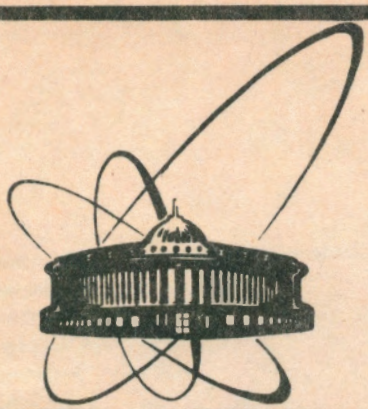

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# THE NEW FOURIER DIFFRACTOMETER AT THE IBR-2 REACTOR: DESIGN AND FIRST RESULTS

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On June 11 this year the High Resolution Fourier Diffractometer (HRFD) at the IBR-2 pulsed reactor was put into operation and the very first high-resolution diffraction spectra were measured (Fig.1). This took place just before the scheduled summer shutdown of the reactor and only a short time left for the test of the diffractometer units, modes of operation, measurement of parameters and diffraction patterns of some standard samples.

This paper presents the results after briefing the reasons for HRFD construction and the details of the diffractometer's design.

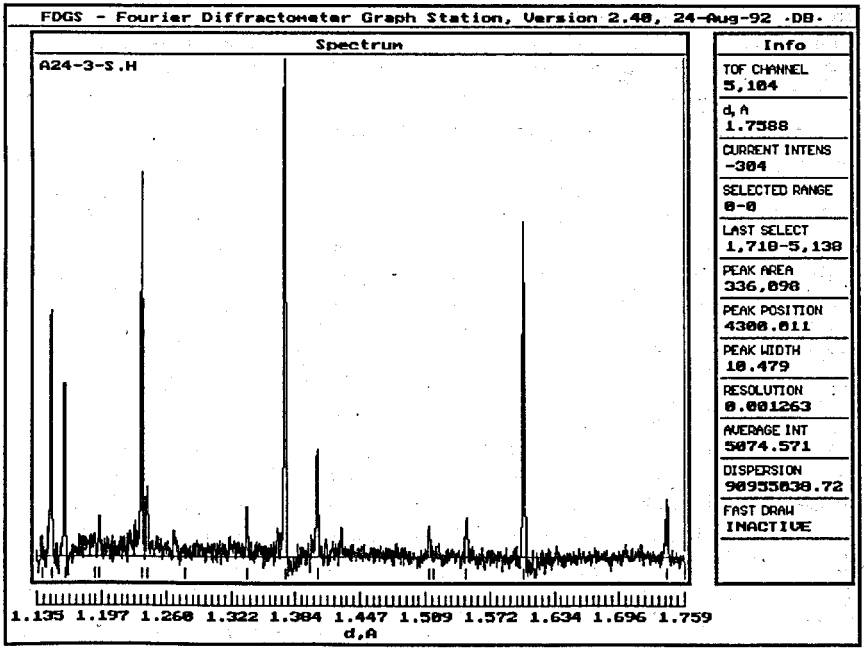


*Fig.1. First high-resolution diffraction pattern from the $Al_2O_3$ standard sample measured with HRFD as it was seen on the computer display.*

# 1 Why HRFD at the IBR-2 ?

It is well known that a powder diffractometer can be optimized in two alternative modes: either high resolution or high intensity. The precision structural studies or the 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 world's highest peak flux of thermal neutrons from the moderator surface - $10^{16}$ n/cm$^2$/s. However the resolution for the conventional TOF-experiments is poor due to the large width of the initial reactor pulse of 230 $\mu$s. After thermalization the pulse width grows up to 320 $\mu$s. So for the flight path of about 25 m the resolution is no better than 1% at d=2 Å. As the result good prospects exist for the experiments that require high intensity, but at the same time high level structural studies are impossible to be carried out with the IBR-2.

The Fourier technique is the only way from the practical viewpoint to improve the resolution of the TOF diffractometer at the IBR-2 to $\Delta d/d$ as small as $5\cdot10^{-4}$ at the intensity loss by a factor of 4 only[2]. In this case the total flux on the sample can be made as high as $10^7$n/cm$^2$/s.

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

$$I(t) \sim \int R_s(t-\tau)\sigma(\tau)d\tau + B(t) \ , \qquad (1)$$

where in rough approximation neglecting all geometrical contributions $R_s$ is the source pulse, $\sigma$ 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

$$I(t) \sim \int R_c(t-\tau)\sigma(\tau)d\tau + c\int \sigma(\tau)d\tau + B(t) \ , \qquad (2)$$

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. It can be shown that the width of $R_c$ is inversely proportional to the maximum beam modulation frequency $\Omega_{max}$, which is equal to $N_s V_m$, where $N_s$ is the number of chopper slits and $V_m$ is the maximum chopper speed. For $N_s$=1024 and $V_m$=9000 rpm, $\Omega$=150 kHz, so the time width of diffraction maxima can be as small as 7 $\mu$s.

The situation with the Fourier chopper at a pulsed neutron source is, in some sense, the combination of the two previous cases:

$$I(t) \sim \int R_s(t-\tau)R_c(t-\tau)\sigma(\tau)d\tau + c\int R_s(t-\tau)\sigma(\tau)d\tau + B(t) \ . \qquad (3)$$

One can see, that in (3) both resolution functions $R_s$ and $R_c$ are present, thus the diffraction peaks are very narrow ($\Delta t \approx 7\mu s$), the correlation background is not constant



and is proportional to the resolution function of the source. The additional modulation with $R_s$ is important to decrease 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 $\Delta t$ is equal to the width of $R_s$, i.e. 320 $\mu$s for the IBR-2 reactor.

The correlation background is the principal difference between the conventional TOF and Fourier diffractometer. Its level is strongly sample dependent, moreover it depends on the wavelength range in wich the pattern is measured. 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.

# 2 The HRFD design (Fig.2)

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. The chopper 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 wall the Fourier-chopper is placed. The focusing guide tube acts both as a forming element of the neutron beam and a filter of fast neutrons and $\gamma$-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 movable 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 the 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, the master clock unit and eight correlator modules. Two these 8192-channel analyzers operate simultaneously with the Fourier chopper pickup signals in opposite phases. Thus the difference between two spectra measured with these two analyzers is the high-resolution spectrum, whereas their sum is the low resolution spectrum.

The RTOF analyzers are connected to the PC-computer (IBM-386) by the BITBUS standard serial interface. PC is used as the interface between the user and the measuring unit.

# 3 The HRFD performance

### 3.1. The wavelength and d-spacing range

The wavelength range is limited by the contrast in neutron transmission through the chopper slits (lower limit) and the thickness of the chopper disk (upper limit).

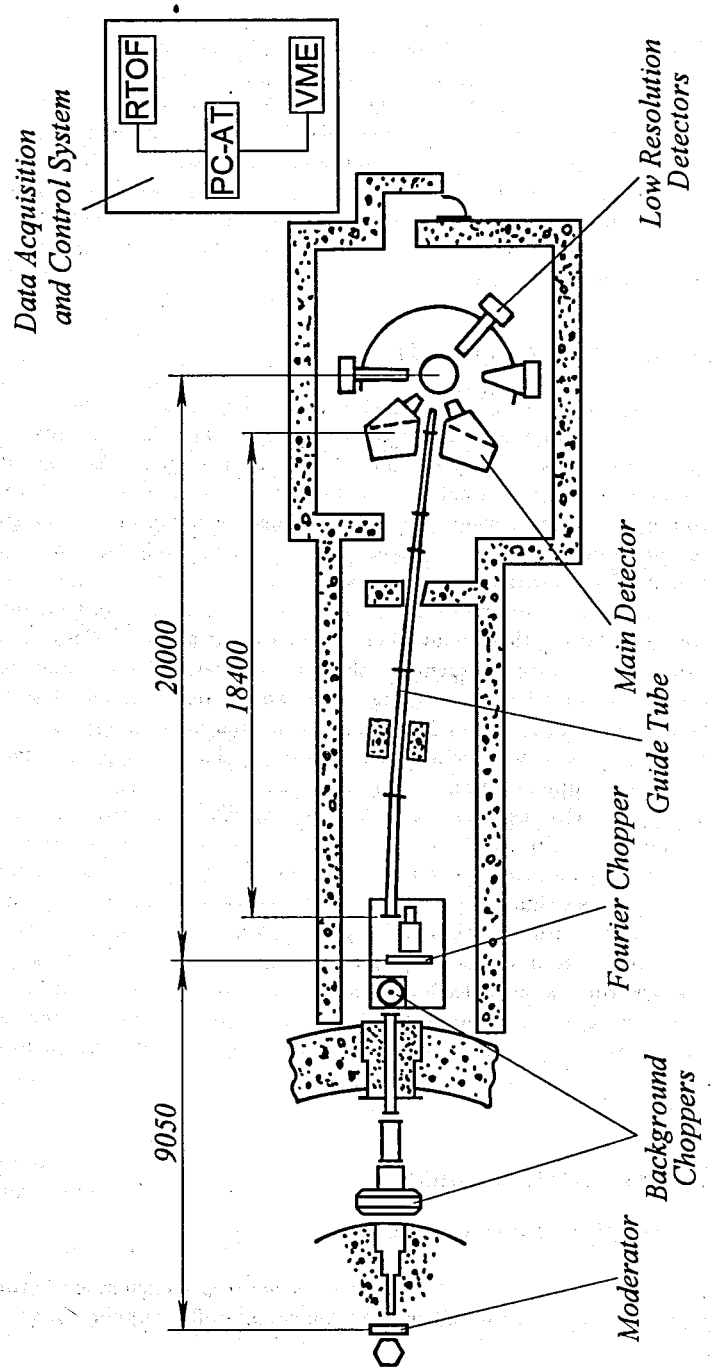


*Fig.2. Layout of HRFD.*



The calculations and the mini-SFINKS operation experience give for the lower and upper limit the value 0.9 Å and 12 Å, respectively. This allows measuring the diffraction pattern with the main detector in the d-spacing range between 0.5 and 6 Å. At larger d the high resolution is unnecessary, and one can use the detectors placed at lower angles. These detectors will give the diffraction information in the range between 4 and 60 Å.

### 3.2. The neutron flux

At the IBR-2 reactor the total thermal neutron flux into $2\pi$ sr is equal to $10^{13}$n/cm$^2$/s. The visible area of the moderator is 400 cm$^2$, so at the distance of 28 m the flux at the sample position will be $\sim 6 \cdot 10^7$n/cm$^2$/s. Taking into account the transmission of the chopper and a guide tube one can hope to have $\sim 10^7$n/cm$^2$/s on the sample.

### 3.3. The resolution

The resolution of the time-of-flight powder diffractometer is (to the first approximation):

$$R = \Delta d/d = [(\Delta t_o/t)^2 + (\gamma ctg\theta)^2]^{1/2} \ , \qquad (4)$$

where $\Delta t_o$ is the width of the thermal neutron pulse, in the $\gamma$ all geometrical uncertainties are included. For HRFD the first term in (4) is:

$$\Delta t_o/t = \Delta t_o/253 L\lambda = 6.6 \cdot 10^{-4}/d \ , \qquad (5)$$

where d is in Å. The divergence of the initial beam, sample dimensions and the difference between the real detector surface and the exact time-focused locus contribute to the second term in (4). The calculated value for this term is equal to $3.3 \cdot 10^{-4}$, so the total width of the resolution function is about $5 \cdot 10^{-4}$, if d equals 2 Å.

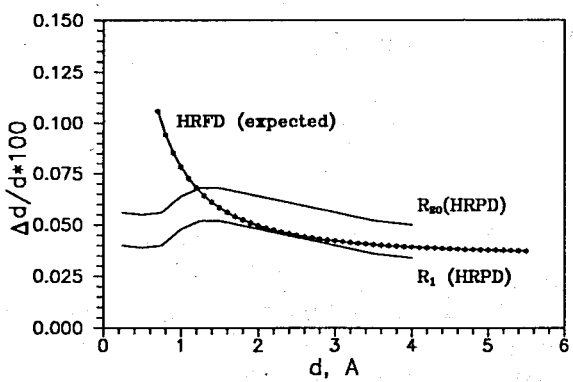


*Fig.3. The comparison of the resolution function (full width of diffraction peak at half maximum height) of HRFD (expected) and HRPD. $R_1$ and $R_{20}$ are the first and twentieth rings of the HRPD detector.*

In Fig.3 the expected resolution function of HRFD is compared with the resolution function of HRPD[3], the best at the moment neutron powder high-resolution diffractometer. In the large d-interval the resolution of these two instruments is very close.

The Table lists the instrument details.

**Table. Instrument details for HRFD (proposed)**

| | |
|---|---|
| Beam | $^{58}$Ni-covered guide tube |
| Guide aperture | 10 mm x 100 mm, variable |
| Moderator - sample distance | 30 m |
| Chopper - sample distance | 2000 cm |
| Fourier-chopper (disk-type) | Ti-Zr-alloy |
| outside diameter | 540 mm |
| slit width | 0.6 mm |
| number of slits | 1024 |
| max speed | 9000 rpm |
| max beam modulation frequency | 150 kHz |
| Thermal neutron pulse width: | |
| low-resolution mode | 320 $\mu$s |
| high-resolution mode | 7 $\mu$s |
| 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 - 12 Å |
| d-spacing interval: | |
| high-resolution | 0.5 - 6 Å |
| low-resolution | 4 - 60 Å |
| Flux at sample position | $10^7$n/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 |

## 4 The experimental results

In Fig.4 two diffraction patterns of aluminum oxide are compared. The lower one was measured at the IBR-2 with the existing diffractometer DN-2, conventionally used for the structure studies. The resolution of this diffractometer is about 1% for d=2 Å. In the upper part the same d-region is shown measured on the new Fourier diffractometer. A short d-interval of this pattern is shown in Fig.5. It contains three diffraction peaks. In the bottom, the DN-2 spectrum is shown. In the middle part, the raw data from HRFD are shown. Here one sees two spectra, the first measured for the case of the positive sign high resolution term, the second for the negative. After subtraction the high resolution spectrum is obtained. The curves in the middle part of this figure can be considered as the correlation background.



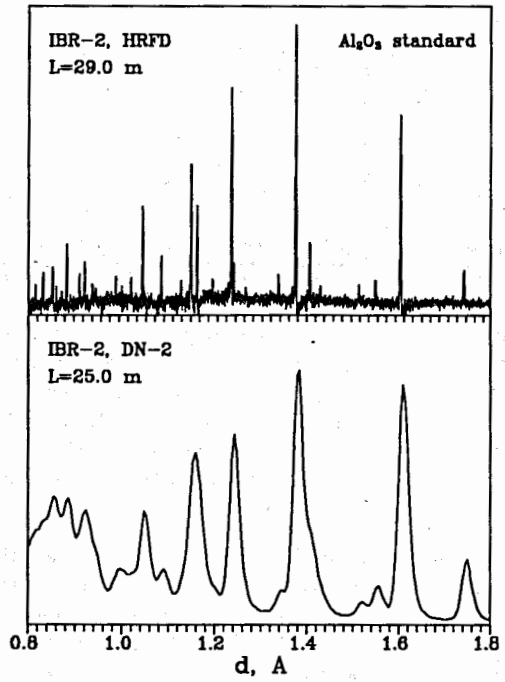


*Fig.4. The comparison of $Al_2O_3$ diffraction patterns measured at the IBR-2 reactor with the DN-2 TOF-diffractometer and the new Fourier diffractometer. L is the total flight path equal to 25 m and 29 m for these two instruments, respectively. The resolution of HRFD is approximately 10 times better than that of DN-2.*

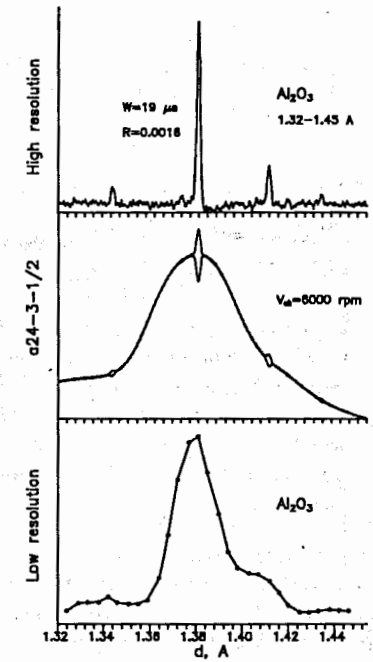


*Fig.5. A small section of the $Al_2O_3$ diffraction pattern measured in the conventional TOF regime (bottom), the raw data - two spectra - from HRFD (middle) and high resolution spectrum (upper). The measurement was done at the chopper speed of 6000 rpm. For the strong peak the resolution R is equal to 0.0016.*

The shape of the diffraction peak is very like a gaussian (Fig.6). However some small difference exists and the reasons of it should be clarified.

HRFD has a very linear d scale. The observed and calculated positions of the diffraction maxima coincide with good accuracy without any corrections being introduced (Fig.7). The relation for the total width of the diffraction peaks $W_{tot}$ can be obtained from the formula (4):

$$W_{tot}^2 = W_t^2 + W_\theta^2 \cdot N^2 \ , \qquad (6)$$

where $W_t$ and $W_\theta$ are the TOF and geometrical terms, respectively, N is the TOF channel number. In Fig.8 the experimental points for the diffraction peak widths and the least square line calculated after this relation are shown.

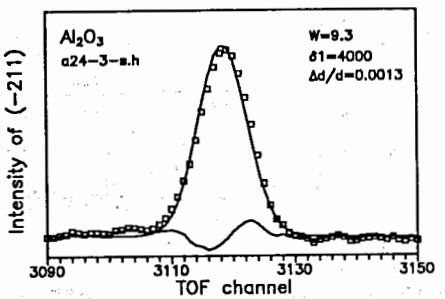


*Fig.6. One of the diffraction peaks of $Al_2O_3$. The experimental points (squares), the gaussian line and the difference curve are shown. The width of the time channel was 2 μs, the delay was equal to 4000 channels.*

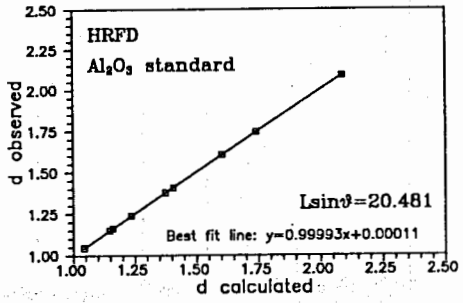


*Fig.7. The observed and calculated $d_{hkl}$ values for the $Al_2O_3$ standard sample. The parameters of the best fit line are 0.99993 (instead of 1) and 0.00011 (instead of 0). The calculation was done without any corrections.*

The resolution was measured at various chopper speed (Fig.9) 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.

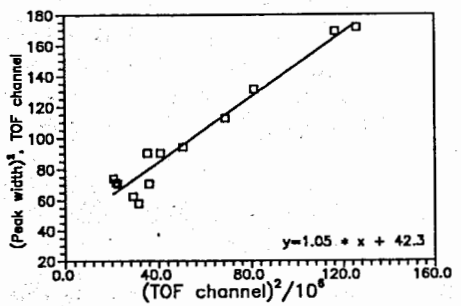


*Fig.8. The fit of experimental values for the total width of diffraction peaks after formula (6).*

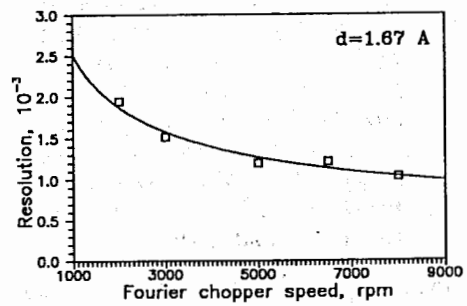


*Fig.9. The dependence of the resolution ($\Delta d/d$) on the speed of the chopper rotation for the diffraction peak at d=1.67 Å.*



Due to a shortage of beam time only very few and preliminary measurements of the scientifically interesting samples were done.
The result for one of them is shown in Fig.10. It is a small portion of the diffraction pattern from the $CuLi_{0.1}V_{0.1}Fe_{1.8}O_4$ sample. At higher temperatures (above 700 K) this compound has the cubic Fd3m symmetry that can be conserved in the quenched sample. But one can see that only peak (444) has a regular shape and a small width, the other peaks have a very complicated shape. It may be due to symmetry disturbance or some internal stress.

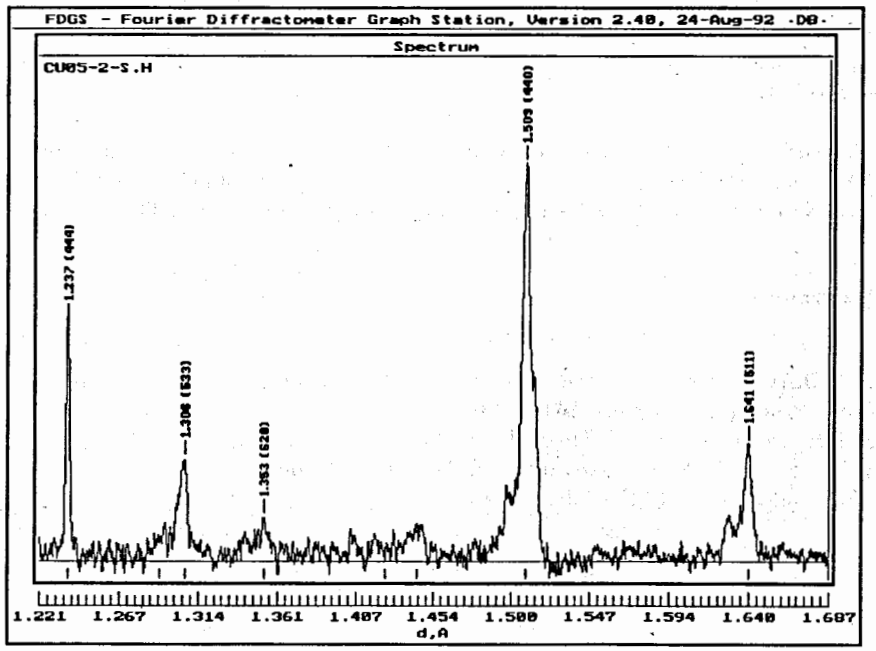


*Fig.10. Part of the diffraction pattern from $CuLi_{0.1}V_{0.1}Fe_{1.8}O_4$. One may see that only the peak (444) has a regular shape.*

## 5 Conclusions

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 both with respect to resolution and intensity. The resolution of 0.0015 in $\Delta d/d$ was achieved at first attempt. The expected value of $\Delta d/d$ equal to 0.0005 can be reached after the diffractometer units are aligned and their assembly completed.

One of the advantages of HRFD - the possibility of changing the relation between the resolution and intensity on a large scale - has been also confirmed.
Now it is obvious that a lot of problems can be solved with the help of such a diffractometer as HRFD. The experience acquired in the diffraction studies with mini-SFINKS, D2B, HRPD and some other machines provided diverse data that allow one to conclude that the structure solution and refinement are the basic problems for high resolution neutron diffractometers. The studies of high temperature superconductors give a good example of a very successful application of high resolution neutron powder diffraction.

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