THE NEW FOURIER DIFFRACTOMETER AT THE IBR-2 REACTOR: DESIGN AND FIRST RESULTS

V.L.Aksenov, A.M.Balagurov, V.G.Simkin, Yu.V.Taran
Frank Laboratory of Neutron Physics, JINR, Dubna, Russia

V.A.Trounov, V.A.Kudrjashev, A.P.Bulkin, V.G.Muratov
Petersburg Nuclear Physics Institute, Gatchina, Russia

P.Hiismaki, A.Tiitta, O.Antson
Technical Research Centre of Finland, Espoo, Finland Abstract

Abstract

A high-resolution neutron powder Fourier diffractometer (HRFD) has been realized at the pulsed reactor IBR-2 in Dubna. The HRFD avails of both the high luminosity of the IBR-2 reactor and the high resolution of the Fourier chopper. The resolution very close to 0.001 has been achieved over a wide range of *d*-spacings and the value of $\Delta d/d$ equal to 0.0005 is expected to be reached after the diffractometer units are aligned. Neutron flux at the sample position is about 10^7 n/cm^2/s providing a good data accumulation rate. The new diffractometer is thought to be very suitable for structural refinement of low symmetry crystals and for residual stress studies.

1. TOF diffractometers at the IBR-2 reactor

It is well known that a powder diffractometer can be optimized in two alternative modes: high resolution or high intensity. Precise structural studies and real-time experiments are the examples of the investigations which are destined for these two kinds of instruments. The pulsed reactor IBR-2 /1/ in Dubna has a very high peak flux of thermal neutrons from moderator surface — 10^{16} n/cm^2/s. At the same time the resolution the conventional TOF spectrometers at the IBR-2 is not too good due to the large width of the neutron pulse (320 μs). As a result good prospects exist for the experiments that require high intensity but moderate resolution. For example, the DN-2 diffractometer at the IBR-2 makes it possible to measure the entire diffraction pattern within a fraction of a minute. The examples of such experiments are reported elsewhere /2/. However the precise structural studies of high level seemed impossible to be carried out with the IBR-2.

2. How the resolution can be improved?

The expression for the resolution of the time-of-flight powder diffractometer has two terms (in the first approximation):

$$R = \Delta d/d = \left[(\Delta t_0/t)^2 + (\gamma\, ctg\theta)^2\right]^{1/2}, \tag{1}$$

where Δt_0 is the width of the neutron pulse, γ includes all geometrical uncertainties, $t = 252.778L\lambda$, L is the total flight path, λ is the neutron wavelength, θ is the Bragg angle. It is clear that the time-of-flight contribution can be reduced either by shortening the source pulse or by lengthening the flight path. For example, the HRPD diffractometer /3/ at ISIS exploits both ways: the pulse width is as short as ~15 μs/Å and the flight path is as long as ~100 m. This permits one to get $\Delta t_0/t$ as small as $6\cdot10^{-4}$ in a wide *d*-range.

From the practical viewpoint these two ways are not good, if one wants to the resolution of the TOF diffractometer at the IBR-2 reactor to be of such a small value. Another and more promising possibility is the utilization of the correlation technique, pseudostatistical or Fourier-type. The Fourier method proved itself to be more preferable for elastic scattering because it allows use of narrow chopper slits together with wide neutron beam /4/.

3. The resolution of the neutron Fourier diffractometer

The key part of the Fourier diffractometer, the Fourier chopper, is the ultimate case of a multislit Fermi chopper when the widths of slits and the spacings between them are equal. For a 1 mm slit and 9000 rpm speed of a chopper the neutron pulse width is about 2 μs. But due to the frequency sweeping the width of resolution function is not so small. In the simplest case, this frequency sweep $g(\omega)$ is Blackman distribution from zero frequency up to the maximum one, ω_m. It was shown, for example in /5/, that the time-of-flight part of the resolution function, $R_c(t)$, in this case is equal to:

$$R_c(t) \sim \int_0^{\Omega_m} g(\omega) cos(\omega\, t) d\omega\,, \tag{2}$$

where $\Omega_m = N\omega_m$ is the maximum modulation frequency of neutron beam, with N denoting the total number of chopper slits. Roughly, the full width at the half maximum of $R_c(t)$ is equal to Ω_m^{-1} and can be pushed down to about 7 μs. In this case the first term in (1) is (for backscattering):

$$\Delta t_0/t = \Delta t_0/253L\lambda \approx 7\cdot 10^{-4}/d, \tag{3}$$

where d is in Å . The divergence of the initial beam, sample and detector dimensions contribute to the second term in (1). For the time-focused detector system the calculated value for this term can be equal /6/ to $3.5\cdot 10^{-4}$, so the total width of the resolution function can be about $5\cdot 10^{-4}$, if d equals 2 Å.

4. Existing neutron Fourier diffractometers

At present two neutron Fourier diffractometers are in practical use: mini-SFINKS facility /7/ in Gatchina (Russia) and FSS /8/ in Geesthacht (Germany). They both are installed at steady state reactors, use the Reverse Time Of Flight (RTOF) method /5/ of data acquisition and reveal good prospects for structural and residual stress studies. The mini-SFINKS diffractometer is the first result of Finnish - Russian cooperation aimed at the development of Fourier TOF techniques for diffractometry applications. In improved configuration it has been operating at the 16 MW reactor since 1986 for routine structural investigations. The maximum beam modulation frequency, Ω_m=120 kHz, allows a FWHM of ~8.5 μs to be achieved. For the total flight path of 7.6 m the resolution changes in the useful interval of d-spacings 0.5-2.6 Å from $\Delta d/d \approx 0.005$ to about 0.002.

5. Some special features of Fourier diffractometer operation at a pulsed neutron source

For the better understanding of the situation with the Fourier chopper at a pulsed source let us compare three types of instruments: the conventional TOF machine and the Fourier diffractometer at a steady state and a pulsed source. In the first case the detected intensity is proportional to the convolution integral:

$$I(t) \sim \int R_s(t-\tau)\sigma(\tau)d\tau + B(t), \tag{4}$$

where, in the rough approximation neglecting all geometrical contributions, R_s is the source pulse, σ is the scattering cross section of the sample, B is the conventional background. The situation with a Fourier diffractometer at a steady state reactor is a little more complicated as there is the modulation of the incident neutron beam by the Fourier chopper. Then the relation for the intensity looks like

$$I(t) \sim \int R_c(t-\tau)\sigma(\tau)d\tau + c\int \sigma(\tau)d\tau + B(t), \qquad (5)$$

here R_c is the resolution function of the Fourier chopper, the second term, which can be called "the correlation background" is proportional to the total detected intensity, c is a certain constant. The situation with the Fourier chopper at a pulsed neutron source is, in some sense, the combination of the two previous cases:

$$I(t) \sim \int R_s(t-\tau)R_c(t-\tau)\sigma(\tau)d\tau + c\int R_s(t-\tau)\sigma(\tau)d\tau + B(t). \qquad (6)$$

One can see, that in (6) both resolution functions R_s and R_c are present, thus the diffraction peaks are very narrow ($\Delta t \approx 7$ μs), the correlation background is not constant and it is proportional to the resolution function of the source. Additional modulation with R_s is very important for decreasing the correlation background, which is now proportional to a small portion of the total intensity, namely, the intensity in the time interval $2\Delta t$, where Δt is equal to the width of R_s, i.e. 320 μs for the IBR-2 reactor. Due to the Maxwellian distribution of the incident beam intensity the signal to noise ratio for steady state and pulsed sources is quite different. There is some gain in this ratio in the pulsed source case, being especially high for the long wavelength neutrons.

6. The HRFD design (Fig.1)

The High Resolution Fourier Diffractometer (HRFD) at the IBR-2 pulsed reactor is the spinoff from the cooperation of the Joint Institute for Nuclear Research, Dubna, Nuclear Physics Institute, Gatchina and Technical Research Centre of Finland, Espoo. The project started in 1988, when all calculations were performed. The electronics and mechanical devices were ready for operation late in 1991. The neutron beam was formed in the beginning of 1992 and in June 1992 the first diffraction patterns were measured /9/.

HRFD is located on beam N5 of the IBR-2 reactor. Immediately after the reactor shielding the mechanical filter and the auxiliary neutron guide tube are placed. The filter is a stainless steel disk-chopper of about 1 m diameter with a $\Delta\phi$=60° window. It is to remove the thermal neutron background off the beam. Through the biological shielding the beam is conducted over a straight neutron guide. Immediately after this shielding wall the Fourier-chopper is placed. The focusing guide tube acts both as the forming element of the neutron beam and filter of fast neutrons and γ-rays. Its incoming and outgoing window cross-sections are as large as 30x200 and 10x100 mm^2, respectively, and its total length is about 19 m. Before the sample the Soller collimator is installed to reduce the angular divergence of the neutron beam. The detector assembly consists of two blocks. At a lower scattering angle two position sensitive detectors are placed on the moveable arms. The main detector is located at a larger angle (≈156°), it is time-focused and has a total solid angle of 0.08 sr. The flight path between the chopper disk and the sample position is 20.0 m. The electronics of HRFD is created using the modular RTOF analyzer based on a polarity correlator ASIC circuit of special design. Each Correlator module contains 1024 channels. The analyzer consists of a master module, acting as the signal interface and the master clock unit, and eight correlator modules. Two these 8192-channel analyzers operate simultaneously with the Fourier chopper pickup signals in opposite phases. The RTOF analyzers are connected to a PC/AT-386 by a BITBUS standard serial interface. The PC is used as interface between the user and the measuring unit. Principal technical data and performance characteristics of HRFD are summarized in the Table. At the present they all correspond to the actual ones, except for the high resolution detector at 90° scattering, which will be ready for operation at the end of 1993.

7. Instrument performance and first experimental results

As it follows from eq. (6) the experimental spectrum measured with each of the two analyzers consists of three components: the high resolution diffraction peak, the low resolution diffraction peak and the background. The sign of the term, representing the high resolution peak, is different for the first and second analyzer. Therefore, simple subtraction allows to separate the high resolution spectrum only. One can easily see that in Fig.2, which shows a small part of the spectrum from Al_2O_3. It is natural that dispersion of the resulting spectrum depends on the statistics collected by the analyzers.
The high resolution spectrum of the *Ge* powder is shown in Fig.3, while Fig.4 gives comparative raw diffraction patterns of Al_2O_3 obtained with HRFD (bottom) and HRPD diffractometer at ISIS. One can see that both diffractometers have practically the same resolutions. At the same time it is clear that the HRPD creates better background conditions. The resolution was measured at various chopper speed (Fig.5) and *d*-values. Then the measured and expected resolution functions were compared. The result was that the TOF contribution to the resolution function was very close to the expected value but the geometrical term was three times larger. We believe it is mainly due to the detector contribution, as no careful detector alignment has been done yet. An attempt of Rietveld refinement was also done (Fig.6) with the help of MRIA program /10/. For profile shape function we used one of the diffraction peaks, measured with good statistics, as it is possible with MRIA. Details will be presented in a separate paper.

8. Conclusions

The high resolution Fourier diffractometer has been realized at the IBR-2 pulsed reactor. The most important result of the preliminary measurements performed consists in the fact that the idea has received confirmation that the Fourier technique in combination with a pulsed neutron source, such as the IBR-2 reactor, gives the diffraction patterns of very good quality. The resolution of 0.0015 in $\Delta d/d$ was achieved at first attempt. It is approximately 10 times better than was before achieved at the IBR-2 (Fig.7). The expected value of $\Delta d/d$ equal to 0.0005 can be reached after the diffractometer units are aligned and their assembly completed.

The authors wish to acknowledge the help of many members of the FLNP, PNPI and VTT staff in engineering, fabrication and installation of this instrument.

Special thanks are due to Drs. B.I.Voronov, E.V.Serochkin, V.I.Aparin, V.G.Tishin, V.E.Novozchilov, A.I.Ostrovnoy, I.P.Barabash, A.A.Bogdzel, A.I.Sirotin, A.P.Trofimova, G.F.Zhironkin and V.V.Zhuravlev.

The work was financed by the Joint Institute for Nuclear Research in the frame of the JINR and BMFT (Germany) agreement on co-operation and supported by the Russian State Commission on HTSC materials within projects 90169 and 90287.

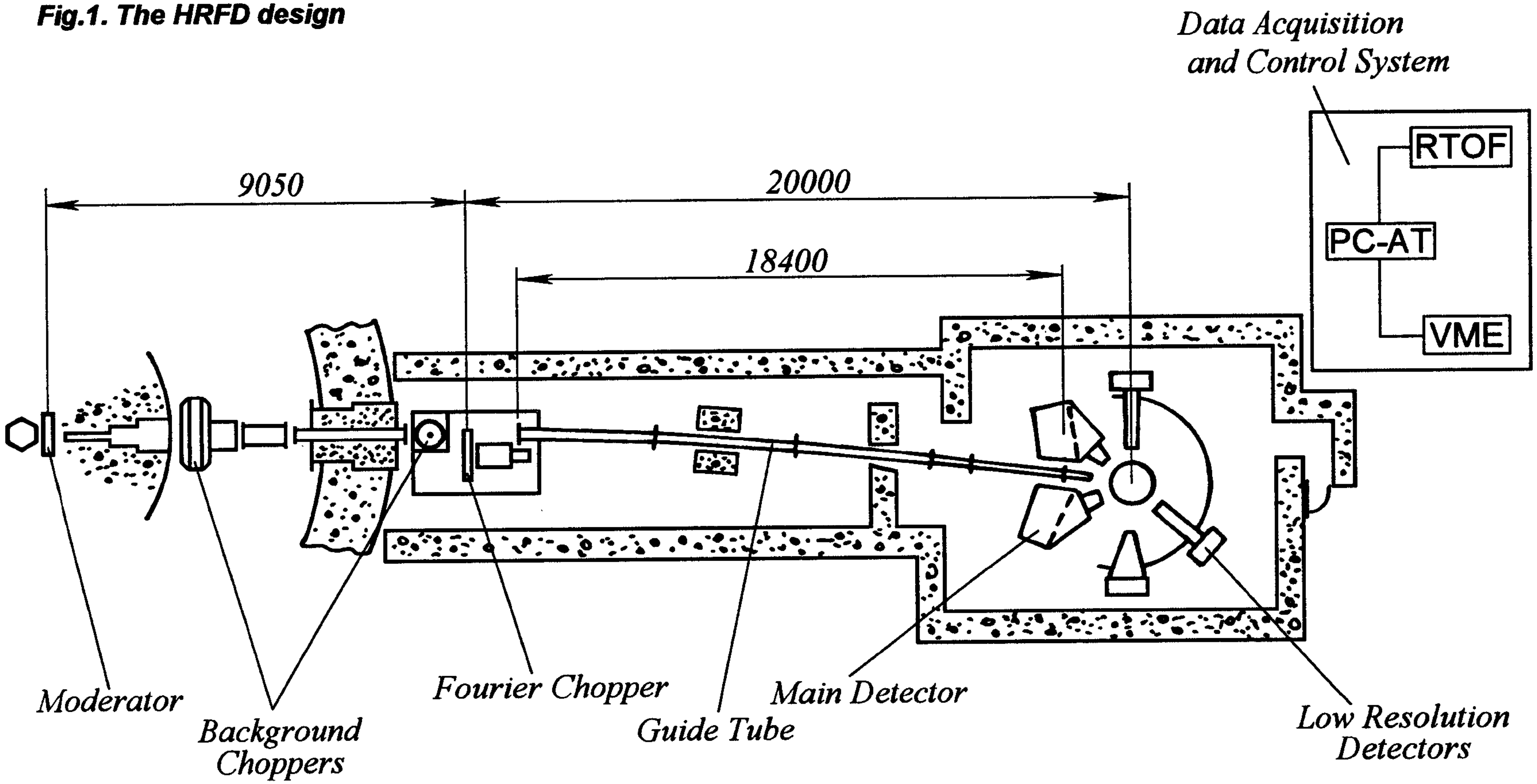
References

[1] V.D.Anan'ev et al, Neutron Scattering in The 'Nineties, Conf. Proc., p.63, Vienna, IAEA, 1985
[2] A.M.Balagurov et al, J.Appl.Cryst., 1991, 24, p.1009
[3] W.I.F.David, W.T.A.Harrison, M.W.Jonson, Communication of RAL, RAL 86-068, Chilton, 1986
[4] P.Hiismaki, H.Poyry, A.Tiitta, J Appl.Cryst. 1988, 21, p.349
[5] H.Poyry, P.Hiismaki, A.Virjo, Nucl.Instr.Meth., 1975, 126, p.421
[6] V.A.Kudryashev et al, Communication of LNPI, 1562, Leningrad, 1989 (on Russian)
[7] P.Hiismaki et al, Neutron Scattering in The 'Nineties, Conf. Proc., p.453, Vienna, IAEA, 1985
[8] H.G.Priesmeyer, J.Schroder, Mat.Res.Soc.Symp.Proc., 166 (1990) 299
[9] V.L.Aksenov et al, Communication of JINR, E13-92-456, Dubna, 1992
[10] V.B.Zlokazov, V.V.Chernyshev, J.Appl.Cryst., 1992, 25, p.447

Table. HRFD details

Beam	^{58}Ni-covered guide tube
Guide aperture	10 mm x 100 mm, variable
Moderator - sample distance	30 m
Chopper - sample distance	20.00 m
Fourier-chopper (disk-type)	Ti-Zr-alloy
outside diameter	540 mm
slit width	0.6 mm
number of slits	1,024
max speed	9,000 rpm
max beam modulation frequency	150 kHz
Thermal neutron pulse width:	
low-resolution mode	320 μs
high-resolution mode	7 μs
High-resolution detectors	^{6}Li, time-focusing
Low-resolution detector	^{3}He, position-sensitive
Aperture of the detectors:	
high-resolution 156°	0.08 sr
high-resolution 90°	0.04 sr
low-resolution 0° - 60°	0.006 sr
Wavelength interval	0.9 - 8 Å
d-spacing interval:	
high-resolution	0.5 - 6 Å
low-resolution	4 - 60 Å
Flux at sample position	10^7n/cm^2/s
Sample volume	~ 2 cm^3
Resolution for:	
$2\theta = 156°$, $d = 2$ Å	0.0005
$2\theta = 90°$, $d = 2$ Å	0.002

Fig.1. The HRFD design



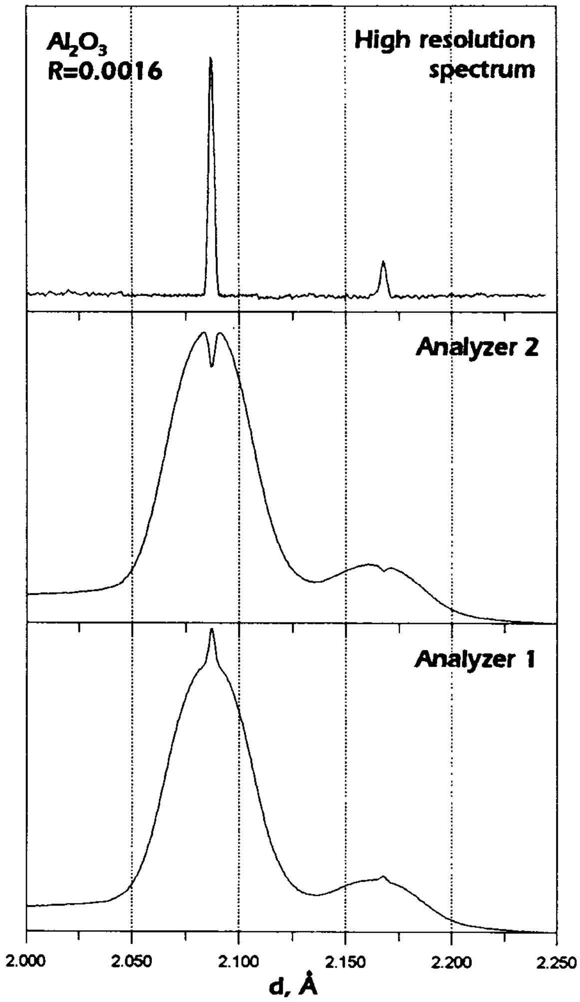


Fig.2. A small section of the Al_2O_3 diffraction pattern measured with analyzers 1 and 2 and their difference (upper).

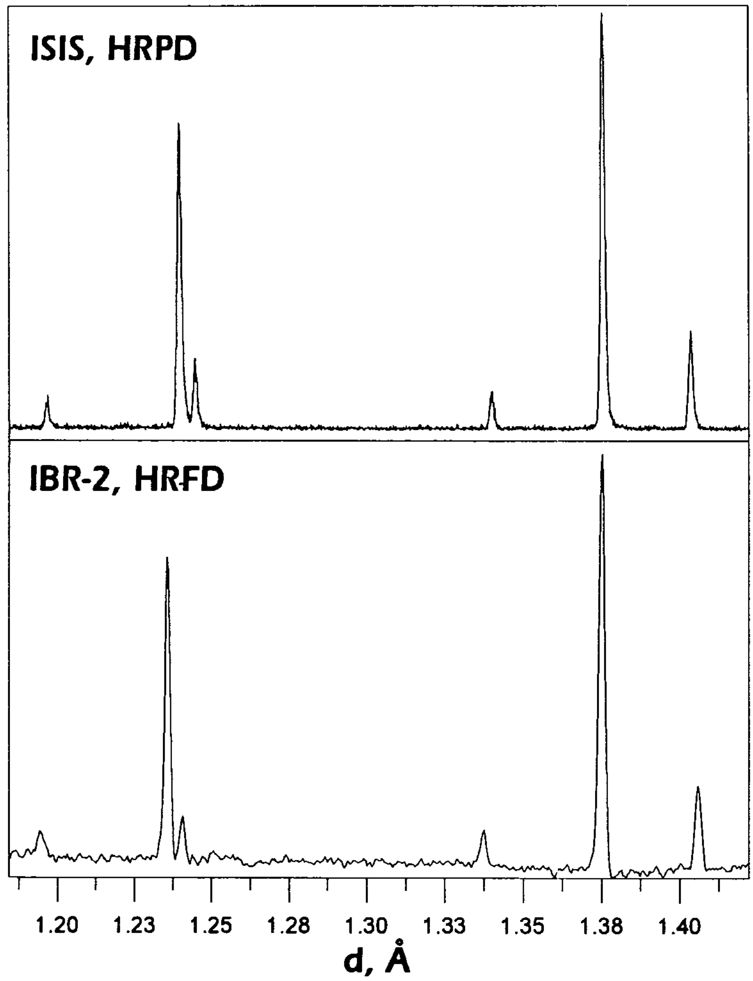


Fig.4. A small section of the Al_2O_3 diffraction pattern measured on the HRPD diffractometer at ISIS (top) and on HRFD (bottom). The horizontal scale is slightly different for the patterns.

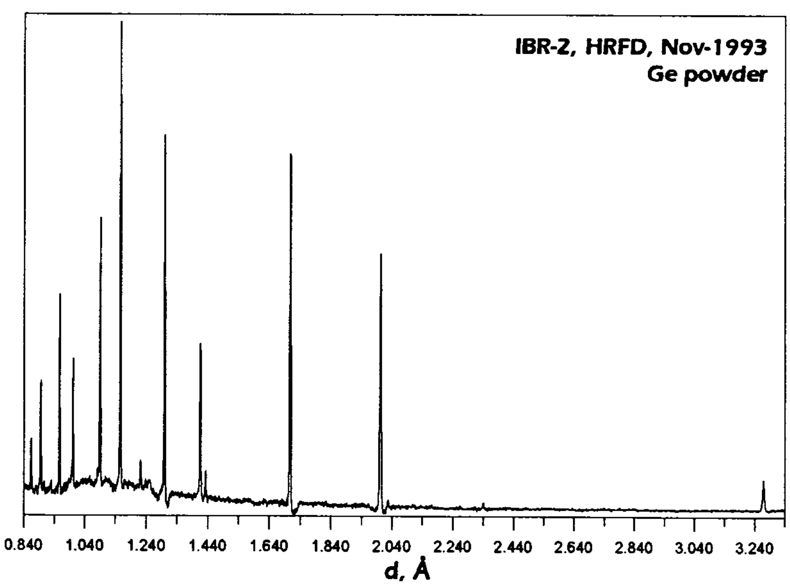


Fig.3. The Ge powder diffraction pattern measured at the IBR-2 reactor with the new Fourier diffractometer. The sample was packed in an Al-foil whose small diffraction peaks are also visible.

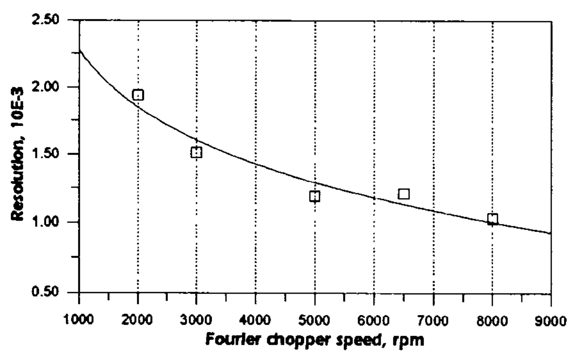


Fig.5. The dependence of the resolution ($\Delta d/d$) on the speed of the chopper rotation for the diffraction peak at $d = 1.67$ Å

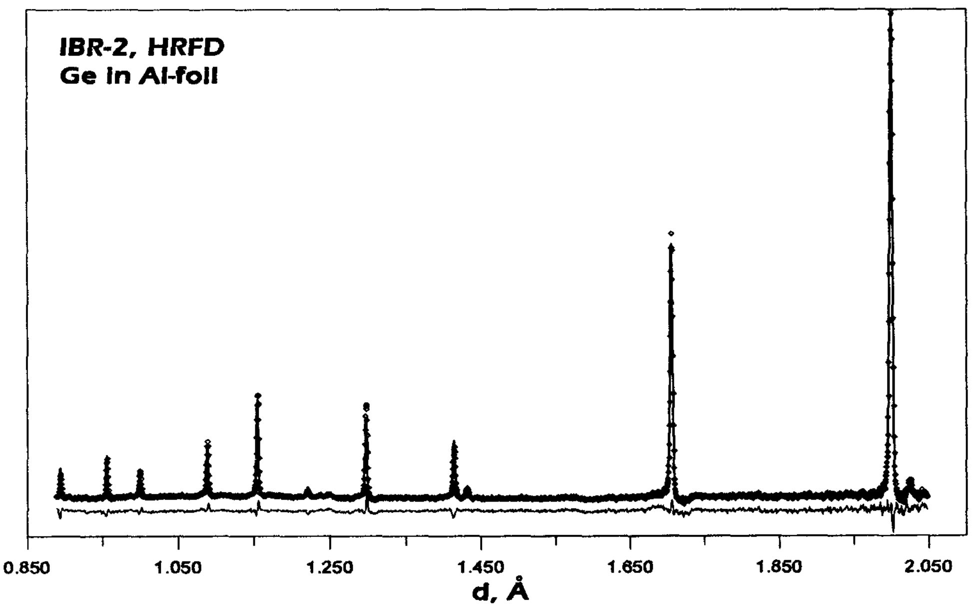


Fig.6. A section of Rietveld refinement pattern for Ge after normalization shown in Fig.3.

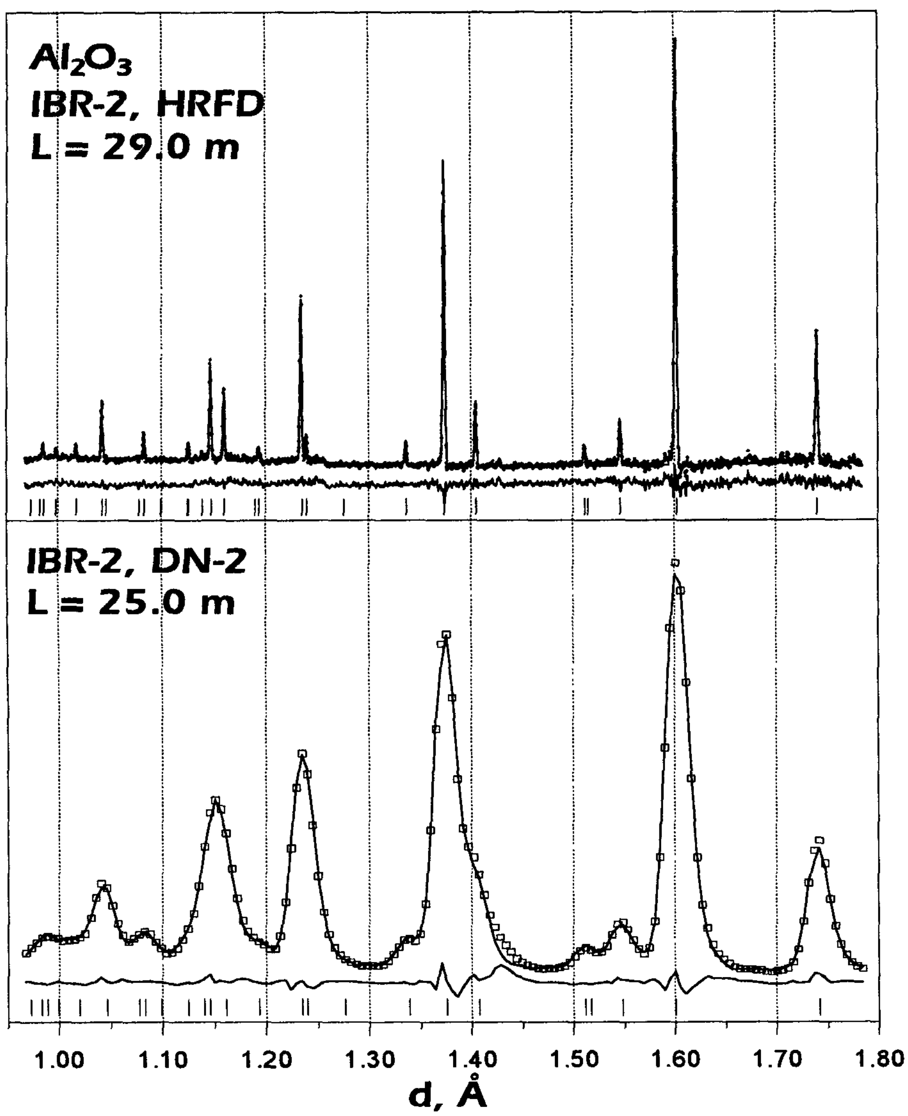


Fig.7. The comparison of Al_2O_3 diffraction patterns measured at the IBR-2 reactor with the DN-2 TOF-diffractometer and the new Fourier diffractometer. *L* is the total flight path equal to 25 m and 29 m for these two instruments, respectively. The resolution of HRFD is approximately 10 times better than that of DN-2. Experimental points, calculated profile and difference curve are shown.