Workshop “Advanced Ideas and Experiments for DNS-IV”
6 – 8 December, 2018, Dubna

Neutron diffraction at DNS-IV

- **A variety of neutron diffractometers**
- **TOF-diffractometers at active pulsed neutron sources**
- **Development trends in TOF-diffractometers design**
- **Diffraction at DNS-IV: basic set and perspectives**

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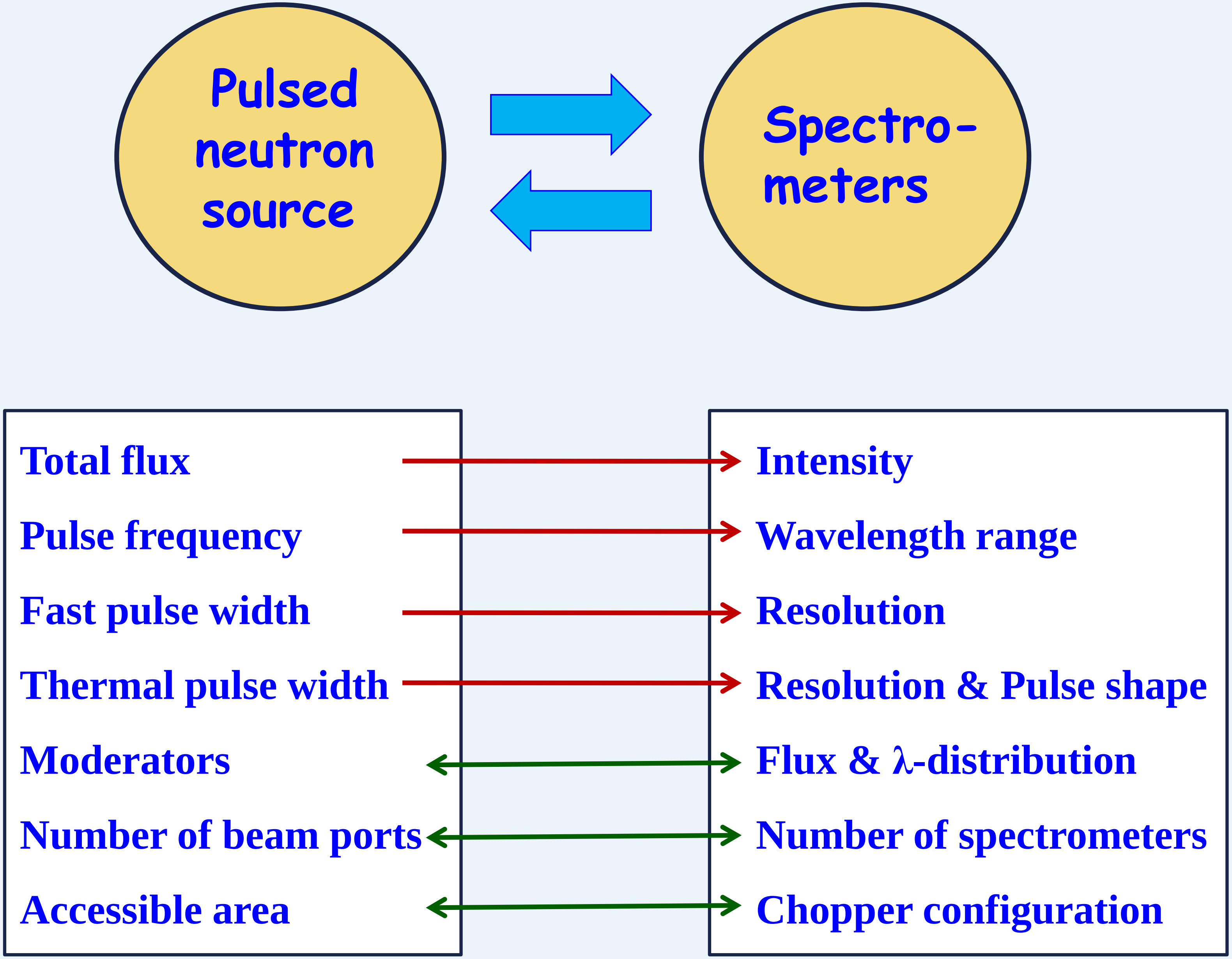

Pulsed neutron source
Spectro-meters
Total flux
Pulse frequency
Fast pulse width
Thermal pulse width
Moderators
Number of beam ports
Accessible area
Intensity
Wavelength range
Resolution
Resolution & Pulse shape
Flux & λ-distribution
Number of spectrometers
Chopper configuration

High-resolution neutron powder diffraction: $(\Delta d/d)_{min} \leq 0.002$

Continuous Sources

W = 10 – 100 MW

$\Delta t_0 = \infty$

λ = const diffractometer
$2\theta_m \geq 120°$
$\alpha_1 \approx \alpha_2 \approx 10'$

D2B (ILL), 0.0005
HRPT (PSI), 0.0009
…

Short Pulse Sources

W = 0.1 – 1.5 MW

$\Delta t_0 \approx (15 - 30)\cdot\lambda$ μs

TOF diffractometer
$L \geq 60$ m
$2\theta \approx 160°$

HRPD (RAL), 0.0005
PG3 (SNS), 0.001
…

Long Pulse Sources

W = 2 – 5 MW

$\Delta t_0 \approx (300 - 3000)$ μs

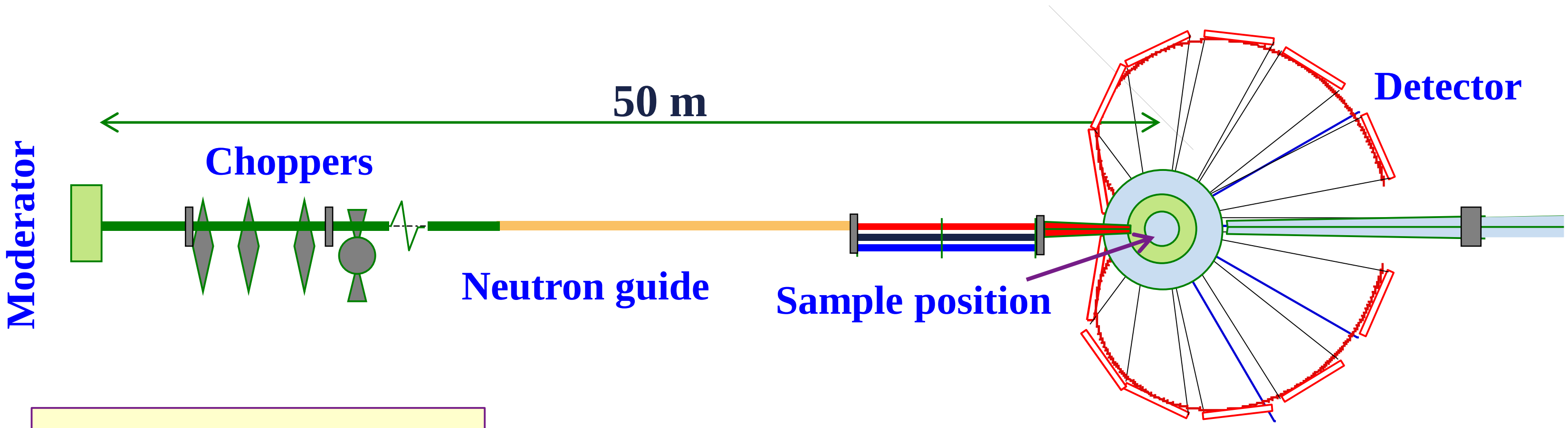
TOF-chopper (Fourier / Fermi) diffractometer
L = 20 - 200 m
$2\theta \approx 160°$

HRFD (FLNP), 0.0005
HEIMDAL (ESS), 0.0006
…

Acting (6) and expected (3) pulsed neutron sources

	Source	Country	Since	W, MW	Φ_0, 10^{13}	Δt_f	Δt_0	ν_0, Hz
1	IBR-2	Russia	1984	2	0.8	215	350	5
2	ISIS-I	UK	1985	0.2	0.07	1	20	50
3	LANSCE	USA	1985	0.1	0.05	1	20	20
4	SNS-I	USA	2006	1	1	1	20	60
5	J-SNS	Japan	2009	1	1	1	20	25
6	ISIS-II	UK	2011	0.05	0.02	1	60	10
7	CSNS	China	2018	0.1	0.05	1	20	25
8	ESS	Sweden	2019	5	30	2860	~3000	14
9	SNS-II	USA	???	0.5	0.5	1000	~1100	10

TOF diffractometer WISH at ISIS (UK)

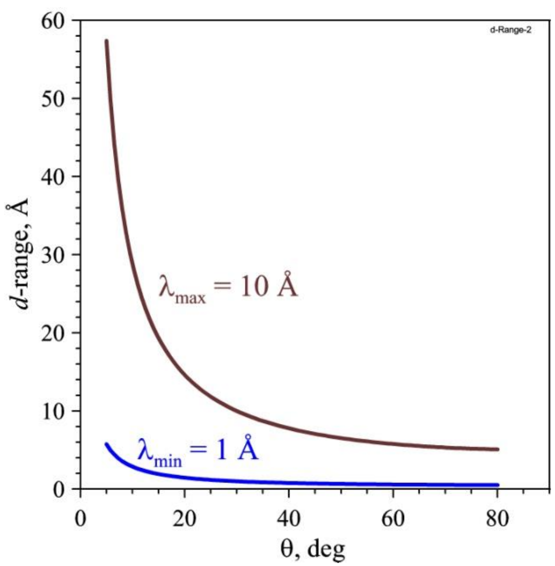


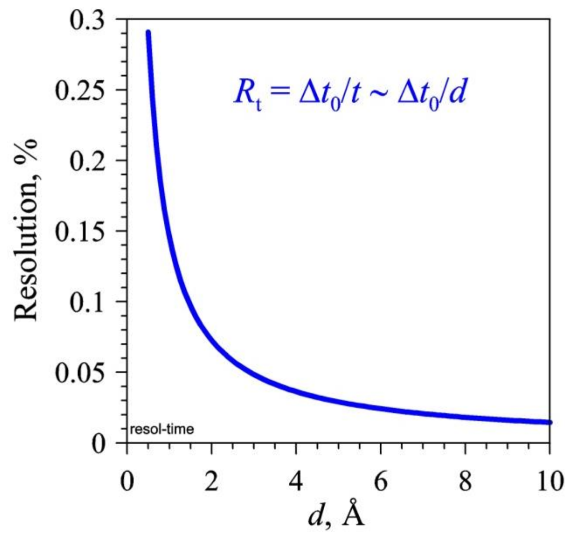
$$d = \lambda/2\sin\theta$$

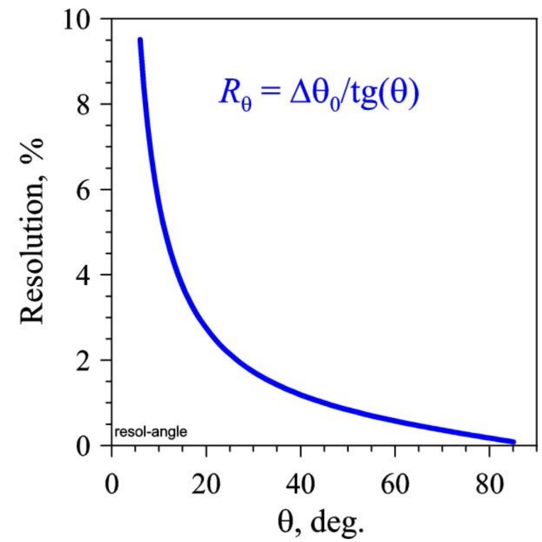
$$d_{min} = \lambda_{min}/2$$

$$d_{max} = \lambda_{max}/2\sin\theta_{min}$$

$$R(t, \theta) = \Delta d/d = [(\Delta t_0/t)^2 + (\Delta\theta/\mathrm{tg}\theta)^2]^{1/2}$$







Specialization of neutron diffractometers

I. Structure: single crystal
2D PSD, $\Delta x < 3$ mm → 4π PSD

II. Structure: polycrystal (powder)
high resolution, $\Delta d/d \approx 0.002$, wide angle PSD

III. Magnetic structure (single- or polycrystal)
medium resolution, $\Delta d/d \approx 0.005$, large (~15 Å) d_{hkl}

IV. *In Situ*, *Real Time* experiment
high intensity (~10^6 н/с), large range of d_{hkl}

V. High pressure, microsamples
high intensity, low background

VI. Long period and macromolecular structures
medium resolution, $\Delta d/d \approx 0.005$, very large (~60 Å) d_{hkl}

VII. Local structure of crystals
high momentum transfer, $Q_{max} \sim 40$ Å^{-1}

VIII. Microstructure of materials (internal stresses, texture, …)
high resolution, $\Delta d/d \approx 0.004$, high intensity

- Intensity
- Pulse width
- λ-range
- λ-distribut.
- Background

TOF-diffractometers at active pulsed sources (~35 in total)

I.	High-pressure	6
II.	Material science	6
III.	Powder, HR	5
IV.	Powder, HI + HR	5
V.	Single-crystal	4
VI.	Macromolecular	3
VII.	Powder, HI	2
VIII.	Magnetic	2
IX.	Texture	1
X.	Nanoscale	1
XI.	Stress	1

Mandatory for use (I.–V.)

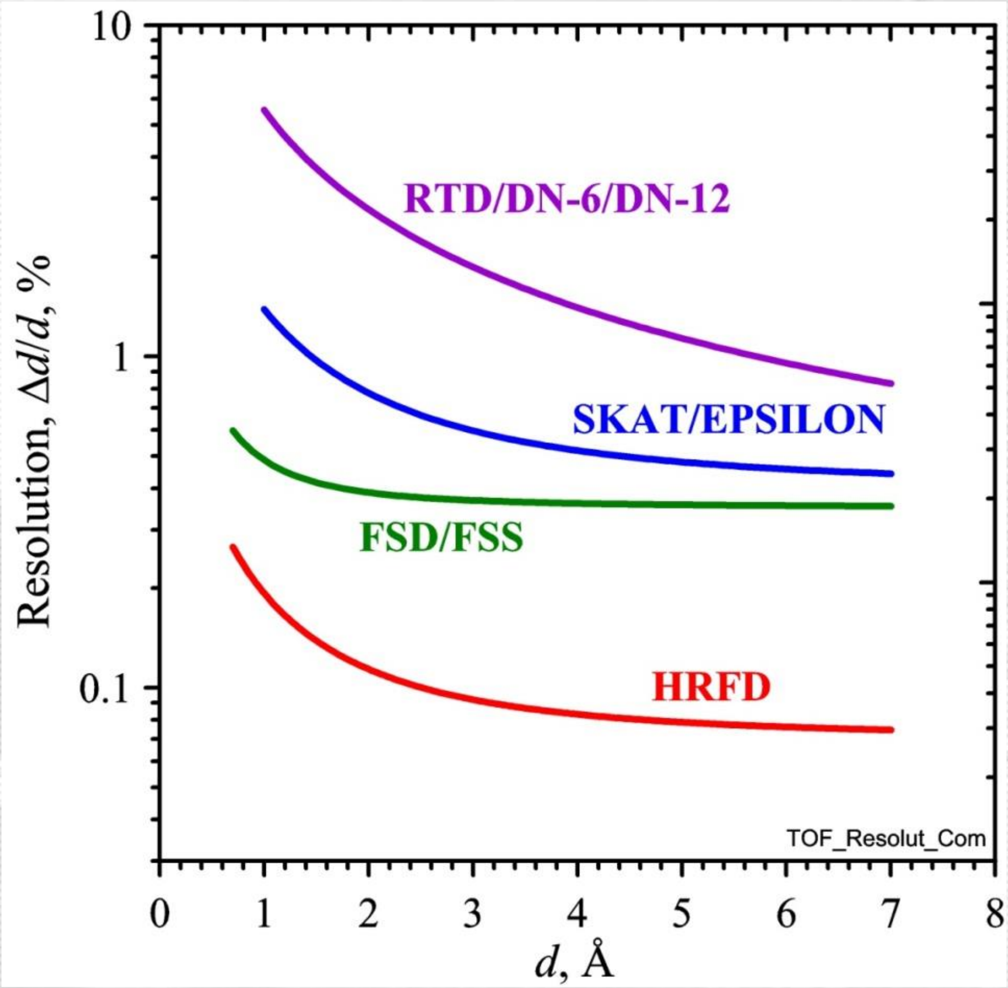
Diffraction at IBR-2

1. **HRFD*** **powders – atomic and magnetic structure**
2. **RTD** **powders, single crystals – real-time, *in situ***
3. **DN-6** **microsamples – high-pressure**
4. **Epsilon**** **rocks, bulk samples – internal stresses**
5. **SKAT**** **rocks, bulk samples – textures**
6. **FSD*** **bulk samples – material science**
7. **DN-12** **microsamples – high-pressure**
8. **FSS*** **bulk samples – internal stresses (setting-up)**

*** Fourier RTOF technique – a special feature of diffraction at the IBR-2 reactor**

**** Long (~100 m) flight pass**

Diffraction at the IBR-2: Resolution

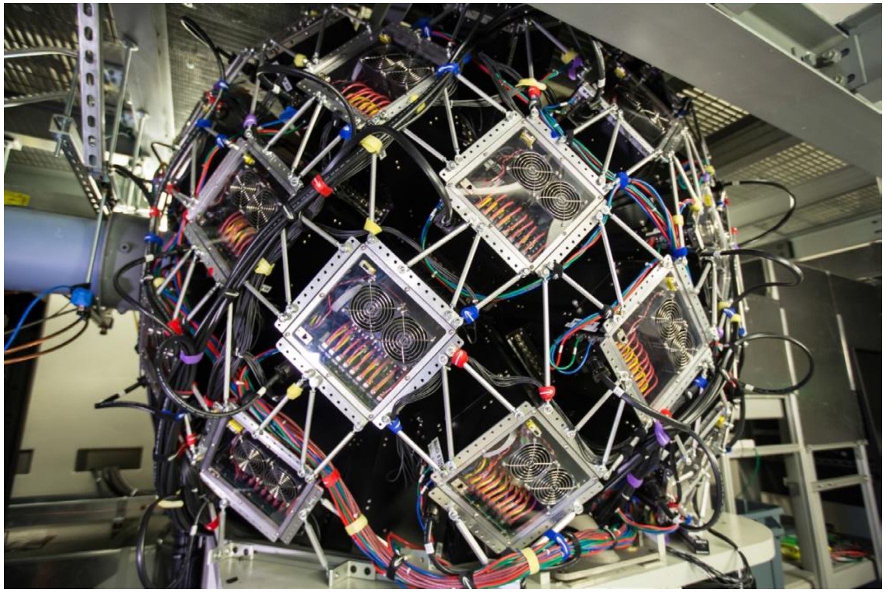


HRFD	**powders**
FSD	**engineering**
RTD	**real-time, multilayers**
DN-6	**high-pressure**
Epsilon	**stresses**
SKAT	**textures**
DN-12	**high-pressure**
FSS	**stresses**

Resolution becomes better for longer *d*-spacing!

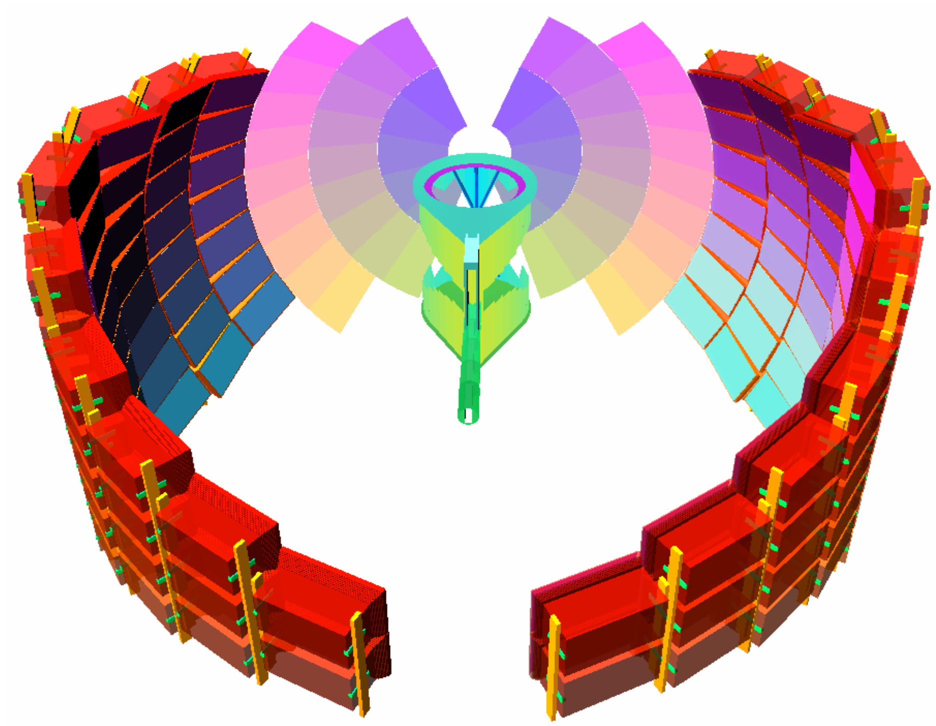
Advanced detectors for TOF diffractometers

MaNDi (SNS), Macromol. single cryst.



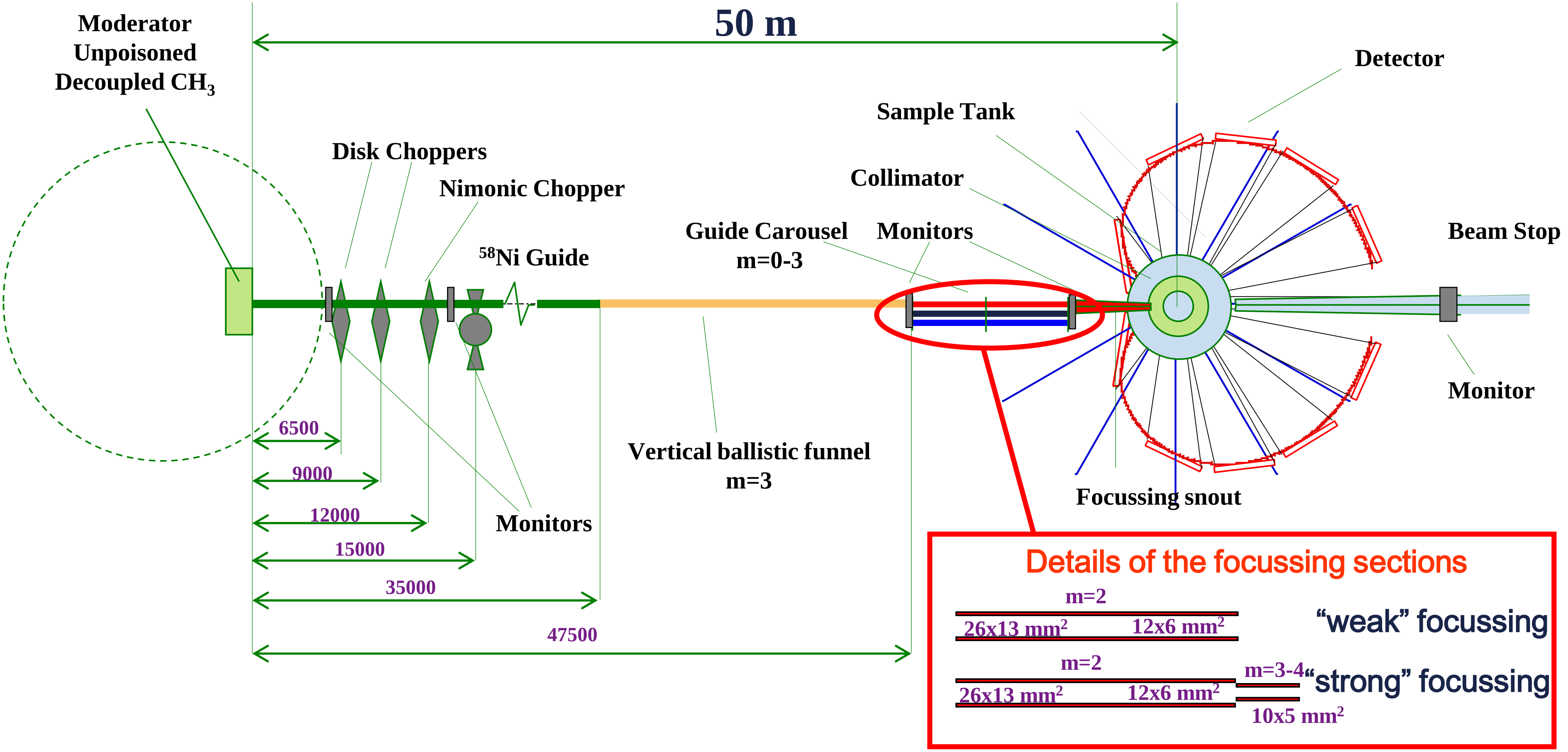
L = 30 m, $\Omega_{det} \approx$ 4.1 sr

Powgen (SNS), HI + HR



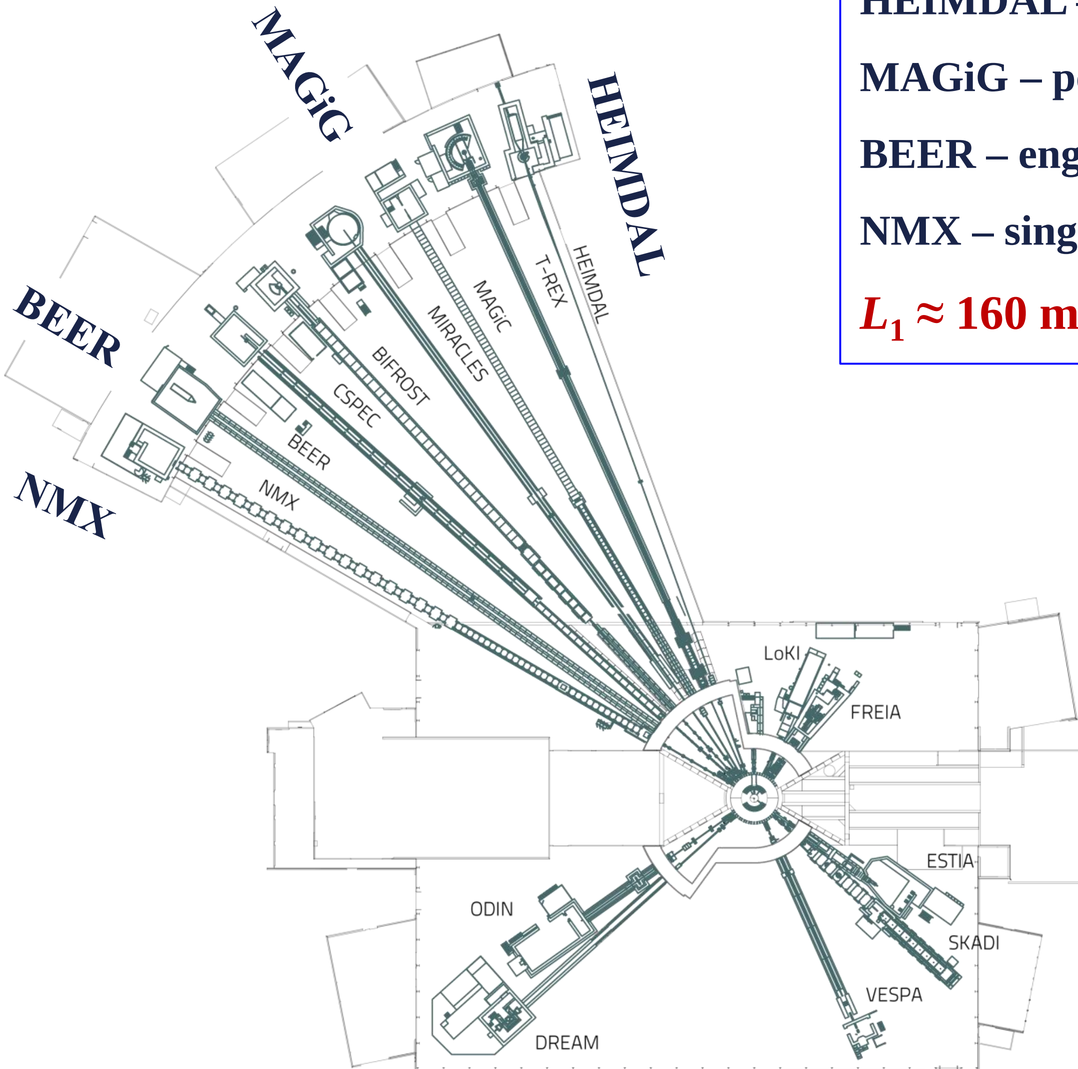
L = 60 m, Ω_{det} = 4.0 sr

Magnetic diffractometer WISH, ISIS, UK



WISH schematic drawing

ESS pulsed neutron sources, ν = 14 Hz, Δt_0 = 2860 μs



HEIMDAL – hybrid, Diff. + SANS + IM

MAGiG – polarized, single crystal

BEER – engineering

NMX – single crystal, macromolecular

$L_1 \approx 160$ m, $\Delta\lambda \approx 1.8$ Å

$\Delta\lambda \approx 282/L$

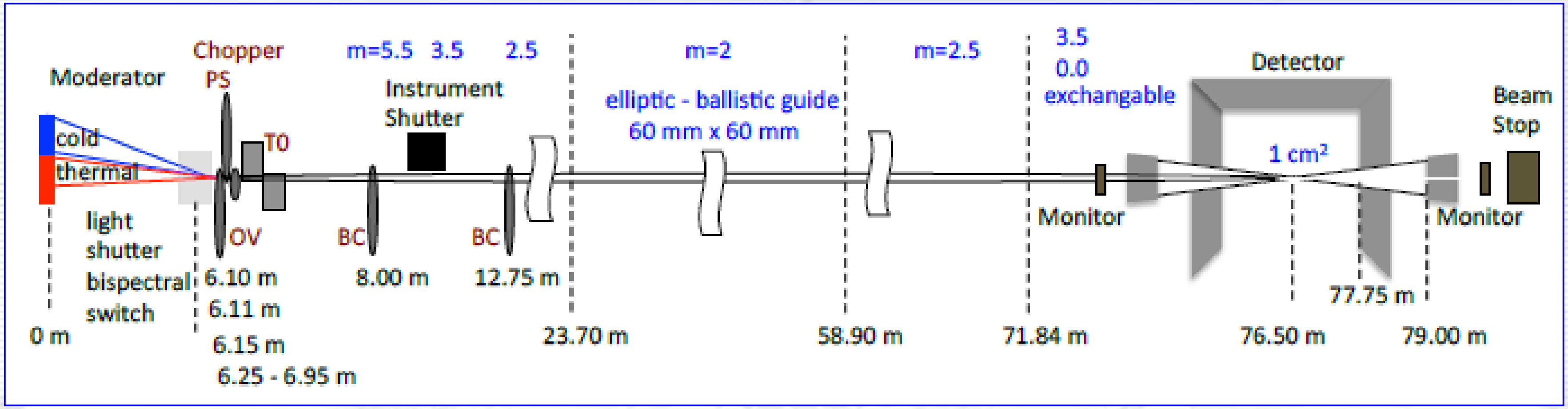
DREAM – powder,
HR + HI, $L_1 = 76$ m, $\Delta\lambda \approx 3.7$ Å

ESS parameters:

Average beam power, MW	5
Peak beam power, MW	125
Proton kinetic energy, GeV	2.0
Pulse repetition rate, Hz	14
Average pulse current, mA	62.5
Macro-pulse length, μs	2860
Number of target stations	1
Number of moderators	2
Number of neutron beam ports	42
Separation between ports degrees	6

HR + HI powder diffractometer DREAM, ESS

(L_1 = 76 m, $\Delta\lambda \approx 3.7$ Å)



<u>DREAM feature</u>: bispectral switch (cold + thermal neutrons)

<u>DREAM choppers</u>: PC – pulse shaping, T0, BC – band control, OV – overlap = 7 ch-s

<u>DREAM costing (kEu)</u>: Design = 1970, Detector + DA = 6620, Optic = 1500,

Choppers = 1120, Shielding = 2120, Infrastr. = 320, … <u>Total</u> = 12 960

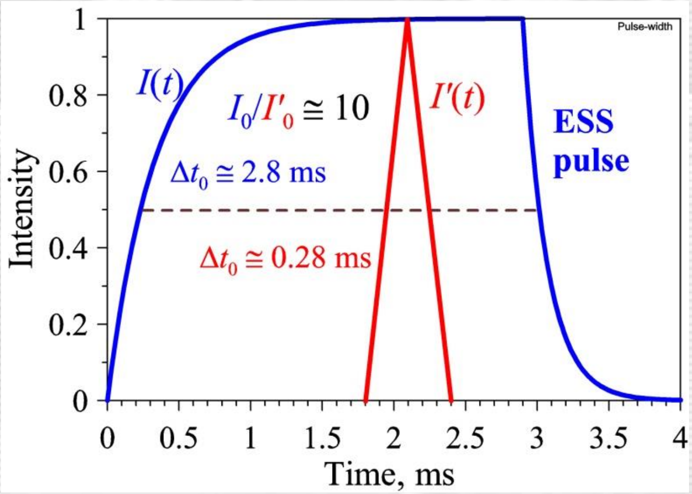
L_1 = 76.5 m, $(\Delta t_0)_{min}$ = 10 μs → $\Delta d \approx 2.8 \cdot 10^{-4}$ Å,

Development trends of TOF-diffractometers at ESS

1. **Bi-spectral extraction:** $(\lambda_1)_{max} \approx 1.2$ Å, $(\lambda_2)_{max} \approx 3$ Å
2. **Very long flight path:** 76 / 160 m
3. **Detector solid angle:** ~4 sr, $\Omega \to 4\pi$ (12 sr)
4. **Combination of diffraction + SANS + imaging**
5. **Focusing on *in situ*, *real-time* mode of data acquisition**
6. **Complicated chopper system:** ~(6 – 11) choppers of different assignments
7. **Extremely high cost:** $(12 \div 20) \cdot 10^6$ Eu

Basic parameters of IBR-2, ESS and DNS-IV (the first stage)

	IBR-2	ESS	DNS-IV
1. Time-average flux density:	$0.08 \cdot 10^{14}$	$3 \cdot 10^{14}$	$2 \cdot 10^{14}$
2. Width of thermal pulse:	~350 μs	3000 μs	~300 μs
3. Pulse repetition rate:	5 Hz	14 Hz	10 Hz
4. Background power:	~7%	<1%	~3.5%
5. Number of beam ports:	14	~40	~25



For TOF-diffractometer: $(\Phi_1/\Phi_2)\cdot(\Delta t_2/\Delta t_1)$

ESS / IBR-2 = $(3\cdot10^{14}/0.08\cdot10^{14})\cdot(350/3000) \approx 4.5$

(without frame multiplication system, $K \sim 2 \div 3$)

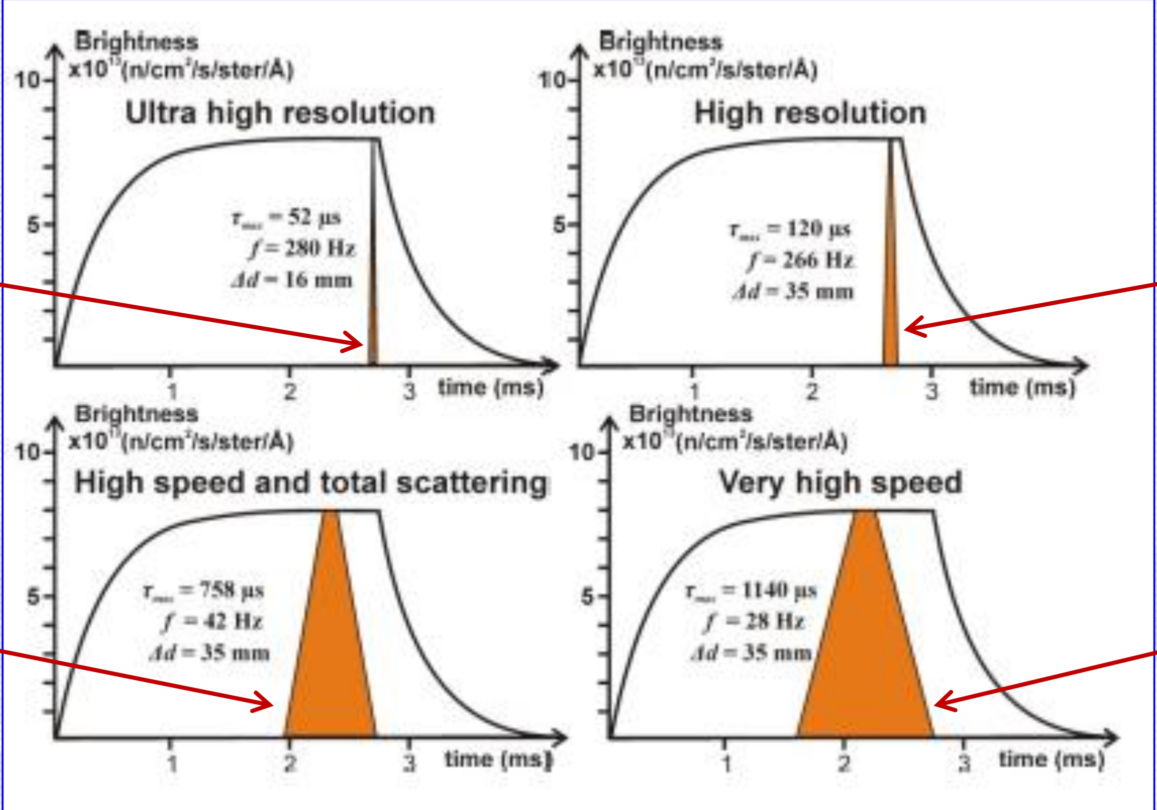
DNS-IV / IBR-2 = $(2\cdot10^{14}/0.08\cdot10^{14})\cdot(350/300) \approx 30$

Hybrid diffractometer HEIMDAL, ESS

(Diffraction + SANS + Imaging, L_1 = 167 m, $\Delta\lambda \approx$ 1.7 Å, $\lambda_{min} \approx$ 0.6 Å)

Δt_0 = 52 µs
f = 280 Hz
Δx = 1.6 cm
R_t = 0.06%

Δt_0 = 120 µs
f = 266 Hz
Δx = 3.5 cm
R_t = 0.14%



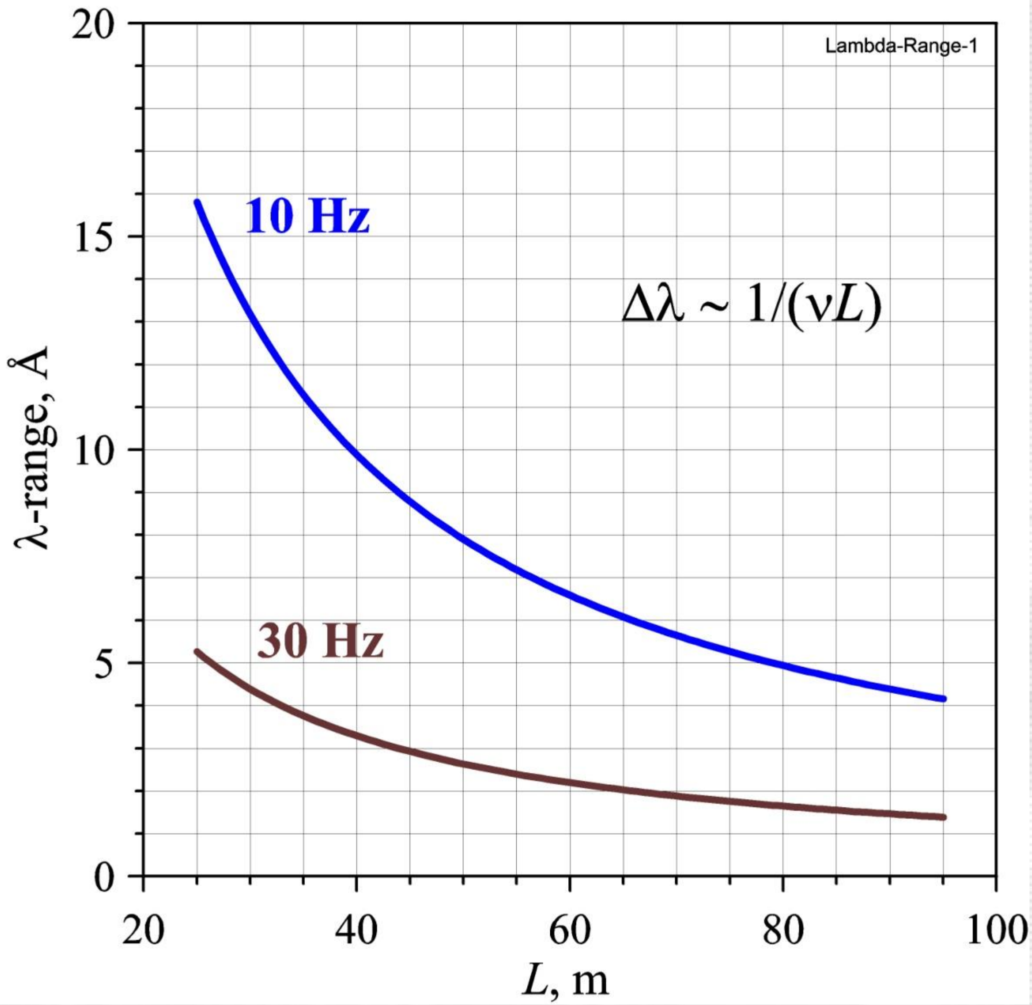
Δt_0 = 758 µs
f = 42 Hz
Δx = 3.5 cm
R_t = 0.87%

Δt_0 = 1140 µs
f = 28 Hz
Δx = 3.5 cm
R_t = 1.3%

Modes of the pulse shaping chopper operation

Flux at a sample: from 3.8x10^6 (HR) to 2.0x10^9 (HI)

Wavelength range as a function of frequency



Fast choppers for TOF neutron diffraction

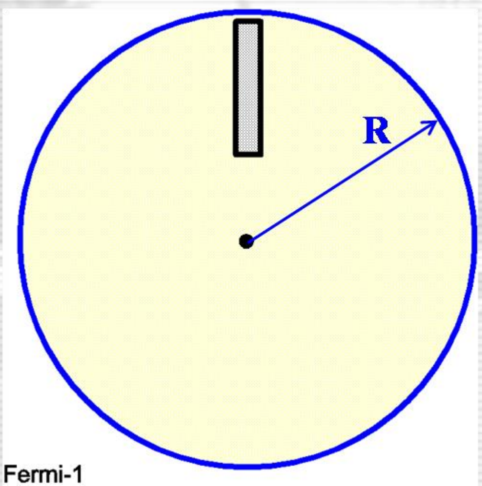
Steady state or long pulsed neutron source

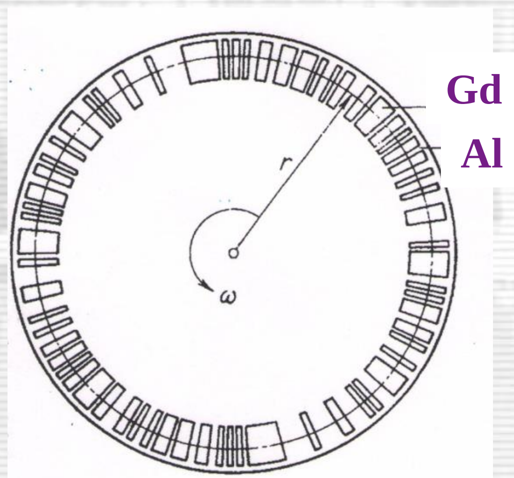
TOF conventional | **TOF correlation**

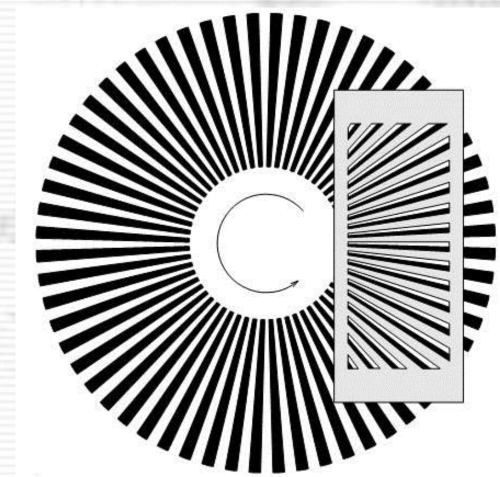
Fermi chopper
$S_{beam} \approx 10$ cm^2, $T \leq 1\%$

Statistical chopper
$S_{beam} \approx 3$ cm^2, $T \approx 25\%$

Fourier chopper
$S_{beam} \approx 30$ cm^2, $T \approx 25\%$







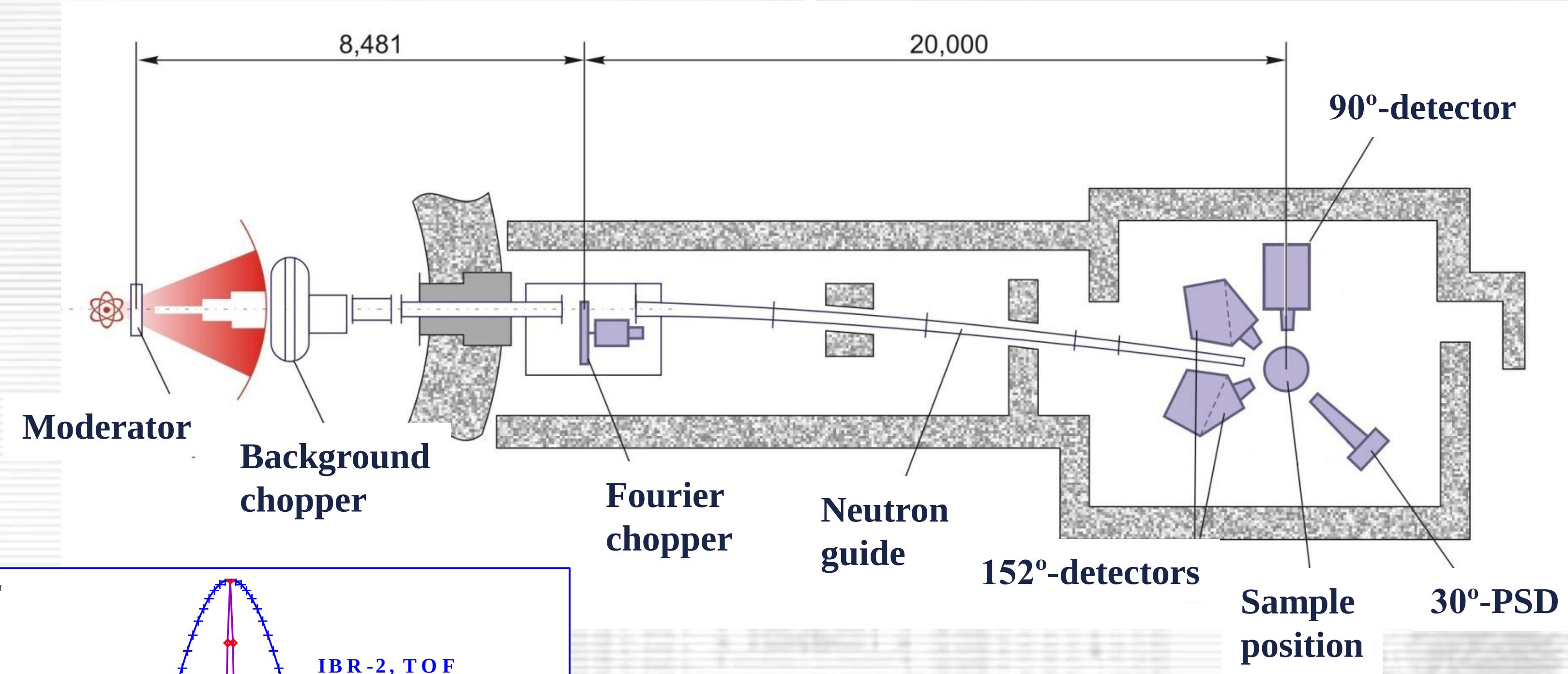
ESS

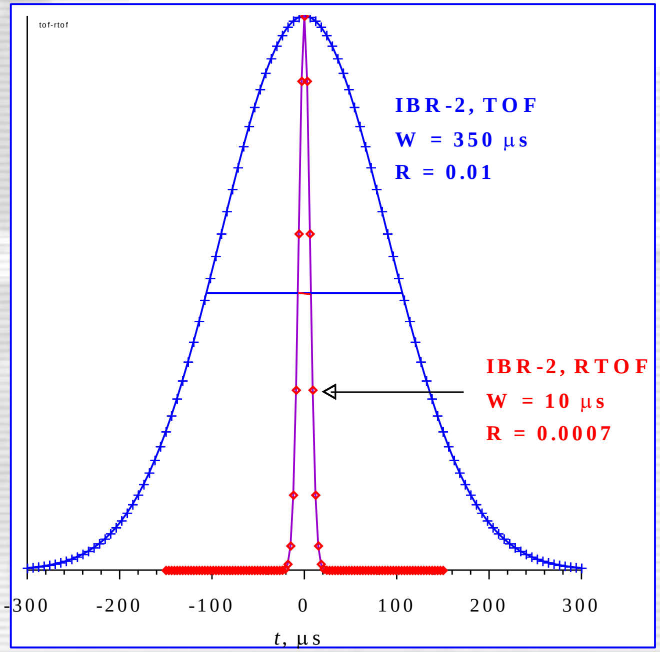
SINQ: POLDI

SNS: CORELLI

IBR-2: HRFD, FSD

Since 1994: High Resolution Fourier Diffractometer at IBR-2





Collaboration:

FLNP (Dubna) - PNPI (Gatchina) - VTT (Espoo)

HRFD (IBR-2): $\Delta t_0 \approx$ Const $\approx$ 10 µs, $L_{tot} \approx 21$ m,

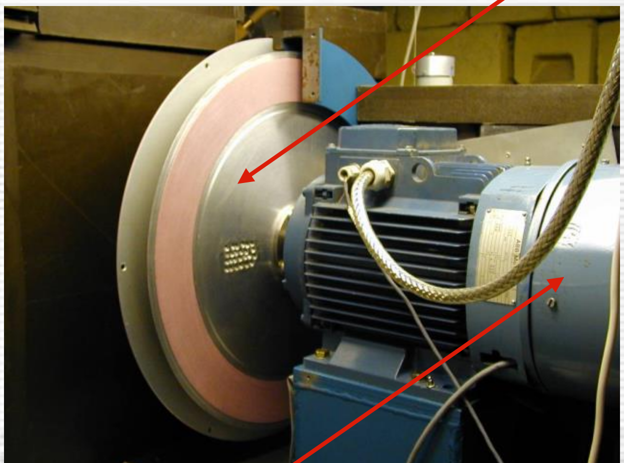
$\Delta t_0/t \approx 1\cdot10^{-3}/d = 5\cdot10^{-4}$ for $d = 2$ Å

Fast Fourier chopper at HRFD (IBR-2)

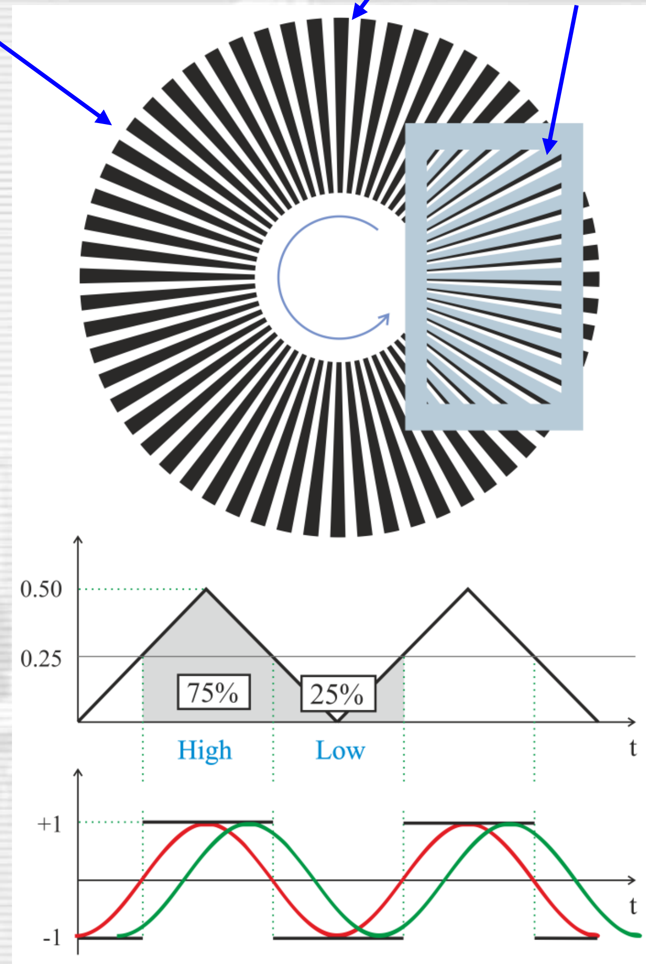
Rotor ($\varnothing$ = 50 cm)

0.7 mm

Stator



7.5 kW motor



Triangular chopper transmission function:
$T(t) \approx 1 + \sin \omega t$

Pick-up signals for RTOF analyzer: binary or sinus-like

<u>Dubna chopper</u>:

Al-alloy

$\varnothing$ = 50 cm

N = 1024

Δx = 0.7 mm

V_{max} = 6000 rpm

Ω = 100 kHz

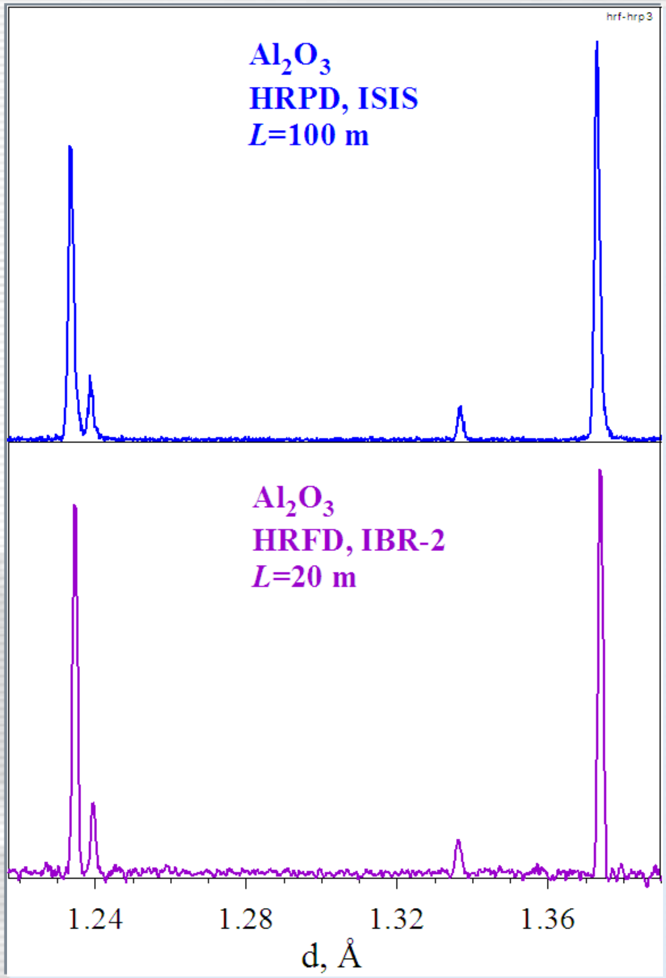
Δt_0 = down to 10 μs

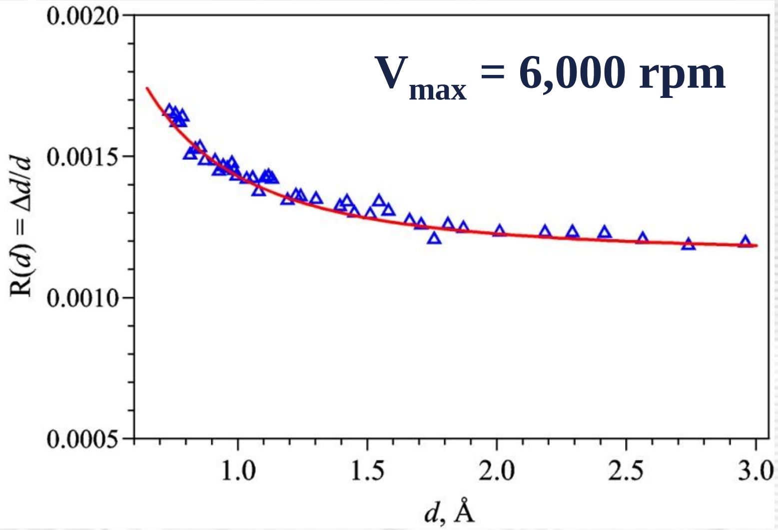
S_{beam} = 3x20 cm^2

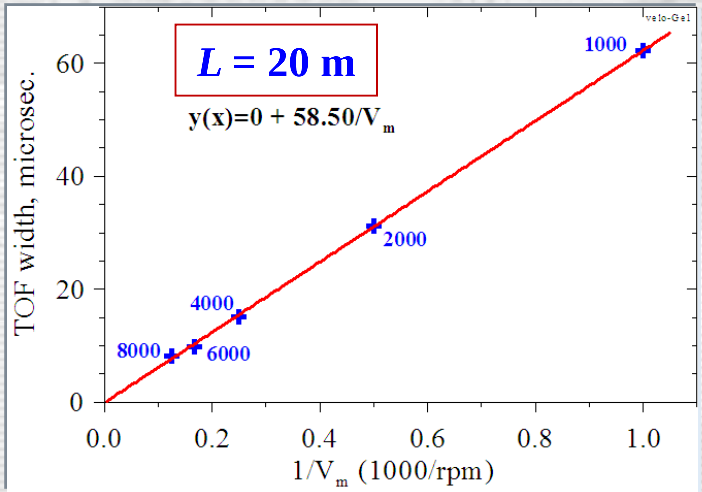
Transmission = 25%

Price = 120 kEu

HRFD resolution



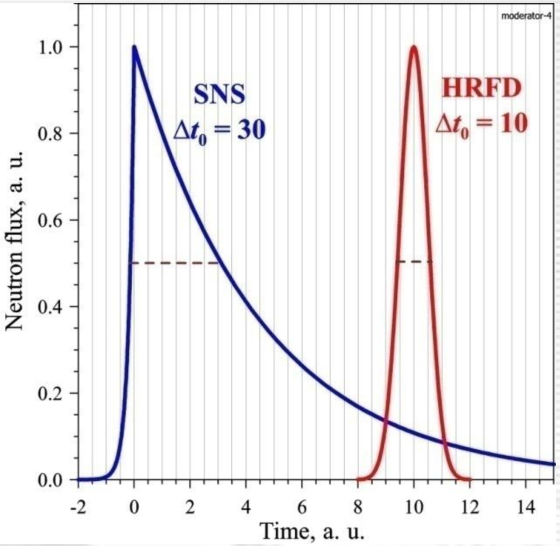




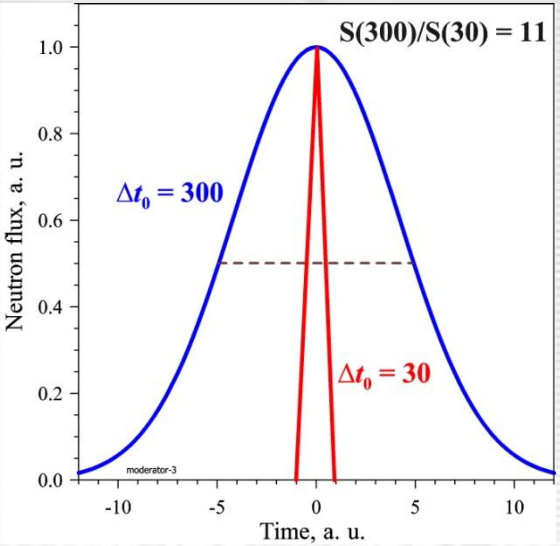
Diffraction patterns of Al_2O_3 measured at ISIS (UK) and IBR-2 (Dubna). Resolution is the same, despite *L* is 5 times longer at ISIS.

For *L*=30 m, V_{max}=11,000 rpm: R_t = 0.0002, $R = (R_t^2 + R_g^2)^{1/2} \approx$ 0.0003

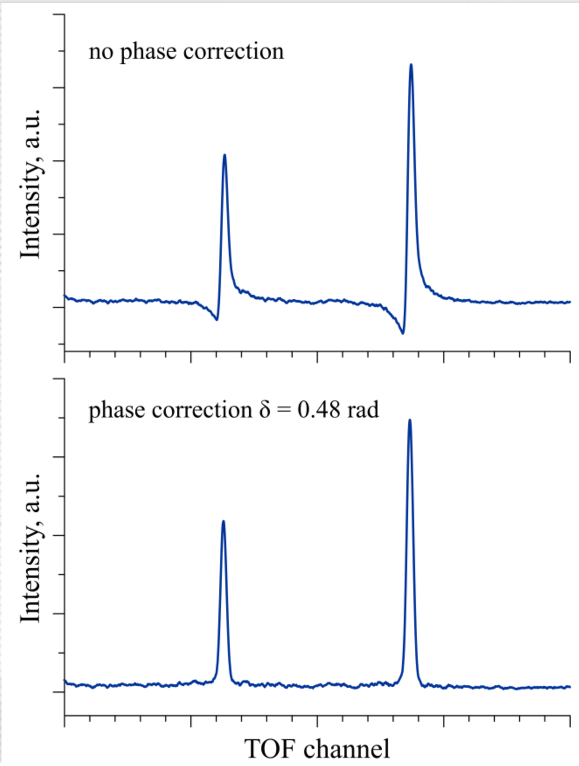
Resolution and shape of diffraction peaks



Comparison of SNS (30 μs) and HRFD (10 μs) peak shape



Pulse shaping 300 → 30 μs



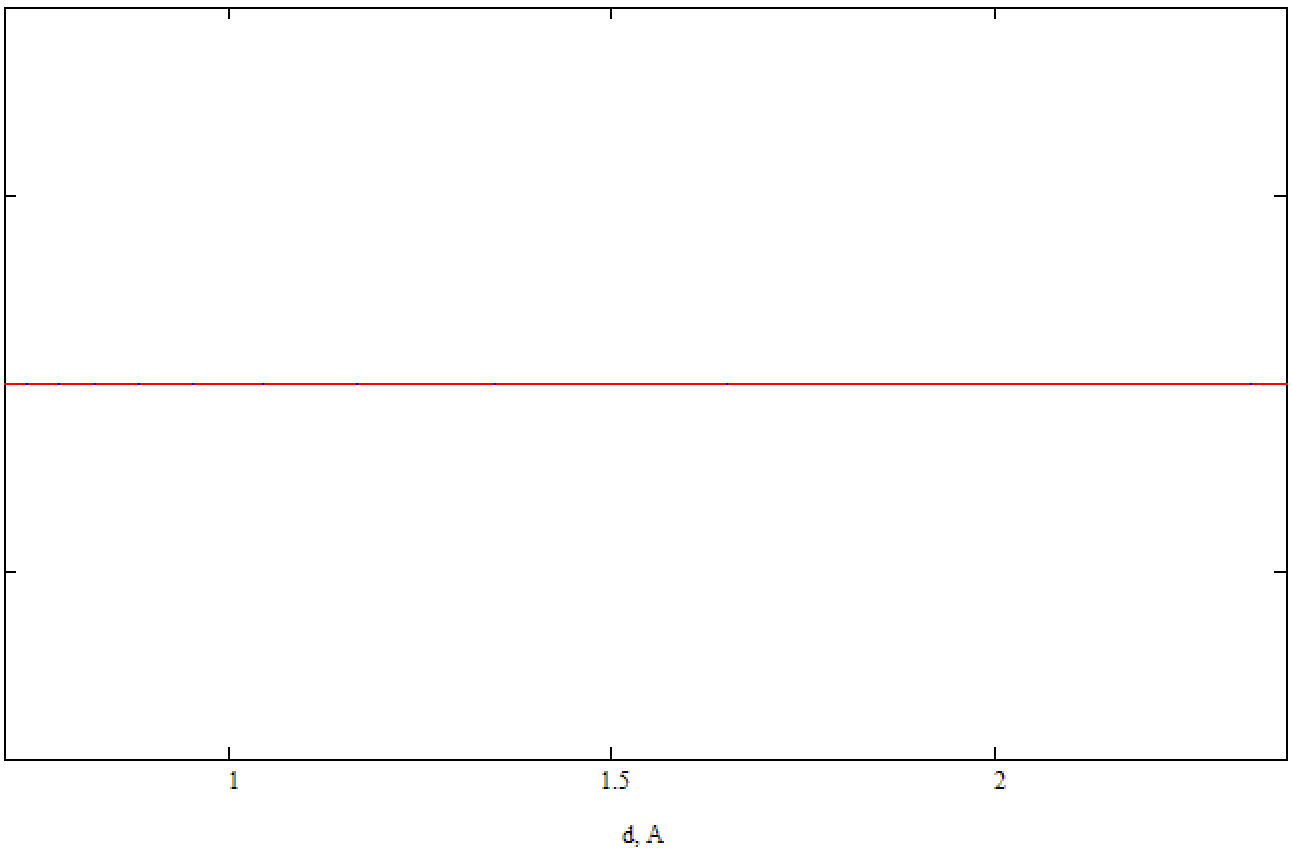
For TOF: $I(300)/I(30) = 11$, Δx = 14 mm, $L_1 = 60$ m, $f = 248$ Hz

For RTOF: $I(300)/I(10) = 5$, Δx = 0.7 mm, $L_1 = 20$ m, $f = 100$ Hz

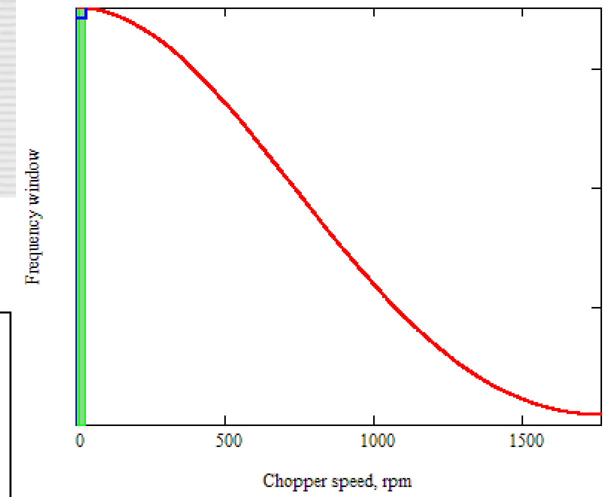
Correction of the peak profile on the phase mismatch.

Simulation: RTOF data acquisition

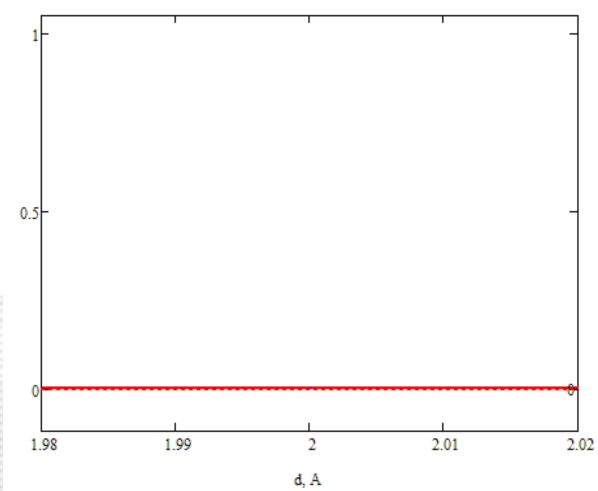
Current chopper speed is 0 rpm or 0 % of max. speed 1760 rpm



Elapsed time / Total time: 0 %



Current chopper speed is 0 rpm or 0 % of max. speed 1760 rpm



Courtesy Gizo Bokuchava

TOF-diffraction: Acceptable characteristics of DNS-IV

1.	Time-average flux density:	$\Phi_0 \sim 2 \cdot 10^{14}$ n/cm^2/s
2.	Half-width of fast neutrons:	$\Delta t_0 \sim 200$ µs
3.	Pulse repetition rate:	ν = 10 Hz
4.	Moderators (at least three):	thermal + cold (~80 K) + very cold (~30 K)
5.	Background power:	< 4%

The basic set of neutron TOF-diffractometers:

	Instrument	Main issue	Moderator	Resolution
1.	Material science*)	internal stresses	300 K	High
2.	High-resolution*)	structure	80 K	High
3.	High-intensity	*in situ*, real-time	80 K	Medium
4.	High-pressure	micro samples	80 K	Medium
5.	Magnetic	structure	30 K	Medium

*) Fourier chopper must be used

TOF-diffraction at DNS-IV: Conclusions

1. **Prospects for TOF neutron diffraction at DNS-IV are looking very promising. The level of the main parameters (intensity, resolution, *d*-spacing range, …) can be extremely high.**

2. **The basic set of diffractometers can include 5 the most called-for instruments, with 3 - 4 additional as the second stage.**

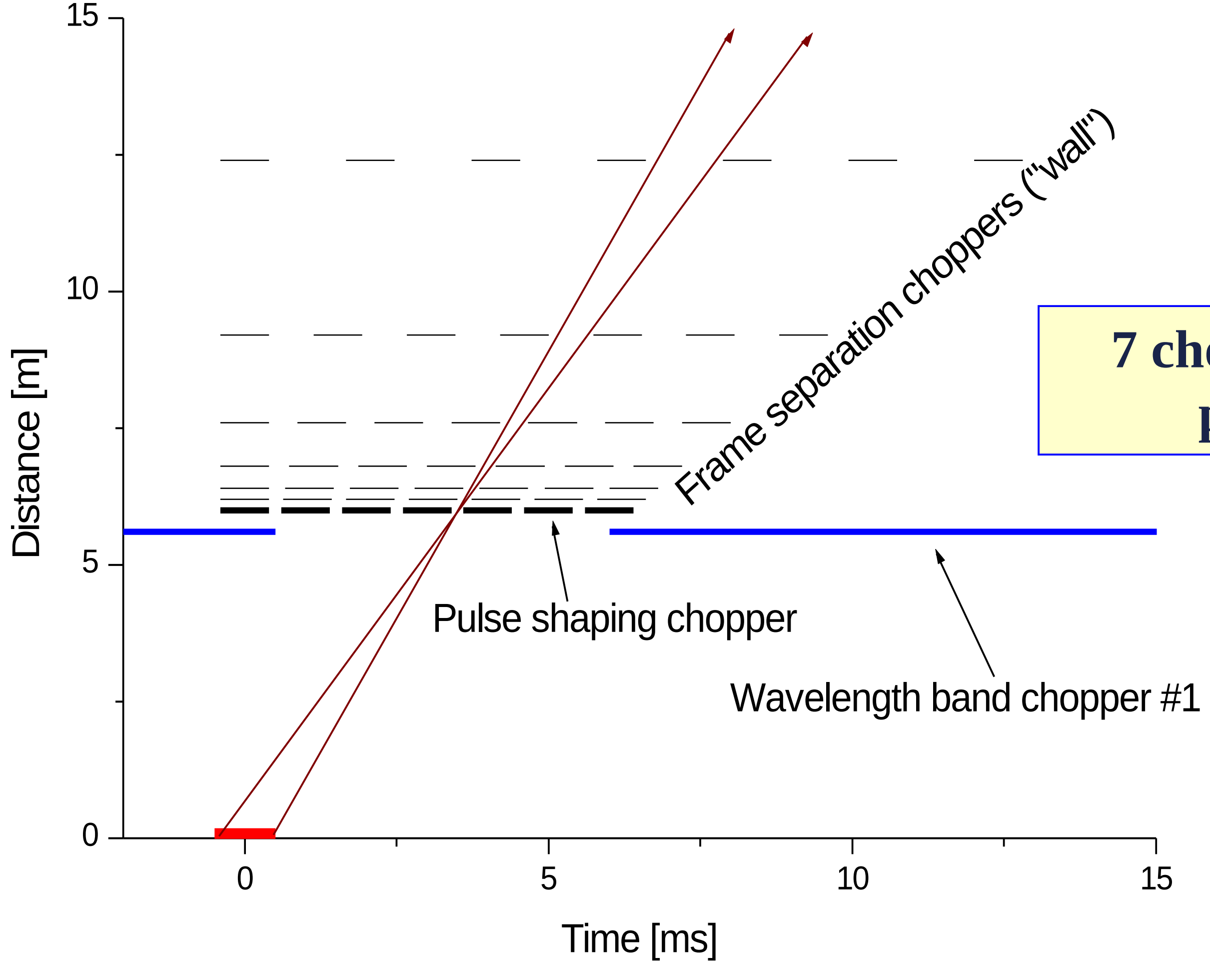
3. **Adequate moderators, neutron guides, detectors, data acquisition systems must be used in design.**

4. **Open question: Radial or tangential geometry of beam-lines?**

The second stage may include:

	Instrument	Main issue	Moderator	Resolution
5.	Single crystal	structure	80 K	Medium
6.	Texture	multi phase	80 K	High
7.	Long period	macromolecular	30 K	Medium

Pulse shaping technique for diffraction at long pulse sources



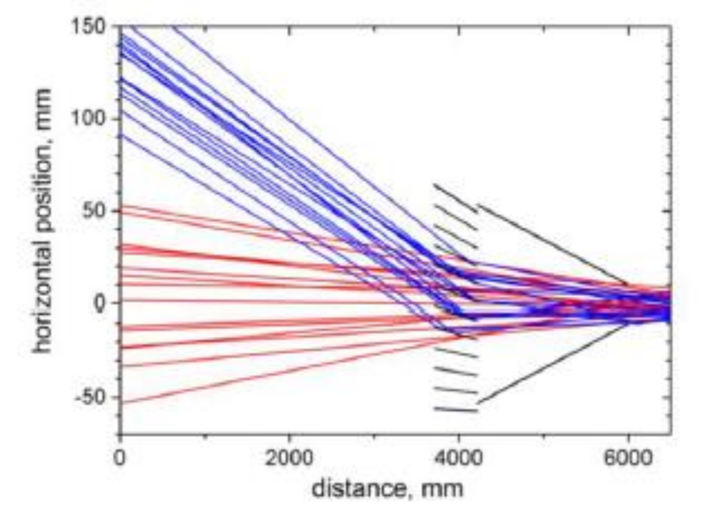
7 choppers in phase

Multiplexing chopper system (with phase slewing to source)

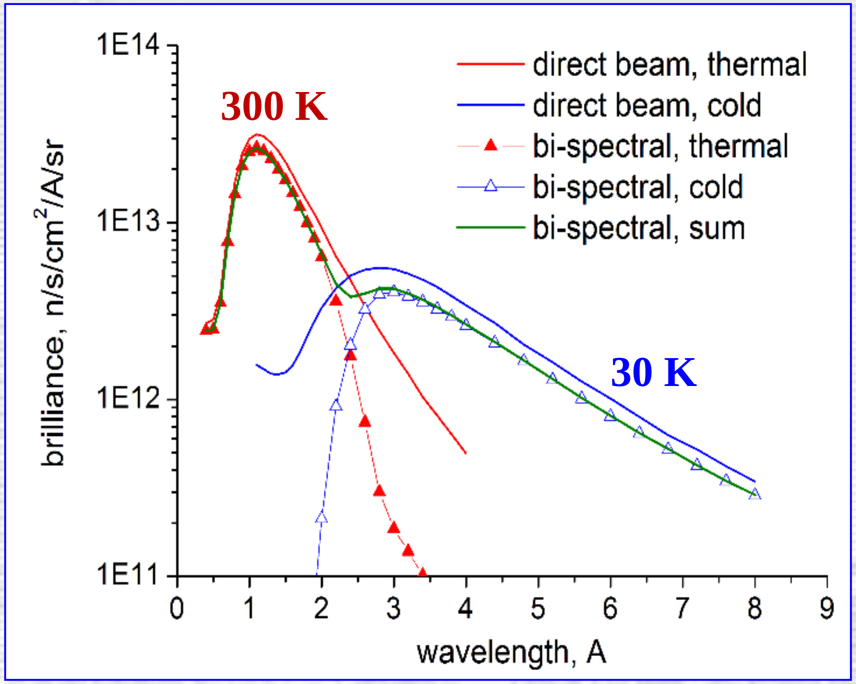
from Feri Mezei & Peter Tindemans (ESS)

Materials engineering diffractometer BEER, ESS

(Diffraction + SANS + Imaging)



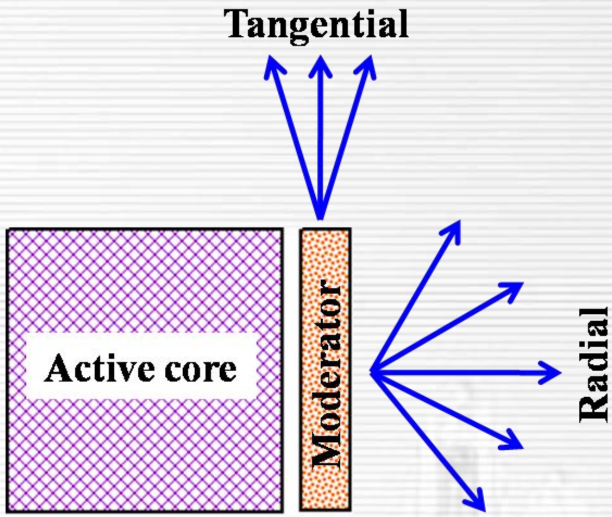
Bi-spectral extraction multichannel (m=4) guide



Simulated neutron spectra at the sample position

Mode	flux	wavelength	resolution	*d*-range
Diffraction	1.6×10^7	1.2 … 2.9 Å	Δ*d*/*d* ~ 0.4%	0.7 ... 2.3 Å
SANS	5.6×10^6	4.7 … 6.3 Å	ΔQ ~ 0.003 $Å^{-1}$	20 … 350 Å

Moderators for DNS-IV

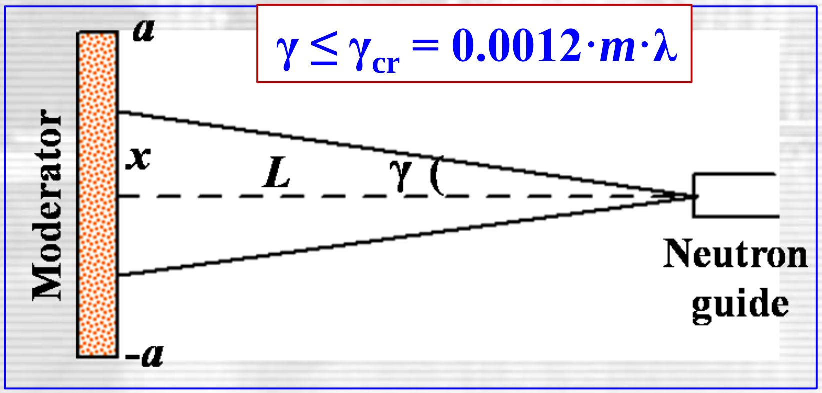


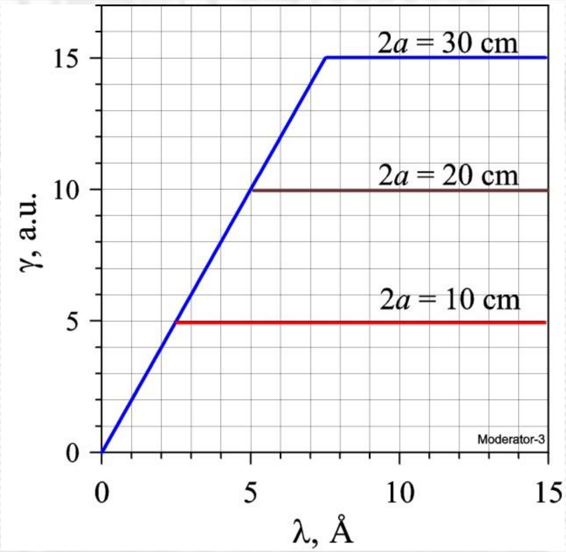
Tangential:

- **Lower fast neutrons background**
- **Higher neutron flux**
- **Smaller distance between moderator and neutron guide**

Radial:

- **Larger number of beam-lines**
- **Larger area**





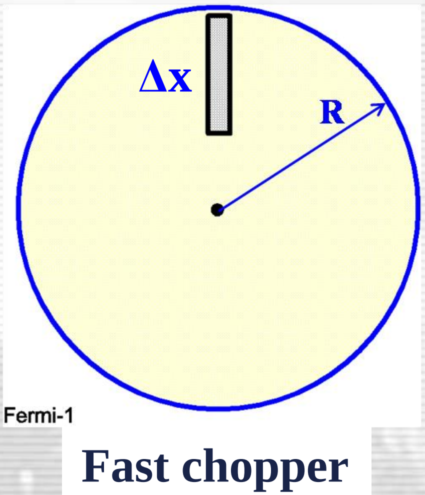
Starting from some λ_{cr} neutron flux does not grow up.

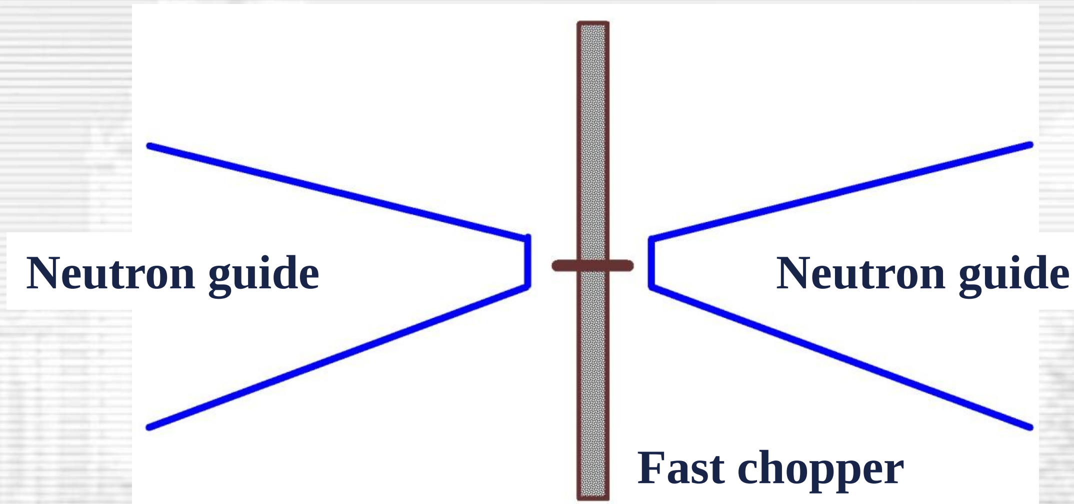
Calculated for:
***L*=8 m & Ni-Ti, m=2 supermirror**

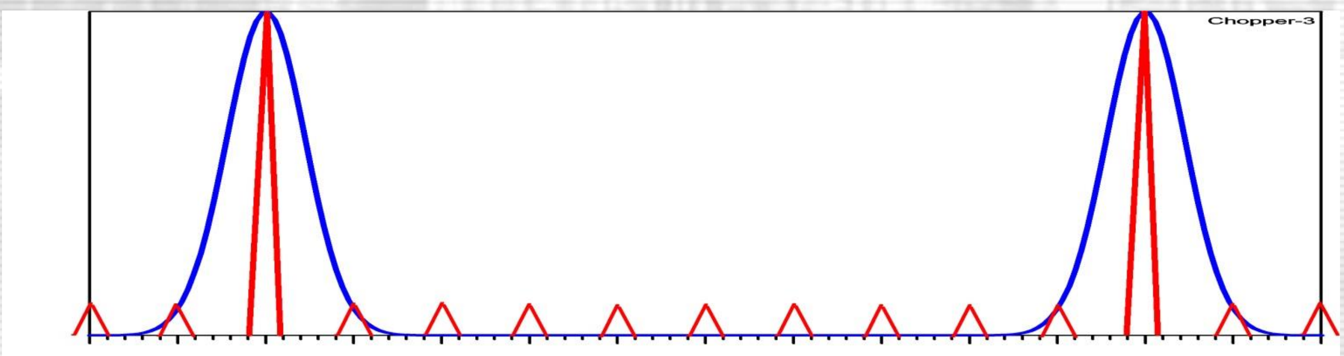
How to transform 300 µs into 30 µs with Fermi chopper

$V = 2\pi R/T \approx 26{,}400$ cm/s for $R = 30$ cm and $f = 140$ Hz = 8,400 rpm

$\Delta t_0 = \Delta x/V \approx 30$ µs for $\Delta x = 0.80$ cm

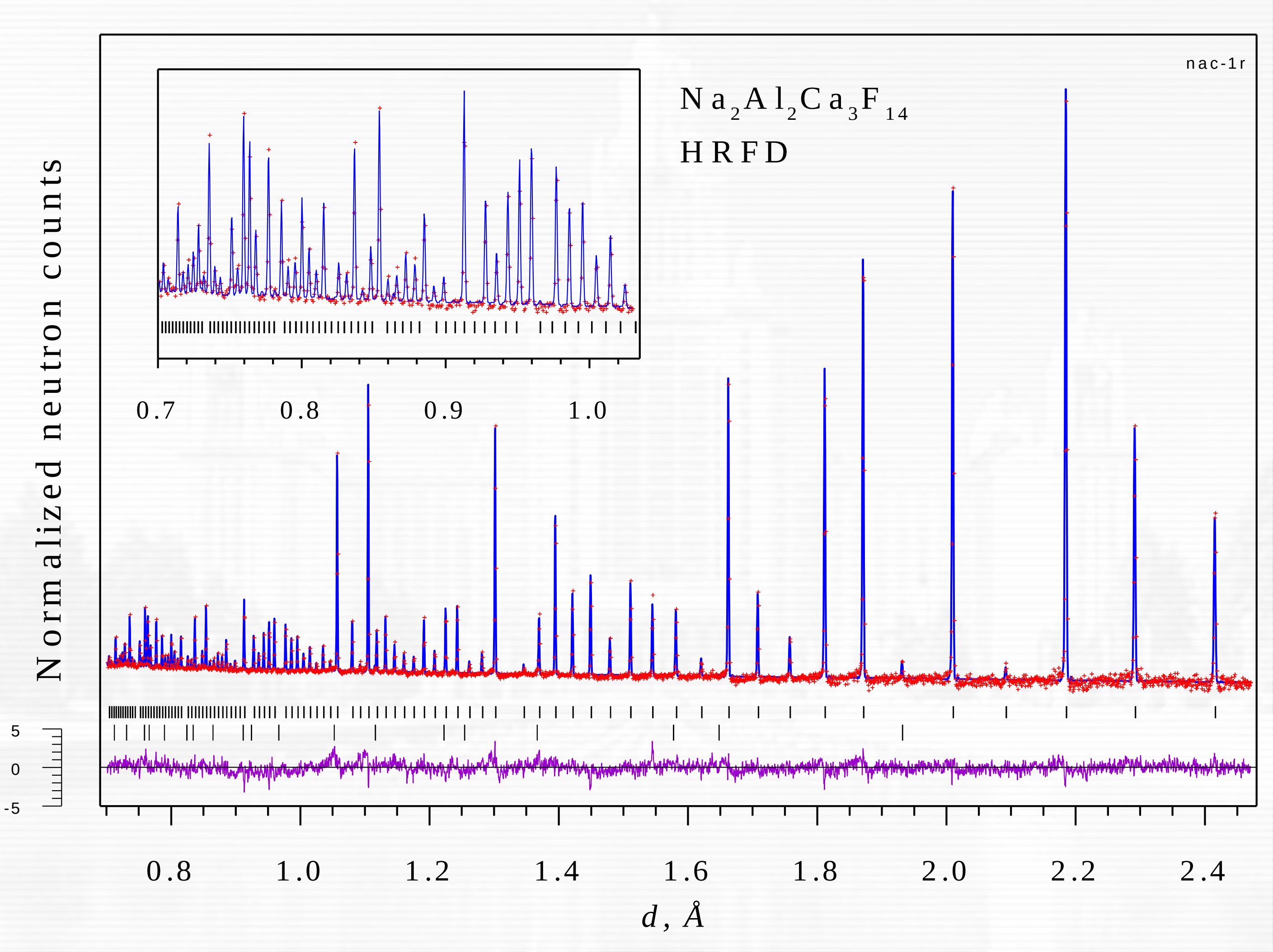






$f/f_0 = 10$ $W/W_0 = 0.1$ $I/I_0 \approx 10$

Rietveld analysis of the HRFD data (MRIA package)



Diffraction pattern obtained with NAC-standard