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PERFORMANCE OF THE HIGH RESOLUTION FOURIER DIFFRACTOMETER AT THE IBR-2 PULSED REACTOR; LATEST RESULTS

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ABSTRACT

The High Resolution Fourier Diffractometer (HRFD) was designed for neutron powder structural studies and was realized at the IBR-2 pulsed reactor in Dubna. The RTOF method is used at HRFD for gathering diffraction spectra. High $\Delta d/d$ resolution, close to 0.001, and high neutron flux at the sample position, ~10^7 n/cm^2/s, make HRFD one of the best diffractometers in the world intended for the structural studies of polycrystalline materials and studies of internal stresses in bulk samples.

1. INTRODUCTION

HRFD is a neutron reverse time of flight Fourier-diffractometer intended for diffraction experiments with polycrystalline materials at a resolution level of about 0.001 or better [1]. It operates at the high flux pulsed reactor IBR-2 in Dubna and is a spin-off of the cooperation between JINR (Dubna), PNPI (Gatchina) and VTT (Espoo). The main special feature of HRFD, distinguishing it from other Fourier-diffractometers, such as mini-SFINKS [2] and FSS [3], operating at steady state reactors, is the analysis of the triple correlations between signals from the neutron source, Fourier-chopper and detector. As a result, the neutron intensity measured with HRFD is:

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

where R_c is the resolution function of the Fourier chopper, R_s is the source pulse, σ is the scattering cross section of the sample, B is the conventional background and c is a certain constant close to 1. The second term, which can be called "the correlation background" is proportional not to the total detected intensity as in the case at steady state reactors, but to the intensity measured in short time intervals equal to the width of the source pulse (~350 μs for IBR-2). This leads to a substantial decrease of that

correlation background, better quality of the diffraction patterns and permits the useful wavelength interval to be extended.

The first experiments with HRFD were performed in 1992 and the results from them were reported at the ICANS XII conference [1]. Since, the development of HRFD has continued, first of all with the detectors, data acquisition system and data analysis procedure. In early 1994 regular experiments were started at HRFD, mainly in two directions: structural studies of new materials and of residual stress in bulk samples [4]. Here we report the current situation with HRFD performance and some results of recent experiments.

2. THE EXPERIMENTAL SET-UP

The HRFD diffractometer is installed on one of the beam-lines of the IBR-2 pulsed reactor (Fig.1). Immediately behind the reactor shielding a background chopper is placed as a filter for fast neutrons and γ-rays. The distance between the moderator and the Fourier chopper is about 9 m. The neutron beam before the Fourier chopper is formed by a straight mirror neutron guide, and after it, by a focusing, horizontal and vertical, curved neutron guide 19 m in length. This guide tube acts both as the forming element for the neutron beam and an additional filter of fast neutrons and γ-rays. Having passed through the choppers and the neutron guide, the neutron flux at the sample position is close to 10^7 n/cm^2/s. Currently, only two of the four detectors planned for operation are working, at the scattering angles of +152° and +90°. The RTOF electronics of the HRFD is based on a dual-delay-line correlator [2] of special design. At present, two 8192-channel analyzers operate simultaneously with the Fourier chopper pickup signals, in opposite phases. For a back-scattering detector these analyzers cover the 3.2 Å *d*-spacing interval, usually from 0.6 Å to 3.8 Å, if the channel width is equal to 4 μs. For the 90°-detector a new DSP-TMS320C25 based correlation analyzer has recently been designed and tested [5].

3. INSTRUMENT PERFORMANCE

Usual experiments with standard samples have been performed with HRFD. They revealed the main parameters of the diffractometer to be close to the computed ones [6]. The resolution function of HRFD depends on the maximum modulation frequency of the neutron beam and on geometrical uncertainties. In principle, the value of 0.00035 is accessible at HRFD for both terms and then $R=0.00035\sqrt{2}=0.0005$ (for d=2 Å). Usually, however, experiments are performed with R≈0.0010 (Fig.2). The effect of such high resolution is shown in Figure 3. The *d*-spacing scale of HRFD is very linear (Fig.4) and that offers the possibility of obtaining lattice parameters of powders with an accuracy close to 10^{-5}.

HRFD Layout

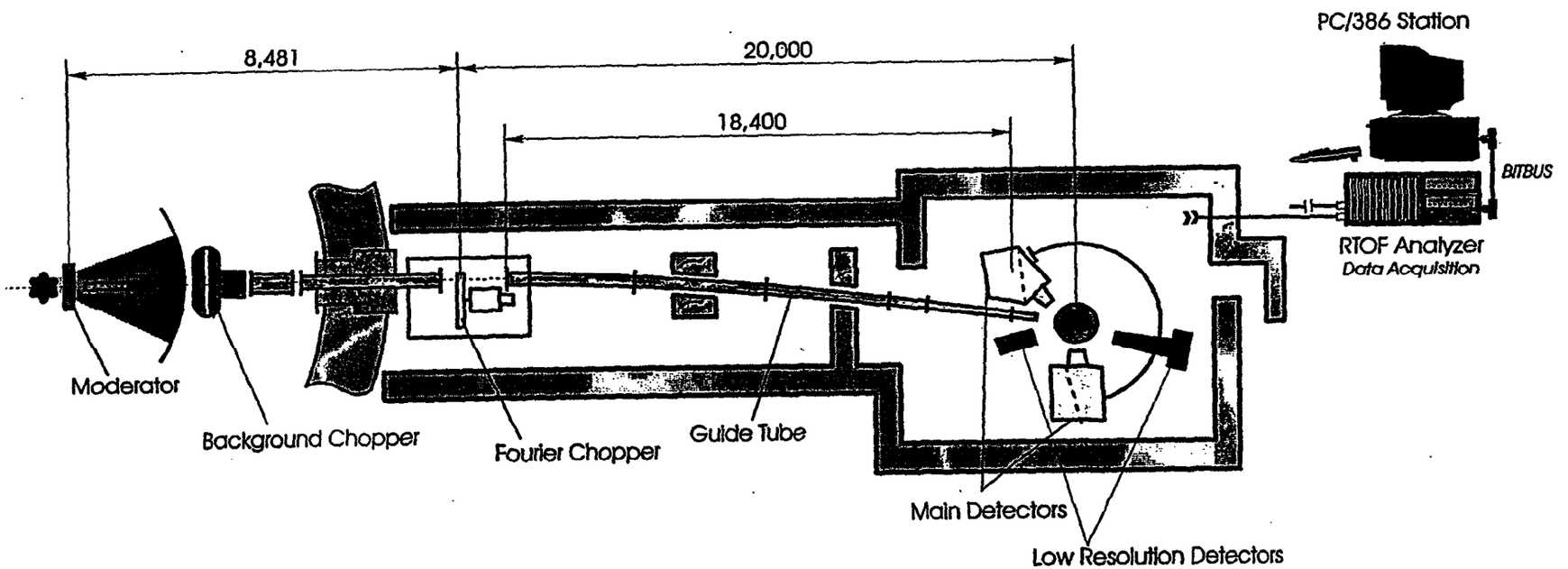

Fig. 1

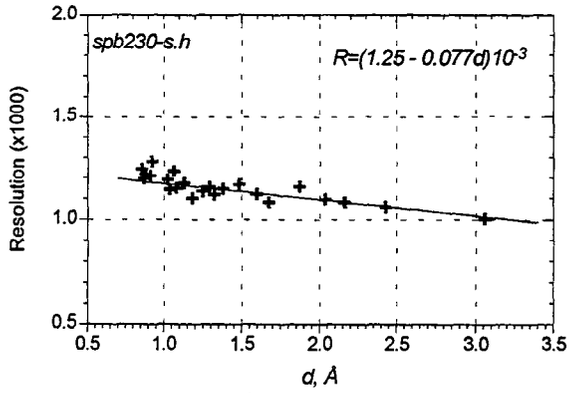

Fig.2. The resolution function of HRFD as a function of d-spacing measured on a high quality powder sample $Bi_{1.7}Mg_{0.3}Nb_{0.7}O_{6.5}$ (SPB) with pyrochlor-type cubic structure.

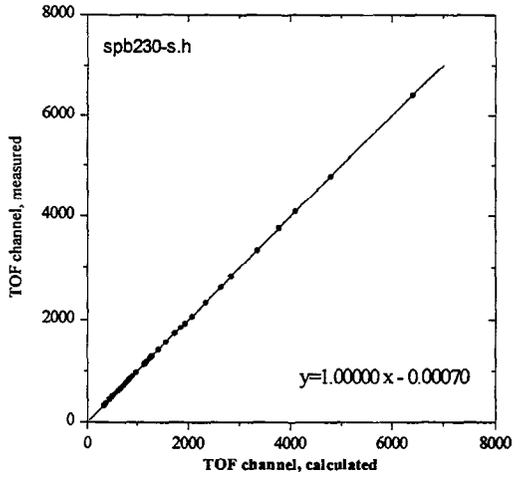

Fig.4. Measured versus calculated TOF channel positions of diffraction peaks of the SPB sample demonstrating the very linear TOF-scale of HRFD.

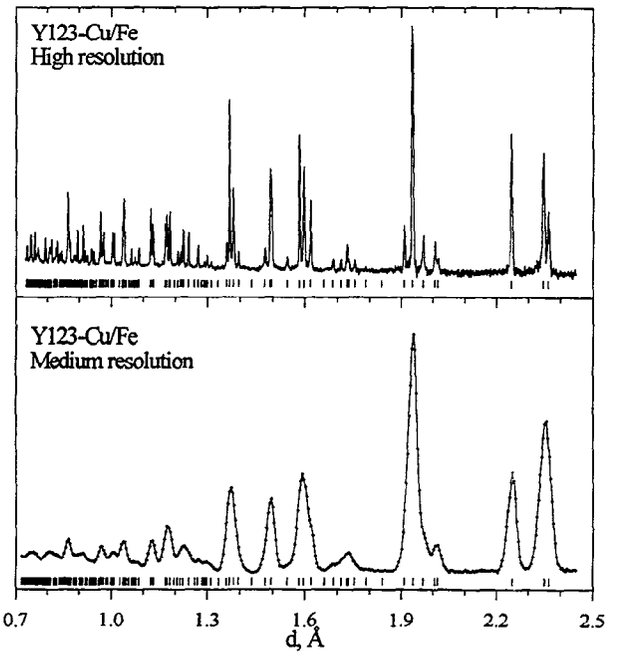

Fig.3. Comparison of $YBa_2Cu_{2.7}{}^{57}Fe_{0.3}O_{6.3}$ diffraction patterns measured with HRFD in conventional and high resolution modes.

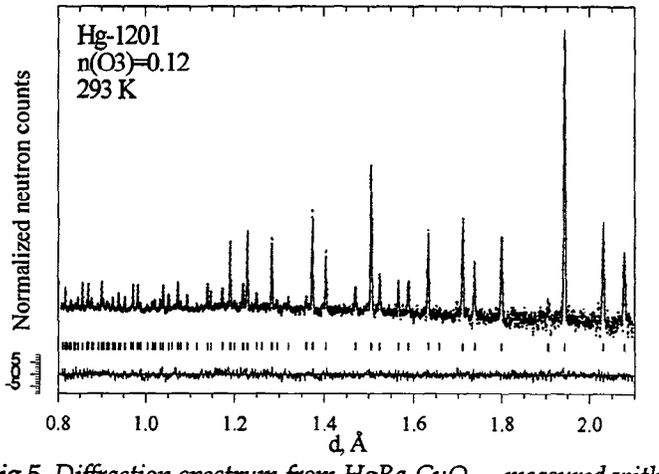

Fig.5. Diffraction spectrum from $HgBa_2CuO_{4.12}$ measured with HRFD and processed by the Rietveld method.

4. EXAMPLES OF EXPERIMENTAL STUDIES

Among the physical experiments performed at HRFD, the following HTSC compounds were studied: $Y(^{44}Ca)Ba_2Cu_4O_8$, $YBa_2{}^{65}Cu_{2.7}Zn_{0.3}O_7$ and several $HgBa_2CuO_{4+y}$ (Hg-1201) samples with various oxygen contents. As an example, in Figure 5 the diffraction pattern of $HgBa_2CuO_{4.12}$ is shown after Rietveld refinement.

In this study [7] the precise structural data for Hg-1201 system were obtained and the real dependence of the superconducting phase transition temperature on oxygen content was found. The very high resolution of HRFD helps the realistic values of experimental structure factors to be obtained for many diffraction peaks of a sample under study and to calculate experimental and difference Fourier maps for the scattering density (Fig.6).

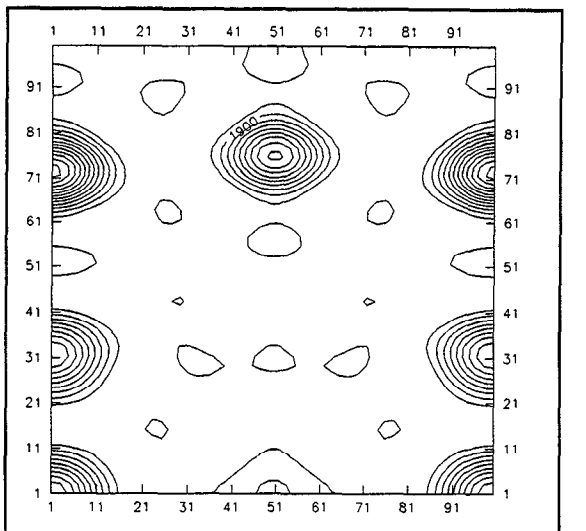

Fig.6. Experimental Fourier map of the y=0 plane of $YBa_2Cu_{2.7}{}^{57}Fe_{0.3}O_{6.3}$, containing Cu1, O4 atoms in chain, Cu2, O2/O3 atoms in plane and apical oxygen O1. The Cu1 and Cu2 sites occupied partly by iron atoms.

5. CONCLUSION

In [2] it was supposed that the Fourier technique at a pulsed neutron source can successfully be used to refine low-symmetry crystal structure with much improved signal-to-noise ratio compared with a steady state source case. The present experience fully confirmed that supposition.

The further development of HRFD includes setting up two large detectors at $2\theta=90°$ and doubling the solid angle of the back scattering detector. The sample environment equipment which includes a cryostat down to 4 K and a He-refrigerator for $8 \leq T \leq 300$ K will be complemented by a furnace up to 900 K and gas high pressure cells up to 1 GPa.

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