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NEW DEVELOPMENTS OF TOF NEUTRON DIFFRACTION AT THE IBR-2 PULSED REACTOR

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Abstract

Development of high-resolution RTOF Fourier technique for powder neutron diffraction studies is being continued at the IBR-2 pulsed reactor in Dubna. Besides some technical improvements in the operating HRFD instrument, a new dedicated instrument, Fourier Strain Diffractometer (FSD), for investigation of residual stresses in bulk materials has been constructed at IBR-2 in 1999. With a new HRFD Fourier chopper smaller than 10 μs TOF contribution in a resolution function was obtained in the experiment with perfect Si single crystal. A series of diffraction experiments with the beams from a new methane cold neutron moderator installed at the IBR-2 in 1999 is discussed. A comparison with the results obtained with the conventional water comb-like moderator shows that for various types of experiments, which are performed at HRFD and DN-2 diffractometers, the methane cold neutron source provides better conditions.

1. Introduction

The high-flux IBR-2 pulsed reactor in Dubna is operational since 1984 at an average thermal power of 2 MW, peak power in pulse of 1500 MW, and repetition rate of 5 Hz. A high neutron flux (about 10^{16} n/cm^2/s in pulse) and a low repetition rate provide good conditions for various types of neutron diffraction experiments. At previous ICANS-XIV Conference improvements in the performance of the high resolution Fourier diffractometer, HRFD, diffractometer for high pressure studies, DN-12, and multipurpose diffractometer, DN-2, at the IBR-2 pulsed reactor in Dubna were discussed [1]. In this paper, recent developments in Fourier technique for powder neutron diffraction, including measurements of internal stresses in bulk materials, and results of diffraction experiments at a new methane cold moderator, which was installed at the IBR-2 in the end of 1999, are presented.

2. HRFD, high resolution RTOF Fourier diffractometer

In the operating HRFD instrument [2] the rotor and stator of the Fourier chopper were replaced by new ones with much better contrast, which improve considerably the quality of the diffraction patterns. At present, the Fourier chopper contrast, which can be defined as $R=I_{max}/I_{min}$, where I_{max} is intensity for fully open state and I_{min} is intensity for fully closed state of rotor + stator, is more than 20. It means that a portion of non-correlated intensity is not higher than 5%.

With a new Fourier chopper, the measurement of the TOF contribution in a resolution function of HRFD was performed. It was done with thin silicon wafers, which were found to be suitable as focusing monochromator and analyzer for high-resolution quasielastic neutron scattering [3]. For using them at TOF instruments in a high-resolution mode the adequate neutron pulse width should be smaller than 10 μs. Neither TOF spectrometer at short-pulse neutron source, nor TOF spectrometer with conventional Fermi chopper can provide such short pulse with appropriate intensity. Our experiment confirmed that it is possible with Fourier chopper. In Fig.1, the diffraction spectrum of commercial 6″ diameter thin Si [111] plate (provided by M.Popovici, Missouri) measured at $2\theta=152°$ with 6000 rpm Fourier chopper maximal speed is seen as four orders of reflection: 111, 333, 444, and 555 (also some peaks from Al jacket are seen).

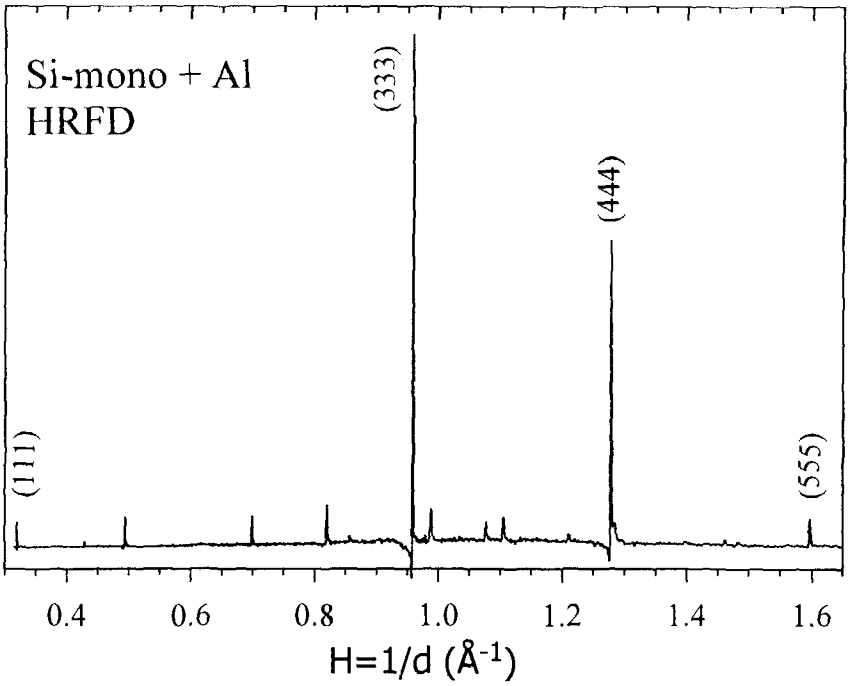

Fig.1. Diffraction pattern from Si-monochromator (hhh-plane) in Al-jacket measured at HRFD for 6000 rpm maximal chopper speed and $2\theta=152°$.

Taking into account that good approximation for diffraction peaks full width (FWHM), $W(d)$, dependence on d-spacing is $W(d)^2 = (W_{TOF})^2 + (W_{ang})^2 \cdot d^2$, where W_{TOF} is the contribution connected with maximal chopper velocity (W_{TOF}~1/V), W_{ang} is geometrical contribution (W_{ang}~$\Delta\theta/\mathrm{tg}\theta$), and measuring $W(d)$ for several chopper speeds, we were able to determine W_{TOF}. In Fig.2, W_{TOF} as a function of 1/V is shown. One can see that the TOF contribution into the full width is indeed inversely proportional to chopper velocity. If Fourier chopper speed V>7000 rpm, TOF contribution is lower than 10 μs, in particular for V=10000 rpm, W_{TOF} could be as low as 6.8 μs.

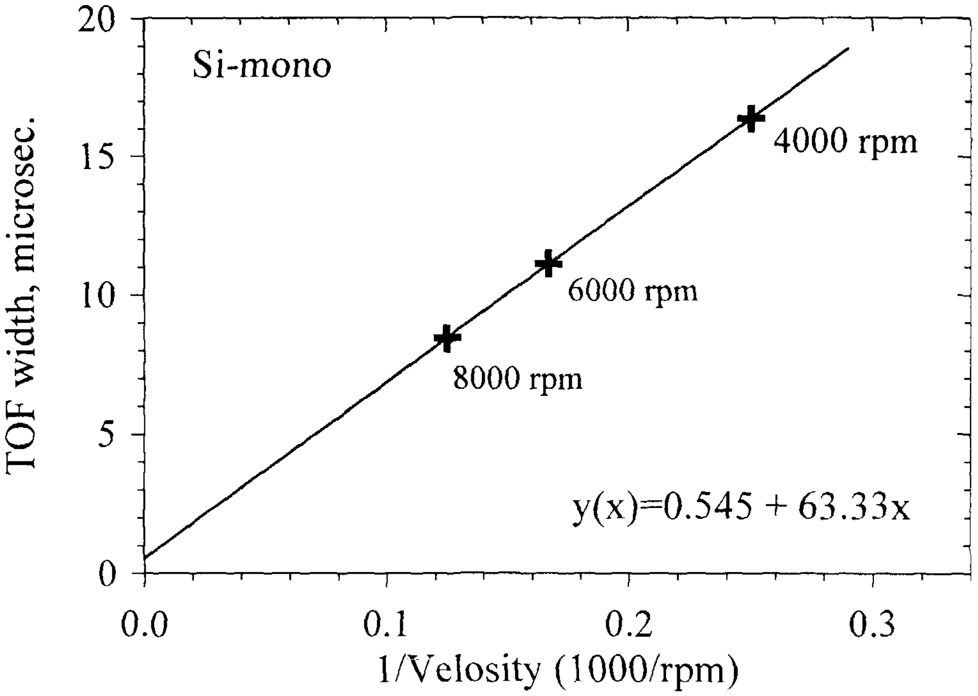

Fig.2. TOF contribution in the full width as a function of 1/V. The points were measured at maximal Fourier chopper speed V=4000, 6000, and 8000 rpm. The line is the least-square fit.

3. FSD, a new RTOF Fourier diffractometer for strain analysis

A new high-resolution neutron powder Fourier diffractometer for strain analysis in bulk materials, FSD, has been constructed at the IBR-2 pulsed reactor. The FSD design satisfies several requirements: high luminosity, high resolution, specific sample environment, wide range of d_{hkl}, and fixed scattering angles $2\theta = \pm 90°$. The collaboration around FSD includes FLNP JINR (Dubna), PNPI (Gatchina), ISSP (Budapest) and IfZP (Drezden). The most important units of the diffractometer (Fig.3) are:

- 19 m long neutron-guide tube with vertical focusing,
- fast Fourier chopper for the modulation of the neutron beam intensity,
- 5.5 m straight neutron-guide to form the thermal neutron beam at the sample position,
- MultiCon detector system with both geometrical and electronic focusing,
- mechanical devices, including 5-axis goniometer "Huber" and loading machines,
- RTOF (VME-based) analyzer for experimental data acquisition,
- supervising VME electronics.

This was accomplished on a basis of experience of carrying out the residual stresses investigations in bulk samples with Fourier correlation technique in PNPI, Gatchina (mini-SFINKS diffractometer [4]), GKSS, Geesthacht (FSS diffractometer [5]), and FLNP JINR, Dubna (HRFD diffractometer [2]).

In May 1999, the first test experiments were carried out with FSD. From the results of measurements with vanadium sample a comparison of neutron intensity spectral distribution was performed for FSD and HRFD diffractometers. Due to larger radius of neutron guide curvature FSD's spectrum is shifted to shorter wavelengths by $\Delta\lambda \approx 0.22$ Å (Fig.4). Correspondingly, the measurement of smaller d-spacings, which are important in the studies of simple metals, is possible with FSD.

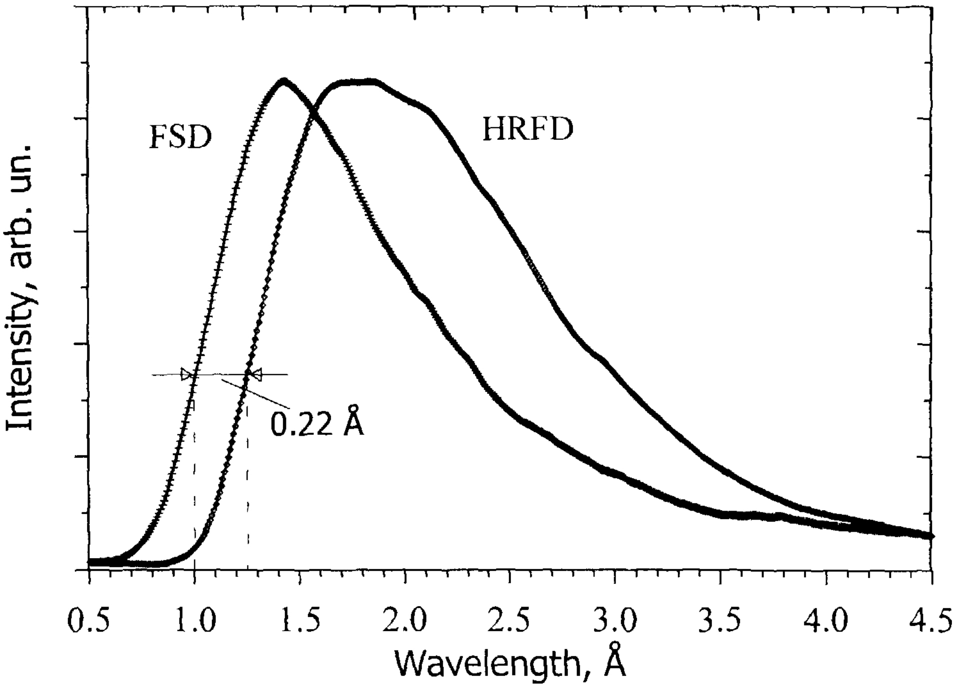

Fig.4. Incident neutron flux distributions for HRFD and FSD instruments. The FSD flux is shifted to the shorter wavelengths due to larger radius of neutron guide curvature.

As the FSD detector, we are going to use a MultiCon system [6] based on ZnS(Ag) scintillators with combination of electronic and time focusing (Fig.5). In final FSD version we are going to use two MultiCon detectors at scattering angles $2\theta=\pm90°$ (8 elements each) with total solid angle of about 0.16 sr and two MultiCon detectors at $2\theta=\pm160°$.

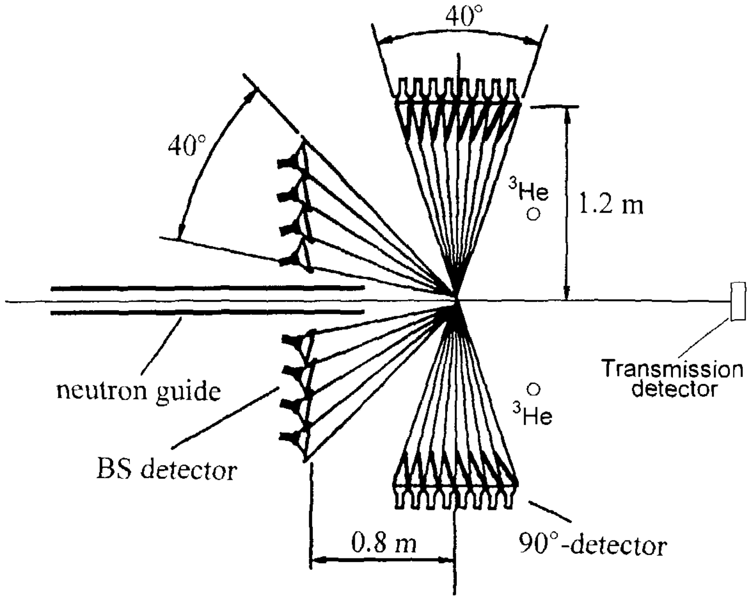

Fig.5. The FSD detector system. For main ±90° and ±160° *detectors combined geometrical and electronic time focusing is used, thus providing high resolution and large solid angle.*

The diffraction spectra from standard samples Ge, α-Fe, and Al_2O_3 were already measured (Fig.6) and the results show that FSD has indeed sufficiently high resolution

$\Delta d/d \approx 0.004$ for 90° detector, which is optimal for the studies of residual stresses in simple metals and alloys (Fig.7).

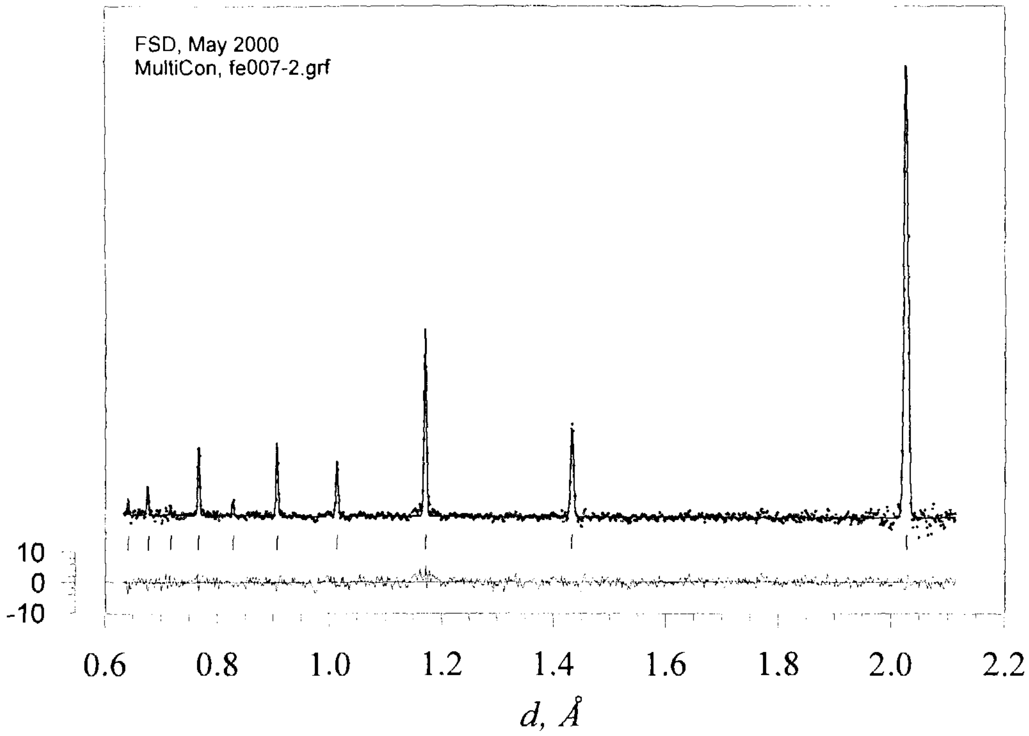

Fig.6. Example of α-Fe diffraction pattern measured with FSD by the 1^{st} *MultiCon element at* $2\theta=\pm90°$ *and 6000 rpm Fourier chopper maximal speed. Experimental points, calculated profile and difference curve are shown. The difference curve is normalized on the mean square deviation.*

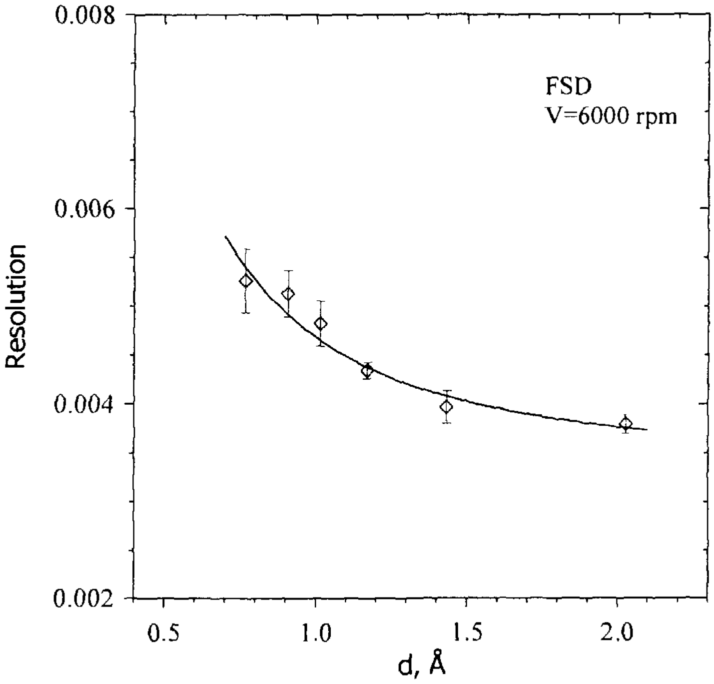

Fig.7. FSD resolution function measured with α-Fe at $2\theta=\pm90°$ *and 6000 rpm Fourier chopper maximal speed.*

4. A new solid methane cold source: results of diffraction experiments

In late 1999, a new methane cold neutron source (CNS) has been installed at the IBR-2 pulsed reactor [7]. CNS is a very flexible machine; it can operate at ~30 K or ~65 K with solid methane and also at 300 K with water pre-moderator (without methane). A switch from one regime to another can be done during a short time. It means that one may think about appropriate distribution of experiments between various regimes of CNS operation. Here we discuss some results of test experiments at HRFD and DN-2 diffractometers, which are situated at the IBR-2 at the CNS side.

At both diffractometers neutron guide tubes with R≈1500 m are used for beam transportation from the source to a sample position. For such an arrangement lowering of moderator temperature leads to the evident gain in total neutron flux. Firstly, it is connected with increase of effective neutron guide solid angle for neutrons with $\lambda>\lambda^*$, where λ^* is neutron guide characteristic wavelength. The second effect is the result of the neutron flux distribution shifting to the larger λ, where a neutron guide transmission function is close to 100%. This shift gives the gain of about 2 in total flux for $\lambda^*\approx1.5$ Å, which is characteristic wavelength for HRFD and DN-2 (Fig.8).

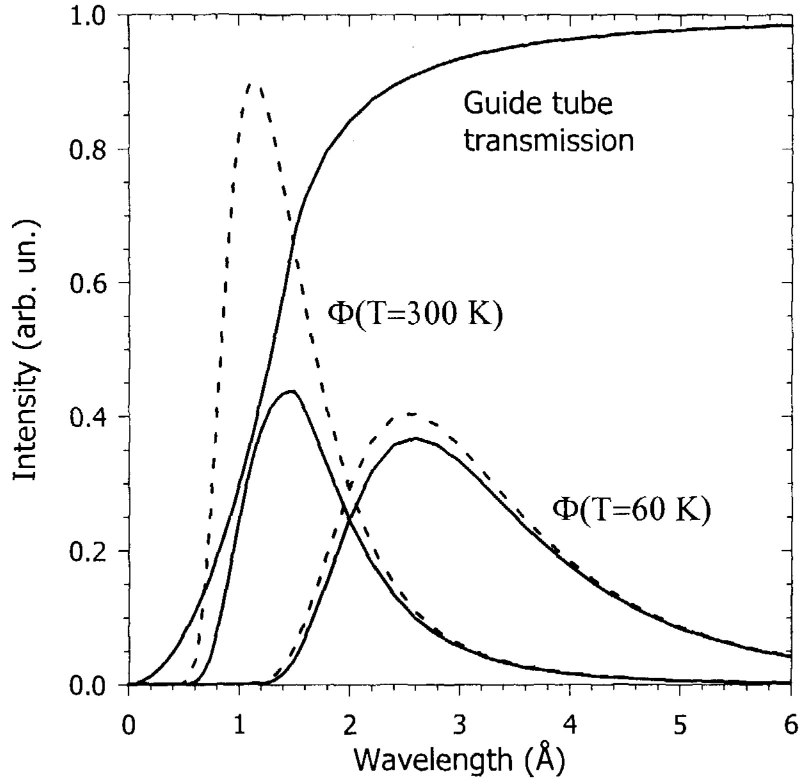

Fig.8. Neutron flux distributions calculated for moderator temperatures T=300 K and 60 K (dashed lines) and the same after neutron guide with transmission function corresponding $\lambda^*\approx1.5$ Å (solid lines). The flux decrease is great for warm spectrum and negligible for cold one.

Another important point, which should be taken into account when diffraction experiments at various moderators are analyzed, is particular dependence of a resolution function on *d*-spacing. In contrast to diffractometers at spallation neutron sources like ISIS, an effective neutron pulse width is practically constant for HRFD (about 15 μs) and DN-2 (about 300 μs), i.e. there is no λ-dependence. It means that resolution becomes worse if the wavelength decreases and the shift to the higher wavelength is useful.

Neutron flux distributions measured at different CNS states are shown in Fig.9. One can see clearly that the shift to the longer λ values follows the increase of the total area due to the above-discussed effect of better neutron guide transmission. The strong deeps at around λ=4 Å are due to thick beryllium reflector included in CNS design. The gain (loss) in the neutron flux at a sample position as a function of λ, which is simply a ratio of intensities measured with CNS at low temperature and with conventional for the IBR-2 water comb-like moderator, are displayed in Fig.10. One can conclude that for T=70 K, the gain for λ≥4 Å is quite good, while the loss for short wavelength (λ≤2 Å) is not too strong, and crystals with complex structures (large unit cell dimensions) can be investigated at this regime quite effectively.

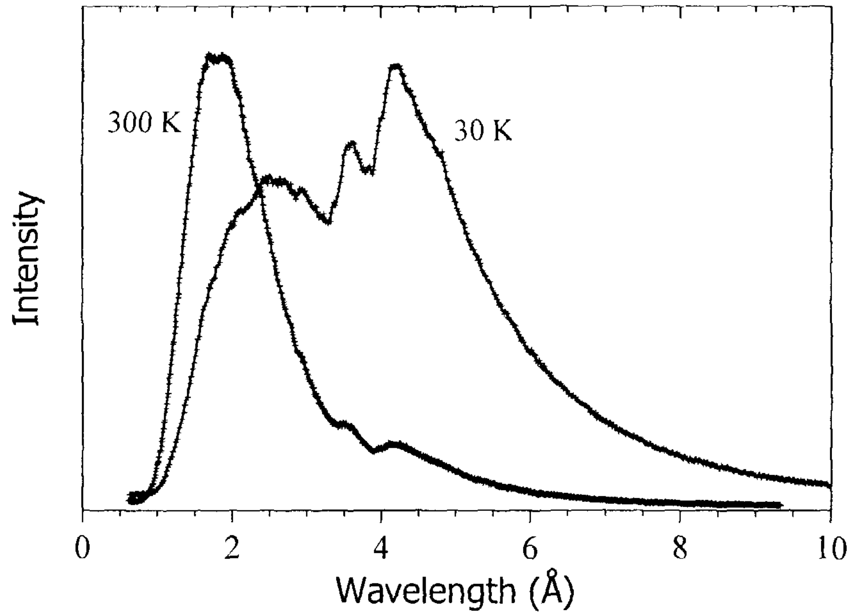

Fig.9. Incident spectra measured at HRFD for CNS temperature 30 K with solid methane and 300 K without methane (water pre-moderator only). The deeps at around λ=4 Å are due to thick beryllium reflector included in CNS design.

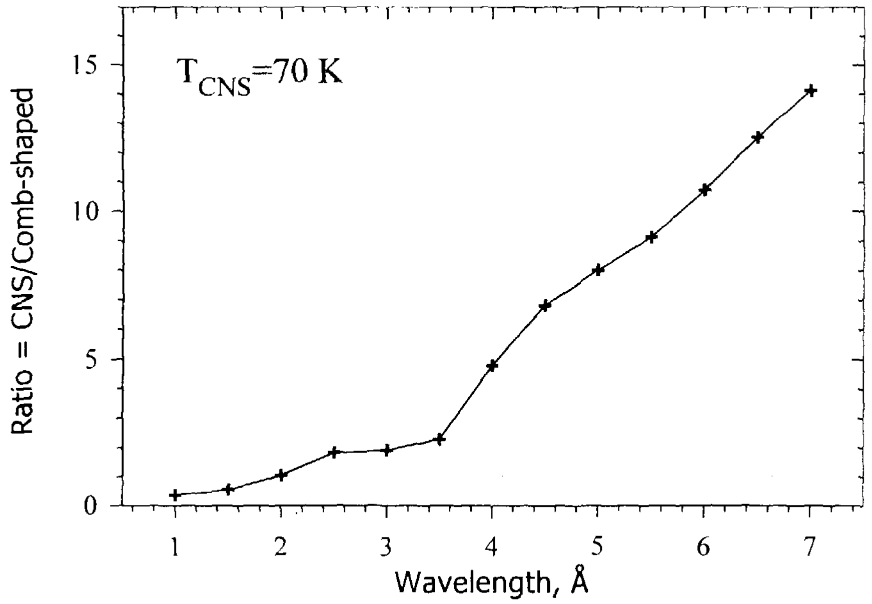

Fig.10. Gain factor in total neutron flux at the HRFD sample position measured as a ratio of intensities with CNS at 70 K and a water comb-like moderator at 300 K.

Direct experiments showed that even for such "simple" structure as Al_2O_3 (V_c=85 Å^3), the quality of Rietveld refinement of atomic coordinates is practically the same for any CNS regime. Of course, thermal parameters can be refined with much smaller uncertainties for "warm" CNS state. For structures with "medium" (V_c≈200 Å^3) and "high" (V_c≥400 Å^3) complexity, the refinement may be performed with better quality if the CNS temperature is 65 K or lower due to better statistics at high *d*-spacings (Fig.11).

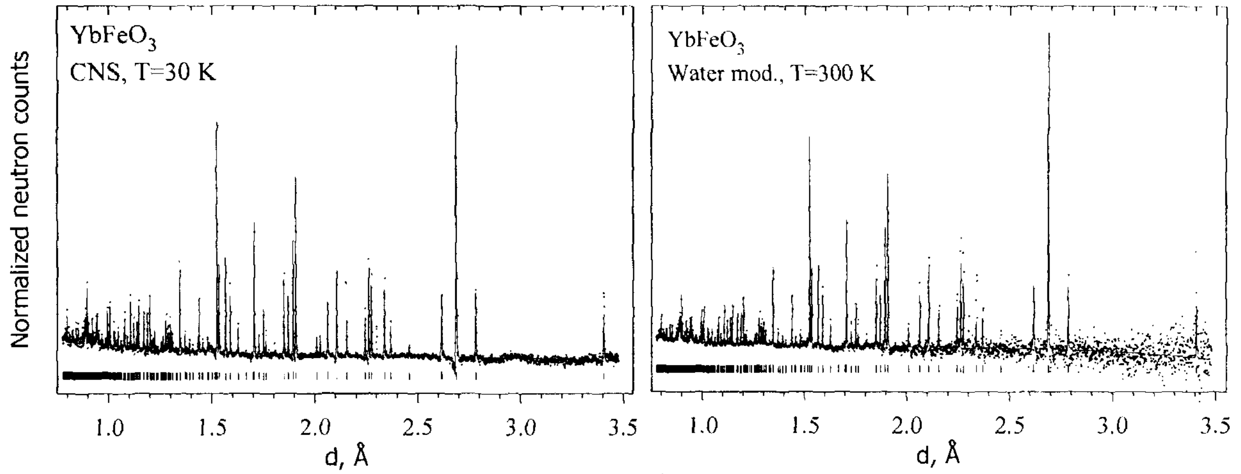

Fig.11. Rietveld refinement of $YbFeO_3$ (V_c=220 Å^3) *diffraction patterns, measured with CNS at 30 K (left) and water comb-like moderator at 300 K (right).*

Another useful consequence of higher intensity of TOF diffractometer at longer wavelengths is much better resolution, which can be obtained for large *d*-spacings by measuring of diffraction patterns at higher scattering angles. This is especially important for experiments in real-time regime, *e.g.* if solid-state chemical reaction is studied. As it was already shown in Ref. [8] the IBR-2 affords the real possibility of measuring the whole diffraction pattern using only one neutron pulse. In this case, a special set-up of the detector assembly would allow studying of irreversible processes in crystals with temporal resolution of about the width of the reactor pulse, *i.e.* 300 μs.

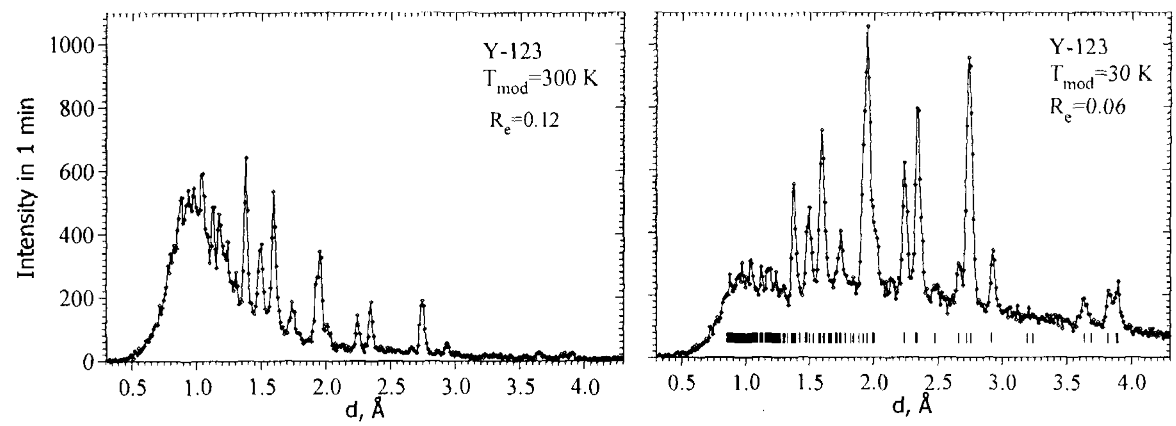

Fig.12. The comparison of the $YBa_2Cu_3O_{6.5}$ *(V_c=170 Å^3) diffraction patterns measured with water moderator (T=300 K) (left) and CNS (T=30 K) (right) in 1 min. Experimental (statistical) R-factor is two times better if CNS is used.*

In Ref. [8], as an example of experiment at DN-2 diffractometer, the real-time diffraction study of the formation of the Y-123 phase through a solid-state reaction of initial oxides was discussed. It was shown that the formation of the final product goes through the intermediate phases and at least three of them were identified: $BaCuO_2$, $Y_2Cu_2O_5$ and Y_2BaCuO_5. That experiment has been performed with conventional water moderator. The CNS provides much better conditions for such kind of experiments. Higher intensity and better resolution at large *d*-spacings (Fig.12) assist the intermediate phases identification or the oxygen content determination in final product. For instance, the mean square deviation for oxygen index is ±0.05 for 1 min spectrum measured with CNS.

Acknowledgements

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