Дифракция нейтронов на DNS-IV

- Разнообразие нейтронных спектрометров (дифрактометров)
- Дифракция нейтронов на ИБР-2
- TOF-дифрактометры на ISIS, SNS, J-SNS, ESS
- Тенденции развития TOF-дифрактометров
- Дифрактометры на DNS-IV: базовый набор и перспективы

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Специализация нейтронных дифрактометров

I. Эксперимент с монокристаллом

2D ПЧД, $\Delta x < 3$ мм → 4π ПЧД

II. Структурный эксперимент на поликристалле

высокое разрешение, $\Delta d/d \approx 0.002$, широкоапертурный ПЧД

III. Магнитная структура (моно- или поликристалл)

среднее разрешение, большие (~15 Å) d_{hkl}

IV. *In Situ, Real Time* эксперимент

высокая светосила (~10^6 н/с), широкий интервал d_{hkl}

V. Высокое давление, микрообразцы

высокая светосила, низкий фон

VI. Длиннопериодные и макромолекулярные структуры

среднее разрешение, очень большие (~60 Å) d_{hkl}

VII. Локальные искажения структуры

большие переданные импульсы, $Q_{max} \sim 40$ Å^{-1}

VIII. Микроструктура материалов и изделий (напряжения, текстура)

высокое разрешение, $\Delta d/d \approx 0.004$, высокая светосила

Neutron sources for condensed matter studies

I. Continuous neutron sources

W = 10 – 100 MW
Const in time

IR-8, Russia
ILL, France
LLB, France
BENSC, Germany
FRM II, Germany
NIST, USA
ORNL, USA
…
SINQ, Switzerland
PIK, Russia

~200 reactors (IAEA data)

II. Pulsed neutron sources

II-*a*. SPS

W = 0.01 – 1 MW
Pulsed in time
$\Delta t_0 \approx (15 - 50)\ \mu s$

ISIS, UK
LANSCE, USA
SNS, USA
J-SNS, Japan
CSNS, China

II-*b*. LPS

W = 2 – 5 MW
Pulsed in time
$\Delta t_0 \approx (200 - 3000)\ \mu s$

IBR-2, Russia
ESS, Sweden

Pulsed neutron sources

Source	Year	W, MW	$\langle\Phi_0\rangle$, 10^{13}	Δt_0, μs	ν, Hz	Diffract.
ISIS	1985	0.2	0.07	20	50	7
LANSCE	1985	0.1	0.05	20	20	2
CSNS	2018	0.1	0.05	20	25	1
SNS	2006	1	1	20	60	6
J-SNS	2009	1	1	20	25	6
IBR-2	1984	2	0.8	320	5	7
ESS	2019	5	30	2860	14	5
NEPTUN	2035	10	50	20 / 200	10	?

TOF-diffractometers at pulsed neutron sources (33 instruments)

I. ISIS (7)

ENGIN-X – engineering
GEM – powder, HR + HI
HRPD – powder, HR
PEARL – high-pressure
POLARIS – powder, HI
SXD – single-crystal
WISH – magnetic

II. SNS (6)

MANDI – macromolecular
NOMAD – nanoscale
POWGEN – powder, HR, HR + HI
SNAP – high-pressure
TOPAZ – single-crystal
VULCAN – engineering

III. LANSCE (2)

HIPPO – engineering
SMARTS – high-pressure

IV. J-PARC (6)

iBIX - macromolecular
iMATERIA - powder, HR + HI
PLANET – high-pressure
SENJU - single-crystal
sHRPD – powder, HR
TAKUMI - engineering

V. IBR-2 (7)

DN-6 – high-pressure
DN-12 – high-pressure
FSD – engineering
HRFD - powder, HR, HR + HI
RTD - powder, HI
EPSILON – stress
SKAT - texture

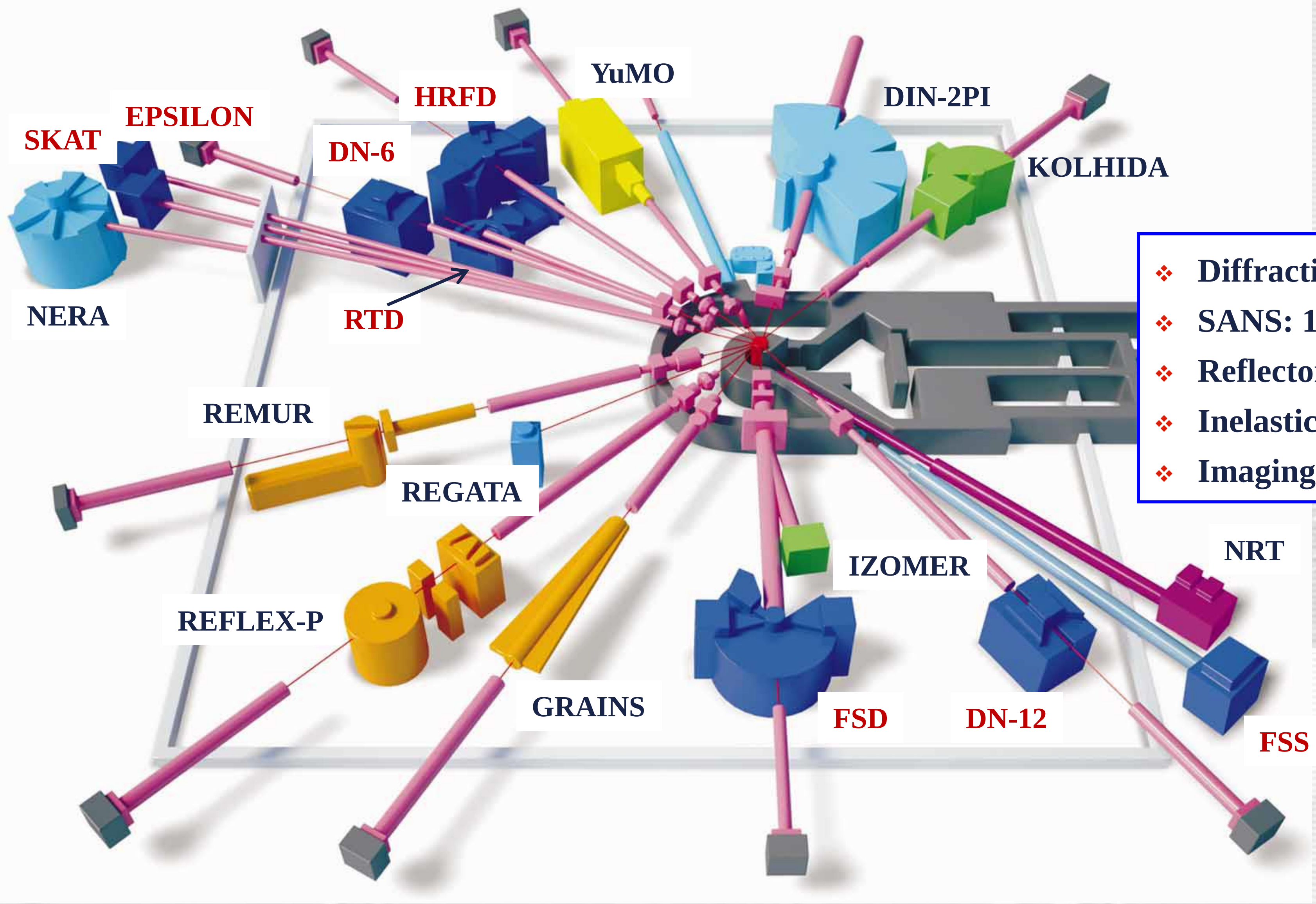
VI. ESS (5)

DREAM – powder, HR, HR + HI
HEIMDAL - hybrid
MAGiG – polarized, single crystal
NMX – macromolecular
BEER – engineering

TOF-diffractometers at 6 pulsed neutron sources

I.	High-pressure	(6)
II.	Engineering	(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)

Spectrometers at the IBR-2 reactor



- Diffraction: 8
- SANS: 1
- Reflectometry: 3
- Inelastic: 2
- Imaging: 1

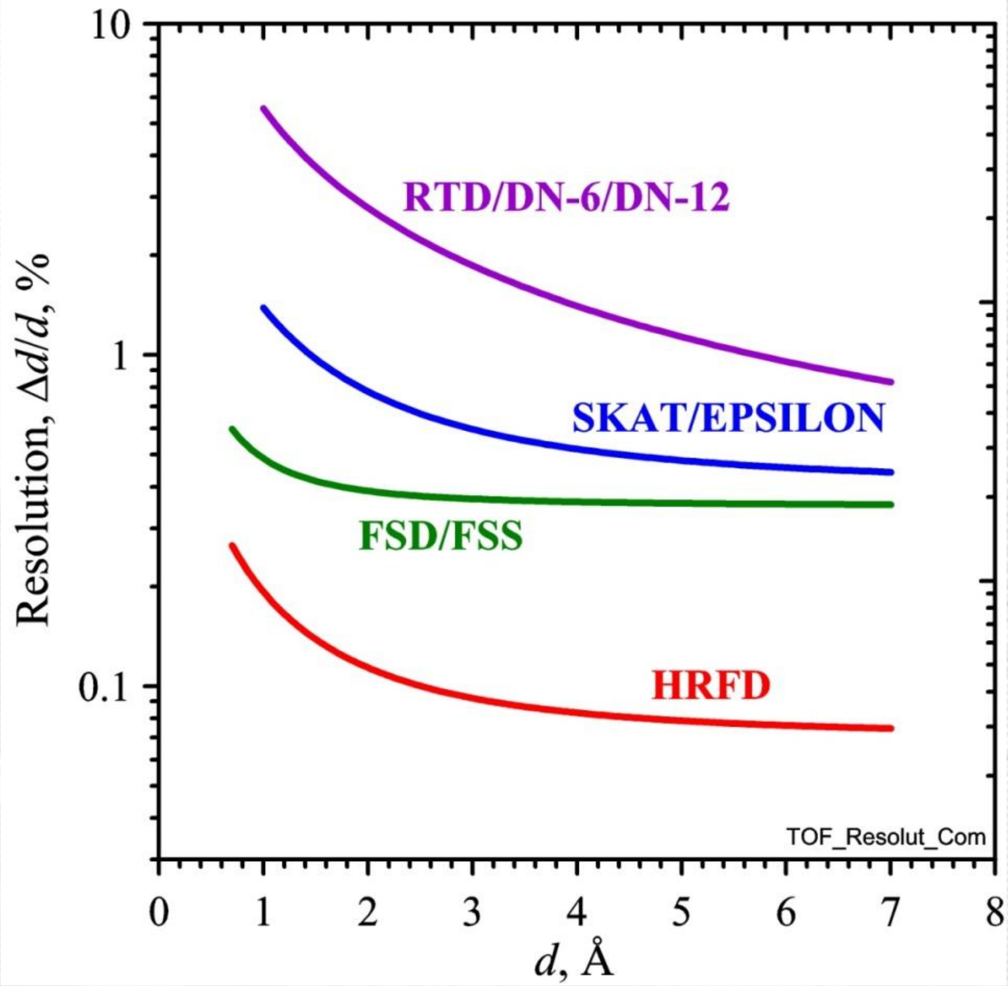
Diffraction at the IBR-2M

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 – engineering
7.	DN-12	microsamples – high-pressure
8.	FSS*	bulk samples – internal stresses (setting-up)

* Fourier RTOF technique

** 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**

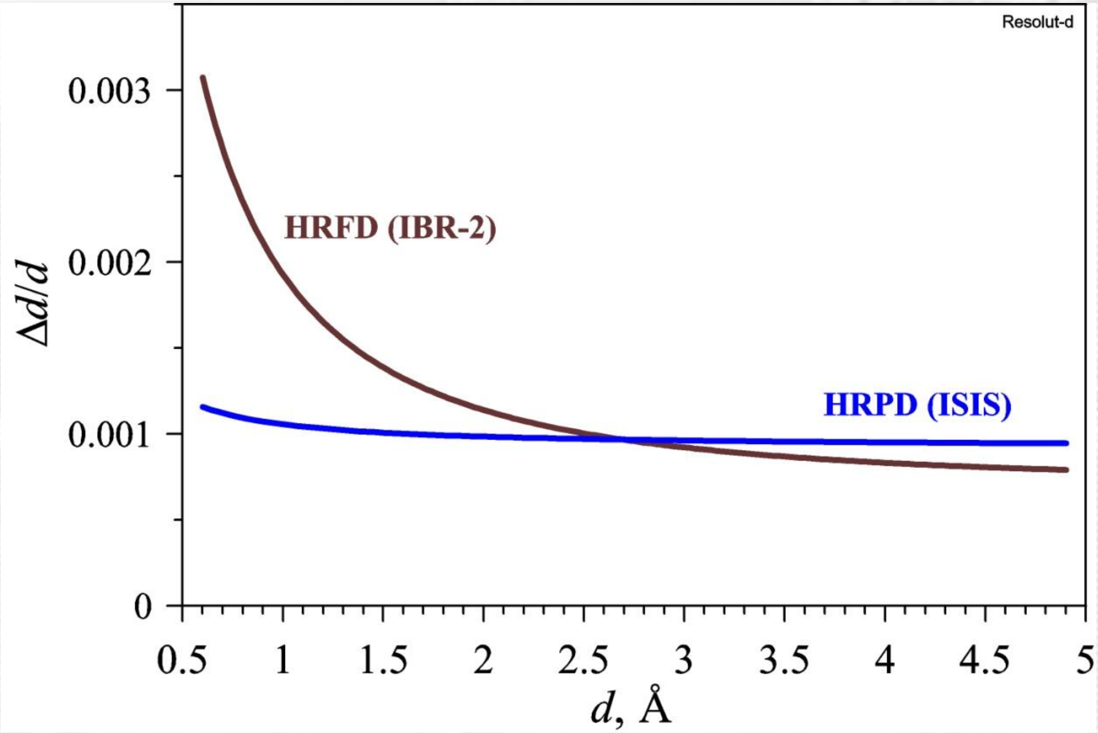
Resolution becomes better for longer *d*-spacing!

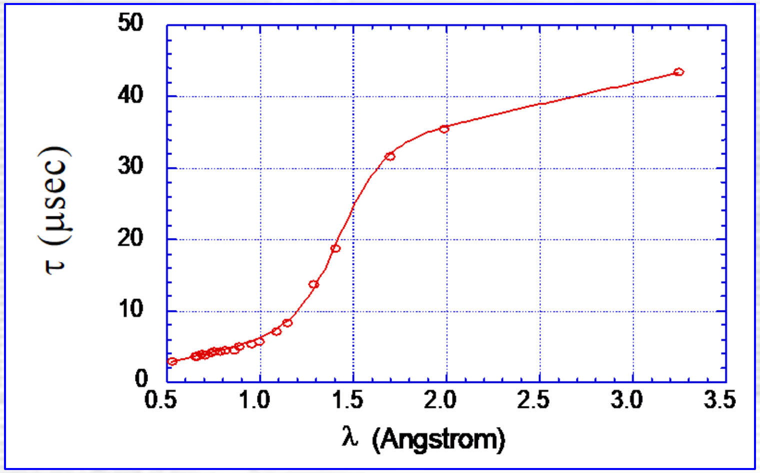
Resolution of a TOF neutron diffractometer

$(\Delta d/d)^2 = (\Delta t_0/t)^2 + (\Delta\theta/\mathrm{tg}\theta)^2$, $t \approx 250 \cdot L \cdot \lambda \approx 500 \cdot L \cdot d \cdot \sin\theta$

1) $\Delta t_0 \sim \lambda$ (SNS) → $R_t(d) \approx \mathrm{Const}$

2) $\Delta t_0 \approx \mathrm{Const}$ (Fourier, ESS) → $R_t(d) \sim 1/d$





$\Delta t_0(\lambda)$ at GEM, ISIS

TOF-diffractometers at the SNS pulsed neutron sources
$\nu = 60$ Hz, $\Delta t_0 = (15 – 40)$ μs (poisoned & de-coupled)

High-pressure (SNAP): $L_1 = 15$ m, $\Delta d/d \approx 1\%$, $\Delta\lambda \approx 0.5 – 3.65$ Å or $3.7 – 6.5$ Å
Detector: 98-150° (hor), ±34° (ver), $P \leq 50$ GPa, $\Delta t \sim 8$ h for 0.15 mm^3

Engineering (VULCAN): $L_1 = 44$ m, $\Delta d/d \approx 0.25\%$ (HR) $\approx 0.45\%$ (HI),
$\Delta\lambda \approx 0.5 – 1.5$ Å (60 Hz), 0.5 – 3.5 Å (20 Hz), Beam = (2 – 12) mm^2,
Detector: 60-150° (hor), ±30° (ver), $V_g = (8 – 20)$ mm^3

Powder, HR (POWGEN): $L_1 = 60$ m, $L_2 = (1 – 6)$ m, $\Delta d/d \approx (0.1 – 1.6)\%$,
$\Delta\lambda \approx 1$ Å (60 Hz), Detector: 6 – 170°, $\Omega_{\mathrm{det}} = 4$ sr

Single-crystal (TOPAZ): $L_1 = 18$ m, $L_2 = 0.5$ m, $\Delta d/d \approx 0.4\%$, 3D Q-space mapping
$\Delta\lambda \approx 3.1$ Å (60 Hz), Detector: 20 - 160° (hor), ±54° (ver), $\Omega_{\mathrm{det}} = 3$ sr

Nanoscale (NOMAD): $L_1 = 19$ m, $L_2 = (0.5 – 3)$ m, $\Delta d/d \approx 0.4\%$,
$\Delta\lambda \approx (0.1 – 3)$ Å (60 Hz), Detector: 3 – 175°, $\Omega_{\mathrm{det}} = 4$ sr (8 sr – full)

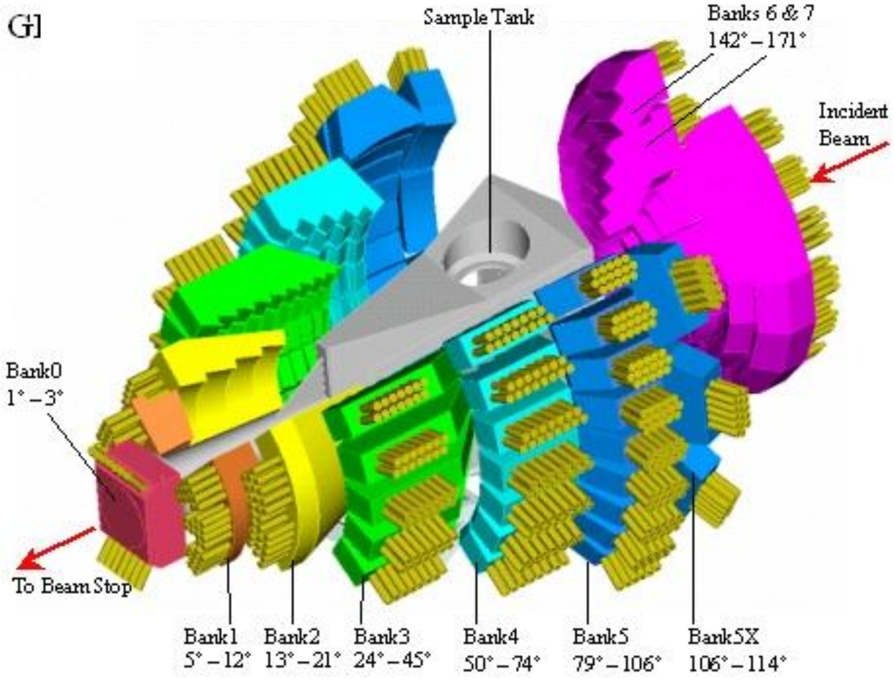
Macromolecular (MANDI): $L_1 = 30$ m, $L_2 = 0.4$ m, $\Delta d/d \approx 0.3\%$,
$\Delta\lambda \approx 2.2$ / 4.3 Å (60/30 Hz), Detector: 20 – 160°, $\Omega_{\mathrm{det}} = 4.1$ sr

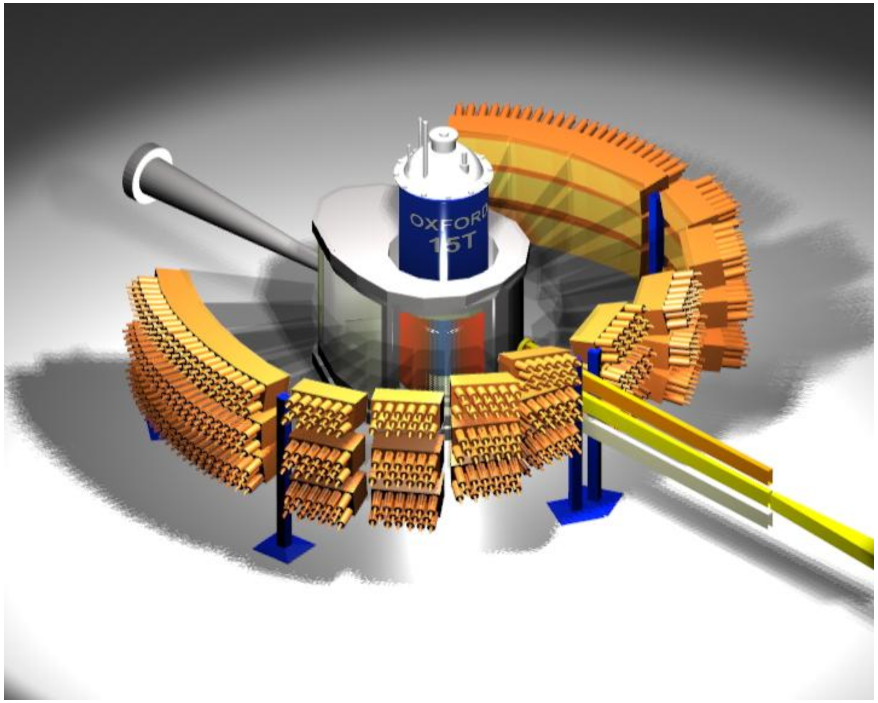
$\Delta d \approx \Delta\lambda/1.5$

Advanced detectors for TOF diffractometers

GEM (ISIS), HI + HR

WISH (ISIS), HI + HR





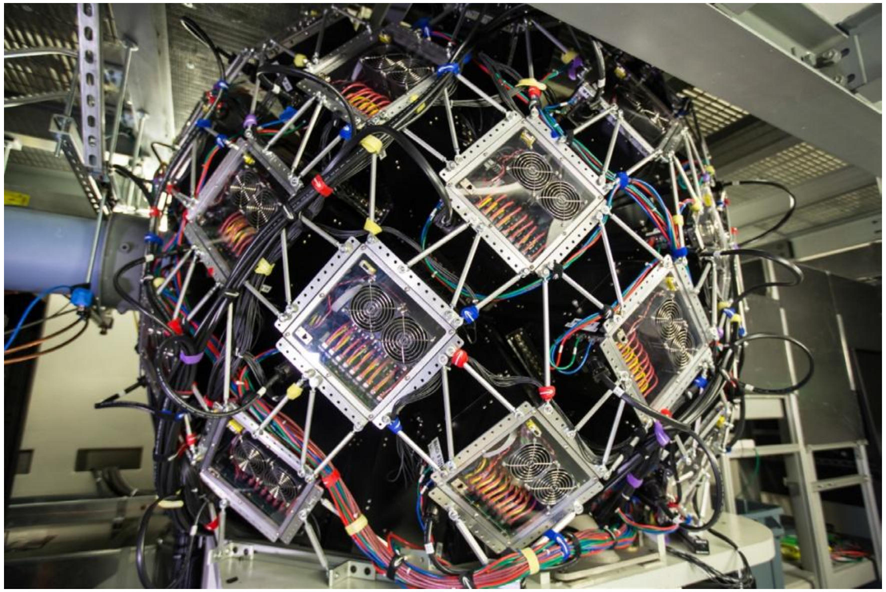
$L = 17$ m, $\Omega_{det} \approx 3.86$ sr

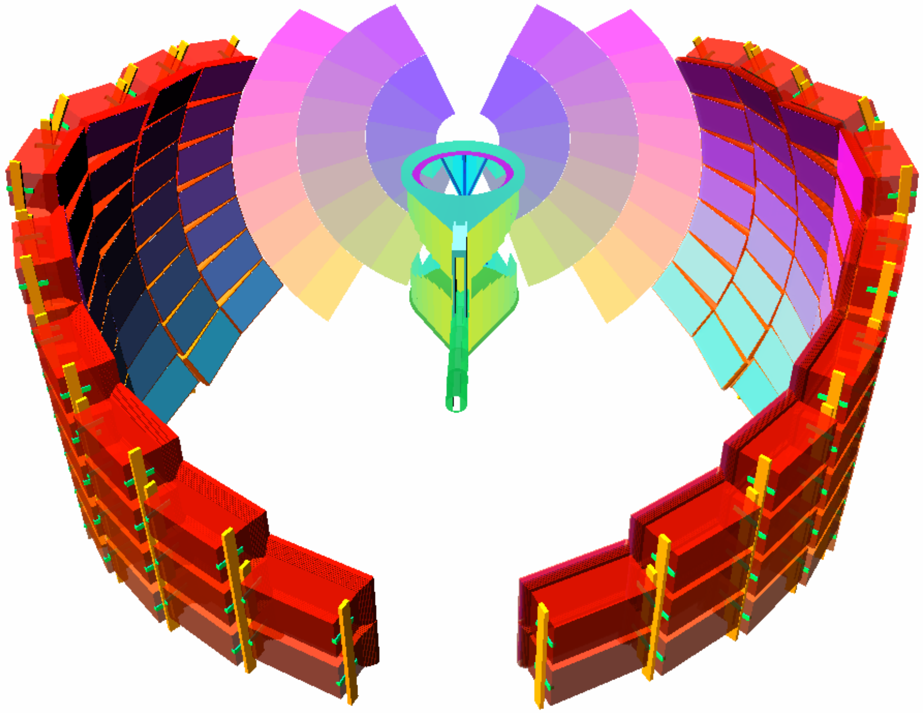
$L = 50$ m, $\Omega_{det} \approx 2$ sr

Advanced detectors for TOF diffractometers

MaNDi (SNS), Macromol. single cryst.

Powgen (SNS), HI + HR

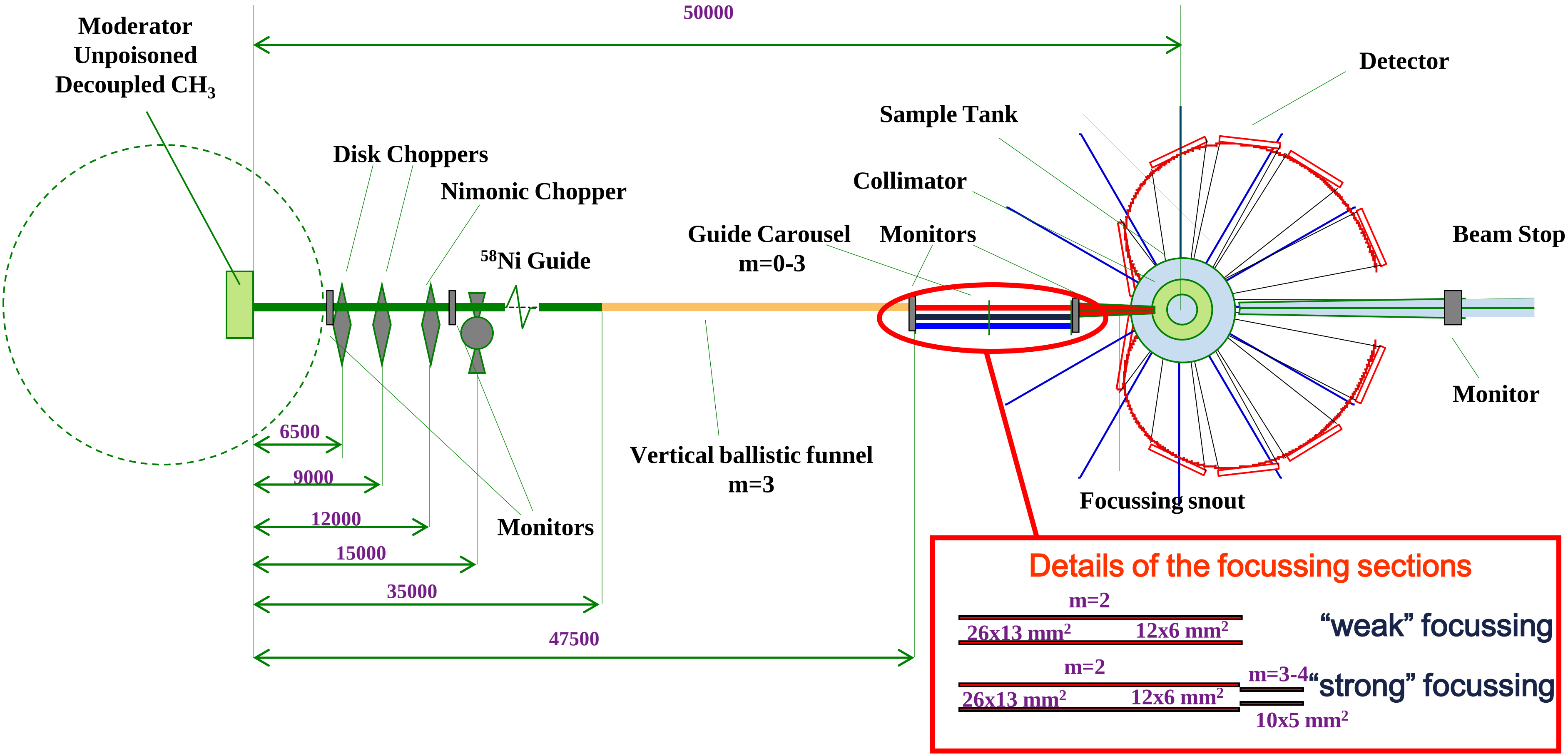




***L* = 30 m, $\Omega_{\mathrm{det}} \approx 4.1$ sr**

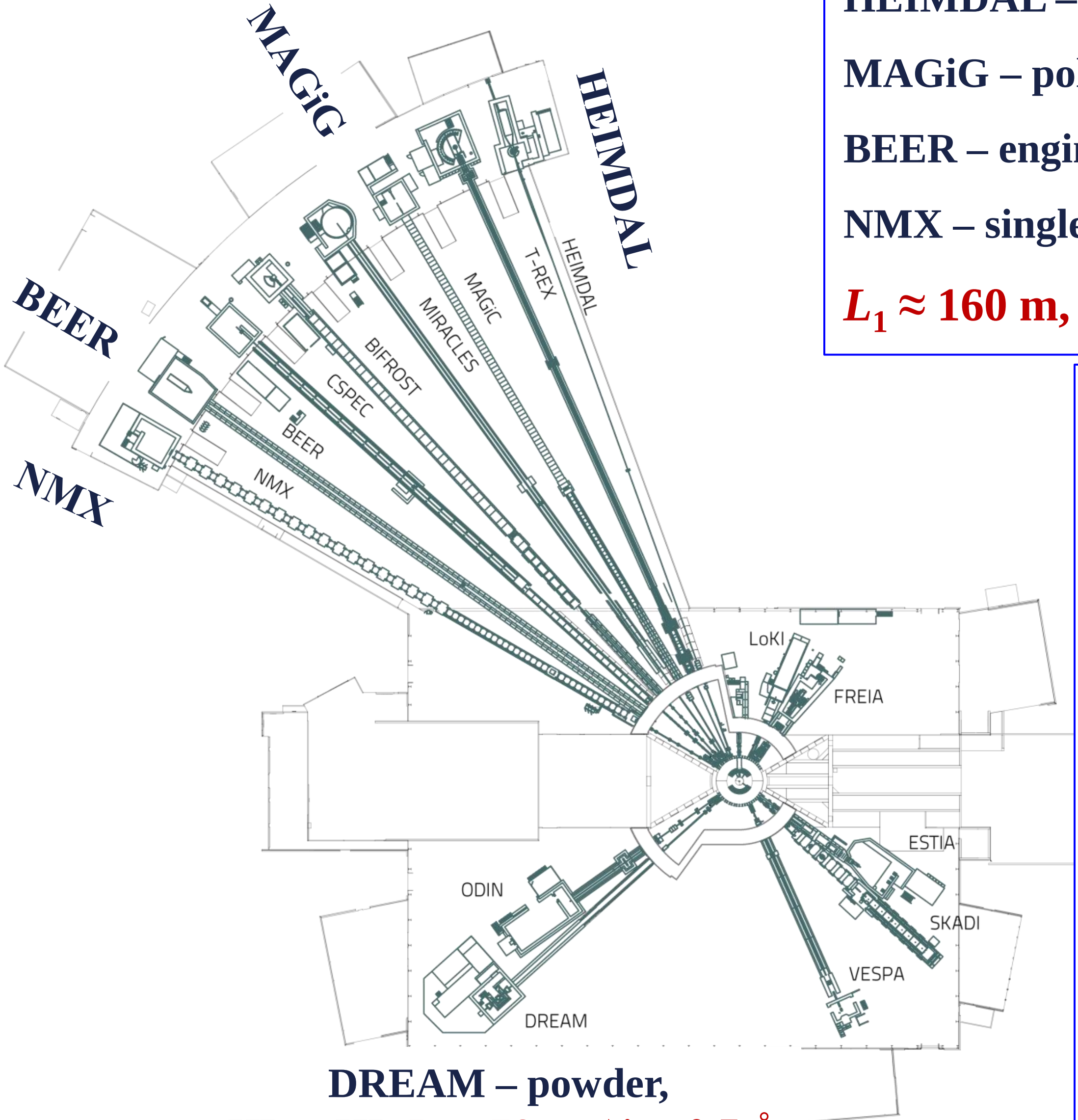
***L* = 60 m, $\Omega_{\mathrm{det}} = 4.0$ sr**

TOF high-resolution magnetic diffractometer WISH, ISIS, UK



WISH schematic drawing

ESS pulsed neutron sources, $\nu = 14$ Hz, $\Delta t_0 = 2860$ 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

$\Delta\lambda \approx 282/L$

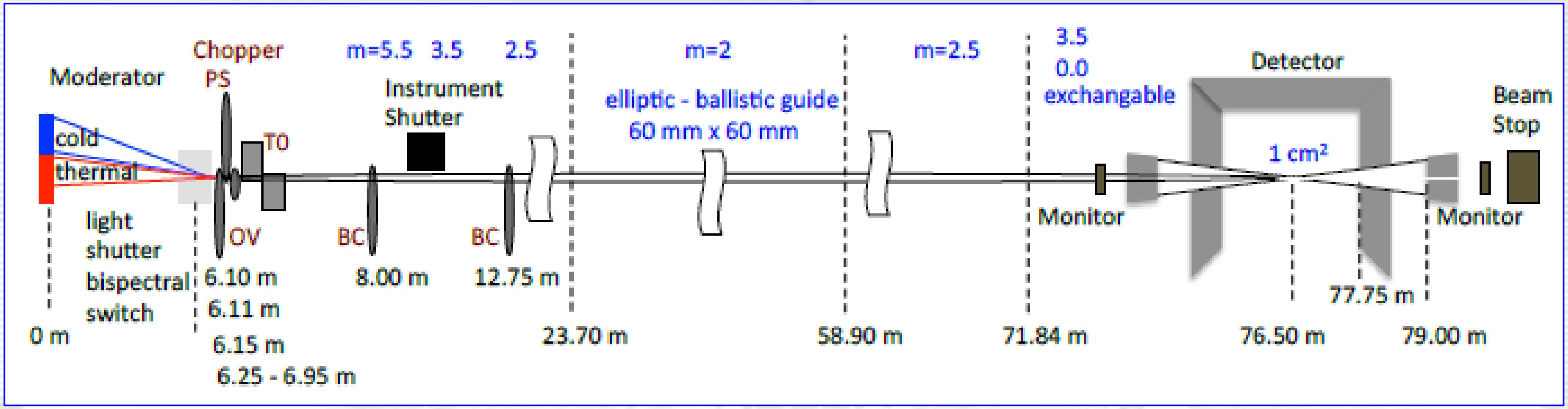
DREAM – powder,
HR + HI, $L_1 = 76$ m, $\Delta\lambda \approx 3.7$ Å

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 instruments	16 (22)
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)



DREAM feature: **bispectral switch (cold + thermal neutrons)**

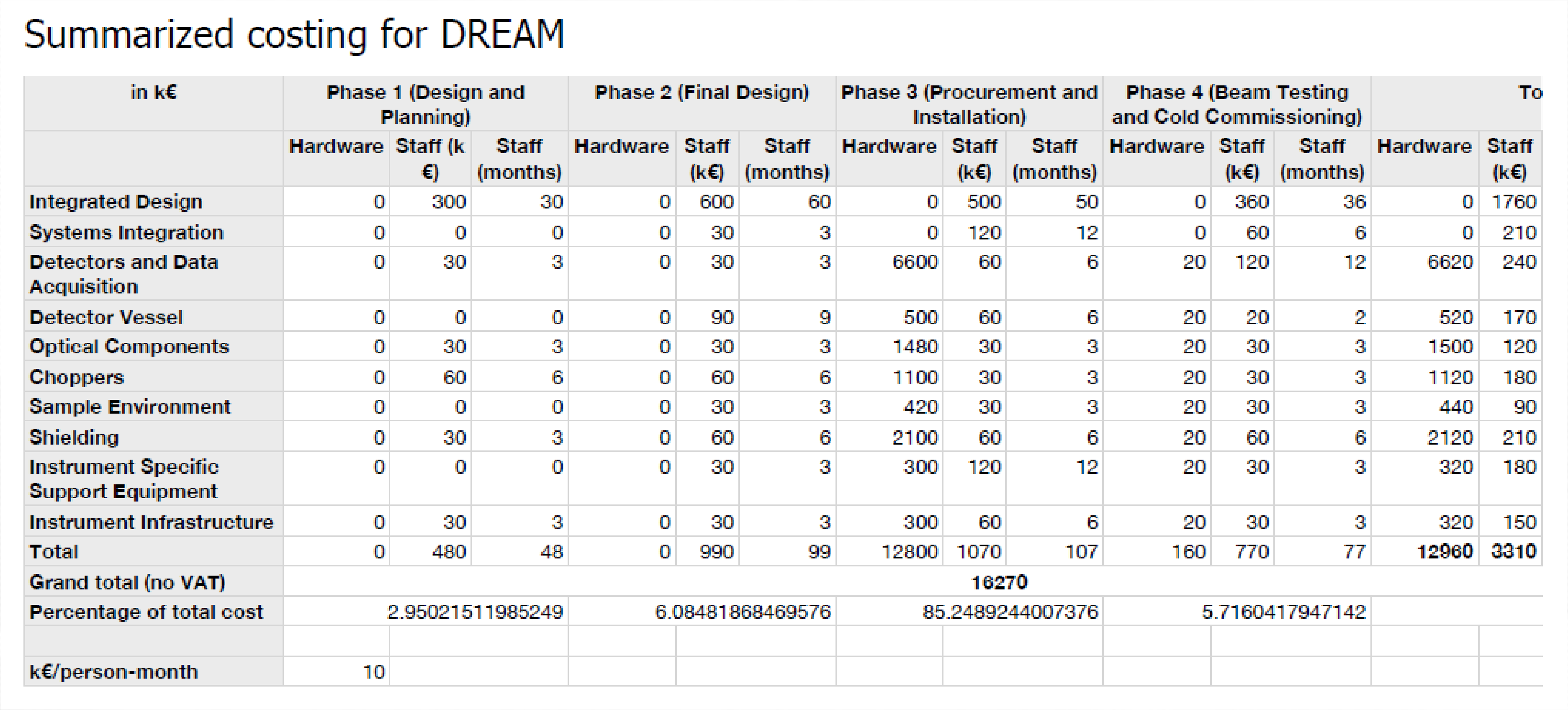
DREAM choppers: **PC – pulse shaping, T0, BC – band control, OV – overlap = 7 ch-s**

DREAM costing (kEu): **Design = 1970, Detector + DA = 6620, Optic = 1500,**

Choppers = 1120, Shielding = 2120, Infrastr. = 320, …   Total = 12 960

L_1 = 76.5 m, $(\Delta t_0)_{min}$ = 10 μs → $\Delta d \approx 2.8\cdot10^{-4}$ Å,

HR + HI powder diffractometer DREAM, ESS



Summarized costing for DREAM

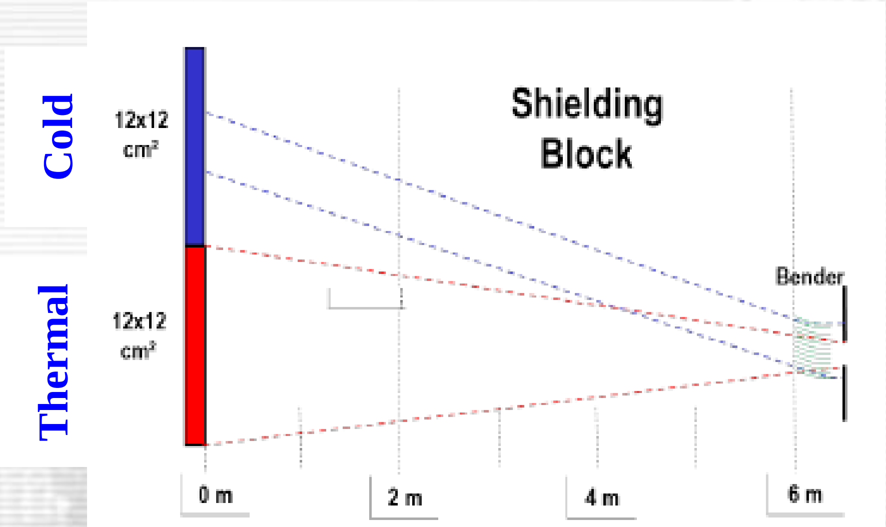
in k€	Phase 1 (Design and Planning)			Phase 2 (Final Design)			Phase 3 (Procurement and Installation)			Phase 4 (Beam Testing and Cold Commissioning)			To	
	Hardware	Staff (k €)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)	Staff (months)	Hardware	Staff (k€)
Integrated Design	0	300	30	0	600	60	0	500	50	0	360	36	0	1760
Systems Integration	0	0	0	0	30	3	0	120	12	0	60	6	0	210
Detectors and Data Acquisition	0	30	3	0	30	3	6600	60	6	20	120	12	6620	240
Detector Vessel	0	0	0	0	90	9	500	60	6	20	20	2	520	170
Optical Components	0	30	3	0	30	3	1480	30	3	20	30	3	1500	120
Choppers	0	60	6	0	60	6	1100	30	3	20	30	3	1120	180
Sample Environment	0	0	0	0	30	3	420	30	3	20	30	3	440	90
Shielding	0	30	3	0	60	6	2100	60	6	20	60	6	2120	210
Instrument Specific Support Equipment	0	0	0	0	30	3	300	120	12	20	30	3	320	180
Instrument Infrastructure	0	30	3	0	30	3	300	60	6	20	30	3	320	150
Total	0	480	48	0	990	99	12800	1070	107	160	770	77	**12960**	**3310**
Grand total (no VAT)	**16270**													
Percentage of total cost	2.95021511985249			6.08481868469576			85.2489244007376			5.7160417947142				
k€/person-month	10													

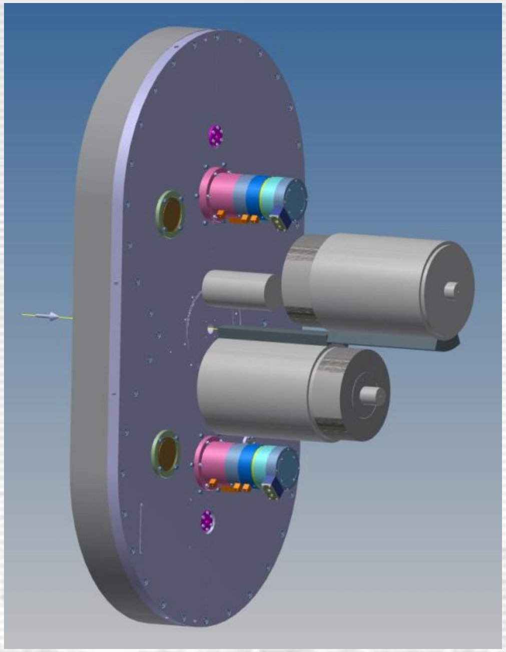
DREAM costing: Grand total = 16,270 kEu

HR + HI powder diffractometer DREAM, ESS

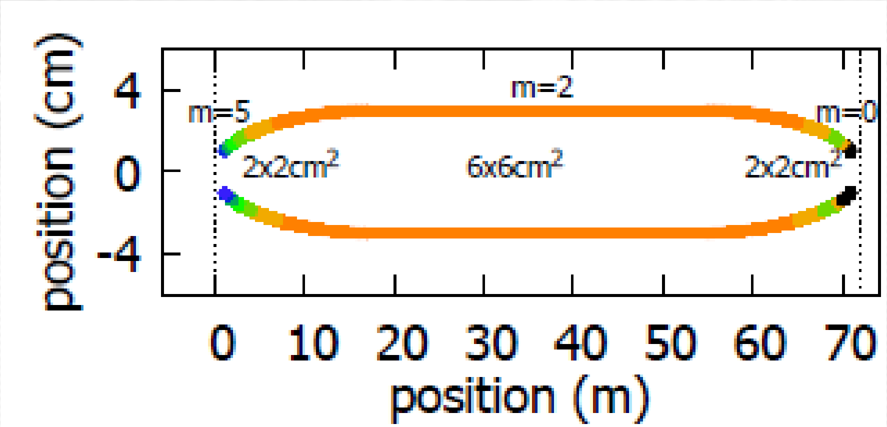
(Diffraction Resolved by Energy and Angle Measurements)

Bispectral extraction system





Chopper arrangement

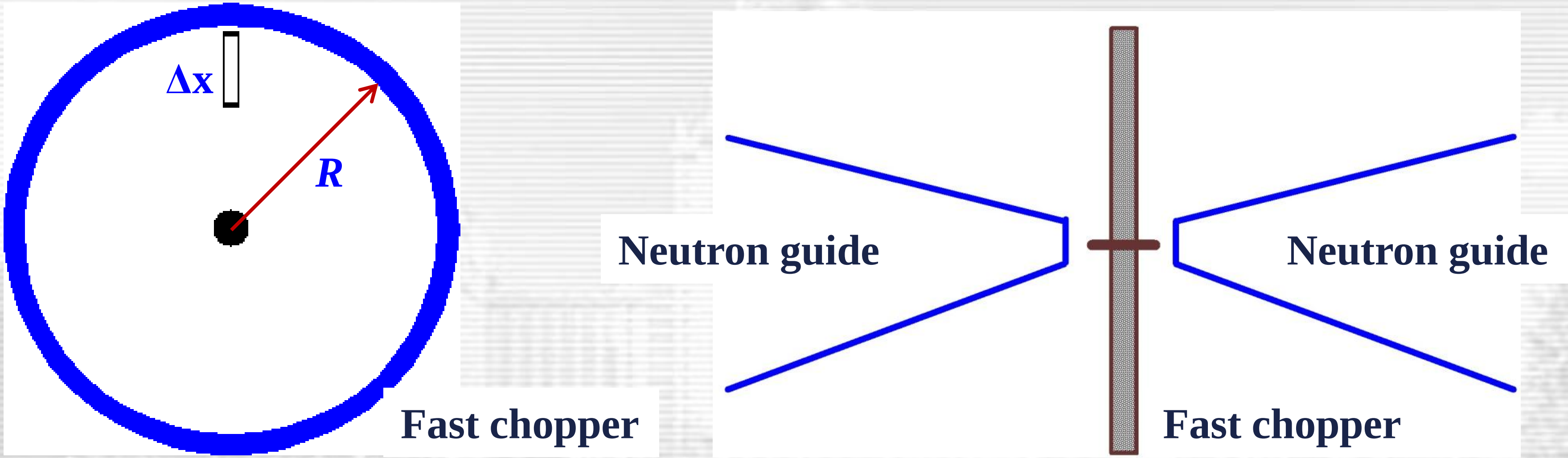


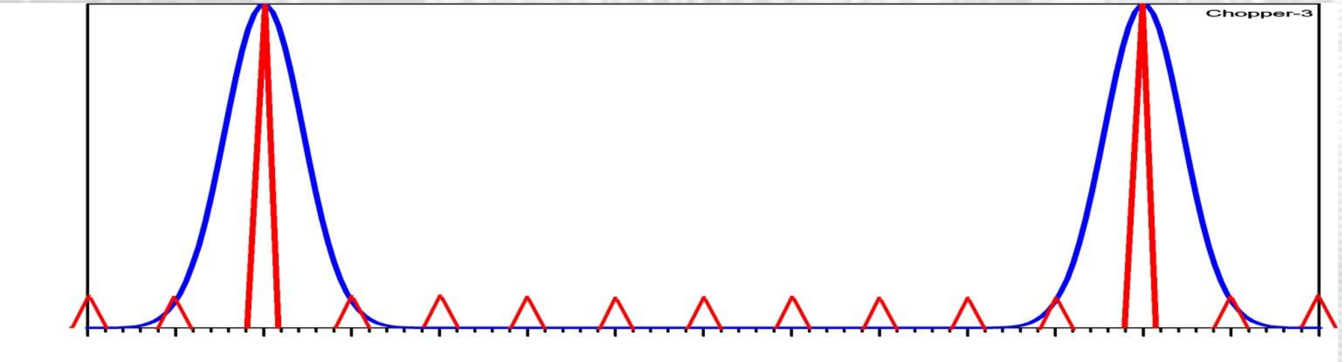
Elliptic guide system

How to transform 300 μs into 30 μs

$V = 2\pi R/T \approx 26{,}400$ 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

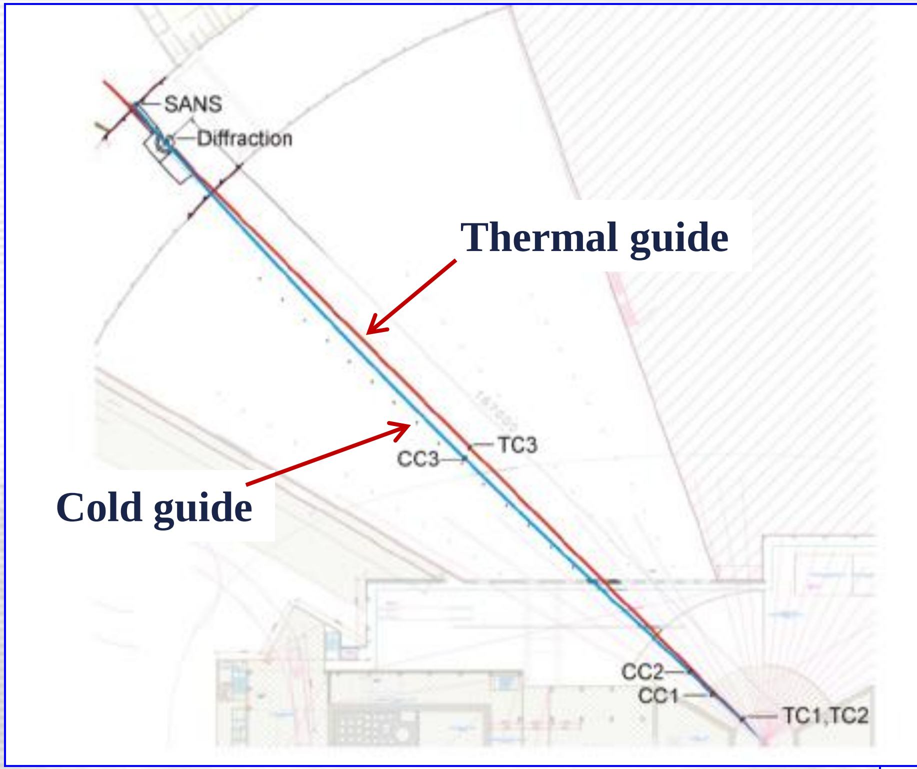




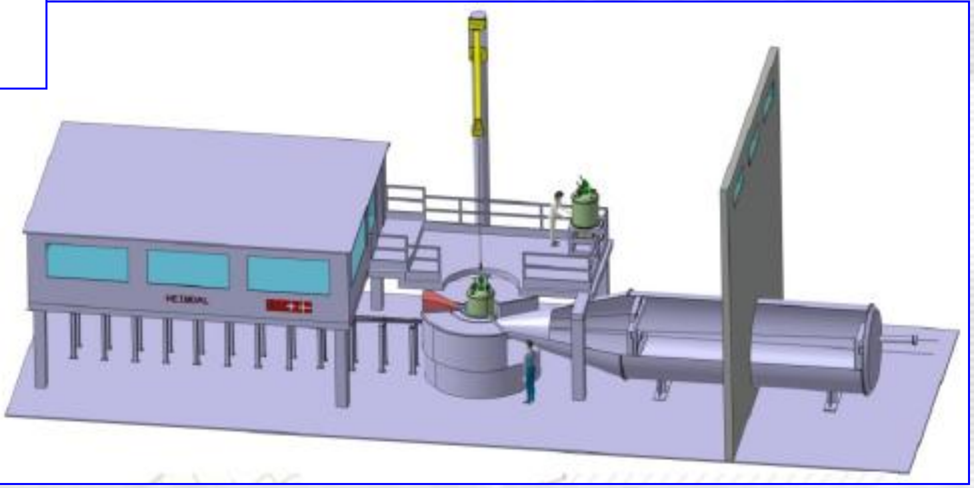
$f/f_0 = 10$ $W/W_0 = 0.1$ $I/I_0 \approx 10$

Hybrid diffractometer HEIMDAL, ESS

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

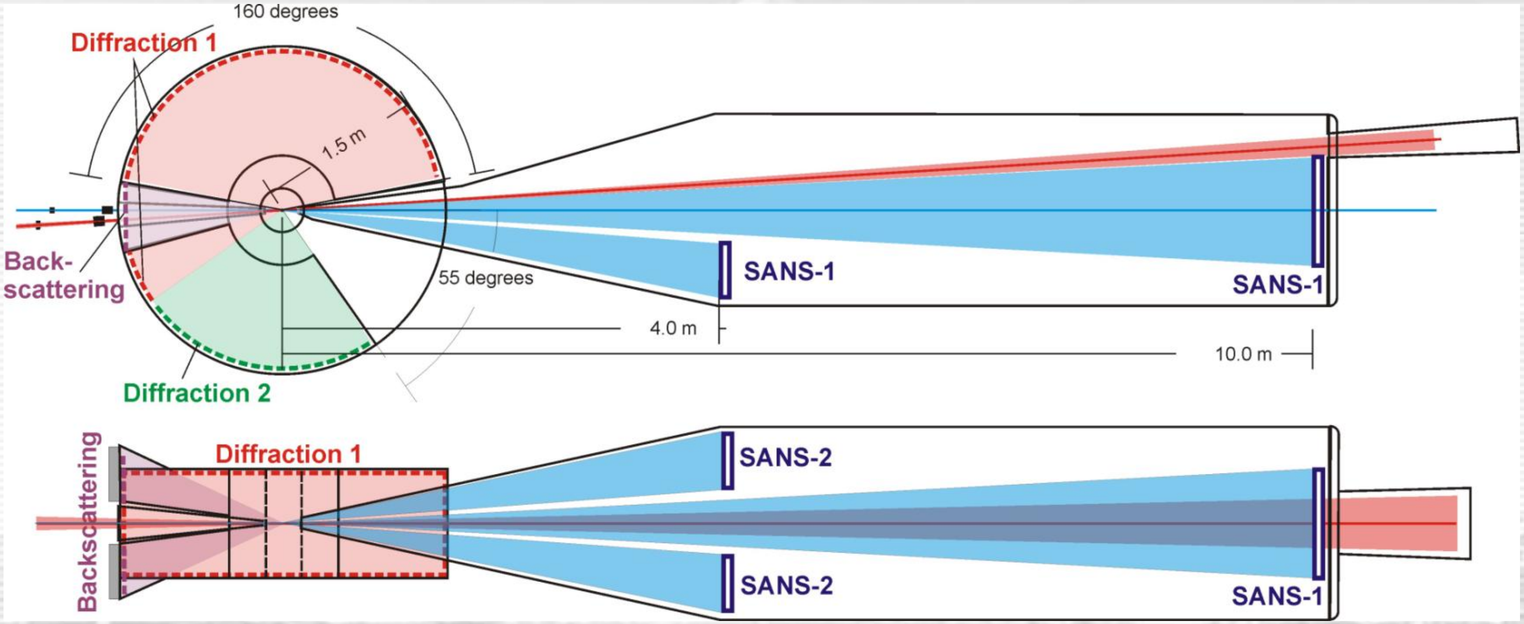


HEIMDAL layout with:
- thermal and cold neutron guides
- thermal (TC1 – TC3) choppers
- cold (CC1 – CC3) choppers
- diffraction and SANS modules



Hybrid diffractometer HEIMDAL, ESS

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



HEIMDAL feature: bispectral switch (cold + thermal neutrons)

HEIMDAL choppers: (pulse shaping + pulse selection + frame overlap) = 6 choppers

HEIMDAL costing (kEu): Detector = 6190, Optic = 5299, Choppers = 600,

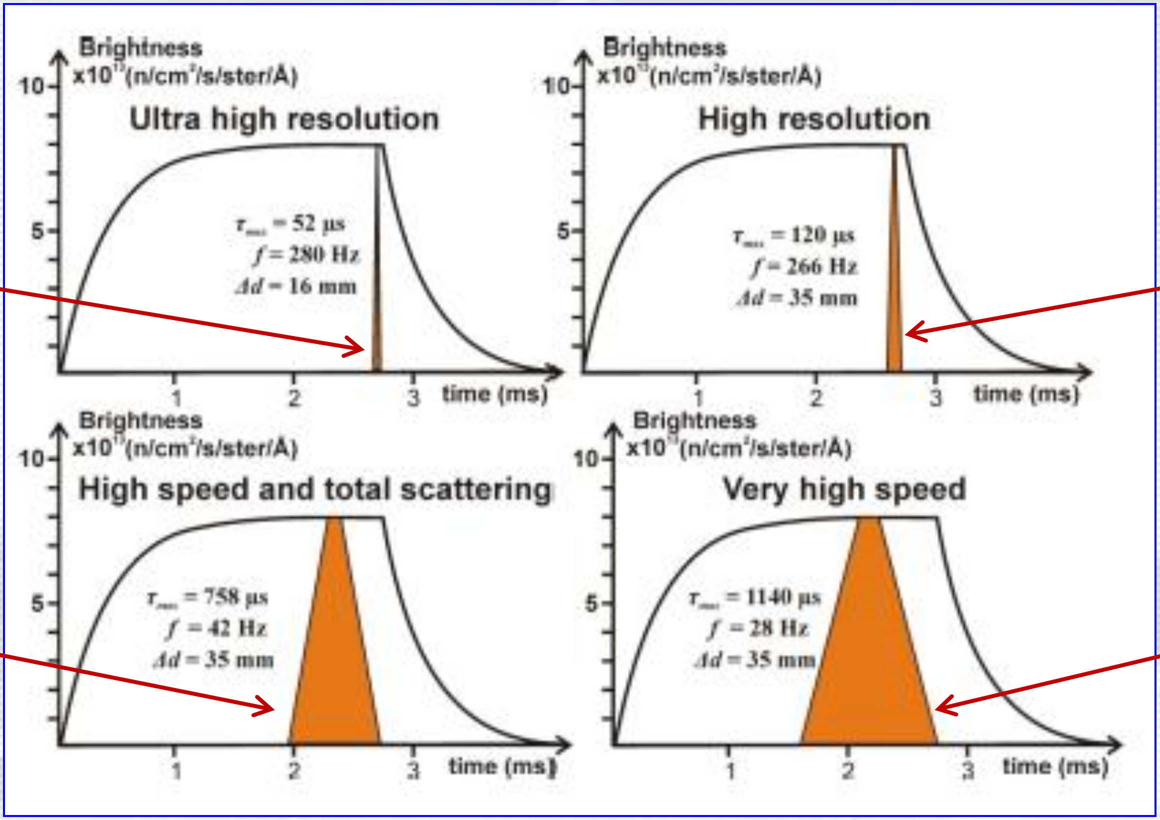
Shielding = 1300, … **Total** = 19 082

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%

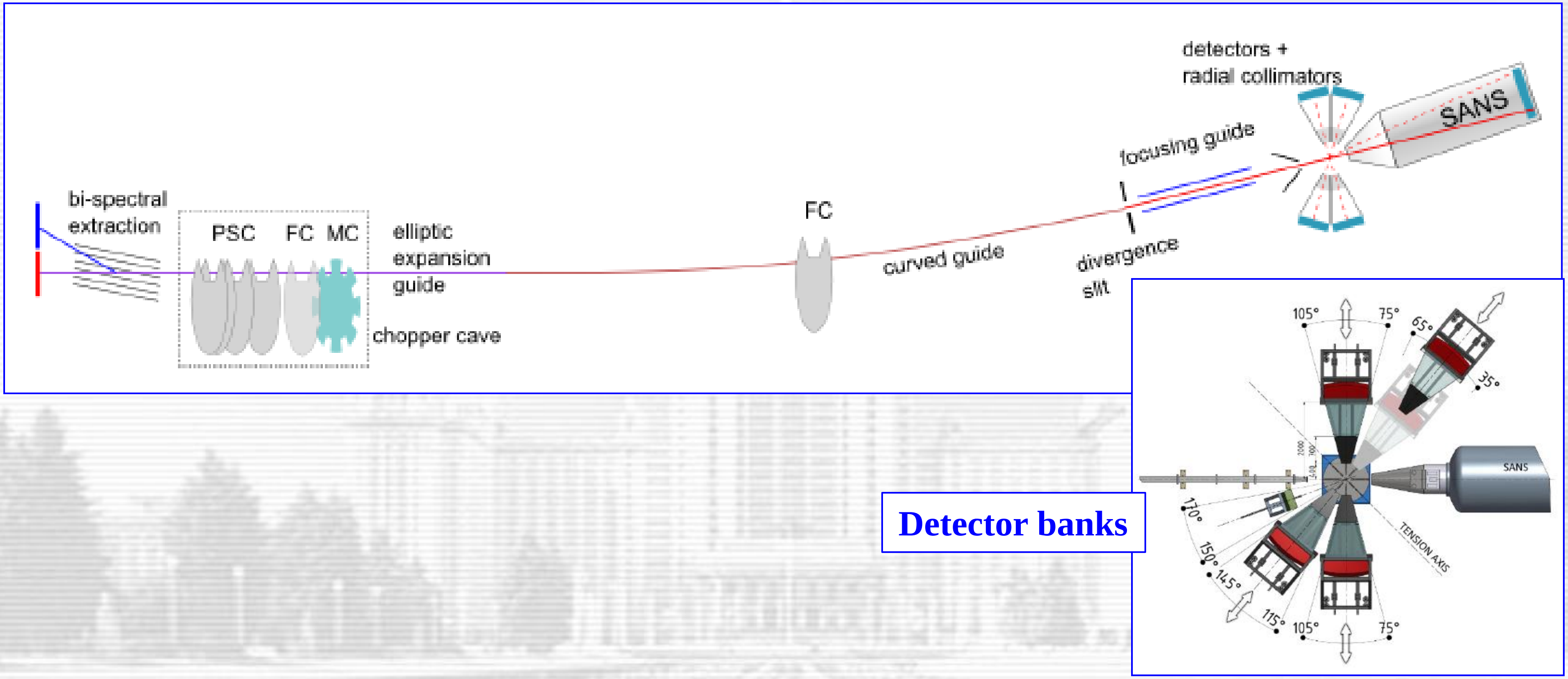
Δt_0 = 1140 μs
f = 28 Hz
Δx = 3.5 cm
R_t = 1.3%

Modes of the pulse shaping chopper operation

Flux at a sample: from $3.8\text{x}10^6$ (HR) to $2.0\text{x}10^9$ (HI)

Materials engineering diffractometer BEER, ESS

(Diffraction + SANS + Imaging, L_1 = 157 m, $\Delta\lambda \approx 1.7$ Å, $\lambda_{min} \approx 0.6$ Å)



Detector banks

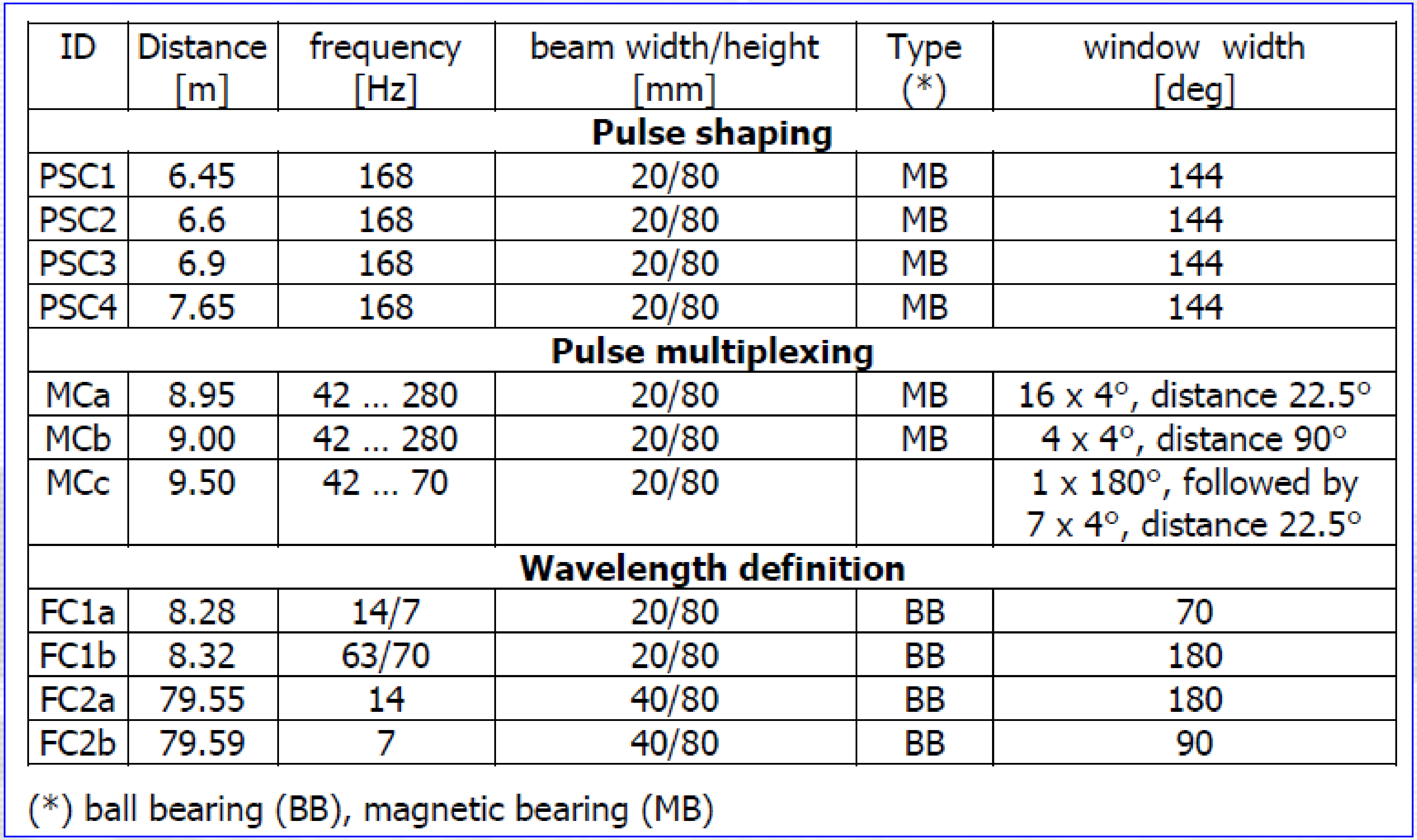
BEER feature: bispectral switch (cold + thermal neutrons)

BEER choppers: (pulse shaping + pulse selection + frame overlap) = 11 choppers

BEER costing (kEu): Detector = 7011, Optic = 3990, Choppers = 1550, Shielding = 700

… Total : Min = 19 701; Max = 21 301

Materials engineering diffractometer BEER: list of choppers)



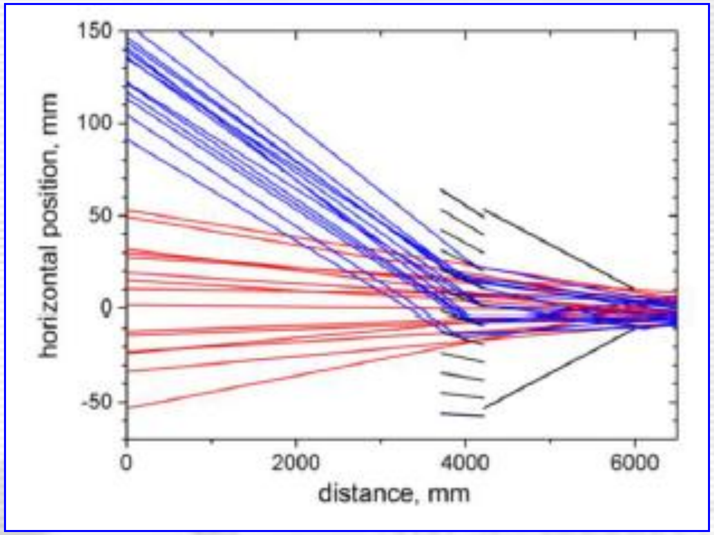
ID	Distance [m]	frequency [Hz]	beam width/height [mm]	Type (*)	window width [deg]
Pulse shaping					
PSC1	6.45	168	20/80	MB	144
PSC2	6.6	168	20/80	MB	144
PSC3	6.9	168	20/80	MB	144
PSC4	7.65	168	20/80	MB	144
Pulse multiplexing					
MCa	8.95	42 … 280	20/80	MB	16 x 4°, distance 22.5°
MCb	9.00	42 … 280	20/80	MB	4 x 4°, distance 90°
MCc	9.50	42 … 70	20/80		1 x 180°, followed by 7 x 4°, distance 22.5°
Wavelength definition					
FC1a	8.28	14/7	20/80	BB	70
FC1b	8.32	63/70	20/80	BB	180
FC2a	79.55	14	40/80	BB	180
FC2b	79.59	7	40/80	BB	90

(*) ball bearing (BB), magnetic bearing (MB)

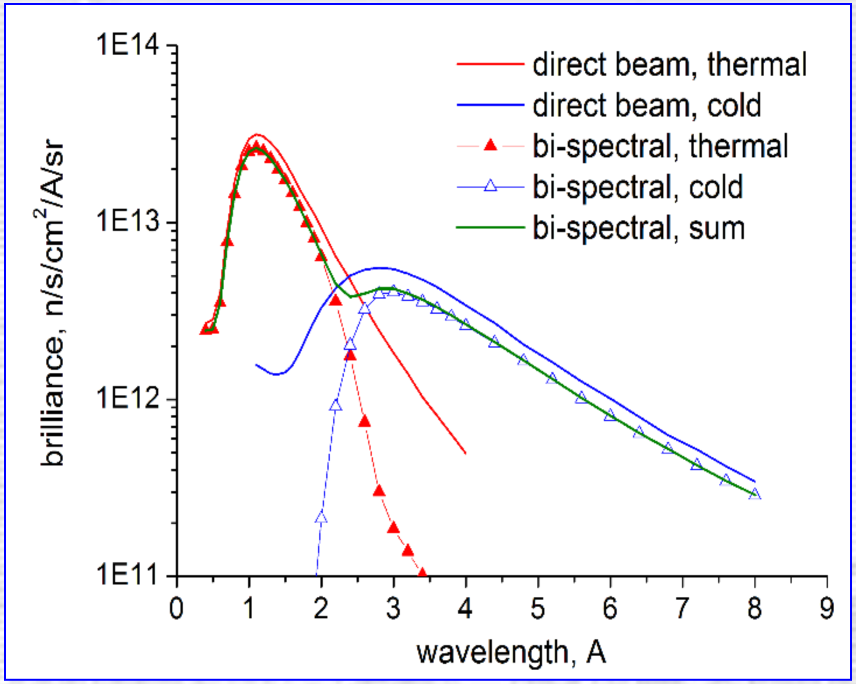
PSC – pulse shaping chopper	**4**
MC – multiplexing chopper	**3**
FC – frame chopper	**4**

Materials engineering diffractometer BEER, ESS

(Diffraction + SANS + Imaging)



Bi-spectral extraction multichannel (m=4) guide

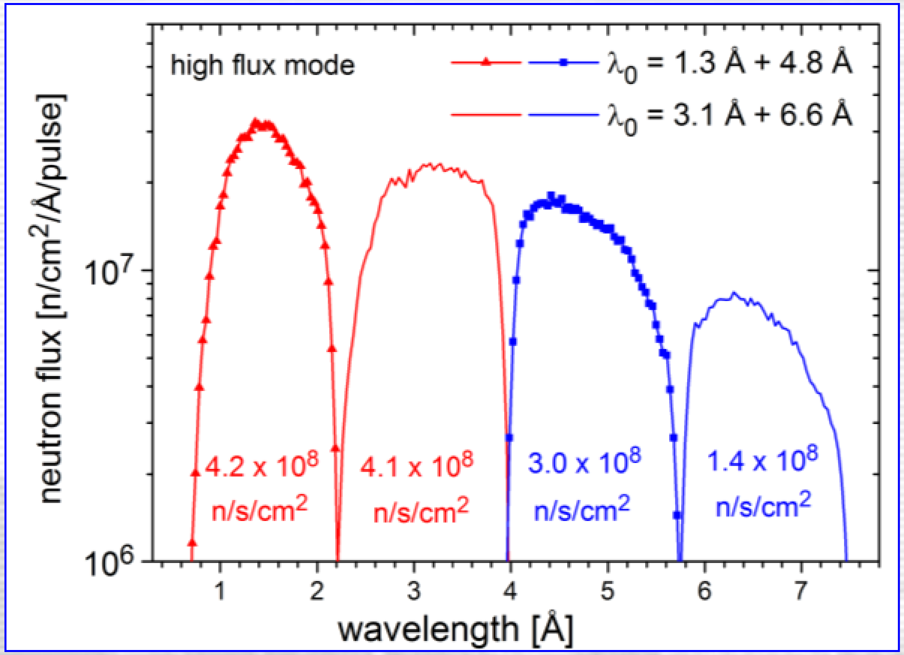


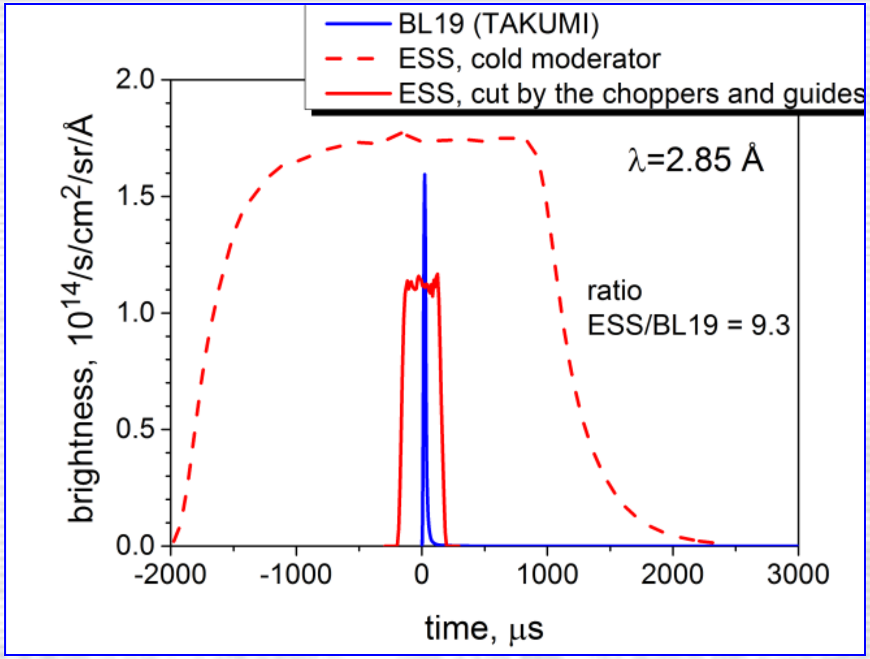
Simulated neutron spectra at the sample position

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

Materials engineering diffractometer BEER, ESS

(Diffraction + SANS + Imaging)

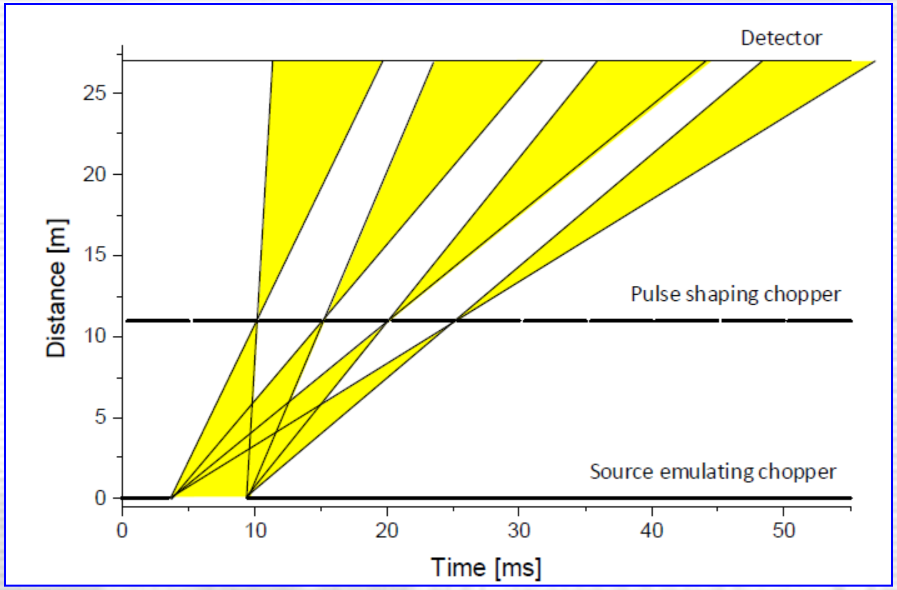


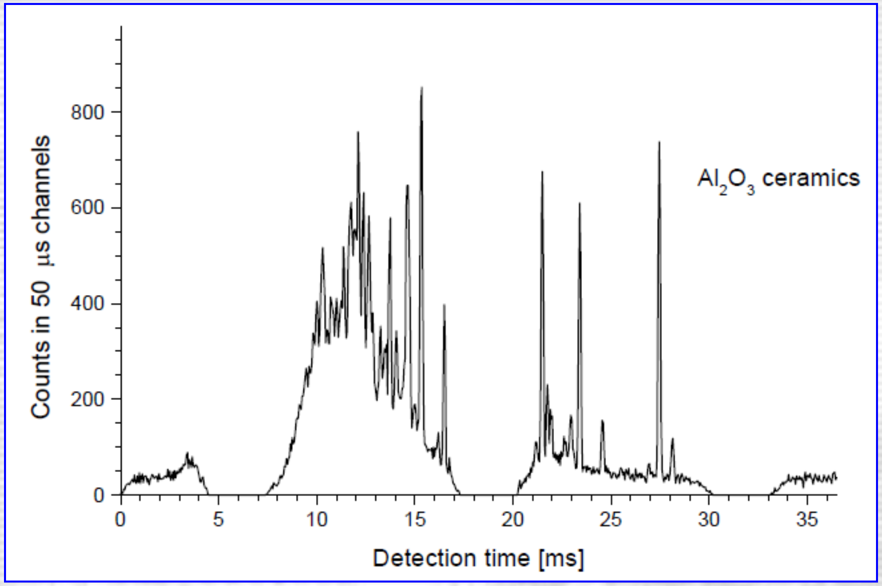


Simulated wavelength distribution of neutron flux in direct beam at the detector distance in the pulse suppression mode. The solid lines show the spectrum after phase shifting of the choppers by one source period.

Brightness and time structure of the J-SNS (BL19) and ESS pulses at λ=2.85 Å. The red dashed and solid lines show the ESS full pulse and a cut made by choppers and guides of the BEER instrument in medium resolution (0.3%) mode, respectively.

Multiplexing technique at pulsed neutron sources of LPS type





Wavelength Frame Multiplication (WFM) diffractometry.

SEC: W_0 = 5200 µs, *f* = 8 Hz

PSC: Δt_0 = 139 µs, *f* = 200 Hz, L = 10.6 m

Time distribution of the neutron counts at the detector in synchronous source – pulse shaping chopper operation. Black-out periods are equal to 2900 µs. Al_2O_3 sample.

Figures from M. Russina at al. J. of Physics (2012)

Diffractometers at ESS (first stage):

1) Powder hybrid
2) Polarized, single crystal
3) Powder
4) Engineering
5) Macromolecular, single crystal

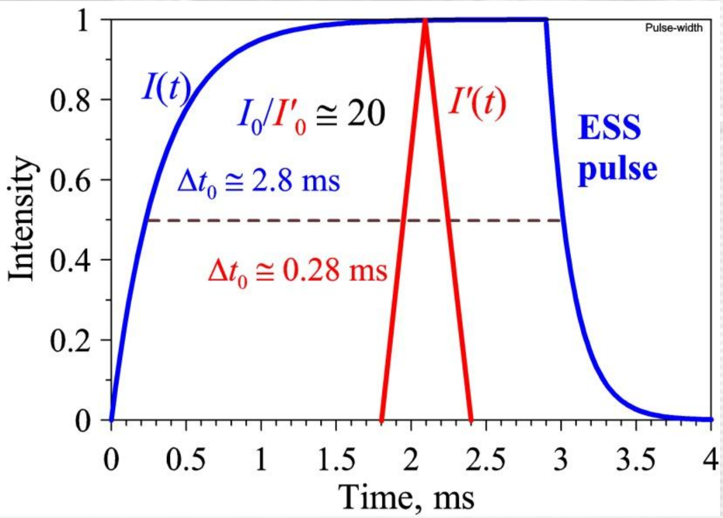
1. Limited number at the 1st stage: 5 altogether
2. Bi-spectral extraction: $(\lambda_1)_{max} \approx 1.2$ Å, $(\lambda_2)_{max} \approx 3$ Å
3. Very long flight path: 76 / 160 m
4. Detector solid angle: ~4 sr, $\Omega \rightarrow 4\pi$ (12 sr)
5. Combination of (Diffraction + SANS + IM)
6. Focusing on *in situ*, *real-time* mode of data acquisition
7. Complicated chopper system: ~(6 – 11) choppers of different assignments
8. Extremely high cost: $(12 \div 20) \cdot 10^6$ Eu
9. Not on the list: High-pressure, Single crystal, Texture, High-Q

Basic parameters of NEPTUN (Booklet, 2018), SNS and ESS

	NEPTUN	SNS	ESS
1. Time-average flux density:	$(0.5 \div 12)\cdot 10^{14}$	$0.1\cdot 10^{14}$	$3\cdot 10^{14}$
2. Half-width of fast neutrons:	(20 ÷ 200) µs	(20 ÷ 50) µs	2860 µs
3. Pulse repetition rate:	(10 ÷ 30) Hz	60 Hz	14 Hz
4. Time-average power:	(5 ÷ 10) MW	1 MW	5 MW
5. Background power:	3.2 %	<1%	<1%
6. Number of beam ports:	20 – 32	22	42

The stock set of neutron diffractometers

	Instrument	Main issue	Moderator	Resolution
1.	**High-resolution**	**structure**	**60 K**	**High**
2.	**High-intensity**	***in situ, real-time***	**60 K**	**Medium**
3.	**High-pressure**	**micro samples**	**60 K**	**Medium**
4.	**Engineering**	**internal stresses**	**290 K**	**High**
5.	**Texture**	**multi phase**	**60 K**	**High**
6.	**Long period**	**macromolecular**	**30 K**	**Medium**



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

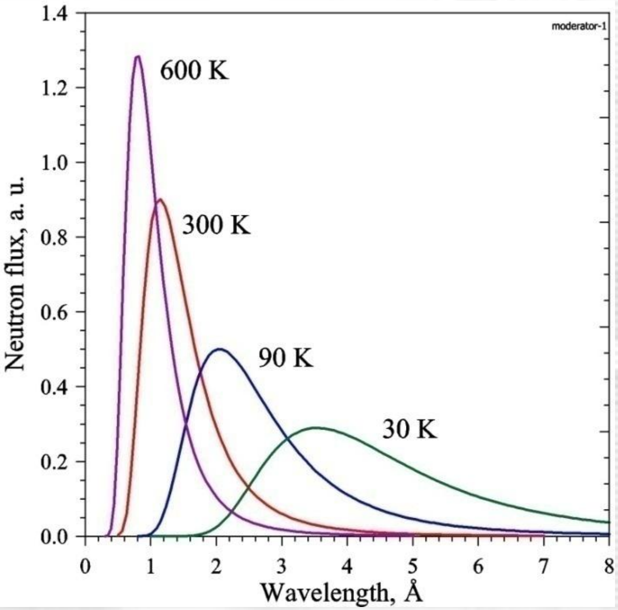
NEPTUN / ESS = $(5\cdot10^{14}/3\cdot10^{14})\cdot(2800/280) = 17$,

if shortening of the ESS pulse to 280 μs is done

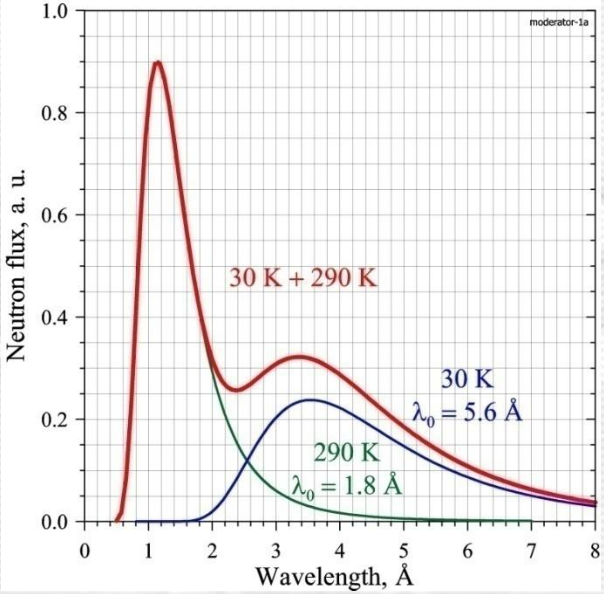
without frame multiplication system

NEPTUN: possible options

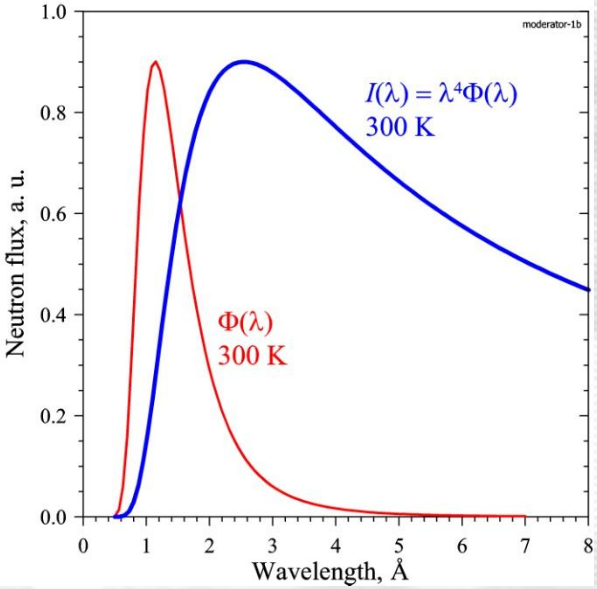
1. **Time-average flux density:** $(0.5 \div 12)\cdot 10^{14}$ → $\Phi_0 = 5\cdot 10^{14}$ n/cm²/s
2. **Half-width of fast neutrons:** $(20 \div 200)$ μs → $\Delta t_0 = 200$ μs
3. **Pulse repetition rate:** $(10 \div 30)$ Hz → $\nu = 10$ Hz
4. **Moderators (at least three):** thermal + cold (~90 K) + very cold (~30 K)
5. **Background power:** 3.2 % acceptable



Maxwellian distributions for (30 – 600) K



Bi-spectral distribution for 30 + 290 K



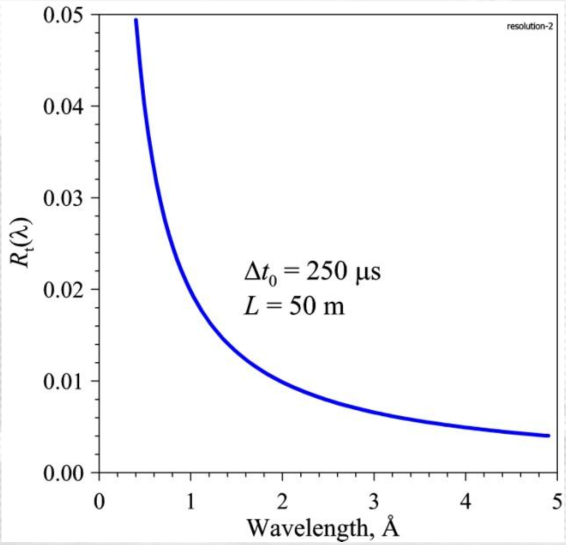
Maxwellian distribution and integral intensity

Resolution of a TOF diffractometer

$$(\Delta d/d)^2 = R_t(\lambda) + R_\theta(\theta) = (\Delta t_0/t)^2 + (\Delta\theta/\mathrm{tg}\,\theta)^2$$

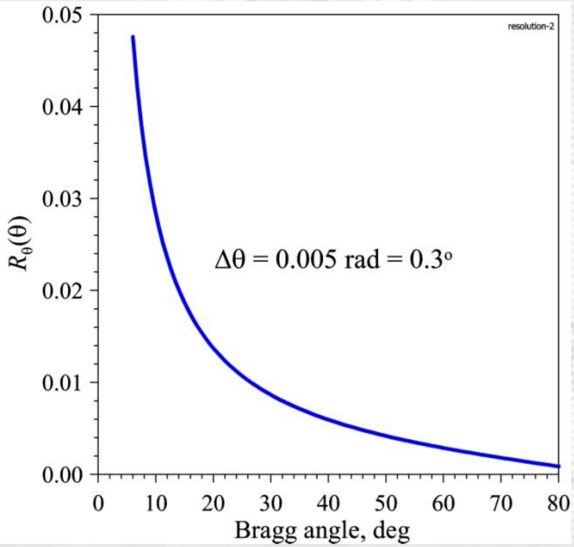
$R_t \Rightarrow 0$ if $\Delta t_0 \Rightarrow 0$ or $L \Rightarrow \infty$

$R_\theta \Rightarrow 0$ if $\Delta\theta \Rightarrow 0$ or $\theta \Rightarrow \pi/2$



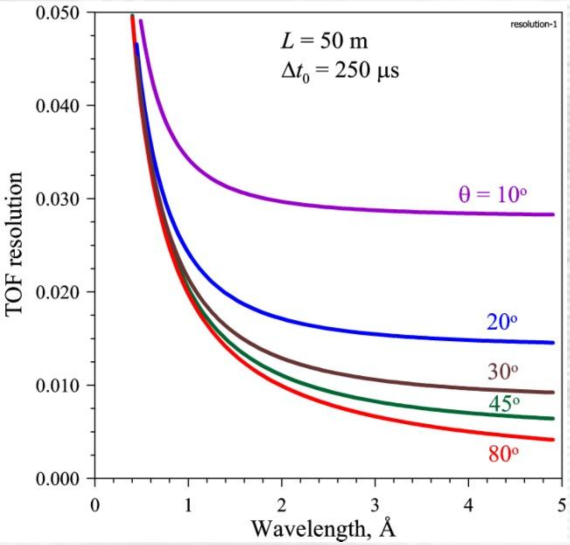
TOF component

+



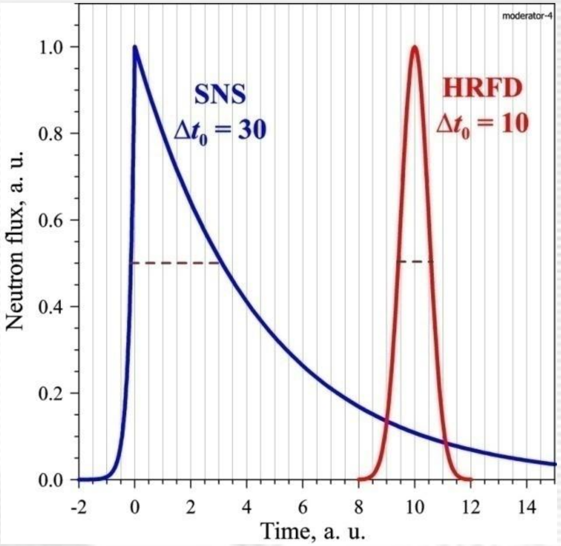
Angle component

→

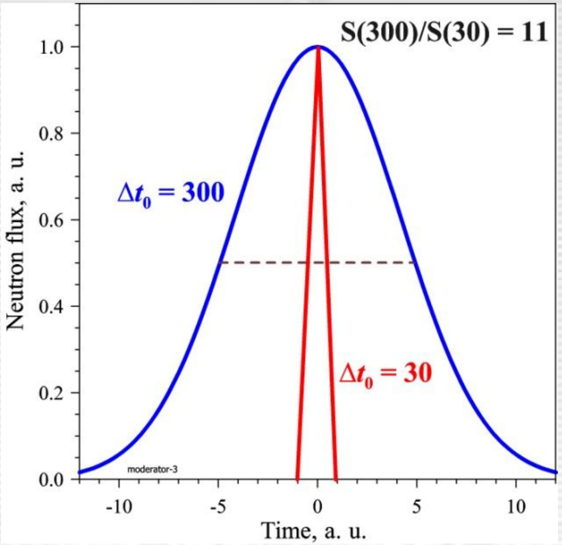


$\Delta d/d$ as a function of λ and θ

Resolution and shape of diffraction peaks



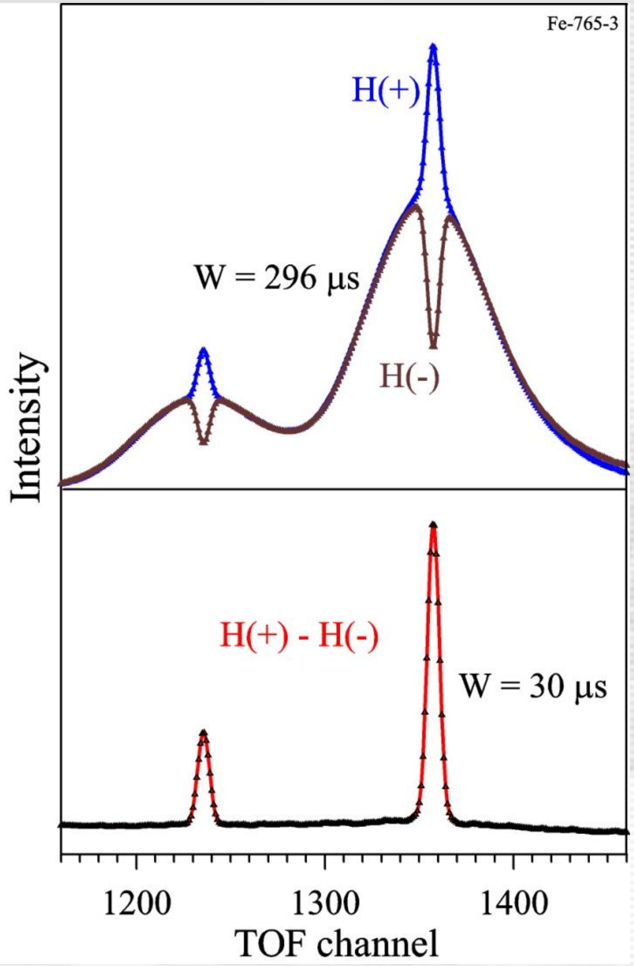
Comparison of SNS (30 μs) and HRFD (10 μs) peak shape



Pulse shaping 300 → 30 μs

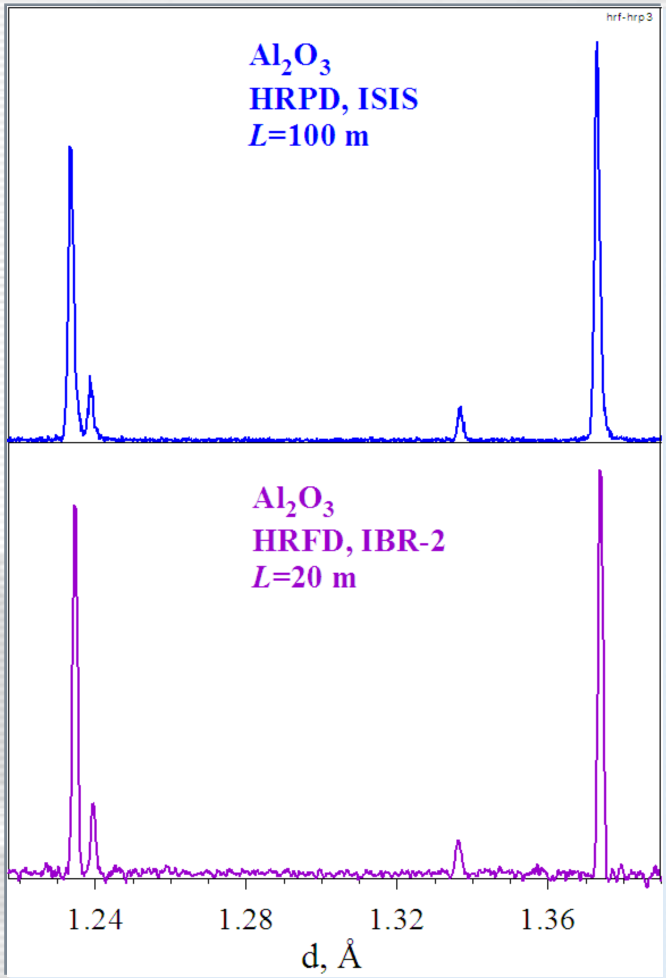
For TOF: $I(300)/I(30) = 11$, $\Delta x = 14$ mm, $L_1 = 60$ m, $f = 248$ Hz

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

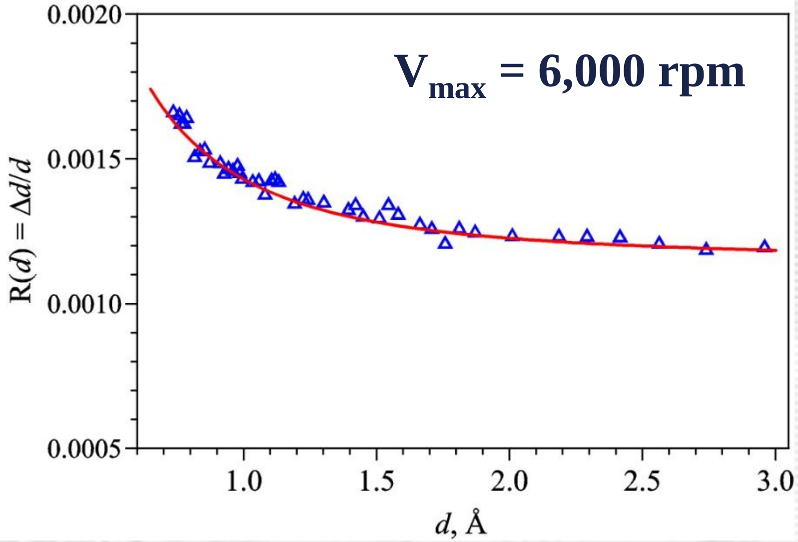


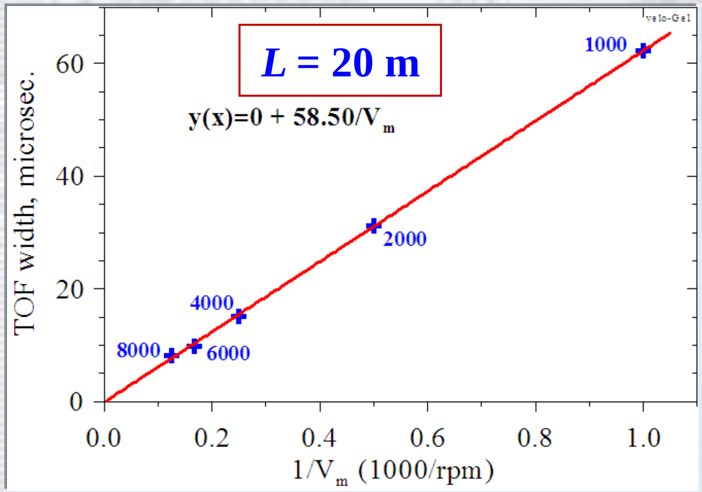
Two closely situated peaks (TOF channel width is equal to 4 μs) measured with HRFD.

HRFD resolution



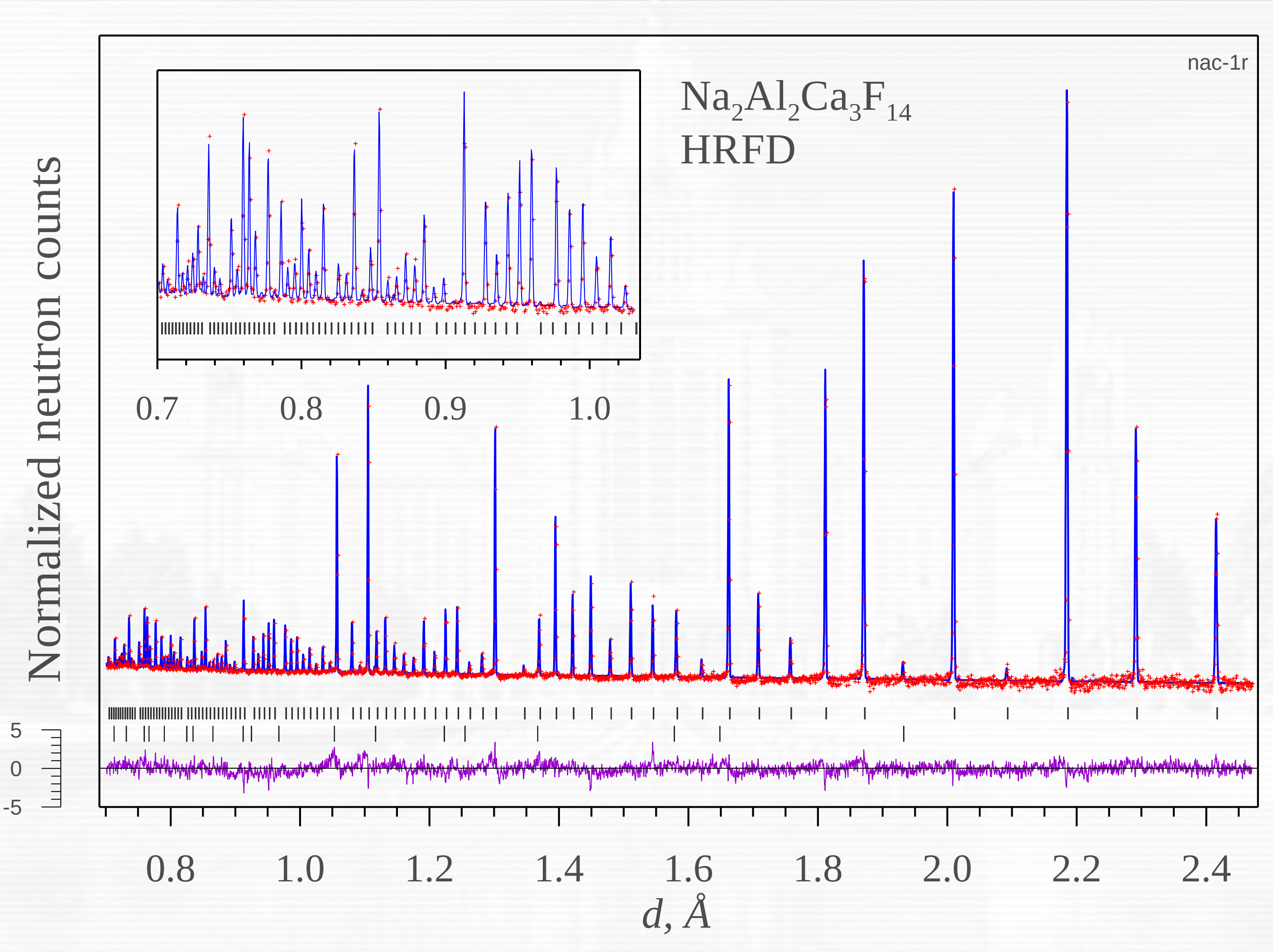
Diffraction patterns of Al_2O_3 measured at ISIS (UK) and IBR-2 (Dubna). Resolution is the same, despite L is 5 times longer at ISIS.





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

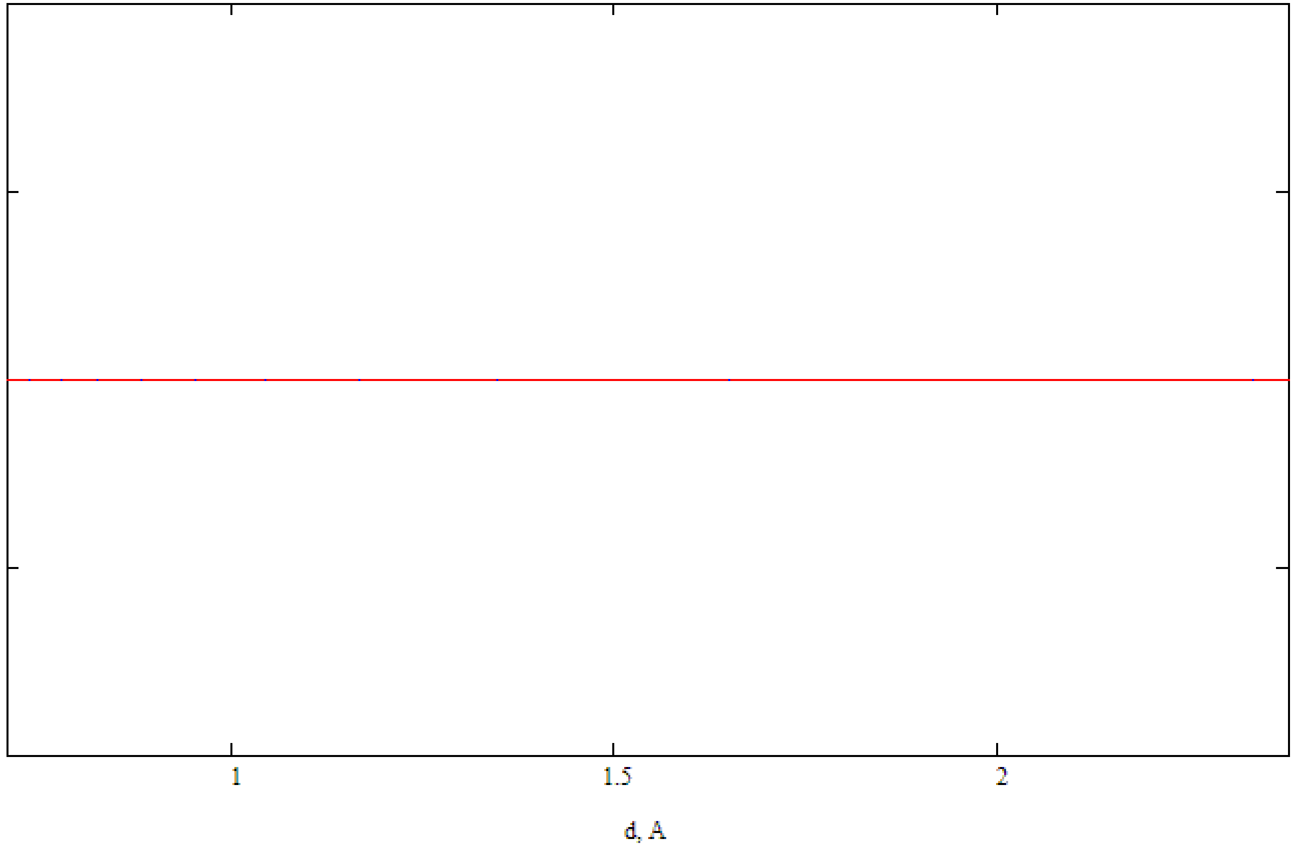
Rietveld analysis of the HRFD data (MRIA package)



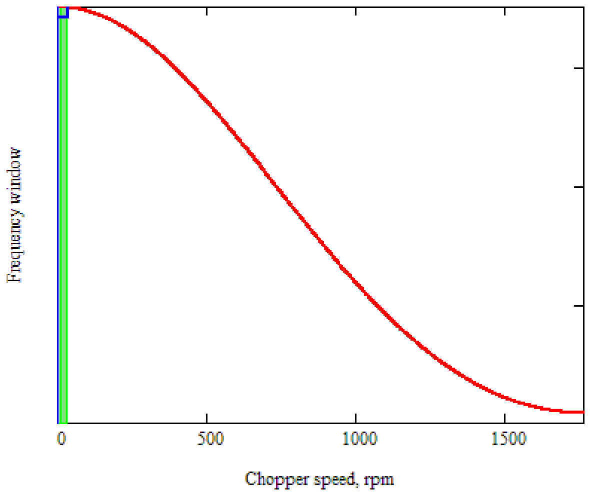
Diffraction pattern obtained with NAC-standard

Simulation: RTOF data acquisition

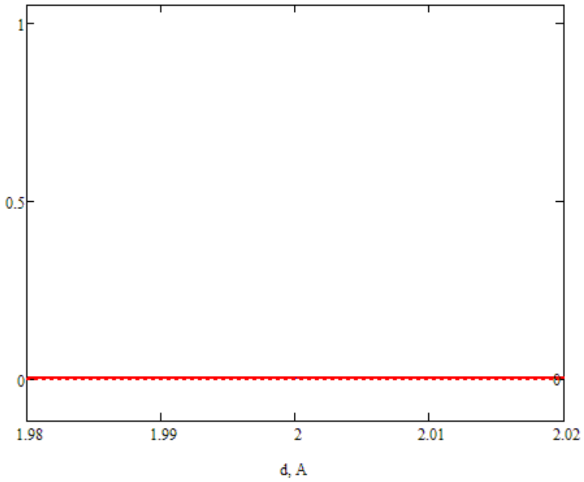
Current chopper speed is 0 rpm or 0 % of max. speed 1760 rpm



Elapsed time / Total time: 0 %



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



Выводы по TOF-дифрактометрам

1. <u>Продолжается бурное развитие</u> (усложнение) систем формирования пучка нейтронов (замедлителей, нейтроноводов, прерывателей) и систем регистрации (детекторов, электроники).

2. <u>Новые тенденции</u>: биспектральные пучки, комбинация дифракции и МУРН, ориентация на эксперименты в реальном времени.

3. В конструкции дифрактометров на ESS <u>не просматриваются</u> ограничения по финансированию.

4. Перспективы дифракции нейтронов на источнике DNS-IV выглядят весьма <u>многообещающе</u>. По совокупности основных характеристик (интенсивности, разрешению, диапазону переданных импульсов) дифрактометры на DNS-IV <u>могут</u> превосходить дифрактометры на SNS, J-SNS и ESS!

5. Многообещающие перспективы будут реализованы <u>только при наличии</u> адекватных детекторов.