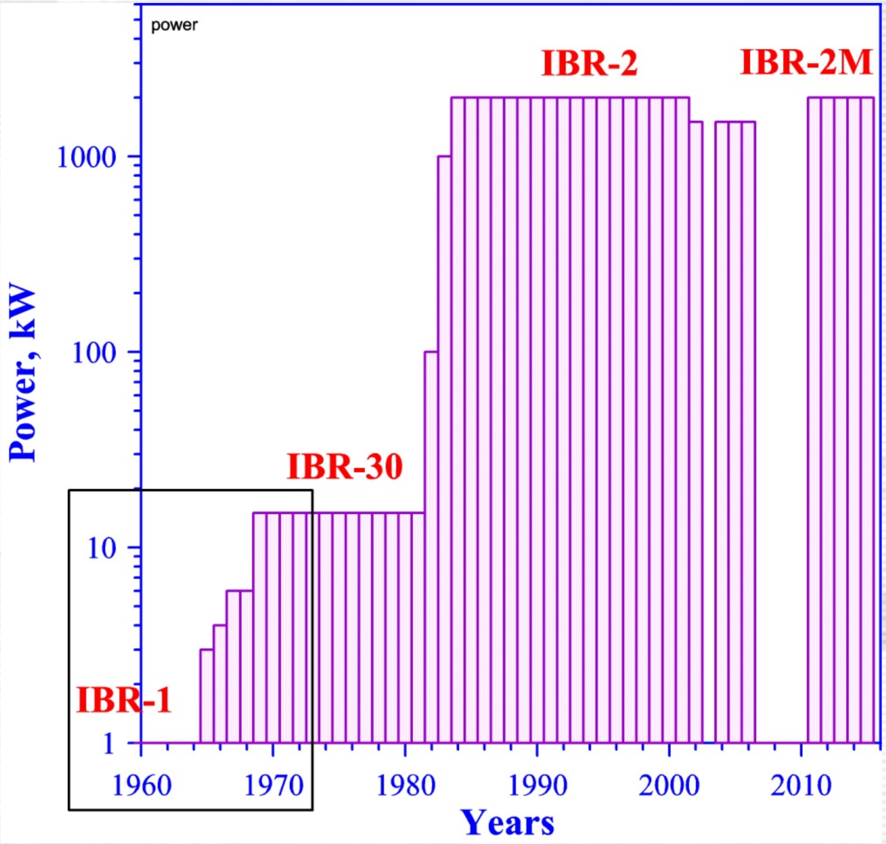
Fyodor L. Shapiro
and neutron scattering condensed matter studies

Fyodor L. Shapiro
1915 - 1973



The IBR-1 pulsed reactor was put into operation in June 1960 at W = 1 kW

bala@nf.jinr.ru

Pulsed neutron sources (reactors and spallation) from 1960 to 2020

I.	JINR, Dubna, Russia, IBR-1 (1960 - 1980, 1 – 15 kW) long pulse	closed
II.	Tsukuba, Japan, KENS, (1980, 4 kW) short pulse	closed
III.	ANL Argonne, USA, IPNS (1981, 7 kW) short pulse	closed
IV.	JINR, Dubna, Russia, IBR-2 (1984, 2000 kW) long pulse	operational
V.	RAL, UK, ISIS (1985, 200 kW) short pulse	operational
VI.	Los Alamos, USA, MLNSC (1985, 50 kW) short pulse	operational
VII.	Oak Ridge, USA, SNS (2008, 1200 kW) short pulse	operational
VIII.	Tokai, Japan, J-SNS (2008, 1000 kW) short pulse	operational
IX.	Beijing, China, Ch-SNS (2017, 200 kW) short pulse	under construction
X.	Lund, ESS (2020, 5000 kW) long pulse	under construction

Twice Dubna was the first: IBR-1 is the first intense pulsed source,
IBR-2 is the first 3rd generation pulsed source with W > 1 MW

Neutron scattering for condensed matter

Neutron scattering in condensed matter is used for studying its structure and dynamics at atomic level. For this low energy neutrons are used: typical energy and wavelength are about of 0.02 eV and 2 Å. The neutron has unique properties that yield distinct information unavailable otherwise.

Main topics:

- atomic structure
- magnetic structure
- soft matter
- atomic and magnetic dynamics
- applied research

Main techniques:

- diffraction
- small angle scattering
- reflectometry
- inelastic scattering
- imaging

$V(\mathbf{r}) = 2\pi\hbar^2 \cdot (b_j/M_j) \cdot \delta(\mathbf{r} - \mathbf{r}_j)$ – potential (Fermi)

$\sigma(\boldsymbol{\kappa}) = (2\pi)^3/V_c^2 \cdot |F(\mathbf{H})|^2 \cdot \delta(\boldsymbol{\kappa} - 2\pi\mathbf{H})$ – cross-section

$\mathbf{b_j}$ is coherent scattering length

$$I_{hkl} \sim |F_{hkl,\,nuc}|^2 + M_{hkl}^2 \cdot |F_{hkl,\,mag}|^2$$

Condensed matter studies with slow neutrons

Main experimental techniques at FLNP's pulsed reactors

- **Inelastic scattering** – **1962**
- **TOF diffraction on crystals** – **1963**
- **Small angle scattering** – **1974**
- **Reflectometry** – **1986**
- **Neutron imaging** – **2012**

Condensed matter studies at IBR pulsed reactors: first steps

1958 – Workshop in Dubna "Science at the IBR pulsed reactor"
I.M.Frank – **F.L.Shapiro** – E. Janik discussions

1959 – visit of I.M.Frank to Poland, invitation of Polish physicists to come to Dubna

1960 - 62 – first Polish physicists at Laboratory (Z.Ogzhewalski, I.Sosnovska, E.Sosnovski, A.Bajorek et al.)

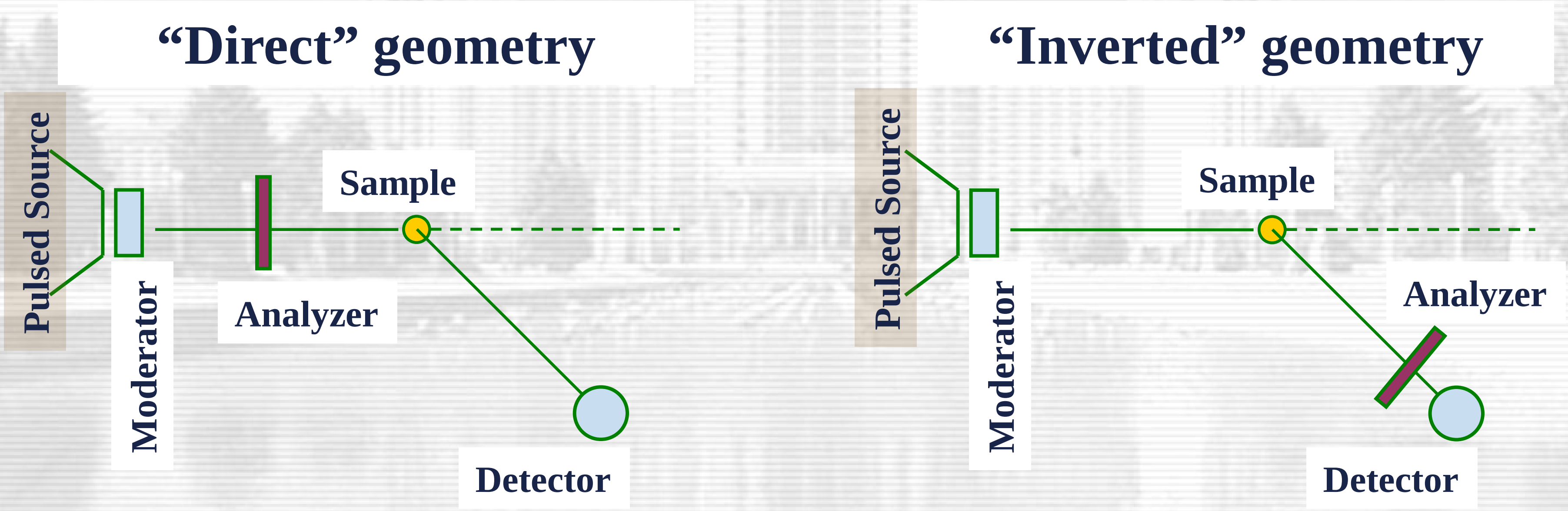
1961 – 62 – first inelastic scattering experiments (T.A.Machekhina, V.V.Golikov, V.V.Nietz)

1962 – first publication T.A.Machekhina, Z.Ogzhewalski, **F.L.Shapiro** "Neutron scattering in graphite and LiF" JINR Communication 952, Dubna, 1962

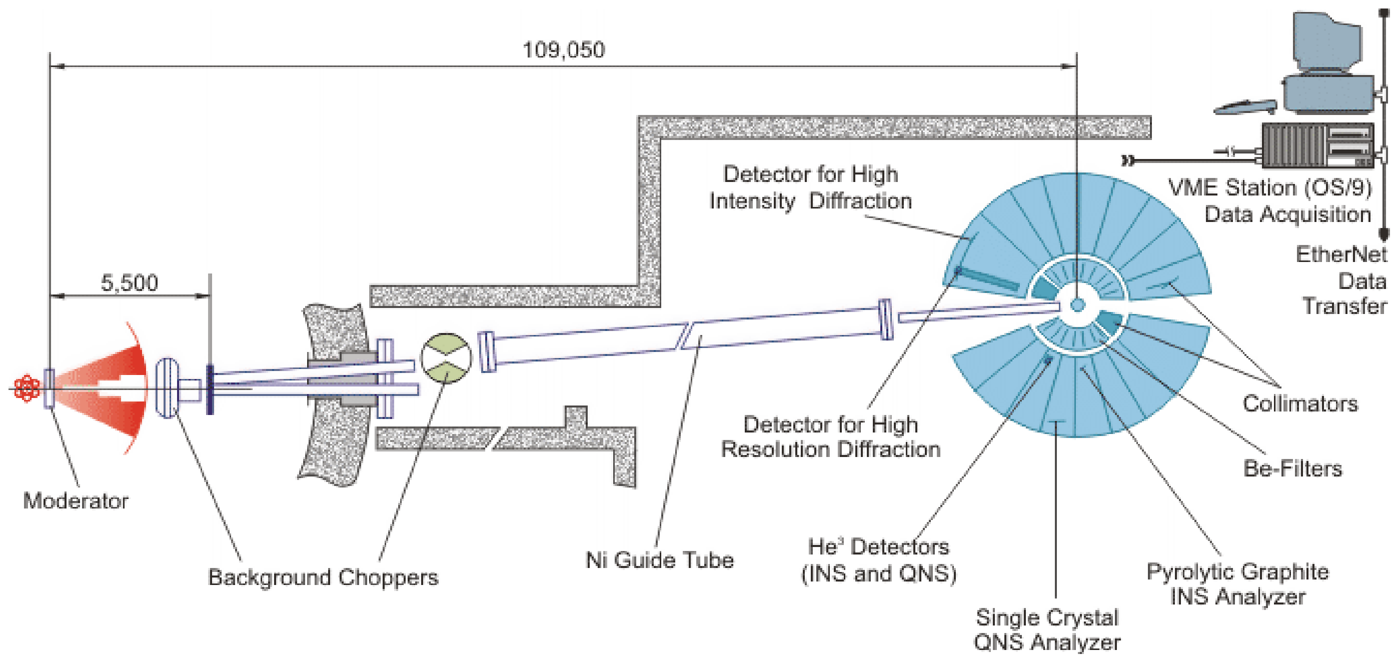
Inelastic scattering: "Direct" and "Inverted" geometry, 1962

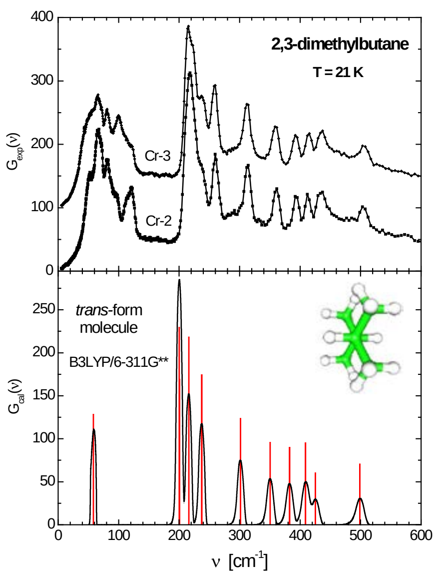
T.A.Machekhina, Z.Ogzhewalski, F.L.Shapiro "Neutron scattering in graphite and LiF" JINR Communication 952, Dubna, 1962. "Inverted" geometry.

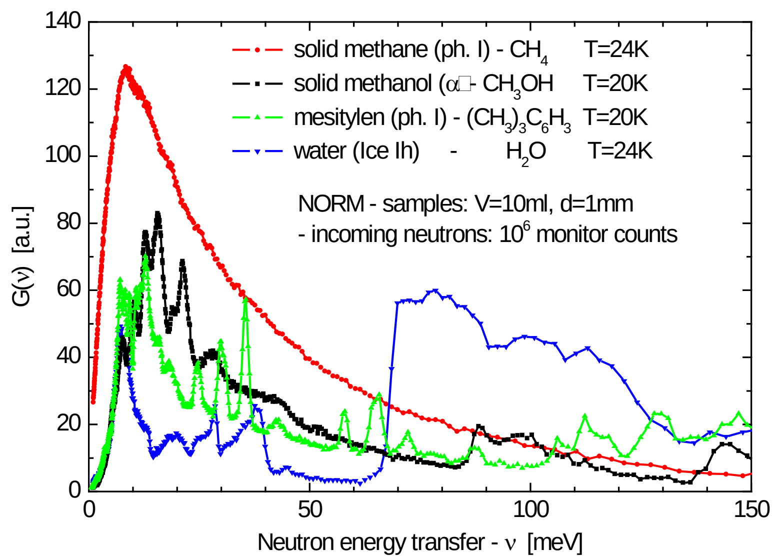
V.V. Golikov, F.L. Shapiro, A. Shkatula, E. Janik "Spectrometer for scattering of cold neutrons" JINR Communication 956, Dubna, 1962. "Direct" geometry.



NERA – inverted geometry spectrometer at IBR-2

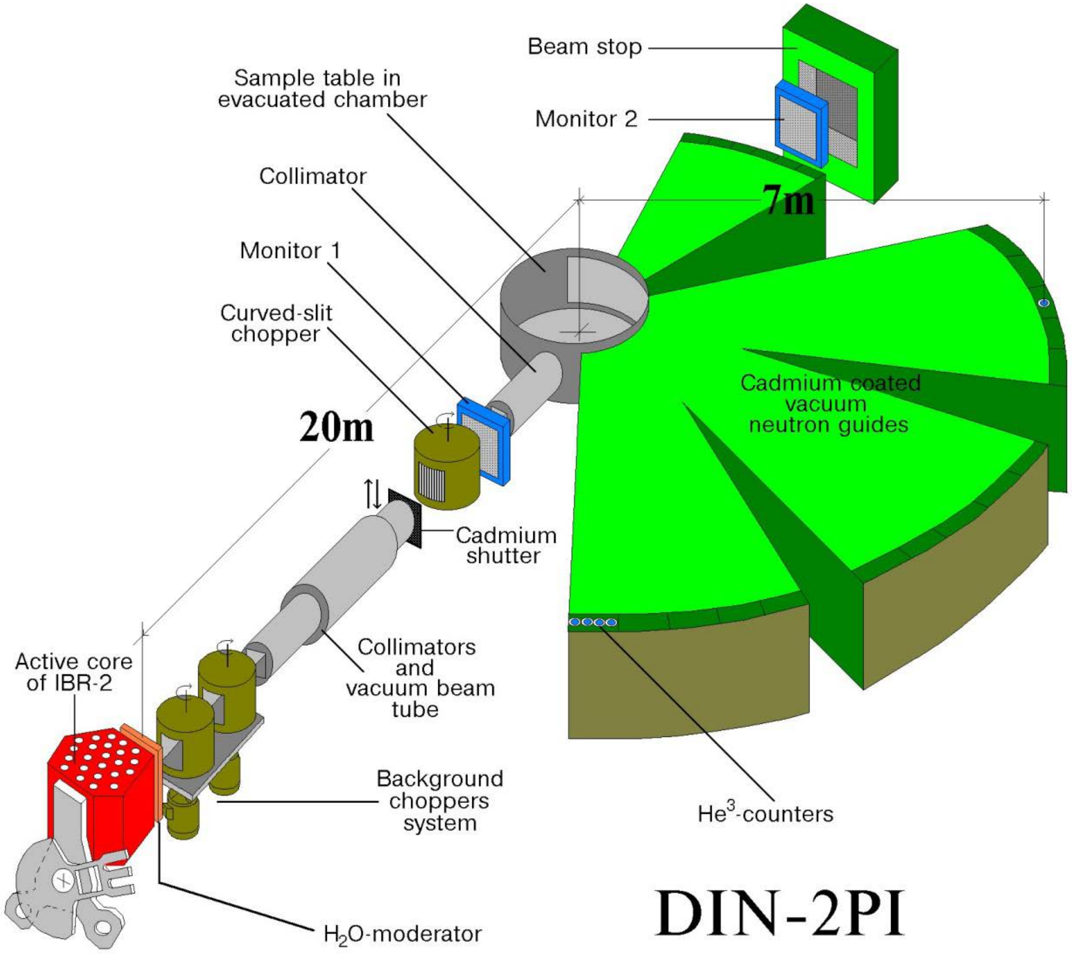


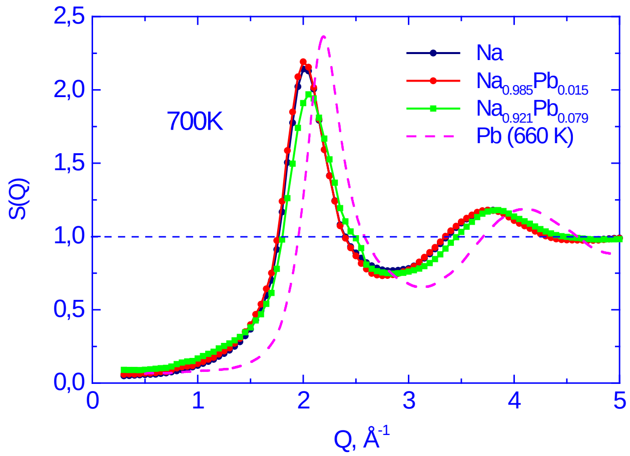




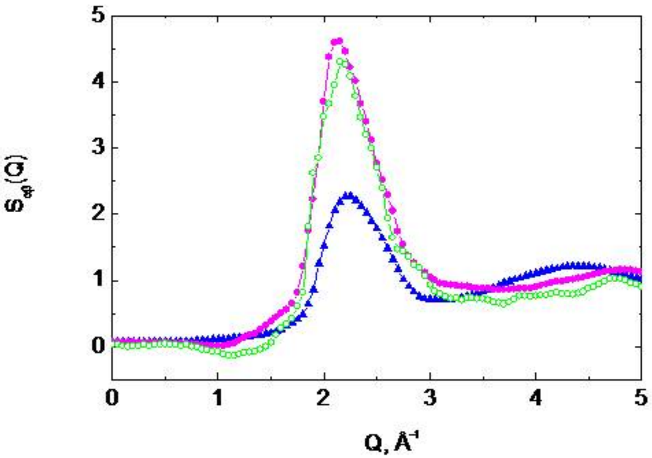
Examples of inelastic scattering spectra of organic compounds measured with NERA spectrometer.

Inelastic scattering spectrometer DIN-2PI at IBR-2





Structural factors of liquid sodium and Na–Pb melts (upper) and partial structural factors of liquid lithium and nitrogen for $Li_{1-x}N_x$ melts (lower) measured with DIN-2PI spectrometer.



TOF-diffraction. Principal steps

1954	P. Egelstaff	The idea of neutron TOF-diffraction, IUCr Congress, Paris
1956	R.D. Lowde	Theoretical justification, Acta. Cryst. 9 (1956) 151
1963	B. Buras	The first experiments at Swierk (Poland)
1963	Dubna	The first experiments at the IBR-1 (Buras, Nietz, Sosnowska, Shapiro)
1967	J. Carpenter & A. Holas	"Focusing of the TOF-diffractometer"
1968	R.M. Brugger	"We need more intense thermal-neutron beams"
1977	ZING-P´	The first spallation source, Argonne (USA) 1980 – KENS, 1981 – IPNS, 1985 – ISIS, 1985 – MLNSC
1984	IBR-2	The first pulsed source of the 3rd generation
1984	mini-SFINKS	The first RTOF-diffractometer, PNPI, Gatchina
1994	HRFD	The first RTOF-diffractometer at pulsed neutron source, Dubna


P. Egelstaff


F.L. Shapiro

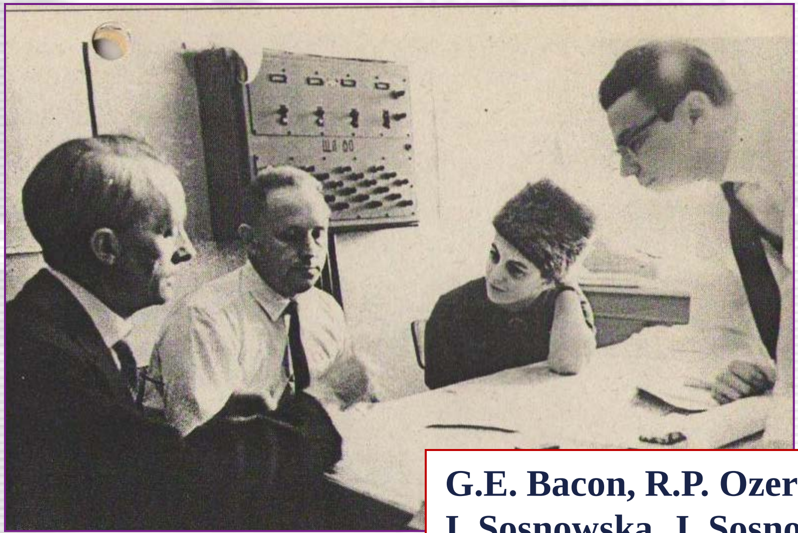

J. Carpenter

F.L. Shapiro: neutron diffraction papers

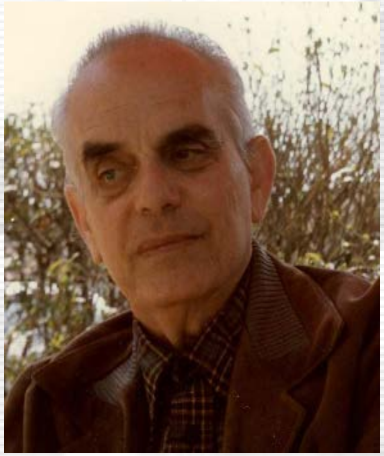
1. "**The Time-of-Flight Method for Neutron Crystal Structure Investigation and its Possibilities in Connection with Very High Flux Reactor**" Nukleonika, **1964**
Together with B. Buras, J. Leciejewicz, V.V. Nitz, I. Sosnowska, J. Sosnowski. Review paper

2. "**Neutron diffraction for crystal structure investigations**" JINR Communication, **1965**
Together with V.V. Nitz, I. Sosnowska, J. Sosnowski. Review paper

3. "**About phases of crystal structure factors**" Physics of Particles and Nuclei, **1972**
A new method of F(Q) phase determination. Original paper



G.E. Bacon, R.P. Ozerov,
I. Sosnowska, J. Sosnowski
(Dubna, 1965)



Bronislaw Buras (1915–1994)
One of the inventors of the time-of-flight (TOF) method for neutron diffraction.

TOF diffraction at the IBR-1 - IBR-30 pulsed reactors

NUCLEAR INSTRUMENTS AND METHODS 69 (1969) 173-180; © NORTH-HOLLAND PUBLISHING CO.

FOCUSING OF A TIME-OF-FLIGHT DIFFRACTOMETER FOR STRUCTURE ANALYSIS. THE EXPERIMENTAL CHECK

A. HOLAS*, J. HOLAS, E. MALISZEWSKI* and L. SEDLAKOVA†

Joint Institute for Nuclear Research, Dubna, USSR

Received 26 October 1968

An experimental check of focusing the neutron time-of-flight diffractometer for crystal structure analysis is presented. The obtained results proved the formulae for focusing. The intensity of the focused diffractometer was increased by an order of magnitude as compared with the previous diffractometer (with collimators) of the same resolution.

Nuclear Instruments and Methods 193 (1982) 617–621 617
North-Holland Publishing Company

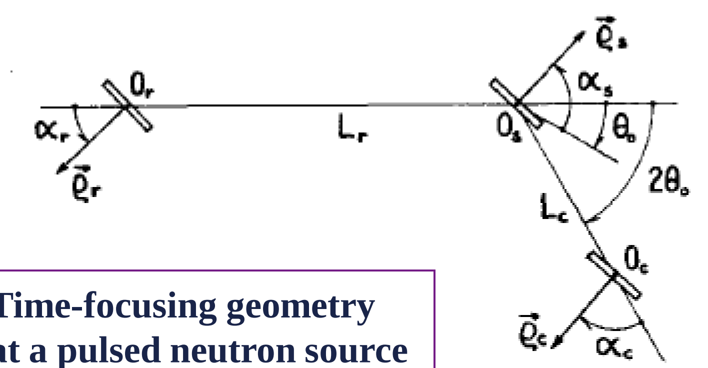
A NEUTRON TIME-OF-FLIGHT DIFFRACTOMETER WITH A ONE-DIMENSIONAL POSITION SENSITIVE COUNTER

A.M. BALAGUROV, V.I. GORDELIY, M.Z. ISHMUKHAMETOV, V.E. NOVOZHILOV, B.N. SAVENKO and V.D. SHIBAEV

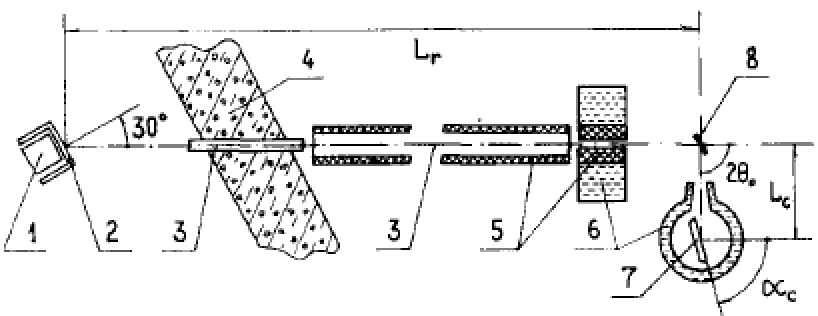
Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, U.S.S.R.

Received 20 March 1981

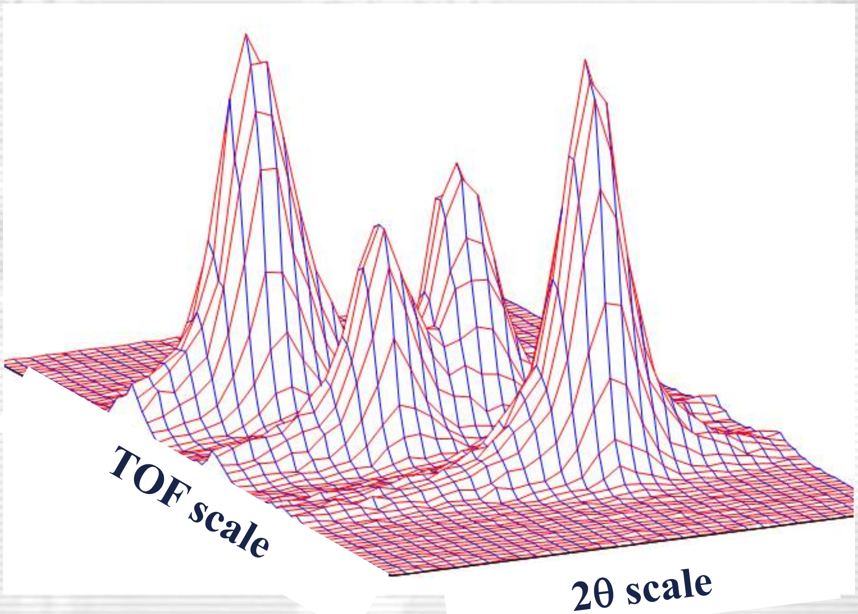
A neutron TOF diffractometer using one-dimensional position-sensitive ^{3}He counter is described. The position readout uses a resistive wire with charge division to detect the position. For calculations a digital processor is used. As an example the ferroelectric KD_2PO_4 is investigated and results are given.



Time-focusing geometry at a pulsed neutron source

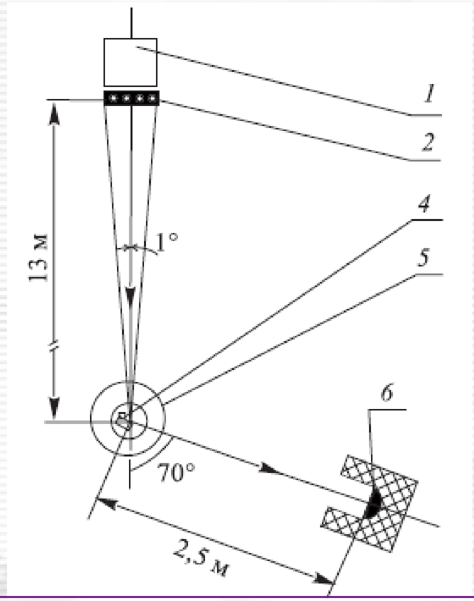


The first time-focused TOF diffractometer at the IBR-1 pulsed neutron source (Dubna)

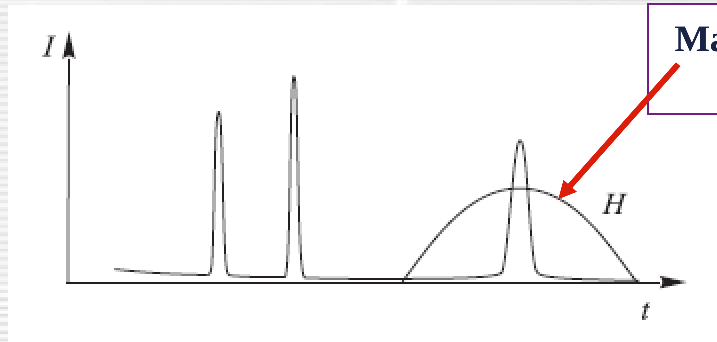


Continuous scanning along TOF and 2θ axes. Several thousands points are measured simultaneously.

Diffraction under pulsed magnetic field

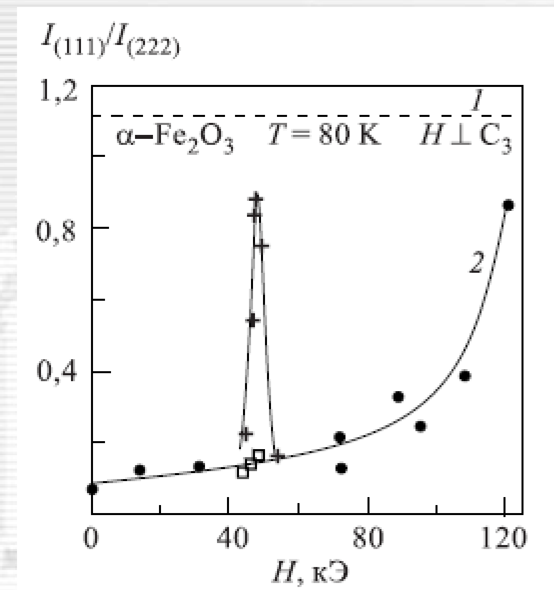


Lay-out of diffractometer with high magnetic field (up to 120 kOe) at the IBR-30 reactor



Magnetic field pulse with amplitude up to 250 kOe (at IBR-2)

Diffraction lines and pulse of magnetic field. Dependence of diffraction intensity on field amplitude is measured.

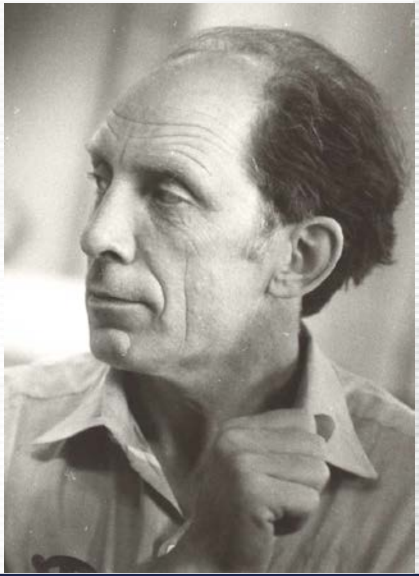


Anomaly increase of diffraction peak intensity for magnetic field amplitude of 49.5 kOe

Up to 1990 neutron diffraction studies with high pulsed magnetic field of about 250 kOe were performed at LNP JINR only. After that such studies were started at КЕК (Japan) and TRIGA Reactor (Vienna) with magnetic field of 160 kOe and 230 kOe, correspondingly.

Condensed Matter Department and spectrometers at the IBR-2

1963 - 64 – discussions about the IBR-2 reactor are started



Yu.M. Ostanevich
25.07.1936 – 01.08.1992

1967 – Condensed Matter Department?

1970 – CMD headed by Yu. Ostanevich is organized

1972 - 1979 – new spectrometers at IBR-30

1980 – 1982 – spectrometers are transferred at IBR-2

1980 – 1984 – first neutrons from IBR-2 – nominal power (2 MW)

1991 – 9 neutron spectrometers are operational at IBR-2

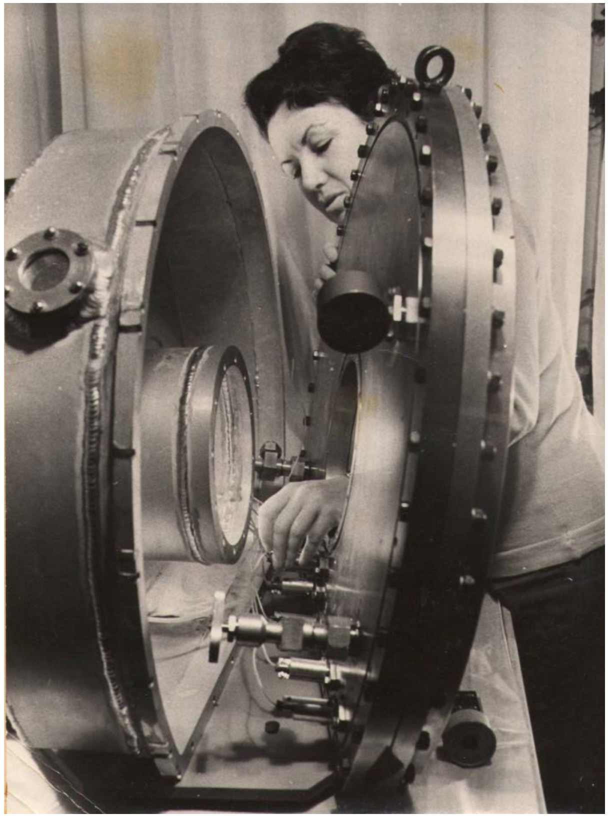
SANS spectrometer at the IBR-30 reactor: slit geometry

Yury Ostanevich and Laslo Cser at the first SANS detector (1974). Slit geometry, ^{3}He-counters inside a tube.

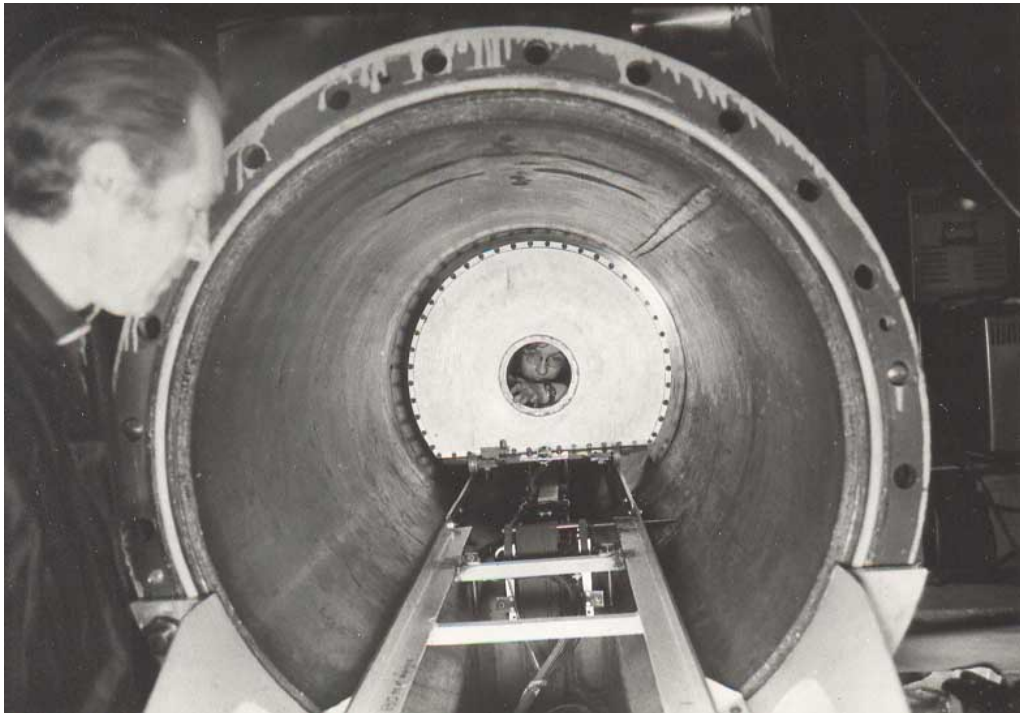
SANS spectrometer with axial geometry at IBR-30 reactor

I.A. Gladkikh, A.B. Kunchenko, Yu.M. Ostanevich, L. Cser

“Investigation of SANS in axially symmetric geometry” JINR, P3-11487, 1978

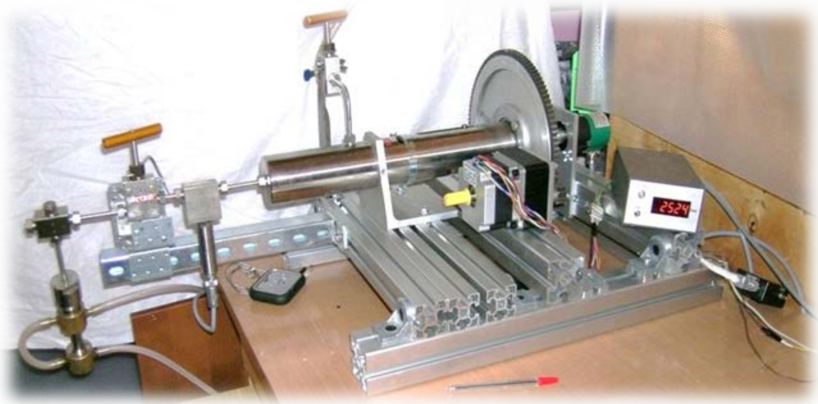


The ring detector is assembled by E. Pikel’ner

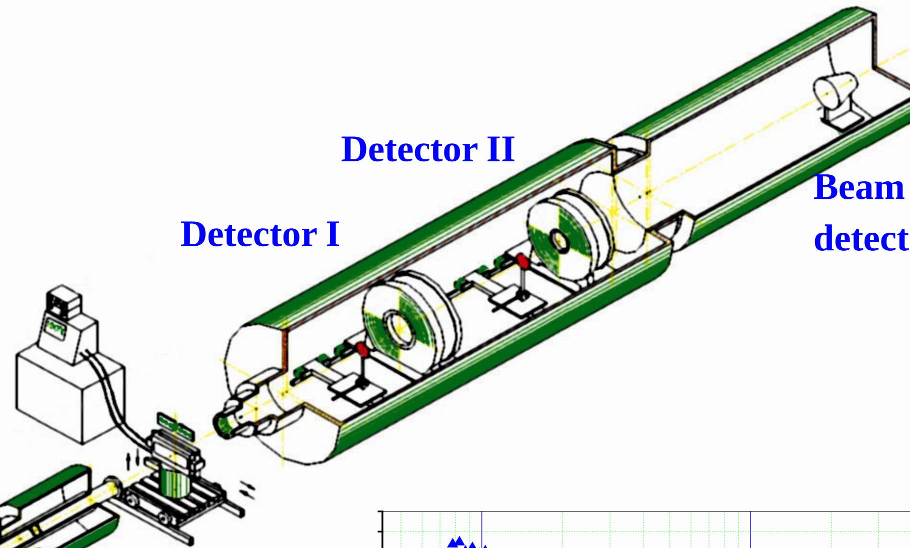


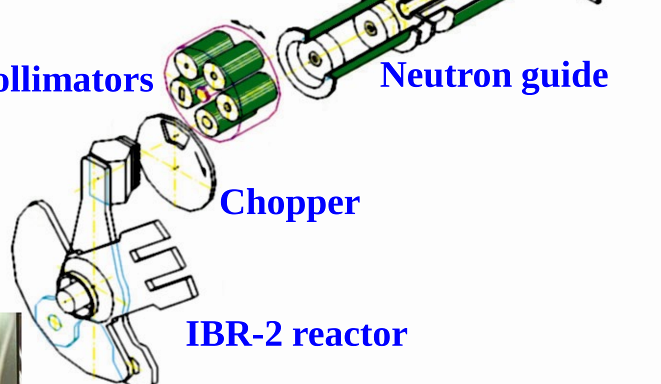
^{3}He ring detector inside the evacuated tube

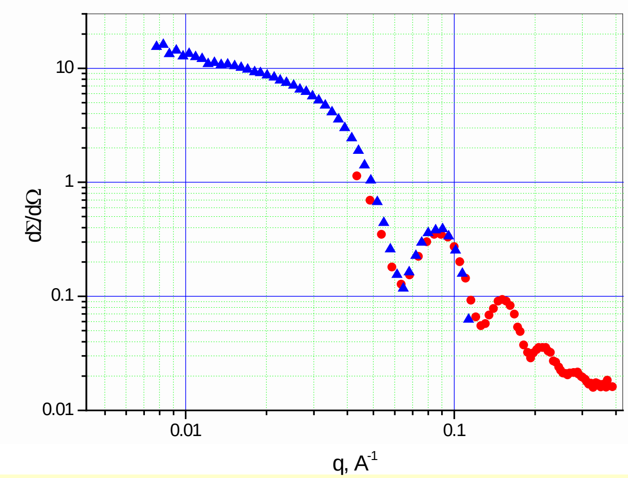
YuMO – small-angle scattering spectrometer:
diverse sample environment, very diverse scientific topics, record number of users



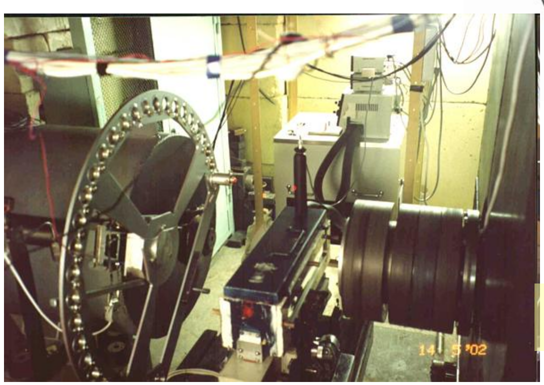
Setup for simultaneous P-V-T and
SANS investigations of liquid samples







SANS from diluted solution of apoferritin (spherical shell with R_{out}= 62 Å and R_{in}= 41 Å). Two detectors at 11.95 m (red) and 5.27 m (blue) were used.



Sample position

Neutron guides and polarized neutron beams at the IBR-2

early 1960s H. Maier-Leibnitz: invention to “conduct” neutrons in totally reflecting and curved tubes

1966 B. Alefeld: the first neutron guides at FRM in Garching

1969 F.L. Shapiro: we need neutron guides at IBR pulsed reactor for conventional and polarized beams

1970 ILL (K. Bendorfer): the first polarized neutron guide. Conclusion: no prospects in future!

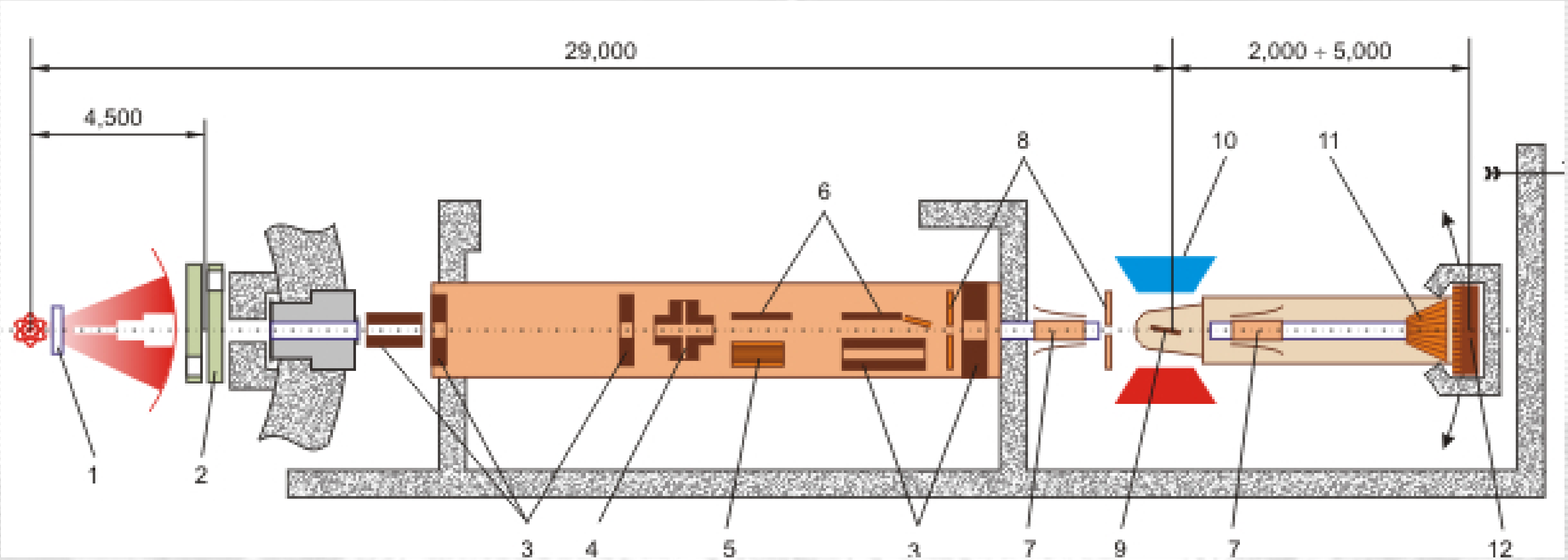
1972 – 76 G.M. Drabkin, D.A.Korneev et al. “Multilayer Fe-Co mirror polarizing neutron guide” for Dubna

1985 250 meters of neutron guides at IBR-2 (V.M. Nazarov)

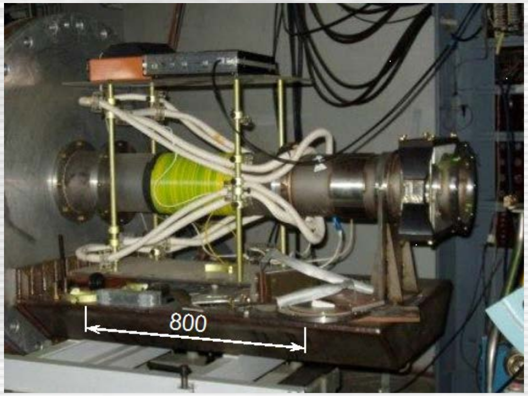
1989, 1995 Reflectometers SPN and REFLEX with polarized neutron beams

D.A. Korneev
08.03.1946 – 22.01.2002

Reflectometer REMUR (former SPN) at IBR-2

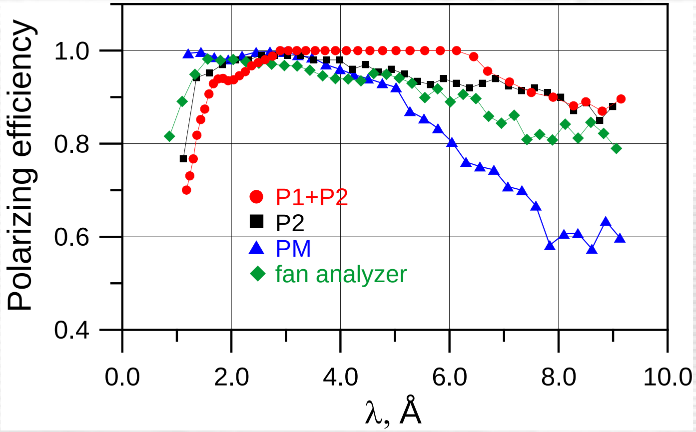


Adiabatic spin-flipper

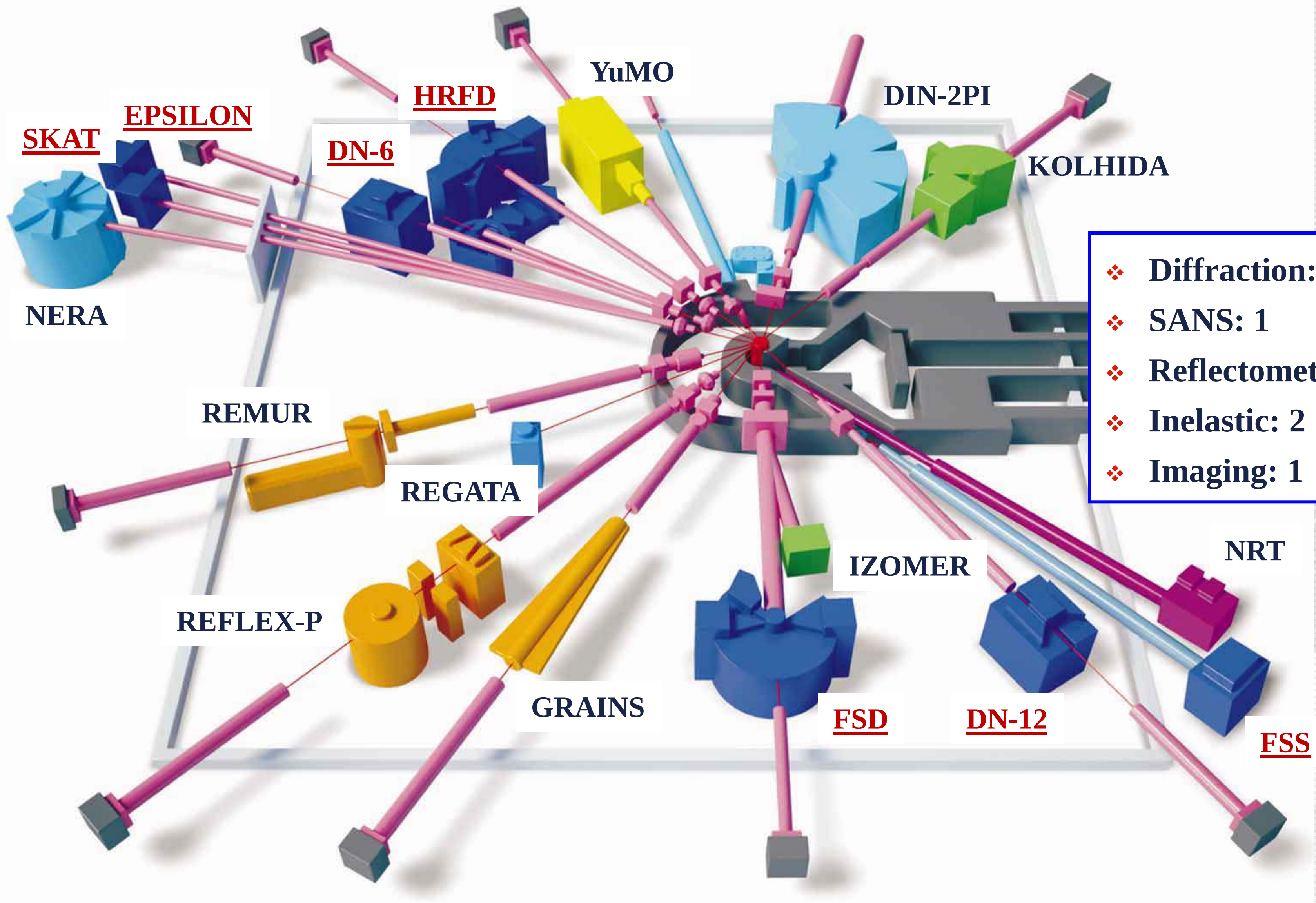


Analyzer of polarization

High polarization efficiency

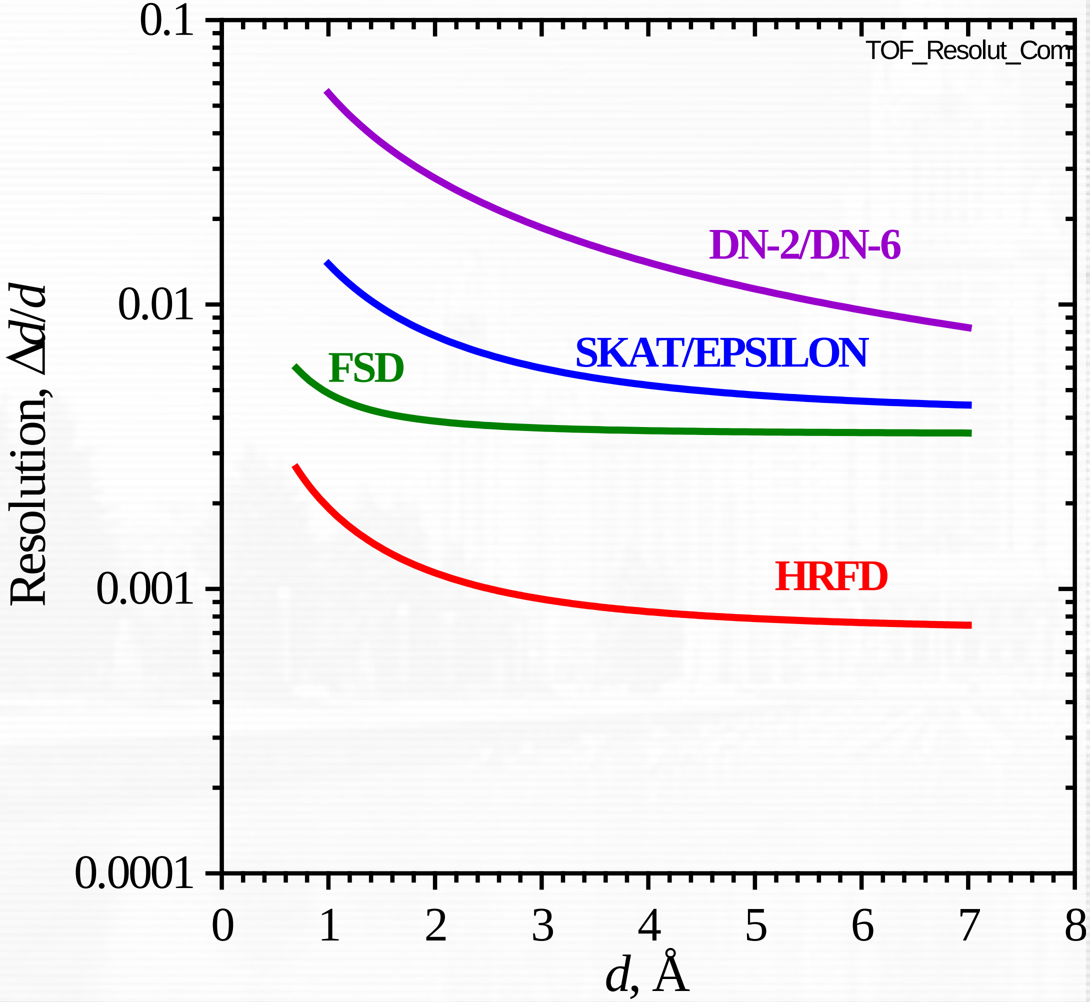


Spectrometers at the IBR-2 reactor



- Diffraction: 7
- SANS: 1
- Reflectometry: 3
- Inelastic: 2
- Imaging: 1

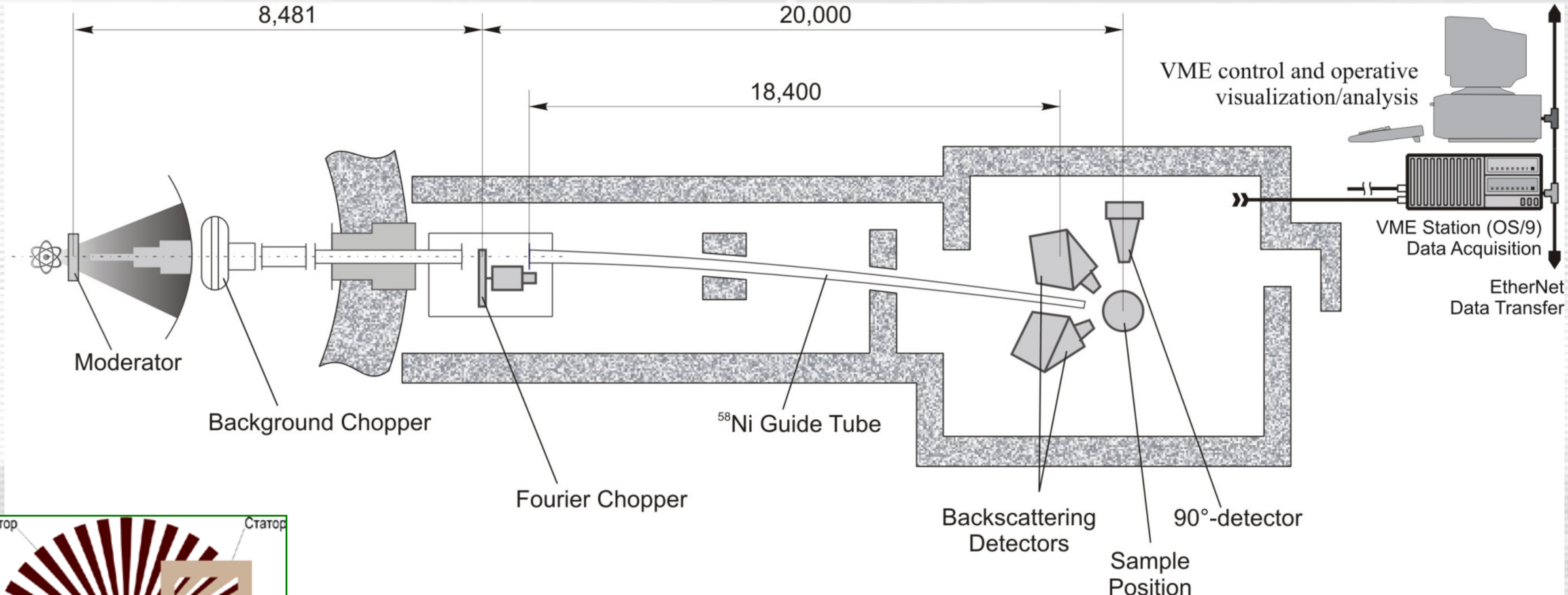
Optimization over resolution

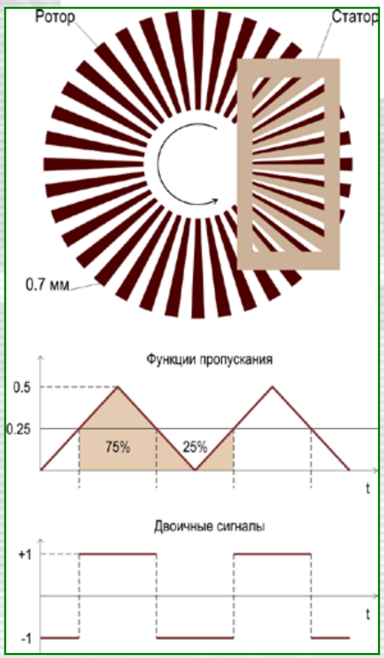


HRFD	**powders**
FSD	**internal stresses**
RTD	**real-time**
DN-6	**micro samples**
Epsilon	**internal stresses**
SCAT	**textures**

Resolution becomes better for longer *d*-spacing!

HRFD – High Resolution Fourier Diffractometer at IBR-2





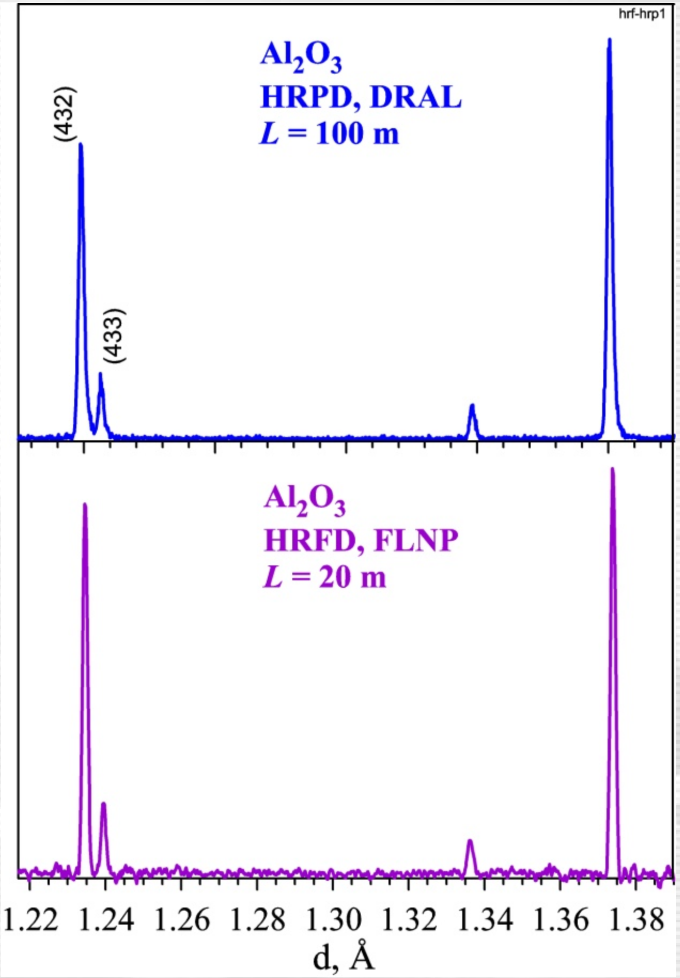
Put into operation in 1994 in collaboration between: FLNP (Dubna), PNPI (Gatchina), VTT (Espoo), IzfP (Drezden)

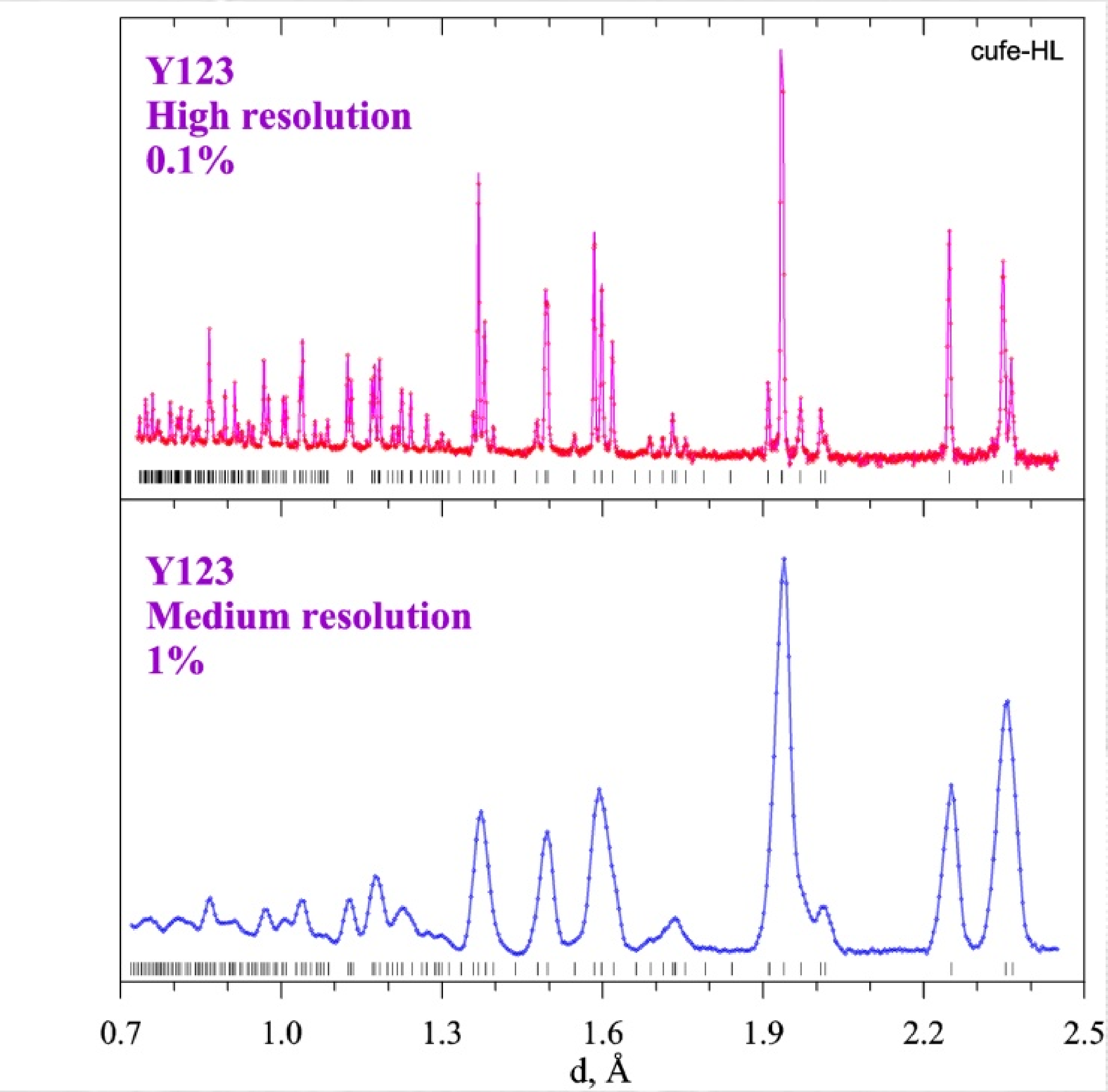
$\Delta t_0 \approx$ Const $\approx$ 10 μs, $L \approx$ 21 m, $\Delta t_0/t \approx 1\cdot10^{-3}/d = 5\cdot10^{-4}$ for d = 2 Å

<u>Advantages</u>: **very short flight path,**

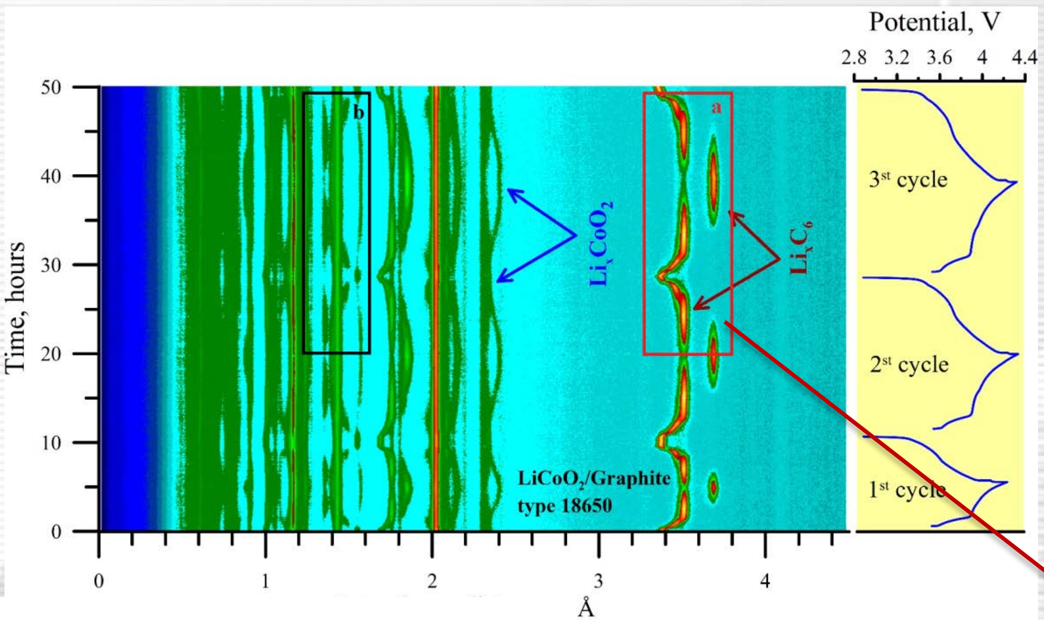
no pulse overlapping

HRPD / HRFD resolution test



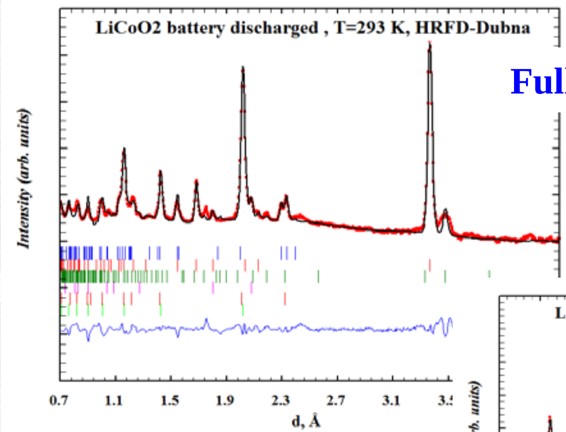


Lithium-Ion Battery of "Robiton" company

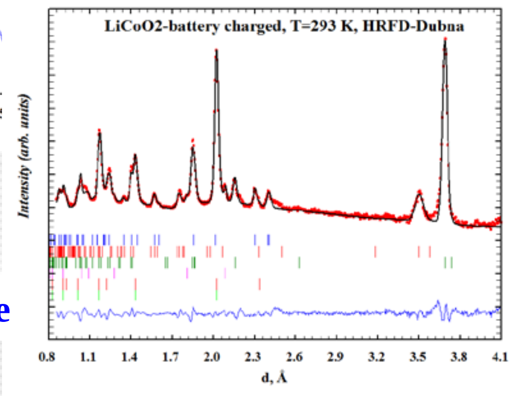


Commercial Li-ion battery of the ROBITON company, 18650 type

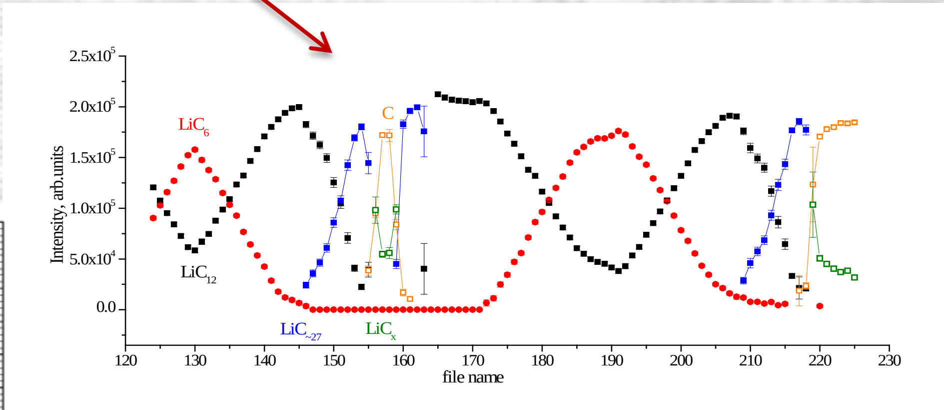
Evolution of neutron diffraction patterns of 18650 type battery in course of three charge-discharge cycles with various speed



Full discharge state



Full charge state



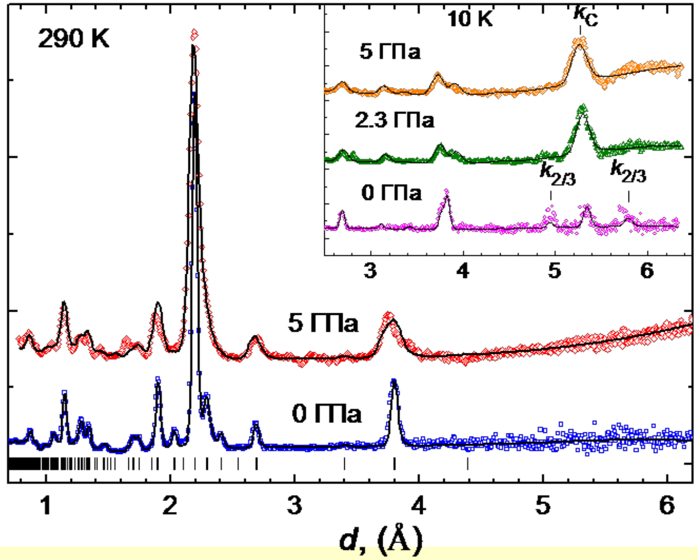
LiC_n phases: diffraction lines intensity changes during charge-discharge processes

High-pressure cells for neutron scattering

DN-12 diffractometer, IBR-2 reactor

Single-crystal anvil cell

P_{max} = 7 GPa (sapphire)
P_{max} = 30 GPa (diamond)
T = 0.1 – 300 K
V_s = 0.5 – 5 mm^3

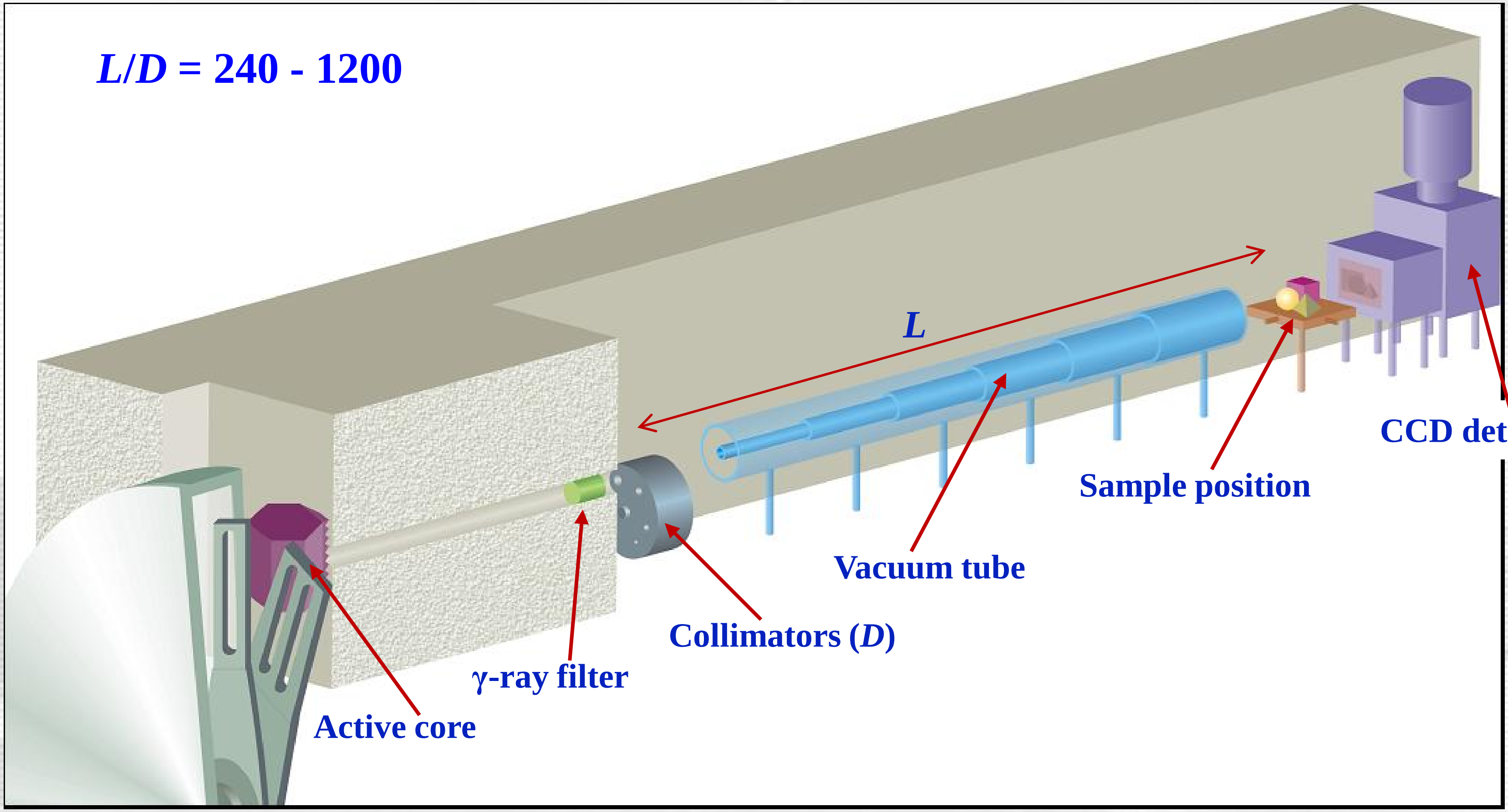


Neutron diffraction patterns of $La_{0.33}Ca_{0.67}MnO_3$ at P = 0 and 5 GPa and T = 290 and 10 K (insert). Sample volume is around 2 mm^3. Exposure time is 24 h. At high pressure and low temperature a complex AFM state is observed.

Paris – Edinburgh press

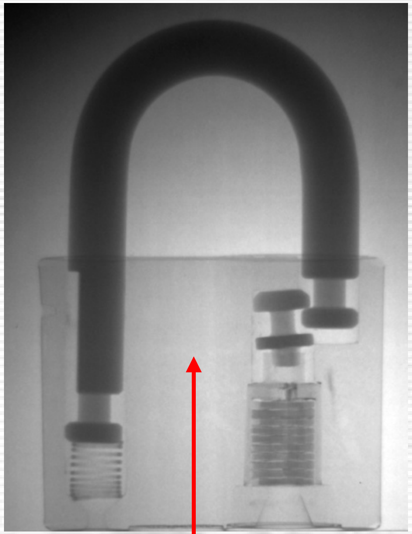
P_{max} = 10 GPa (WC)
P_{max} = 30 GPa (diamond)
T = 90 – 1000 K
V_s = 30 – 100 mm^3

New instrument for energy selective neutron imaging at IBR-2



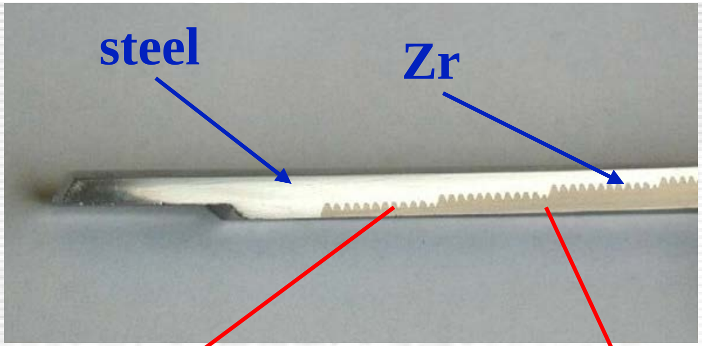
Main feature: wavelength contrast → studies of complex media

First neutron imaging experiments at IBR-2

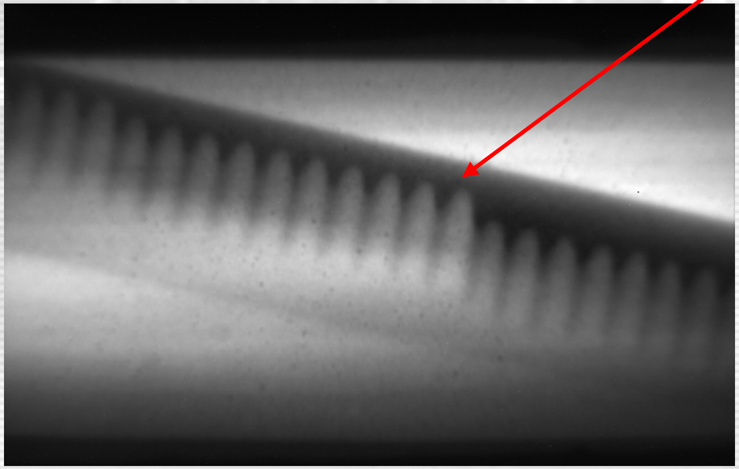


Defective door-lock

Cross-section of steel-zirconium adapter used in RBMK reactor components

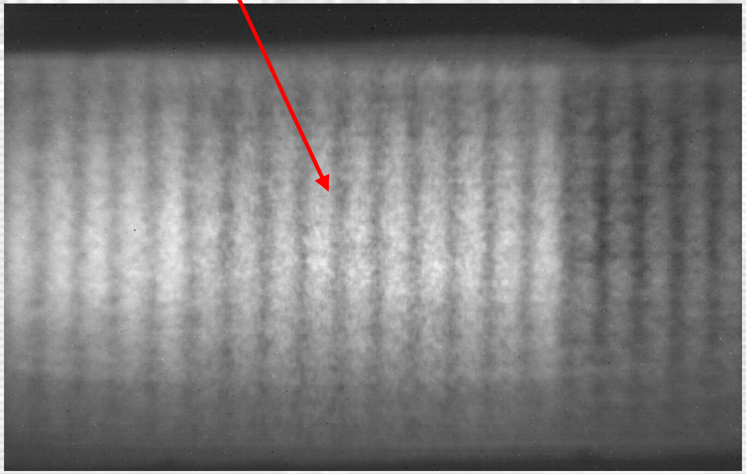


Small CCD camera used for experiments



Zr teeth are clearly seen by NI

$\Delta x \sim 150\ \mu m$



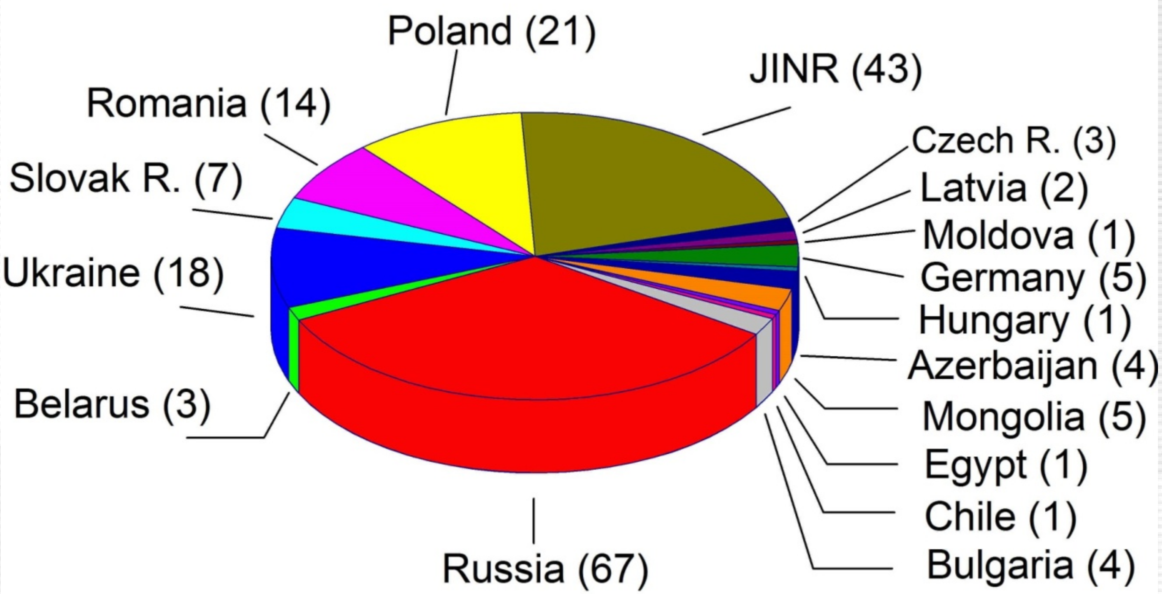
Conferences, Workshops, Schools

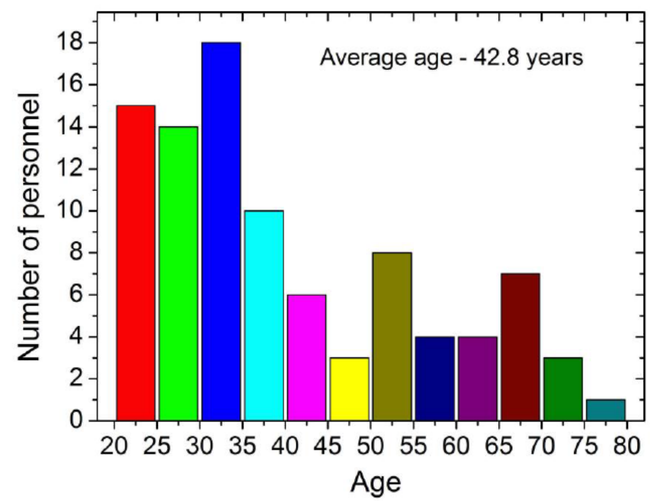
Structural Investigations at Pulsed Neutron Sources, Dubna, September 1 – 4, 1992

3rd IAEA Research Collaborative Meeting "Characterization and Testing of Materials" Dubna, May 13 – 17, 2013

Stress and Texture Investigations by Means of Neutron Diffraction Dubna, June 6 – 9, 2011

IBR-2 proposals in 2013: 200 applications from 17 countries





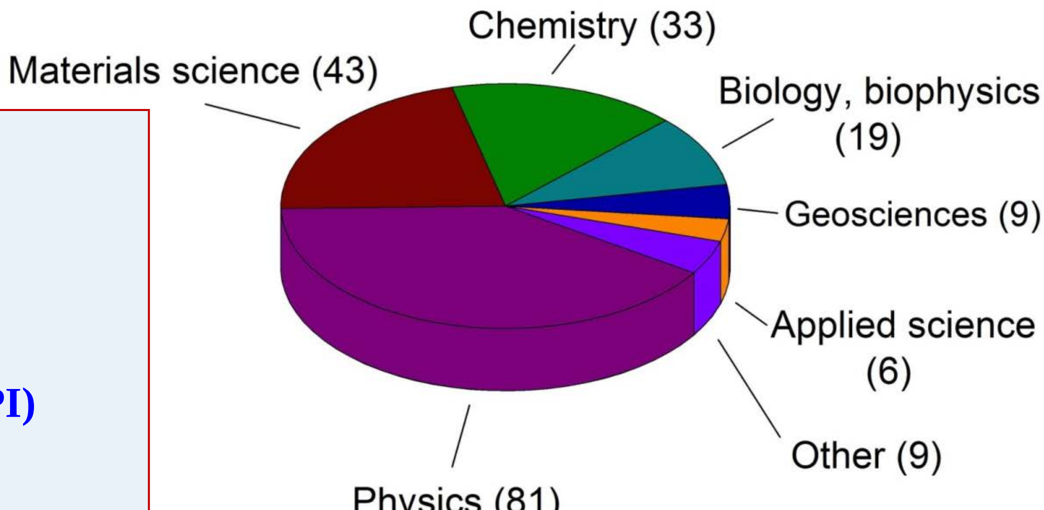
Condensed Matter Department in 2014

76 – Diffraction (HRFD, DN-12, FSD, DN-2, SKAT, Epsilon)

68 – Small angle scattering (YuMO)

35 – Inelastic scattering (NERA-PR, DIN-2PI)

21 – Reflectometry (REMUR, REFLEX)



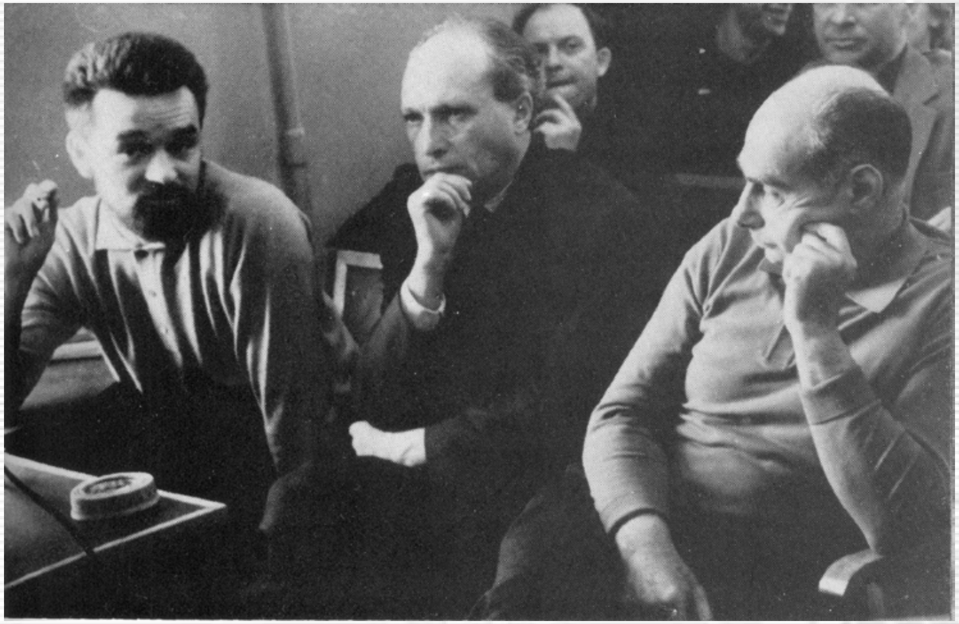
Summary

Neutron condensed matter studies in Dubna has passed an impressive way of development, which has been started in early 1960s by Prof. Feodor L. Shapiro.

Prospects for the further development of neutron spectrometers at the IBR-2 reactor are associated primarily with the improvement of existing experimental techniques: neutron-optical devices, detectors, registration systems.

The IBR-2 spectrometer complex allow to solve almost any of present-day problems neutron scattering studies of condensed matter.

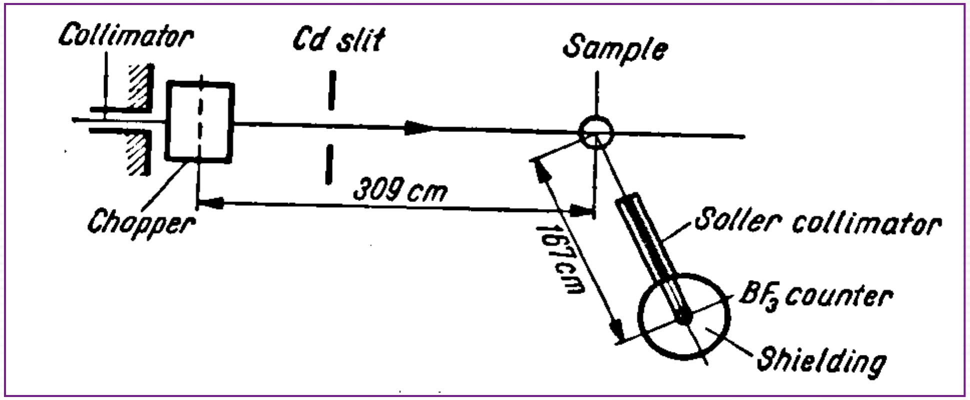
Thank you


At a Seminar

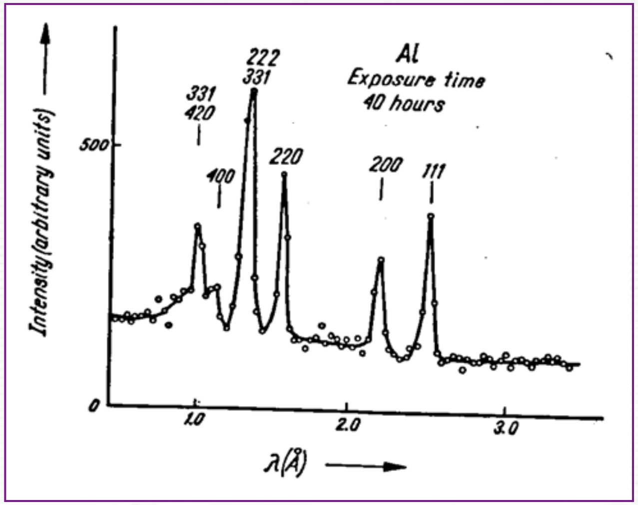

A party, 1970


At the Caucasus, 1963

The first TOF neutron diffraction patterns (1963)

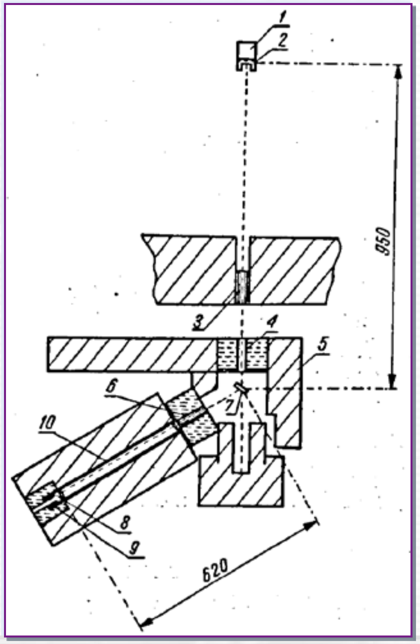


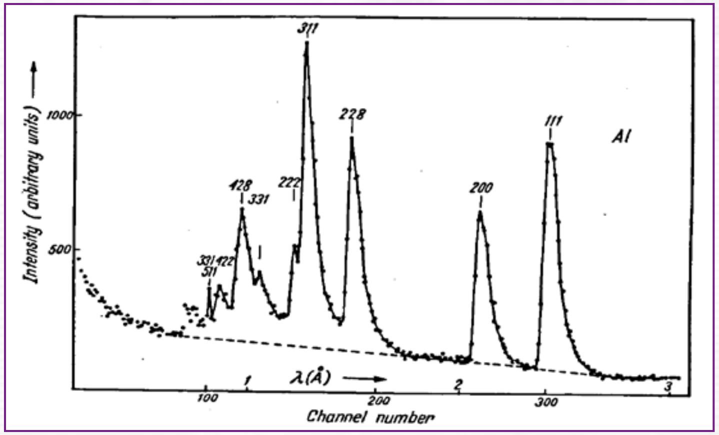
Experimental set-up for TOF neutron diffraction at the EWA, 2 MW reactor (Swierk, Poland) (1963)



The first TOF neutron diffraction pattern measured with Al-powder in 40 hours

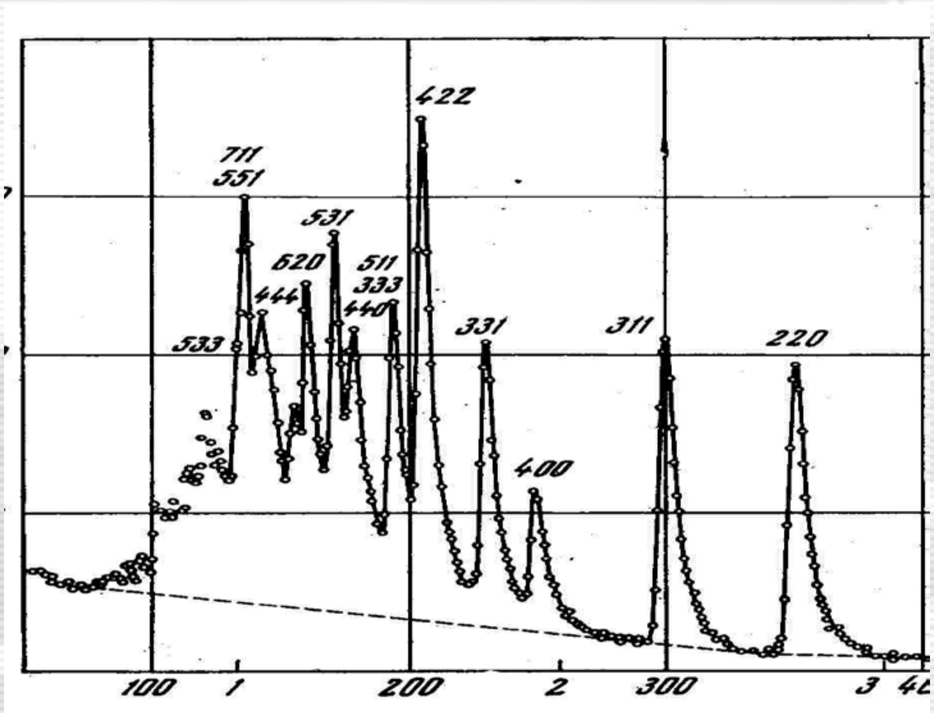
The geometry of the experiment using the pulse reactor IBR-1:
L (reactor-sample) = 950 cm,
L (sample-det.) = 620 cm (1963)

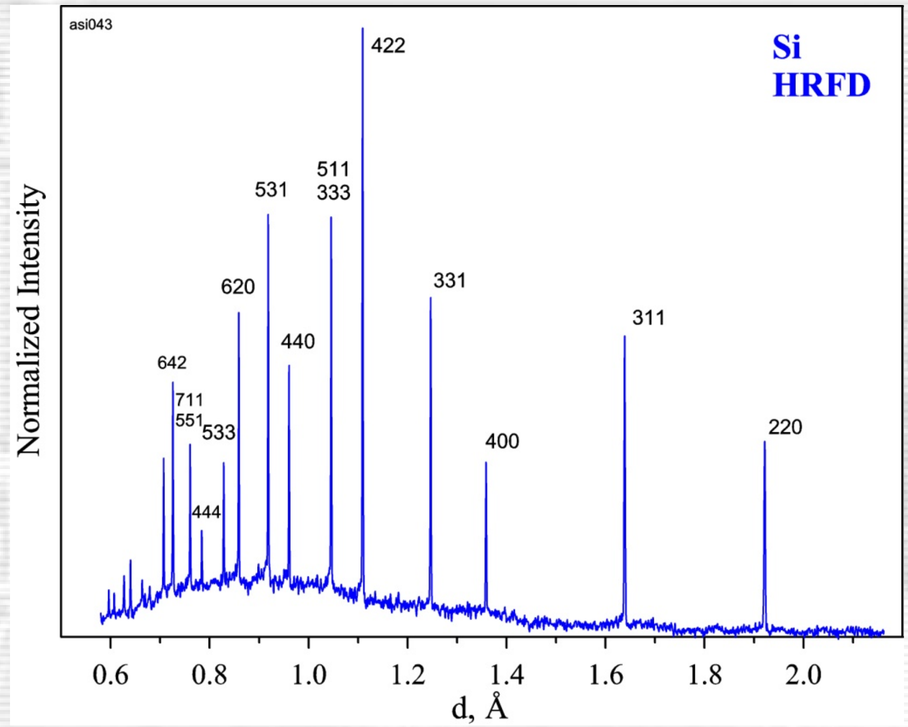




The first TOF diffraction pattern measured at the IBR-1 pulsed reactor with W = 1 kW

Neutron diffraction TOF patterns of Si powder measured at the IBR- pulsed reactors in 1965 and 1994





IBR-1 (1965) Δ*d*/*d* ≈ 0.02

IBR-2 (1994) Δ*d*/*d* ≈ 0.001

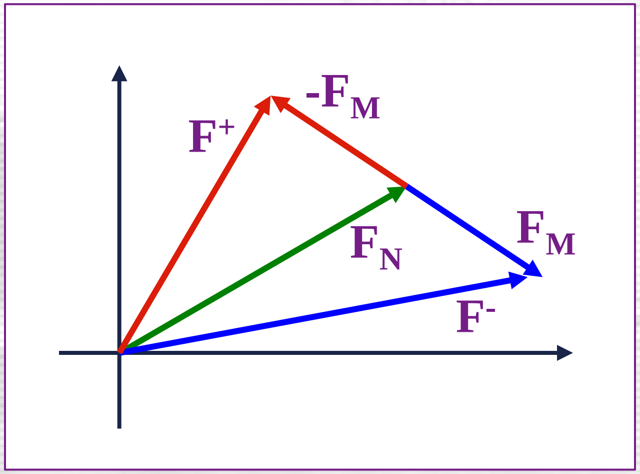
Structure Factor of a Crystal

"About phases of crystal structure factors"

presented at a Workshop in Dubna, 13-16.10.1970

$F(H) = \Sigma\, b_j \exp(2\pi i r_j H) = |F| \cdot e^{i\varphi}$ $|F|^2 \sim I$ (интенсивность пика)

$|F|^2 = |F_N + F_M|^2$ или $|F_N - F_M|^2 = |F_N|^2 + |F_M|^2 \pm |F_N|^2 \cdot |F_M|^2 \cdot \cos(\varphi_N - \varphi_M)$



1. F_N, without magnetic field
2. ± F, with magnetic field of 2 directions
3. $\varphi_N - \varphi_M = \pm \alpha$

"Controlled" scattering amplitude !

Neutron diffraction at the IBR-2 reactor ИБР-2

1982	DN-2 – single crystals, multilayers, real-time studies
1983	DIFRAN – single crystals, dynamical diffraction
1985	SNIM – high pulsed magnetic field
1991 / 99	NSVR / SKAT – texture of geological samples (minerals)
1992	HRFD – powders: atomic and magnetic structure
1993	DN-12 – high pressure studies
2000	EPSILON – stresses in bulk materials
2002	FSD – stresses in bulk materials
2014	DN-6 – atomic and magnetic structure under high pressure

- **Irreversible transition processes in crystals : *real time, in situ* experiments**
- **Fourier diffractometry – crystal structures, microstructure of materials**
- **High pressure in anvil cells – atomic and magnetic structures**
- **Crystallographic textures – minerals, bulk materials**

Experimental hall at the IBR-2 reactor. End of 70th.

Experimental hall at the IBR-2 reactor. End of 90th.

High Resolution Fourier Diffractometer (HRFD) in Dubna