Precision investigation of crystalline materials by neutron diffraction method

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Frank Laboratory of Neutron Physics

- **Neutron-nuclear investigations**
- **Condensed matter physics**
- **Investigations at electrostatic generators and transient radiation**
- **Applied research**

Neutron and X-ray diffraction

- Neutron scattering falls in 2 basic categories:
  - ¾ elastic scattering : info on the structure of gases , liquids and solids since we deal only with scattering processes that do not involve energic excitation of the atoms
  - Inelastic scattering: info on the binding energy within matter due to neutron excitation of atoms

Basic neutron properties used in neutron scattering

1. The energy of moderated neutrons is comparable to the energy of atomic and molecular motions and lies within the MeV to meV energy range.

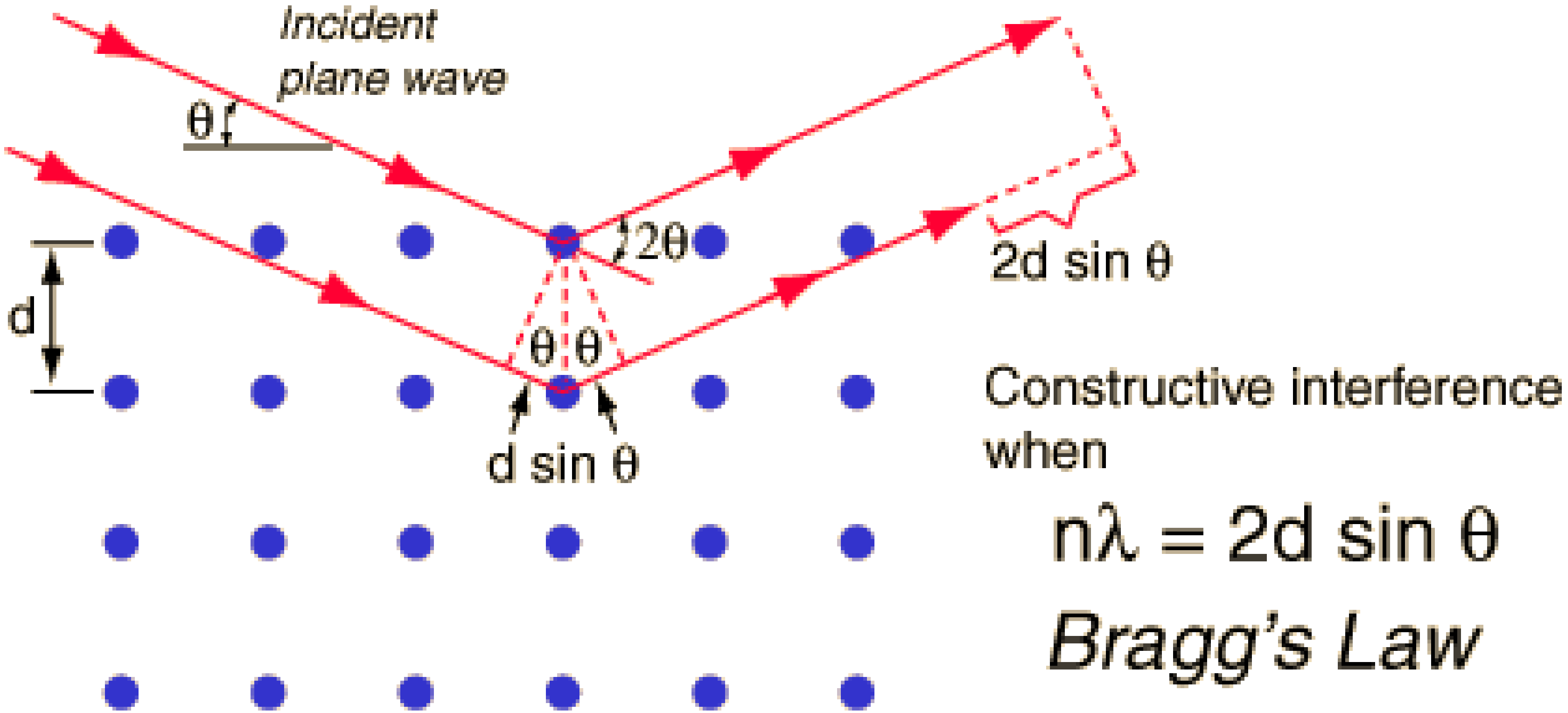
2. The moderated neutron wavelength is comparable to interatomic distances.

3. Since neutrons are neutral particles, they interact with the nucleus of an atom rather than with the diffuse electron cloud.

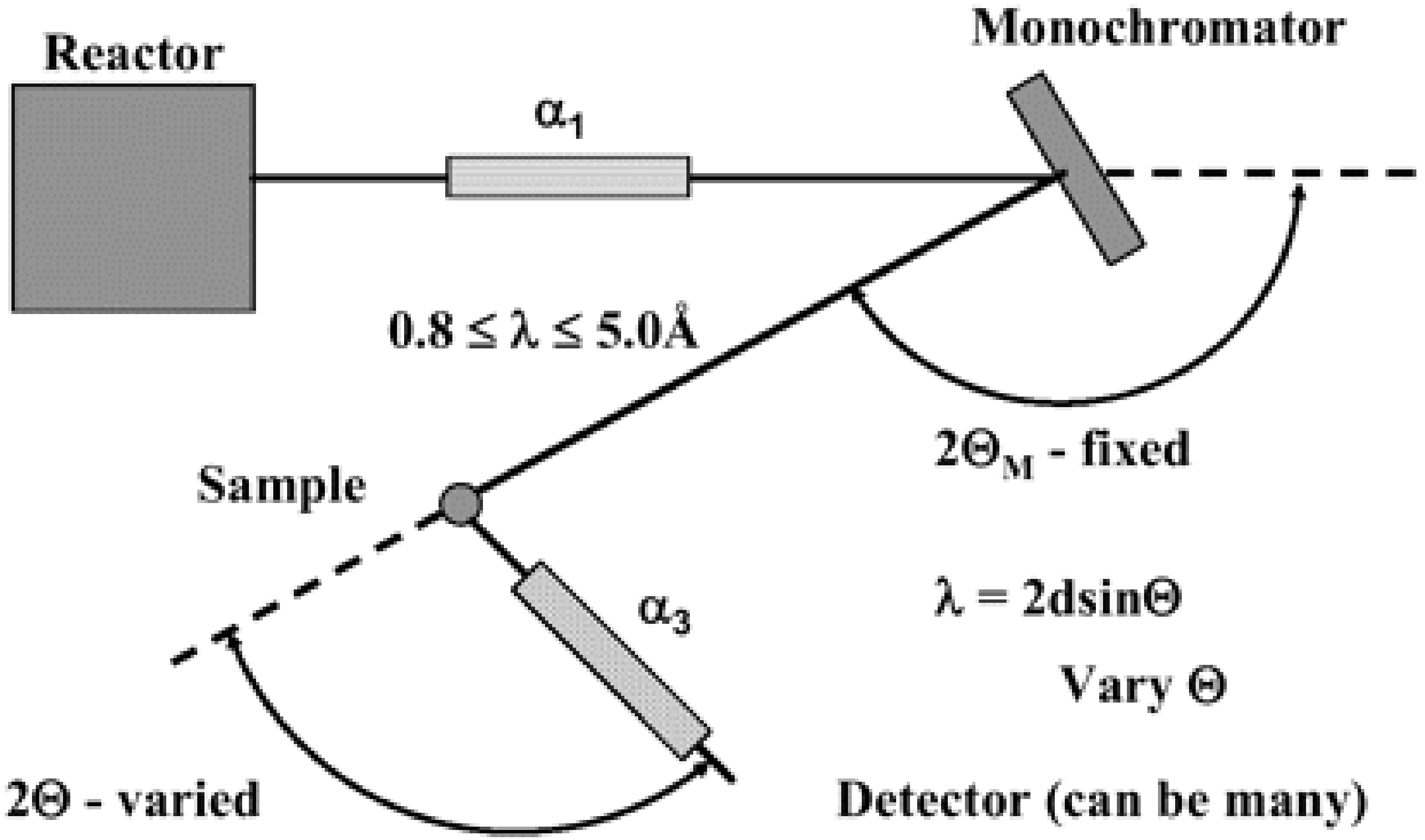
4. Neutrons have a magnetic moment and therefore can be used to study microscopic magnetic structure and magnetic fluctuations, which determine macroscopic parameters of matter.

5.Neutron radiation penetrates deep into materials, thus making it possible to study microscopic properties of bulk samples. Impossible with X ray.

Neutron and X ray diffraction



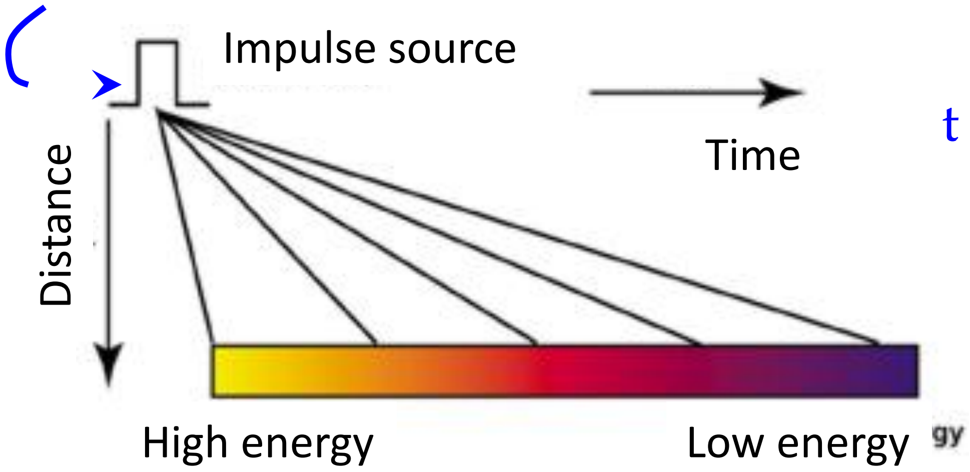
Angle Scan Method



The method time-of-flight (TOF) on pulsed neutron source

Alternatives:

Reactor (steady state source)	W = 10 – 100 MW
Accelerator (pulsed source)	W = 0.01 – 1.5 MW



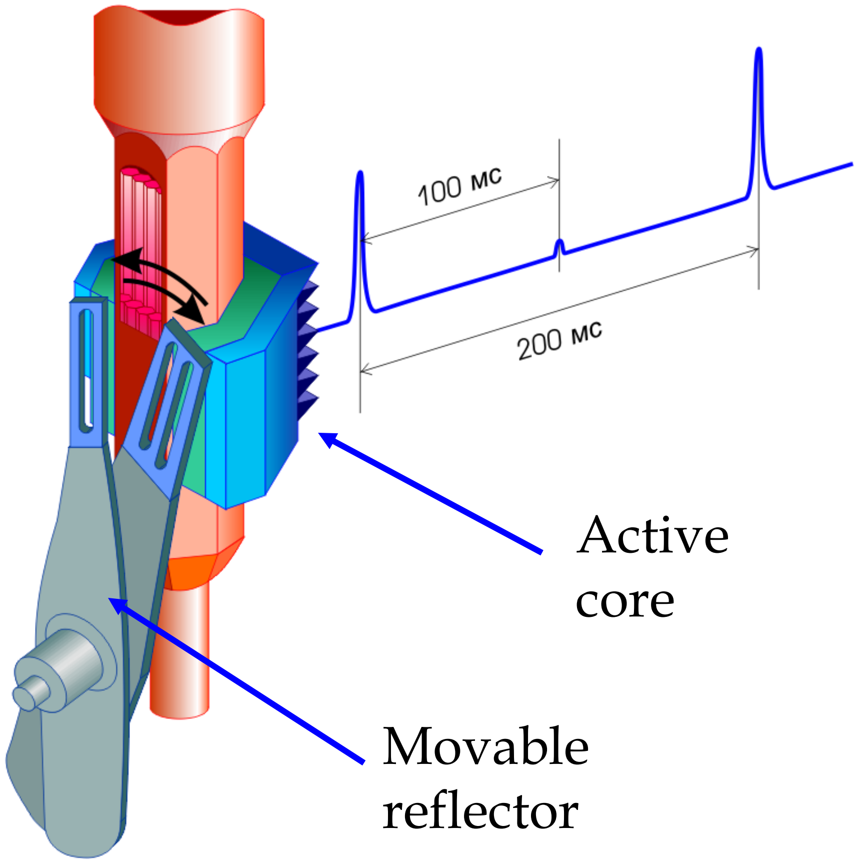
The rate of thermal neutrons is about 2000 m / s.

$t = L/V = 20$ м/2000 м/с = 0.01 с = 10 мс

When the pulse period T = 20 мс (ν = 50 Гц), V_{min} = 1000 м/с, $\lambda_{max} \approx 4$ Å.

The pulse rate at the IBR-2 ν = 5 Гц (T=200 мс), т.е. $\lambda_{max} \approx 40$ Å.

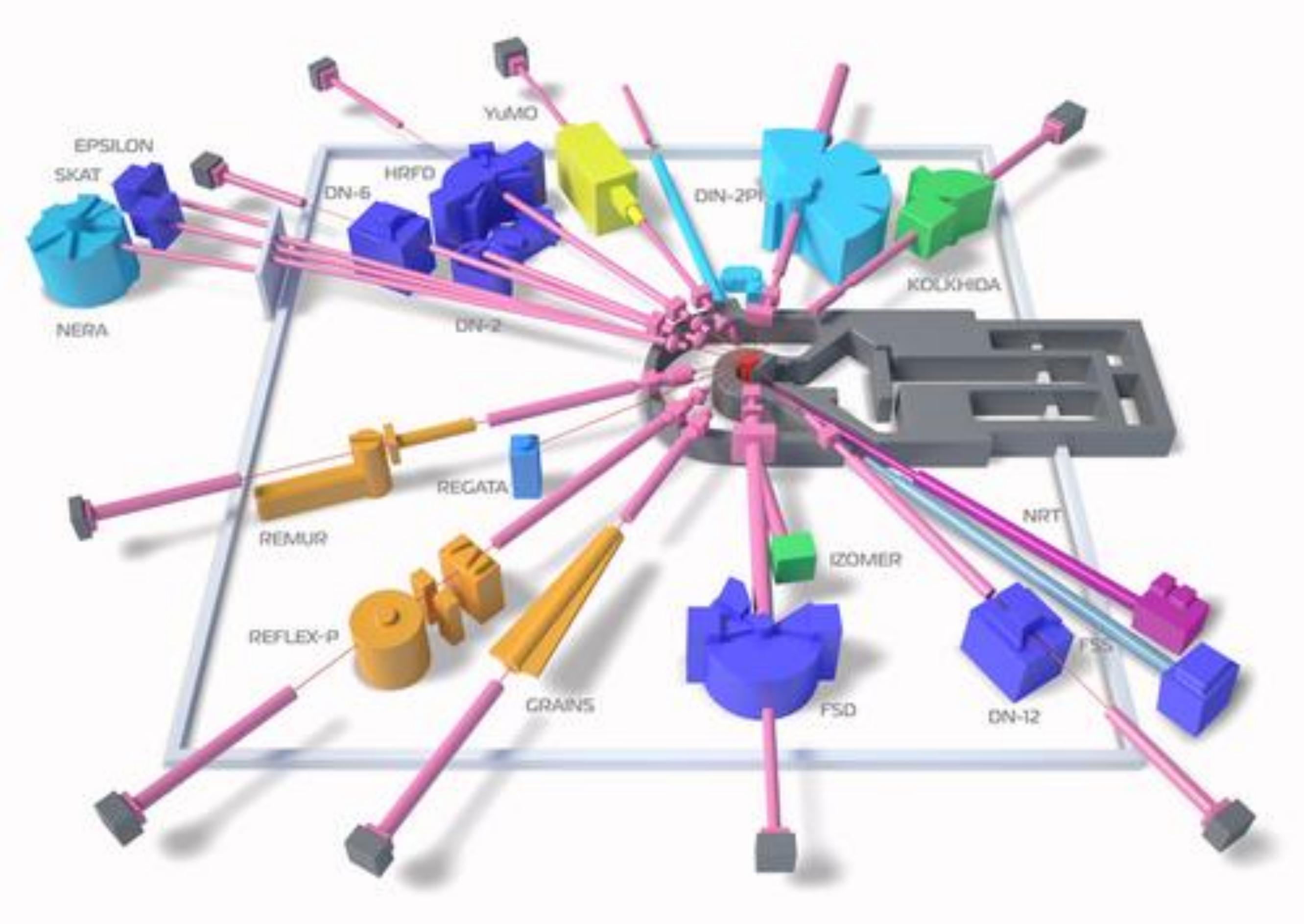
IBR-2 pulsed reactor (1984 – present)



The IBR-2 parameters

Fuel	PuO_2
Active core volume	22 *l*
Cooling	liquid Na
Average power	2 MW
Pulsed power	1850 MW
Repetition rate	$5\ s^{-1}$
Average flux	$8 \cdot 10^{12}\ n/cm^2/s$
Pulsed flux	$5 \cdot 10^{15}\ n/cm^2/s$
Pulse width (fast / therm.)	215 / 320 μs
Number of channels	14

IBR-2 stations



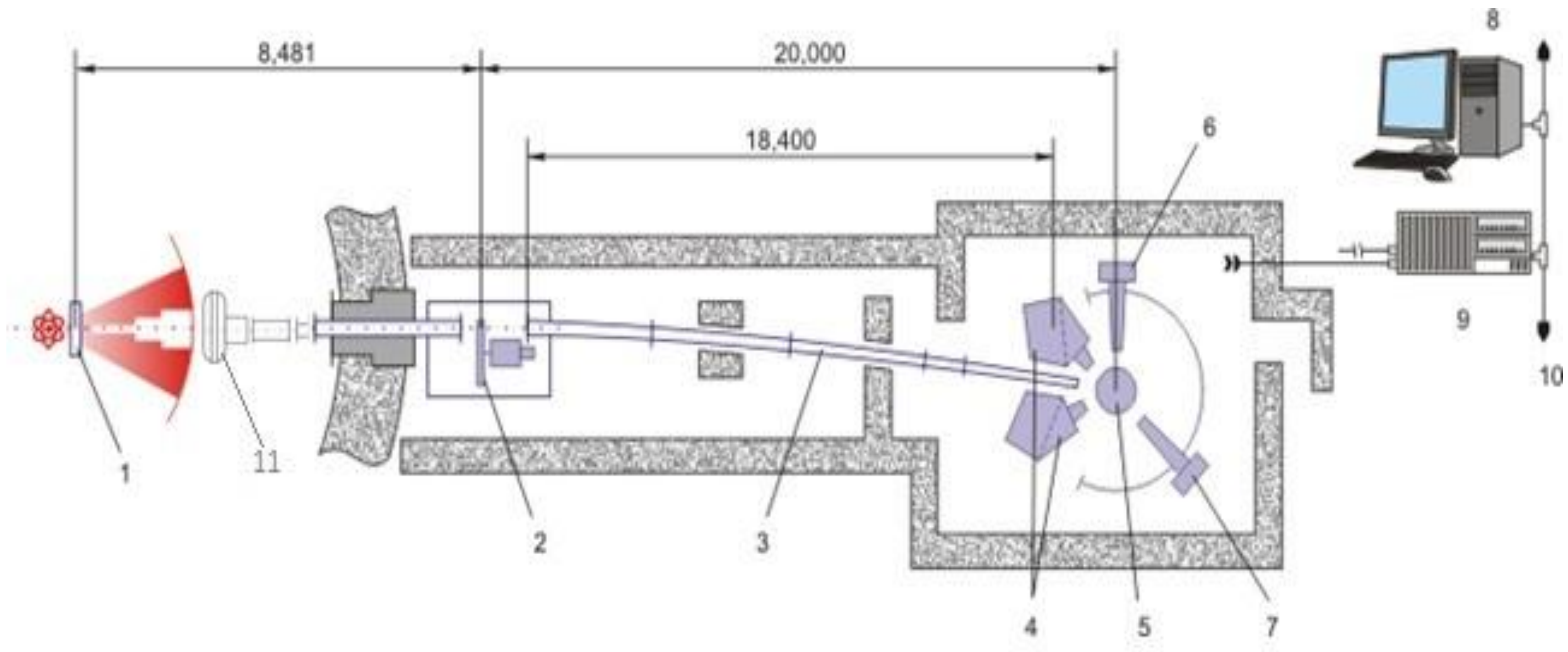
Diffraction (6):
HRFD, DN-2, SKAT, EPSILON, FSD, DN-6

SANS (2):
YuMO, SANS-C

Reflectometry (3):
REMUR, REFLEX, GRAINS

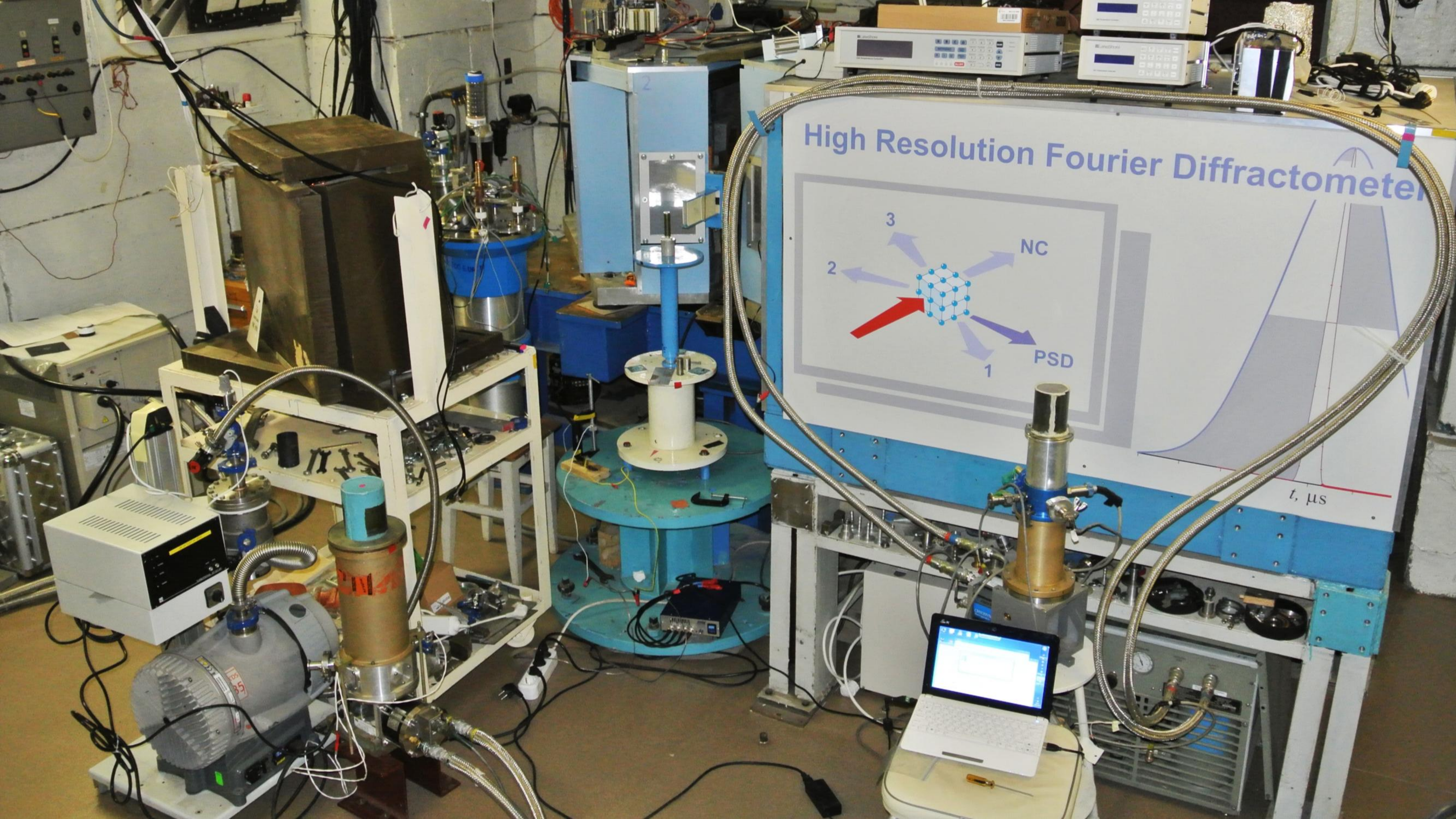
Inelastic scattering (2):
NERA, DIN

13 spectrometers (3 new)



1. Moderator
2. Fourier chopper
3. Guide tube
4. Main detector
5. Sample position
6. 90° Detector
7. Position-Sensitive Detector(PSD)
8.PC
9.Data Acqusition
10. Ethernet
11.Background chopper

High Resolution Fourier Diffractometer



High Resolution Fourier Diffractometer
3
NC
2
PSD
1
t, μs

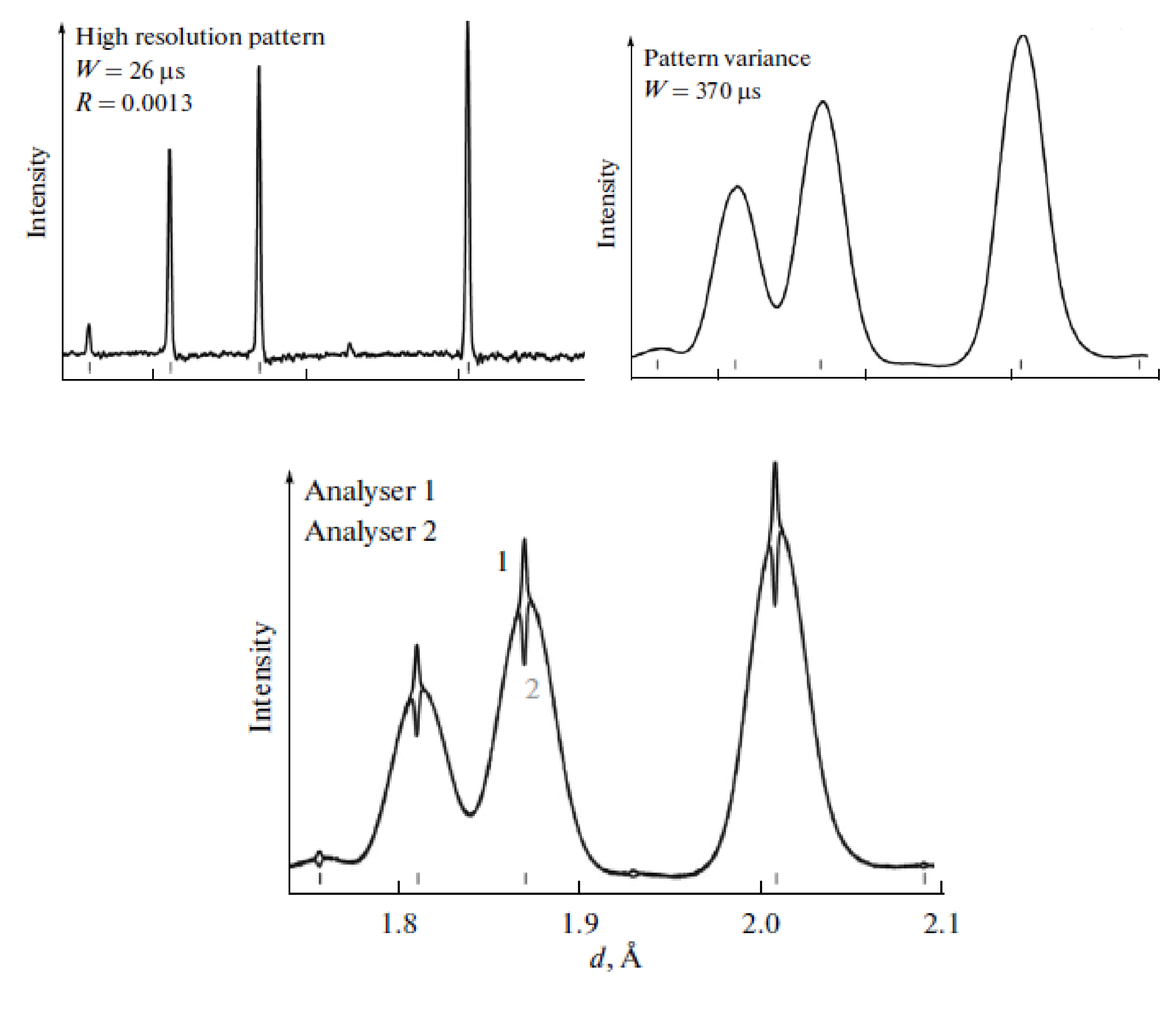
Resolution

R=$\frac{\Delta d}{d}$; d – interplanar spacing

R=$[(\frac{\Delta t}{t})^2+(\frac{\Delta \theta}{\theta})^2+(\frac{\Delta L}{L})^2]^{1/2}$

Δt= effective neutron pulse width

L= fligh path from Fourier chopper to the detector



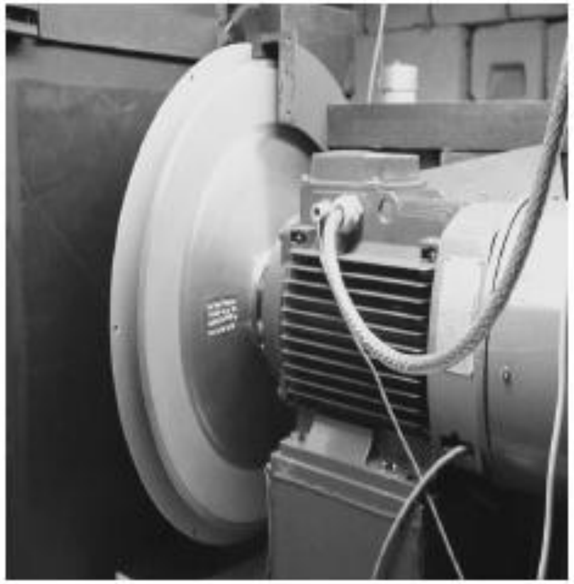
Correlation Method

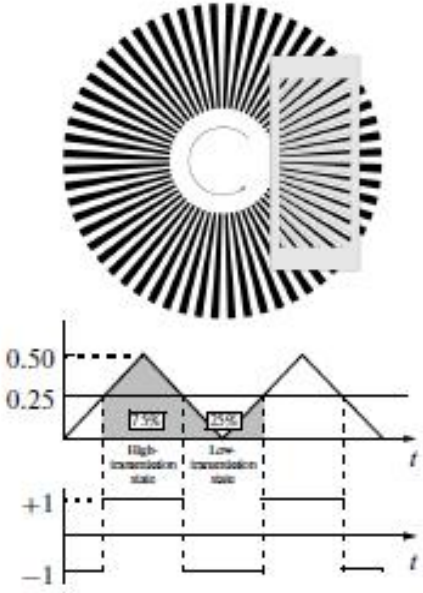
Time Of Flight Method
Spallation sources
High resolution

Drawbacks :
Large flight path means intensity loses
Small flight path (overlapping spectra by generated pulses)

Solution :

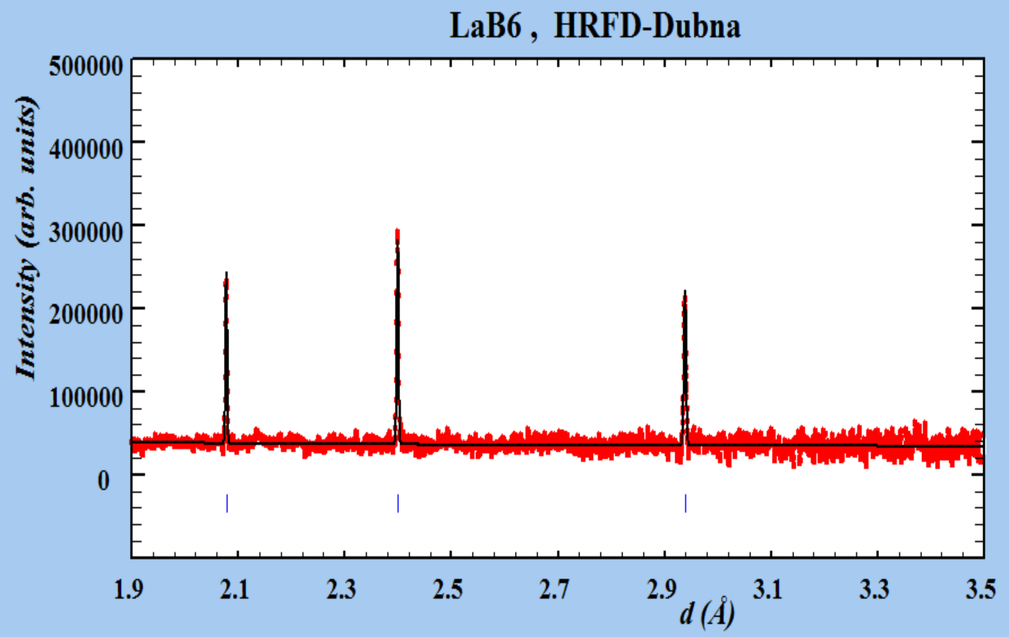
Fourier Chopper





The Rietveld Method

- Unique method
- Uses least squares approach to refine a theoretical line profile until it matches the measured profile



$$y^{calc} = cy^{obs}$$

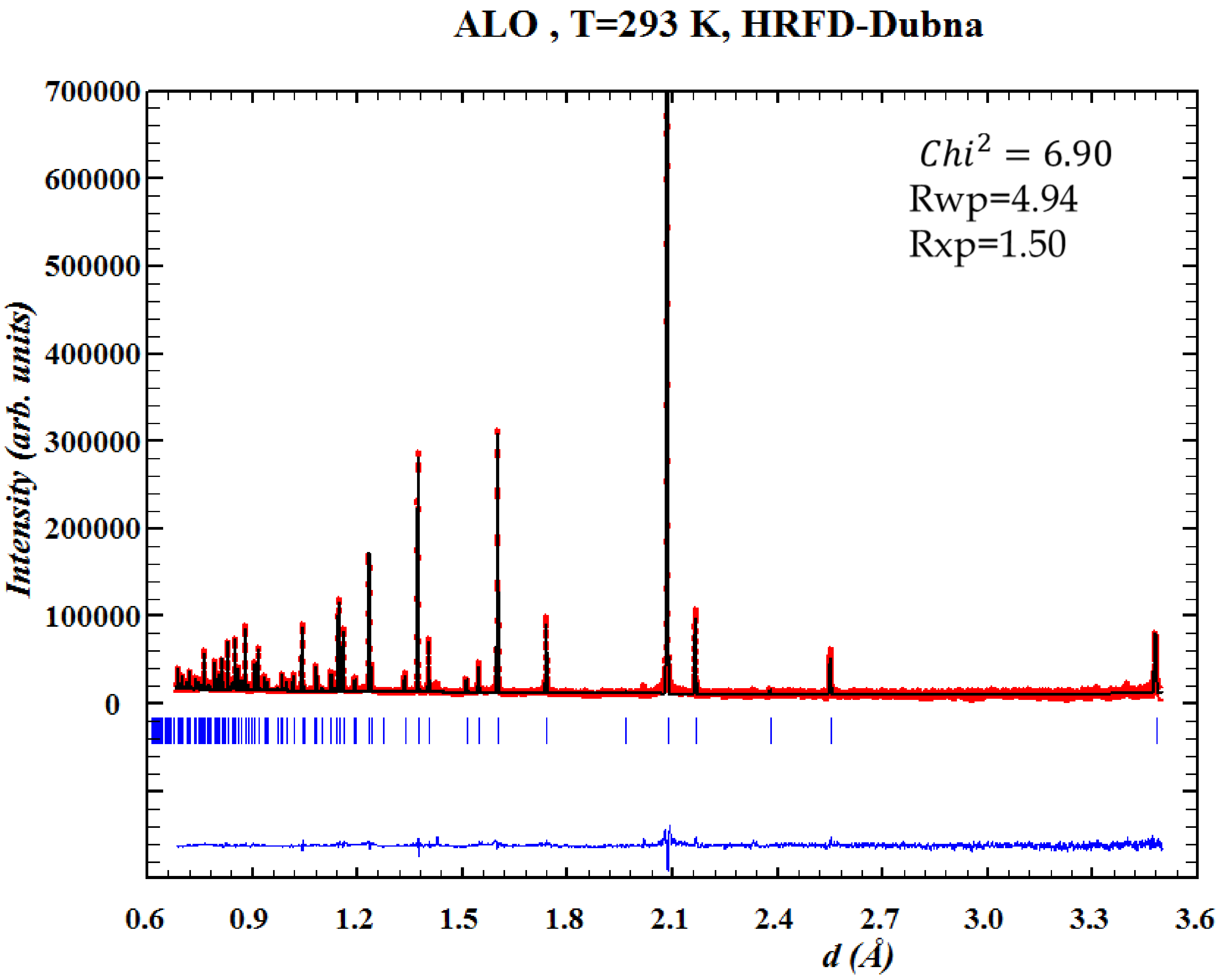
$$M = \sum_i W_i \left\{ y_i^{obs} - \frac{1}{c} y_i^{calc} \right\}^2$$

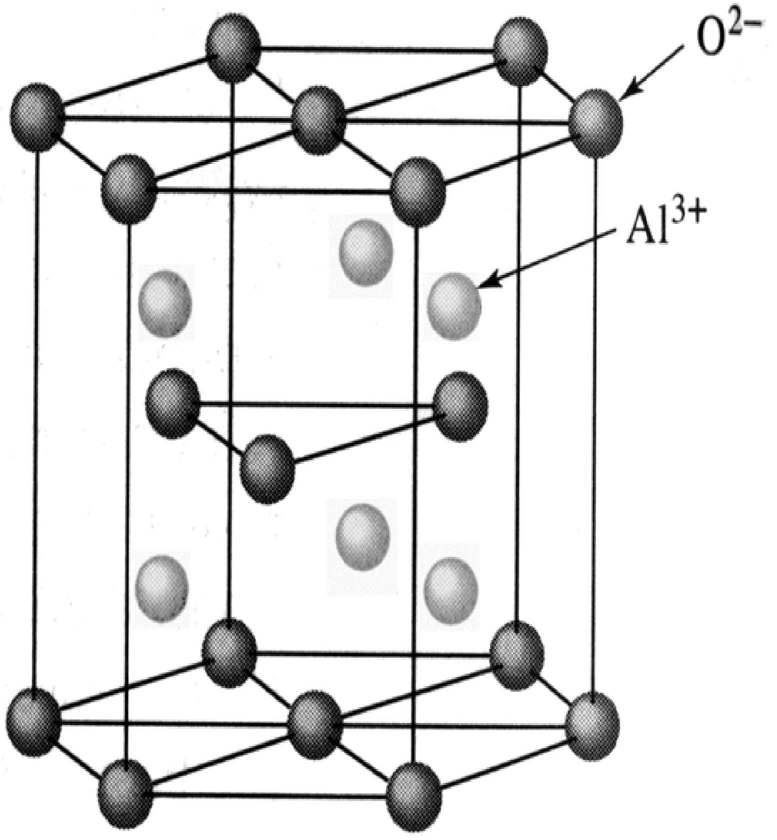
Step intensity

intensity y_i^c at the ith step consists of:

$$y_i^c = y_{bi}^c + S \underbrace{\sum_{k=1}^{n} m_k \, L_k \, |F_k|^2 \, G(\Delta 2\theta_{ik}) \, P_k}_{n \text{ overlapping reflections}}$$

y_{bi}^c	...background
S	...scale factor
n	...number of overlapping reflections (hkl)
m_k	...multiplicity of reflection k
L_k	...Lorentz-, polarisation correction
F_k	...structure factor
$G(\Delta 2\theta_{ik})$	...peak shape function
P_k	...preferred orientation correction





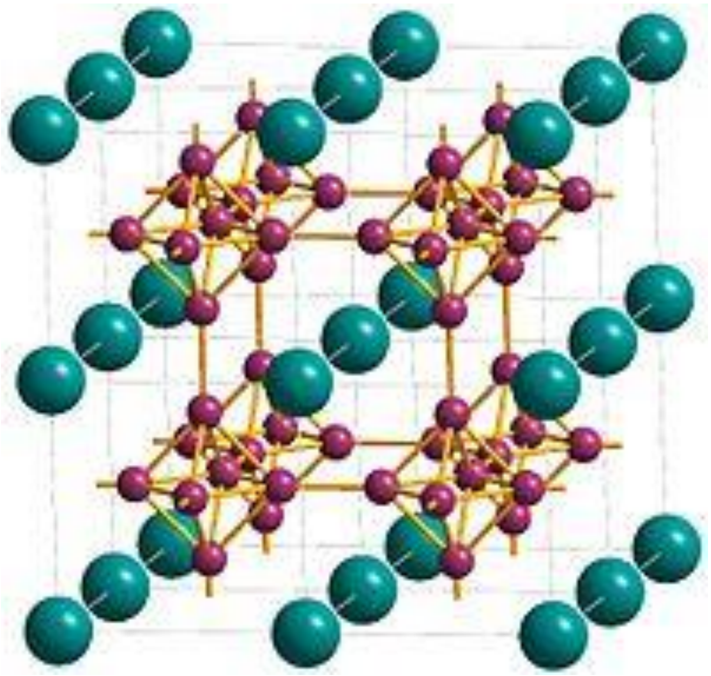
Atomic Position and Cell Info

Al 0,0,0.35233(0.00008)

O 0.30612(11),0,0.25

a=b=4.57906 c=12.99674

α=β=90° γ=120



Atomic position

La 0,0,0

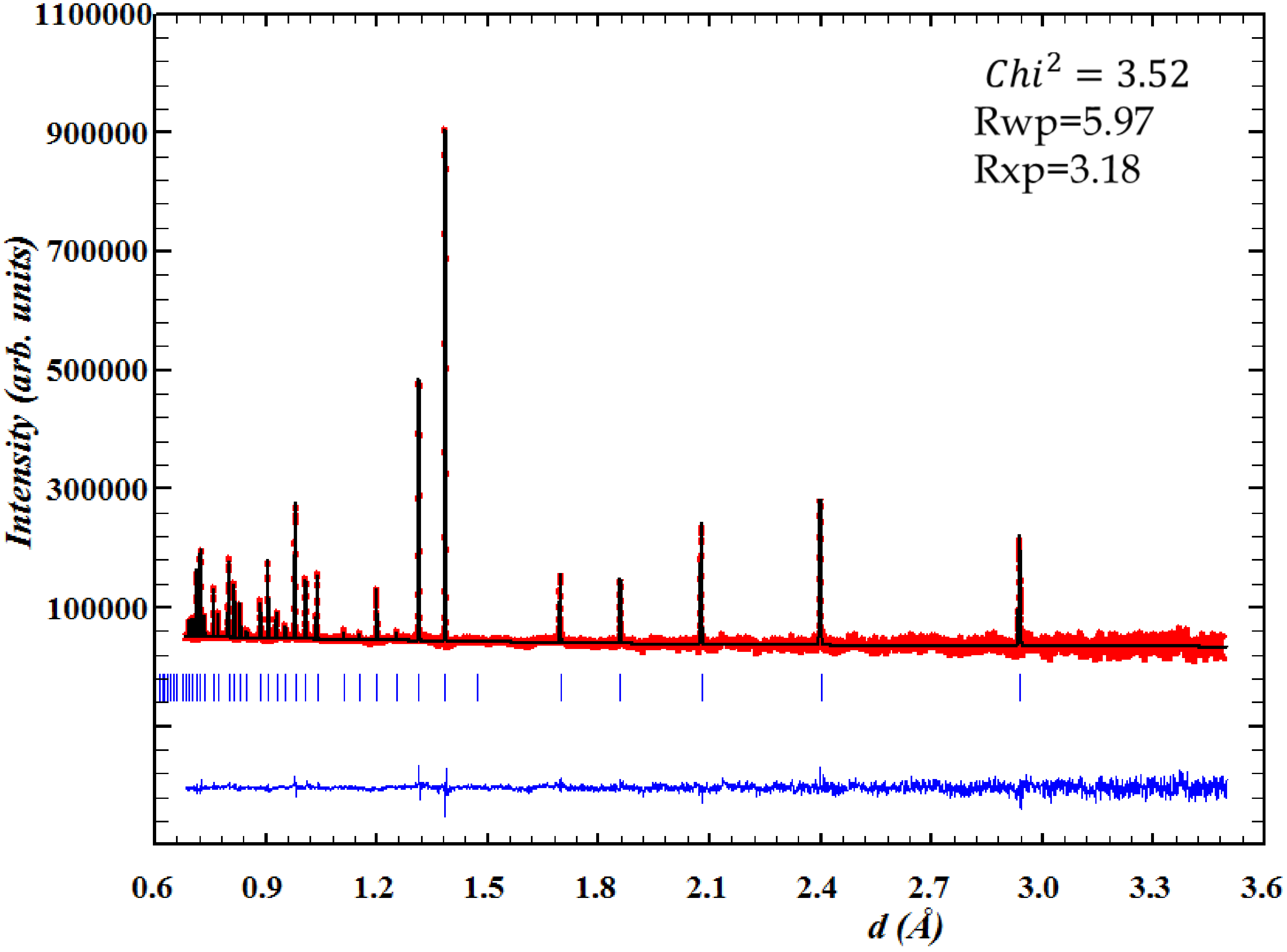
B 0.5,0.5,0, 0.19967(0,00021)

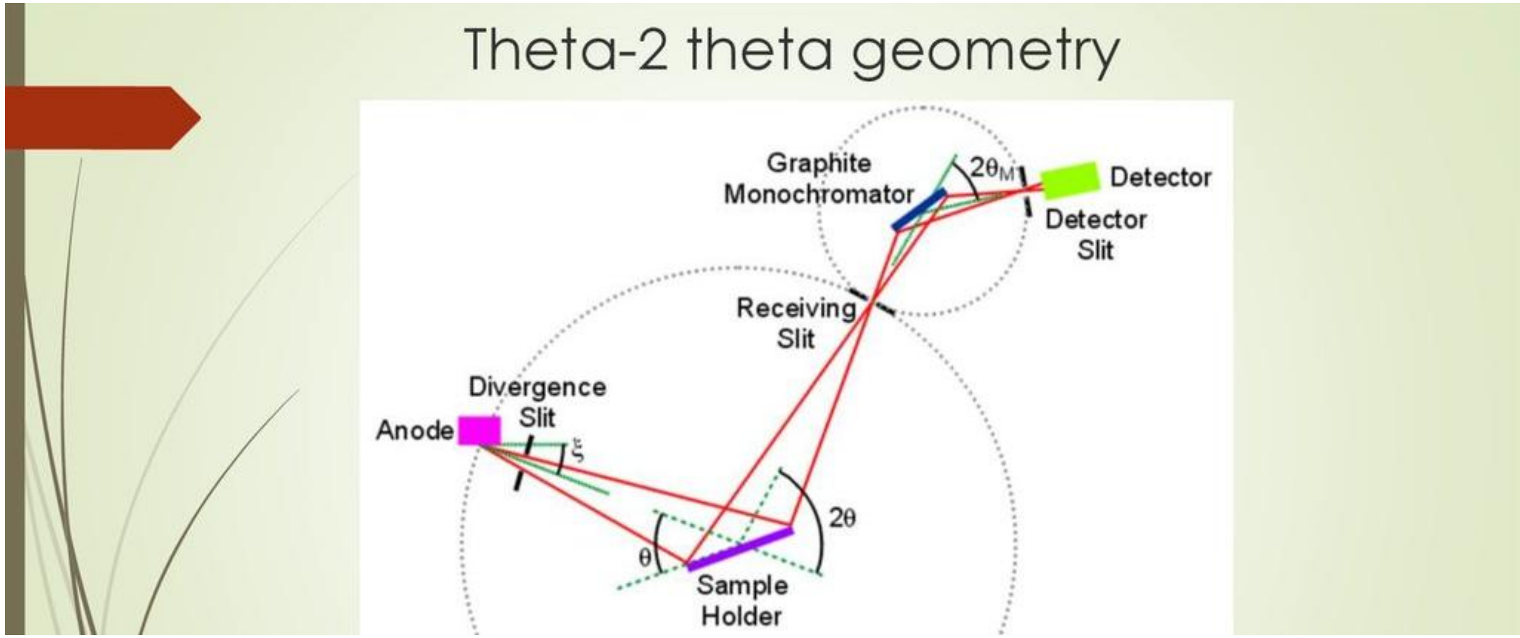
Unit Cell Info

a=b=c=4.15671(0.00006)

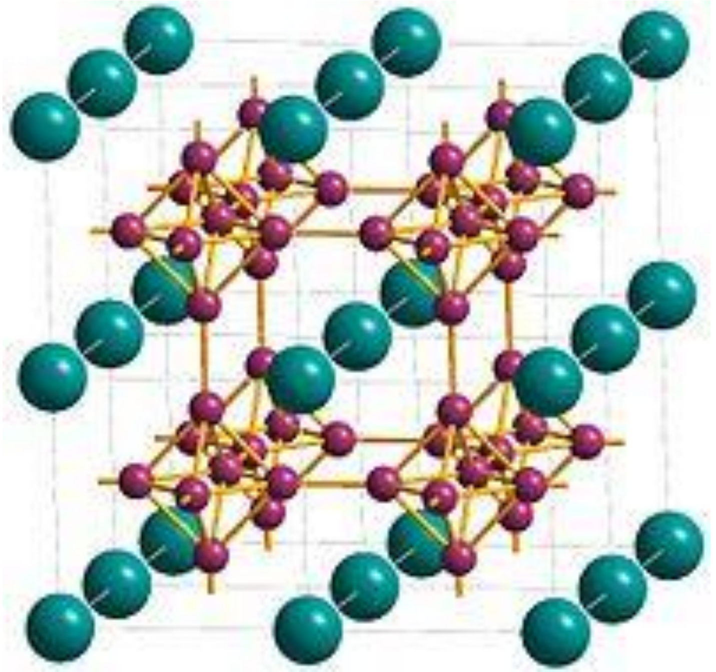
α=β=γ=90°

LaB6，HRFD-Dubna





X-ray diffractometer



Atomic position

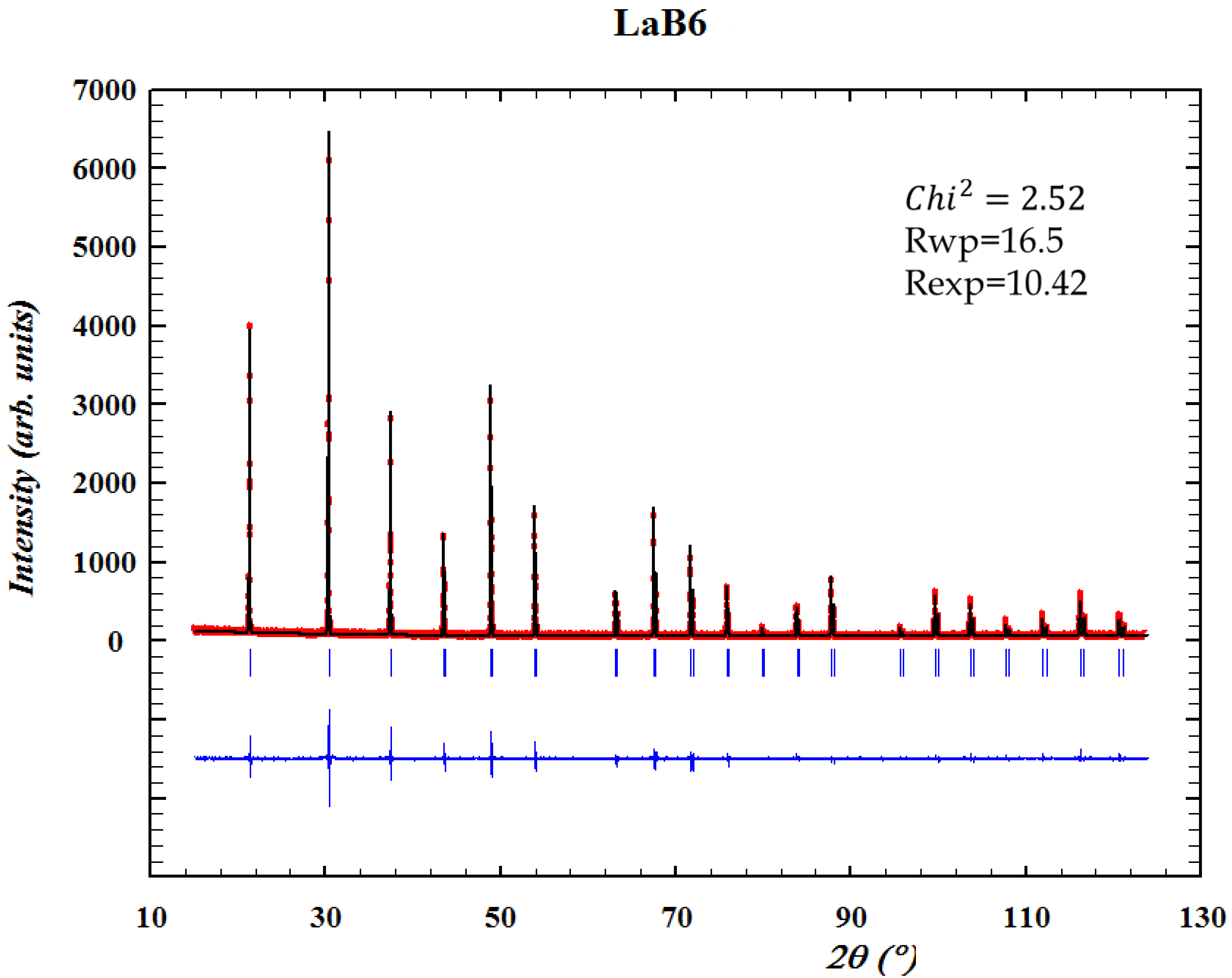
La 0,0,0

B 0.5,0.5,0.20091(0,000344)

Unit Cell Info

a=b=c=4.15773(0.00002)

α=β=γ=90°



Comparison of LaB6

Neutron Diffraction

Atomic position

La 0,0,0

B 0.5,0.5,0, 0.19967(0,00021)

Unit Cell Info

a=b=c=4.15671(0.00006)

α=β=γ=90°

X-ray Diffraction

Atomic position

La 0,0,0
B 0.5,0.5,0.20091(0,000344)

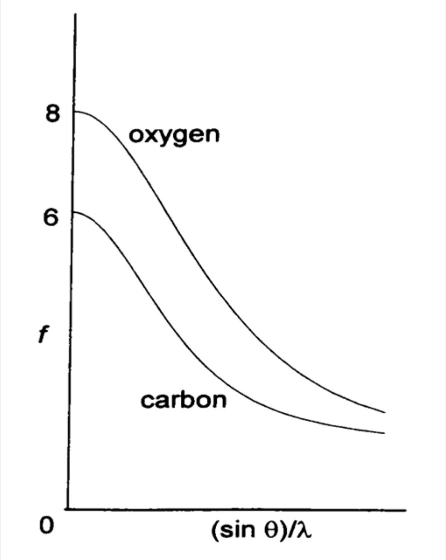
Unit Cell Info

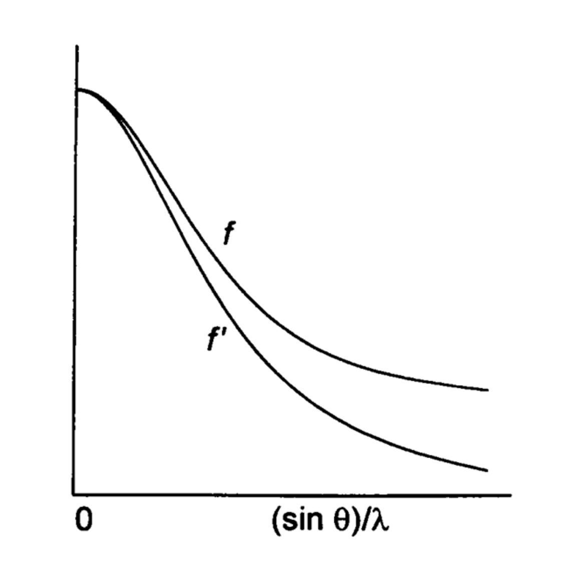
a=b=c=4.15773(0.00002)
α=β=γ=90°

Thank you for your attention !

Structure factor

- $F(hkl) = \sum f_j \exp\left[2\pi i \left(hx + ky + lz\right)\right]$





$$f = 4\pi \int \rho(r) \frac{\sin Kr}{r} \mathrm{dr}$$

$$f_j = {}^0 f_j \exp\left(\frac{-B \sin^2\theta}{\lambda^2}\right)$$

Error

Weighted profile factor:

$$R_{wp} = 100\left[\frac{\sum_{i=1,n} w_i \left| y_i - y_{c,i} \right|^2}{\sum_{i=1,n} w_i y_i^2}\right]^{1/2}$$

Reduced chi-square:

$$\chi_v^2 = \left[\frac{R_{wp}}{R_{exp}}\right]^2 = S^2$$

Expected profile factor:

$$R_{exp} = 100\left[\frac{n-p}{\sum_i w_i y_i^2}\right]^{1/2}$$

R≤10%，$\chi2 \rightarrow 1$