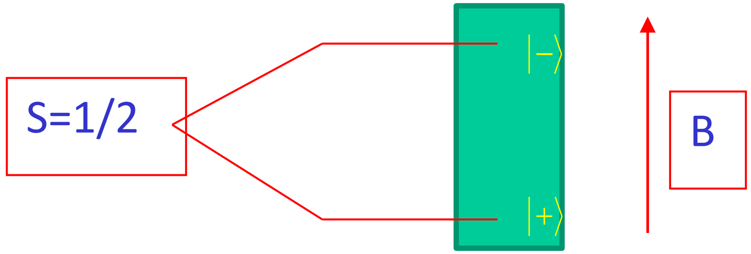


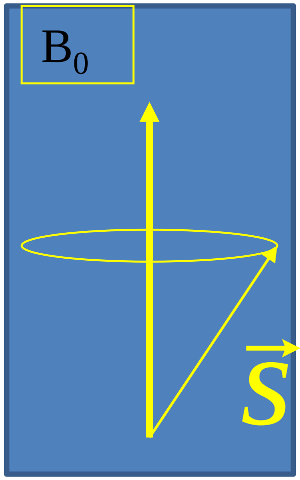
ILL, IN11A

Neutron are polarised, pi/E rotation, then under magnetic field

The measured quantity : the scattered beam polarisation...

Neutron spin , precession ...



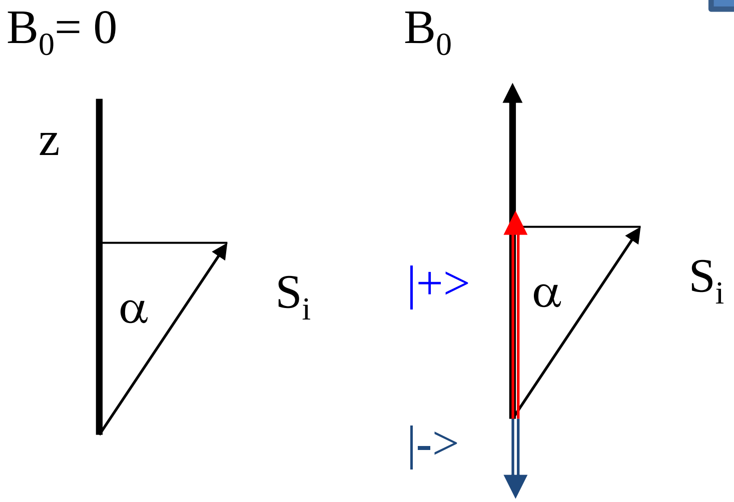


$$\frac{d\vec{s}}{dt} = \gamma\vec{s} \wedge \vec{B}_0$$

$$\omega = |\gamma_n| B_o$$

$$N = \gamma_n B_o t / 2\pi$$

$$\gamma_n = -2913.2\pi\ (Gs)^{-1}$$

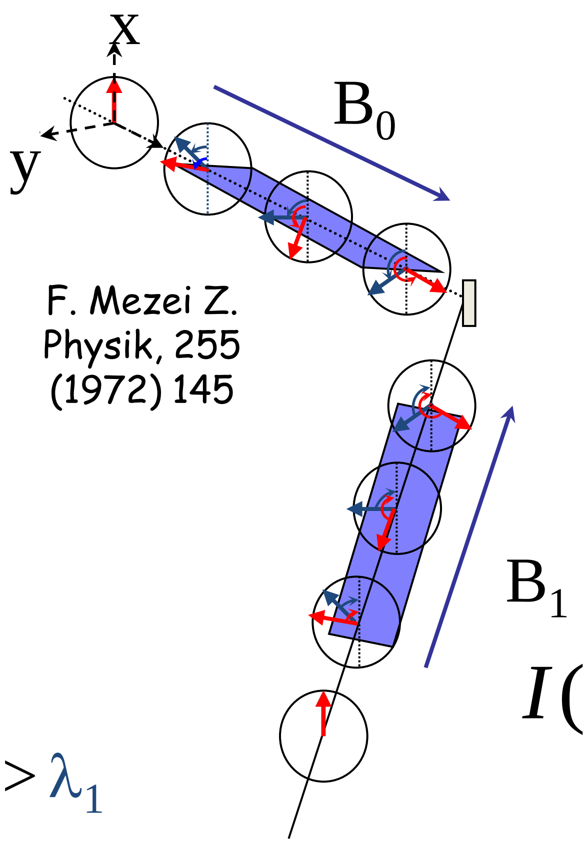


$p_{|+>} = \cos^2(\alpha/2)$

$p_{|->} = \sin^2(\alpha/2)$

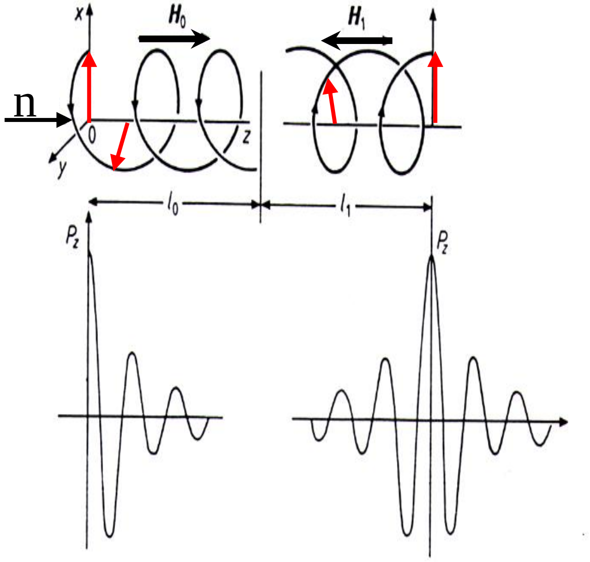
$$\langle P \rangle = \frac{n_+ - n_-}{n_+ + n_-} = \langle p_{|+\rangle} \rangle - \langle p_{|-\rangle} \rangle = \left\langle \cos^2(\frac{\alpha}{2}) \right\rangle - \left\langle \sin^2(\frac{\alpha}{2}) \right\rangle = \langle \cos(\alpha) \rangle$$

to increase the energy resolution without drastic lost of intensity as in QENS-ToF energy transfer *is causing a phase shift of the neutron spin precession angle for* each scattered neutron.

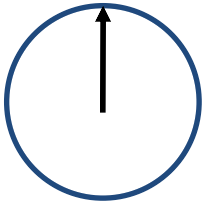


F. Mezei Z. Physik, 255 (1972) 145

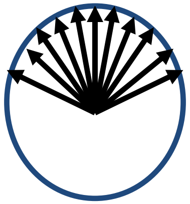
$\lambda_2 > \lambda_1$



elastic



Quasi-elastic



$$I(Q,t) \approx \langle P \rangle \approx \int_{-\infty}^{+\infty} S(Q,\omega)\cos(\omega\tau_{NSE})\,d\omega$$

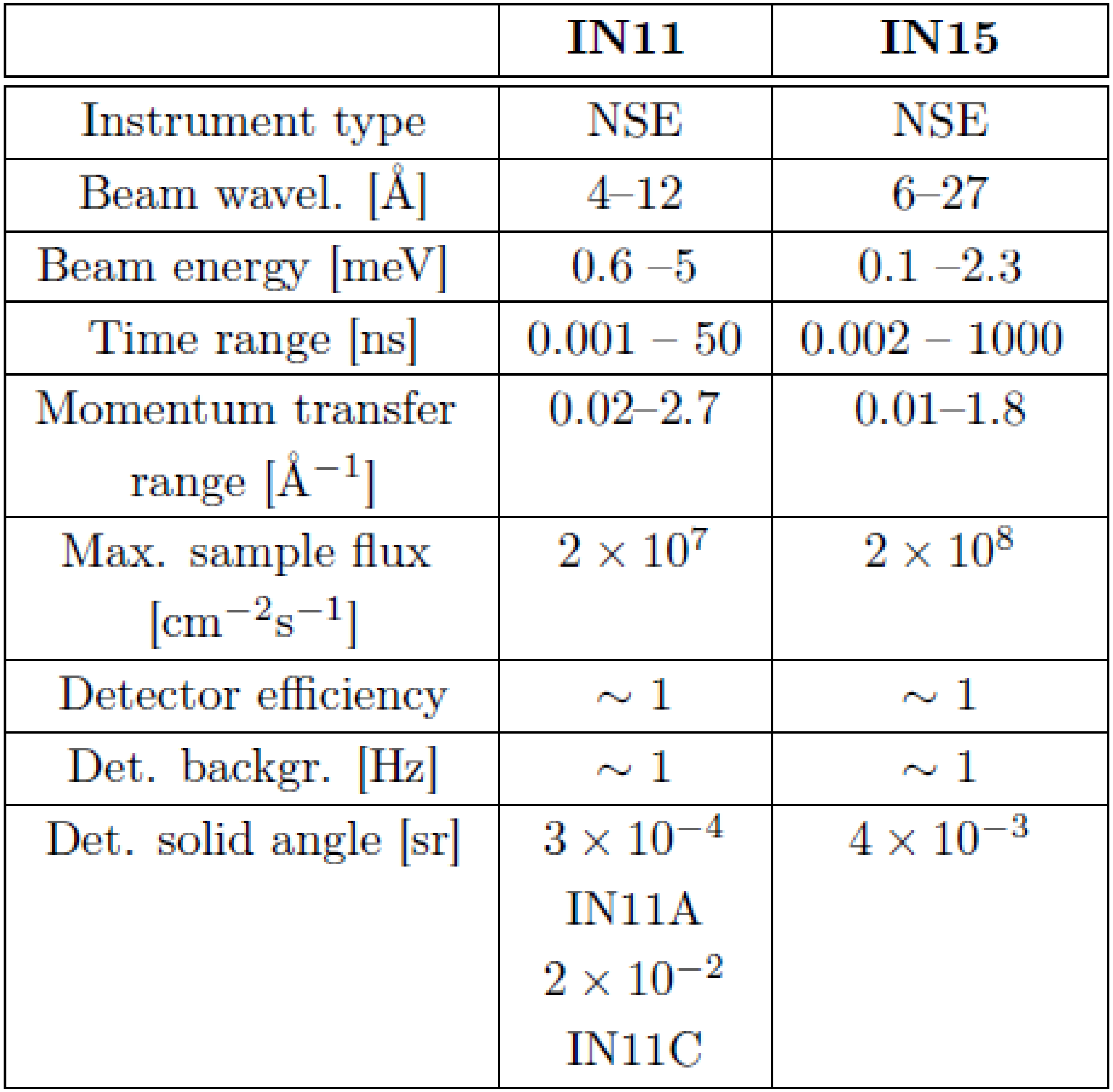
Polarisation

Probability of energy exchange

$= S_{coh}-1/3S_{inc}$ ➔ Attention aux systèm

$$\tau_{NSE} = \frac{m^2\gamma\int B dz}{2\pi h^2}(\lambda_0')^3$$

Spin Echo Spectrometers



	IN11	**IN15**
Instrument type	NSE	NSE
Beam wavel. [Å]	4–12	6–27
Beam energy [meV]	0.6 –5	0.1 –2.3
Time range [ns]	0.001 – 50	0.002 – 1000
Momentum transfer range [Å^{-1}]	0.02–2.7	0.01–1.8
Max. sample flux [cm^{-2}s^{-1}]	2×10^7	2×10^8
Detector efficiency	~ 1	~ 1
Det. backgr. [Hz]	~ 1	~ 1
Det. solid angle [sr]	3×10^{-4} IN11A 2×10^{-2} IN11C	4×10^{-3}

Comparaison TOF-BS et NSE

- Frequence measurement
- Scan at fixed ω
- $S(Q,\omega)=S(Q,\omega)\otimes R(Q,\omega)$
- Self motions
- Shorter dynamical range
- Good Q resolution
- Excitations, vibrations

- Time measurement
- Scan at fixed t
- $S(Q,t)= S(Q,t).R(Q,t)$
- Collective motions
- Large dynamical range (3 to 4 décades)
- $\Delta\lambda/\lambda \sim 15\% = \Delta Q/Q$

Laboratoire Léon Brillouin / Orphée

JOURNAL OF POLYMER SCIENCE **Polymer Physics** WWW.POLYMERPHYSICS.ORG

Recent Progress on Polymer Dynamics by Neutron Scattering: From Simple Polymers to Complex Materials

Juan Colmenero,[1,2] Arantxa Arbe[1]

TABLE 2 Currently operating high-level neutron facilities in the world and the available QENS spectrometers on them

Facility	Instrument	Type
Institute Laue-Langevin (ILL)	IN10	BS
Grenoble, France	IN16	BS
www.ill.eu	IN16b[a]	BS
	IN13[b]	BS
	IN5	ToF
	IN6	ToF
	IN11	NSE
ISIS Facility, Rutherford Appleton Laboratory		
Oxford, United Kingdom		
www.isis.stfc.ac.uk		
Maier-Leibnitz Zentrum (MLZ)		
Garching, Germany		
www.frm2.tum.de	J-NSE	NSE
& www.fz-juelich.de/jcns	RESEDA	NSE
Helmholtz Zentrum Berlin (HZB)	NEAT[a]	ToF
Berlin, Germany		
www.helmholtz-berlin.de		
Paul Scherrer Institute (PSI)	MARS	BS-ToF
Villigen, Switzerland	FOCUS	ToF
sinq.web.psi.ch		
Laboratoire Léon-Brillouin (LLB)	FA#[c]	ToF
Saclay, France	MUSES	NSE
www-llb.cea.fr		
Spallation Neutron Source (SNS)	BASIS	BS-ToF
Oak Ridge, USA	CNCS	ToF
Sydney, Australia		
www.ansto.gov.au		

Quasielastic Neutron Scattering in Biology Part I: Methods

Ruep E. Lechner[1] and Stéphane Longeville[2]

[1]Hahn-Meitner-Institut Berlin, Glienicker Strasse 100, D-14109 Berlin, Germany, and [2]Laboratoire Léon Brillouin, CEA Saclay, F-91191 Gif-sur-Yvette, France