

## RECENT DEVELOPMENTS OF NEUTRON DIFFRACTION INSTRUMENTS AT THE IBR-2 PULSED REACTOR

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### ABSTRACT

Improvements in the performance of the high resolution Fourier diffractometer, HRFD, diffractometer for high pressure studies, DN-12, and single crystal diffractometer, DN-2, at the IBR-2 pulsed reactor in Dubna are discussed. On HRFD, a second back-scattering detector bank started operation and a geometrical contribution to the resolution function was reduced. On DN-12, a new supermirror neutron guide tube was installed, which substantially increases the neutron flux. On DN-2, a new two-dimensional detector is used. That allows registering three-dimensional intensity distributions. Examples of experimental results are also presented.

### 1. Introduction

The IBR-2 pulsed reactor in Dubna is one of the most effective neutron sources for condensed matter studies. A high neutron flux (about  $10^{16}$  n/cm<sup>2</sup>/s in pulse) and a low repetition rate (5 Hz) provide good conditions for various types of neutron scattering investigations. Together with inelastic and small angle scattering, reflectometry and polarized neutron optics, neutron diffraction is an essential scientific area at IBR-2 [1]. All together five diffractometers are in operation: HRFD, for high resolution powder and single crystal studies; DN-2, medium resolution, multipurpose diffractometer; SNIM, diffractometer with a high pulsed magnetic field; DN-12, for experiments with micro samples, and NSVR, diffractometer for texture studies. During the last two years the upgrading of some of them is accomplished and in this paper the new characteristics of HRFD, DN-2, and DN-12 together with examples of recent results are discussed.

### 2. HRFD, high resolution RTOF Fourier diffractometer

HRFD is the TOF correlation spectrometer using a fast Fourier chopper for modulating the intensity of the incident neutron beam and the reverse TOF-method (RTOF) for data acquisition. The detailed description of the method, HRFD design, and the reasons why it is very suitable for high resolution powder diffraction at long pulse neutron sources were earlier presented in papers [2,3] and at ICANS conferences [4,5]. The HRFD *d*-spacing resolution depends on the highest frequency of intensity modulation (highest chopper rotating speed,  $V_m$ ) and on geometrical contribution and could be as small as 0.05%. In reality this level can

be reached only with a special collimator at the incident beam and a sample of very high quality.

In the last two years on HRFD a second back-scattering detector bank at  $2\theta = -152^\circ$  was put into operation, which reduced the time of experiments, and final adjustment of the detector elements was done, which decreased the geometrical component of the resolution function. In Fig.1, one of the diffraction peaks from  $\text{La}_2\text{CuO}_4$  single crystal measured at  $V_m=8000$  rpm is shown. The relative full width of the peak ( $d=1.335$  Å), that can be regarded as the HRFD resolution at this point, is about  $9 \cdot 10^{-4}$ . For powder samples of even very high quality like the  $\text{Al}_2\text{O}_3$  sintered reference sample, the lowest limit in the same conditions is  $11.5 \cdot 10^{-4}$  (at  $d=1.5$  Å). Nevertheless, at this resolution level the famous  $\text{Al}_2\text{O}_3$  doublet (433) / (432) (R3c setting) is quite well separated (Fig.2). The diffraction line width dependence on  $d$ -spacing is linear in a wide  $d$ -range, which makes it possible to use only one model for the line shape [2].

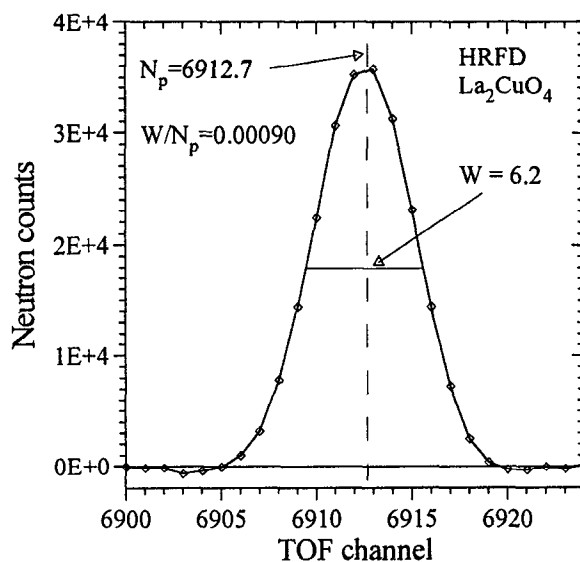


Fig.1. A typical Bragg peak for a  $\text{La}_2\text{CuO}_4$  single crystal measured at  $V_m=8000$  rpm with the TOF channel width of  $2 \mu\text{s}$ . The position and the full width of the peak are equal to 6912.7 and 6.2 TOF channels, respectively, i.e. the relative resolution is about 0.0009. Small negative deeps on both sides of the peak are due to the correlation nature of counts.

At present HRFD is used for structure refinements of powders, for experiments with single crystals if it's a very high  $d$ -spacing resolution is needed, and for residual stress investigations in bulk samples. Recently the results of high-resolution powder studies of  $\text{HgBa}_2\text{CuO}_{4+\delta}$  [6] and  $\text{HgBa}_2\text{CuO}_4\text{F}_\delta$  [7] structures, investigation of phase separation phenomenon in  $\text{La}_2\text{CuO}_{4+\delta}$  [8], and semiconductor – metal phase transition in CMR compounds [9] were published.

Figure 3 shows an example of the Rietveld refinement of the  $\text{La}_{0.8}\text{Ba}_{0.2}\text{MnO}_3$  diffraction pattern. For  $\text{La}_{0.35}\text{Pr}_{0.35}\text{Ca}_{0.30}\text{MnO}_3$ , the analysis of the characteristic bond length (Fig.4) reveals that coherent deviation of oxygen octahedra is not significant at room temperature and in the FM phase ( $T_c \approx 160$  K). The axial distance Mn-O1 is slightly shortening with decreasing of temperature from room temperature to 40 K while equatorial Mn-O21 and Mn-O22 distances significantly and simultaneously change at  $T \approx 200$  K. This fact proves the ordering of static Jahn-Teller distortions in the equatorial plane of  $\text{MnO}_6$  octahedra at this temperature [9].

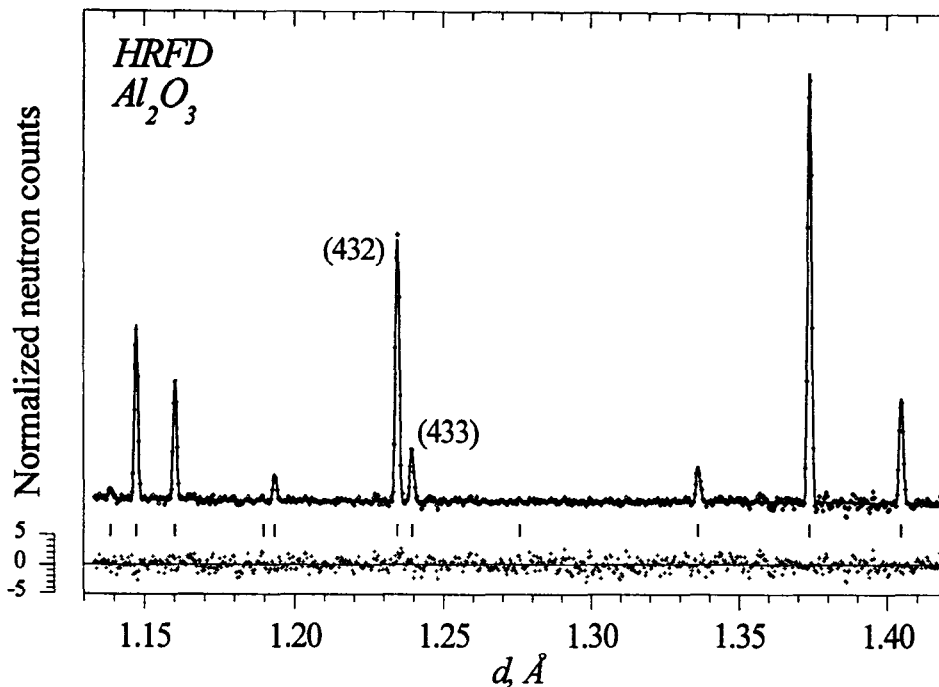


Fig.2. A part of the diffraction pattern of the reference  $\text{Al}_2\text{O}_3$  sample measured with HRFD at  $V_m=8000 \text{ rpm}$  and processed by the Rietveld method. The difference curve is normalized on the mean-square deviation.

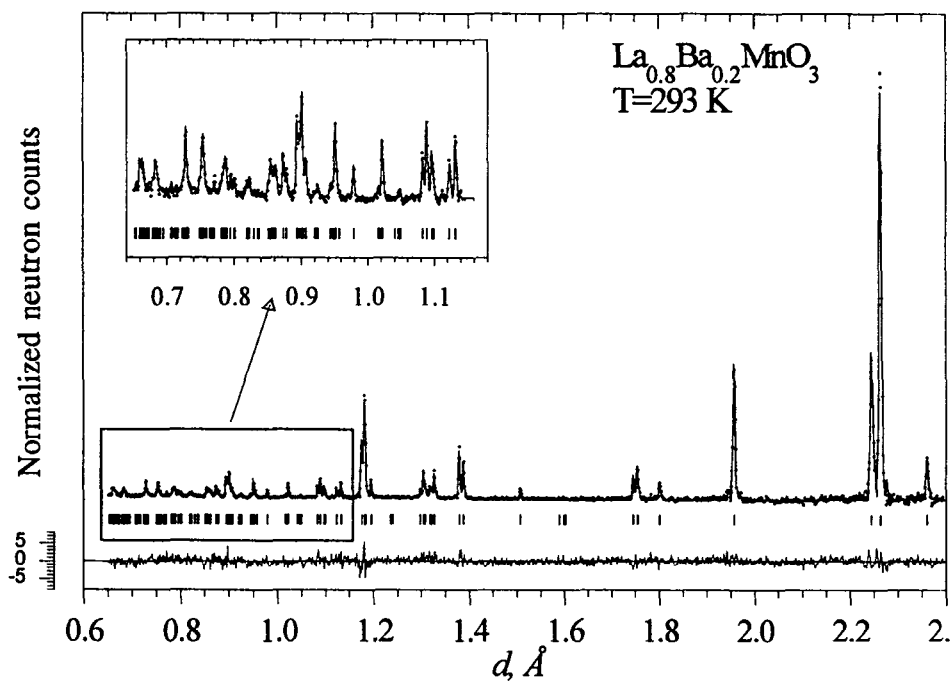


Fig.3. The diffraction pattern of the  $\text{La}(\text{Ba})\text{MnO}_3$  sample measured at  $V_m=8000 \text{ rpm}$  and processed by the Rietveld method. The difference curve is normalized on the mean-square deviation.

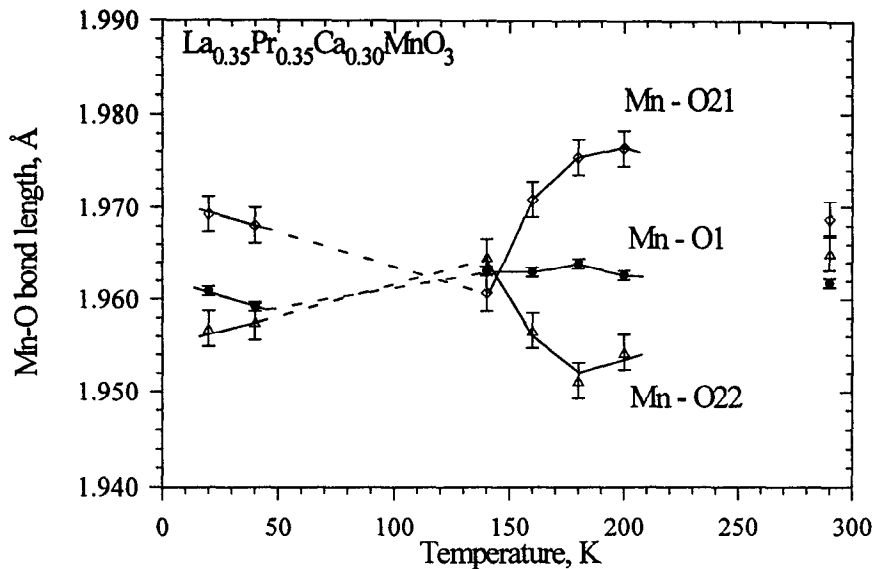


Fig.4. Mn–O bond lengths of  $La_{0.35}Pr_{0.35}Ca_{0.30}MnO_3$  vs temperature.  $T \approx 175$  K is the onset temperature of the semiconductor – metal phase transition. At  $T \approx 140$  K the FM state is fully accomplished.

### 3. DN-12, TOF spectrometer for micro samples studies under high pressure

The first stage of the DN-12 spectrometer for high-pressure studies was put into operation three years ago [10,11]. After the first successful experiments [12] DN-12 developments continued. To increase the neutron flux, improve background conditions, and extend the wavelength range, a curved supermirror neutron guide tube is installed (Fig.5). As a result, the total flux on the sample increases twice and the flux of long wavelength neutrons increases several times (up to 9 for  $\lambda=3.8$  Å) (Fig.6). At present, the DN-12 detector consists of two 800 mm in diameter rings, positioned in the vertical plane. Both rings are assembled of 16  $^3\text{He}$  counters and can be moved along the beam axis providing a scattering angle interval from  $45^\circ$  to  $135^\circ$ . For inelastic neutron scattering investigations a Be filter can be placed in front of one of the rings. The results of the  $\text{NH}_4\text{Cl}$  vibrational spectrum study can be found in Ref. [13].

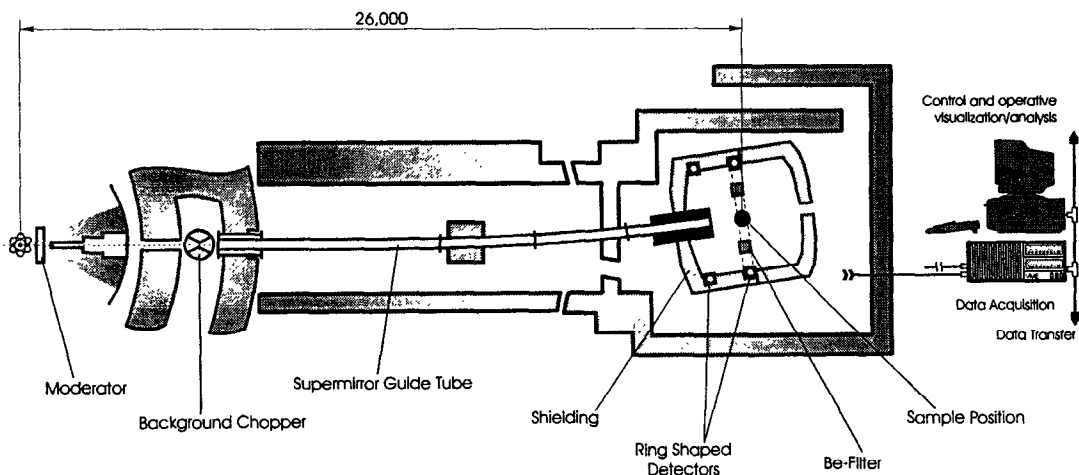


Fig.5. The lay-out of the new design of the DN-12 spectrometer at the IBR-2 reactor.

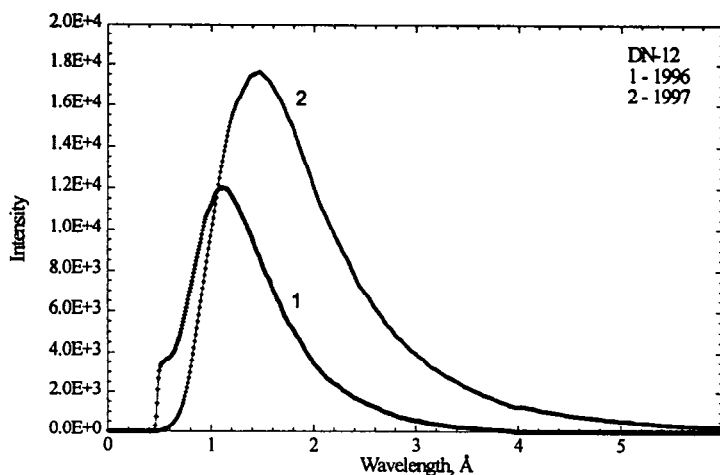


Fig.6. The neutron intensity at the sample position measured before (1) and after (2) installation of a supermirror neutron guide.

Figure 7 shows an example of the  $C_{60}$  diffraction pattern of a  $0.5 \text{ cm}^3$  sample, measured at ambient conditions in 1 h. The hematite  $\text{Fe}_2\text{O}_3$  is studied at pressures up to 3.7 GPa. At  $P=2.5$  GPa in this compound the magnetic phase transition occurs (Fig.8).

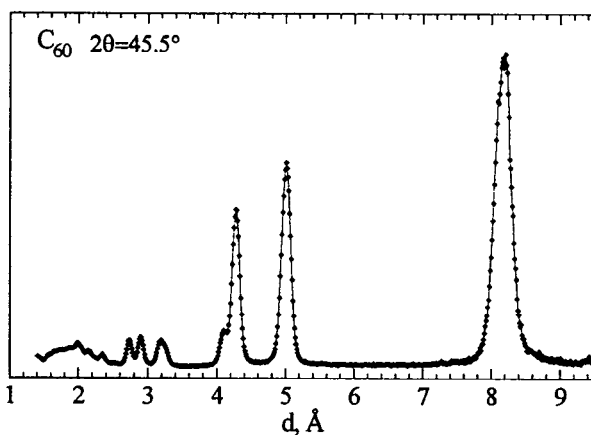


Fig.7. The diffraction pattern of  $C_{60}$  measured from a  $0.5 \text{ cm}^3$  sample at ambient pressure in 1 hour.

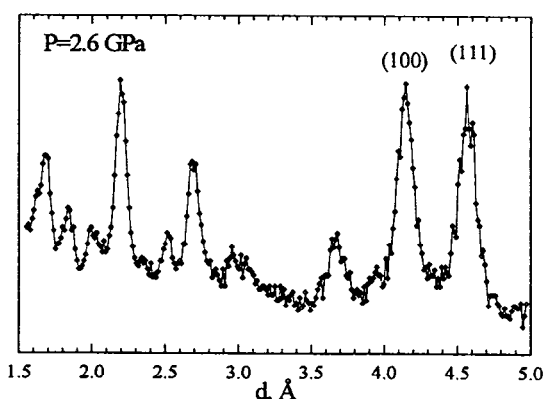
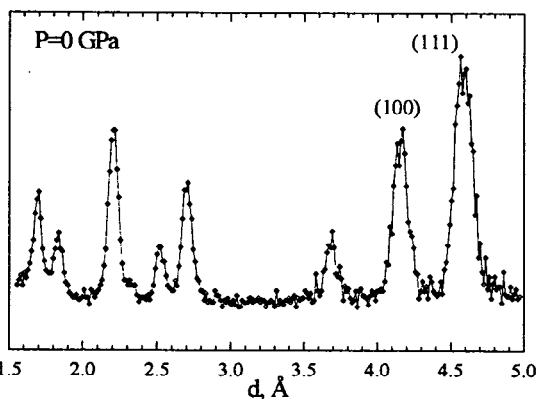


Fig.8. The diffraction patterns of  $\text{Fe}_2\text{O}_3$  measured before (left) and after (right) the magnetic phase transition at  $P=2.5$  GPa. The sample volume is about  $0.5 \text{ mm}^3$ , time of experiment is 24 h.

#### 4. DN-2, TOF diffractometer

DN-2 is a high intensity diffractometer mainly for single crystal but also for real-time studies. Recently, the characteristics of DN-2 were noticeably improved after a two-dimensional detector ( $320 \times 280 \text{ mm}^2$ ) with the position resolution about 2.5 mm in two directions started operation. In addition to time-of-flight analysis, this detector allows registering the three-dimensional intensity distribution in the reciprocal space of the crystal without rotating the sample or the detector. Figure 9 illustrates a two-dimensional intensity distribution in the (220) and (440) reciprocal lattice sites in a  $\text{La}_2\text{CuO}_{4+\delta}$  single crystal following the tetragonal to orthorhombic phase transition and twin structure formation.

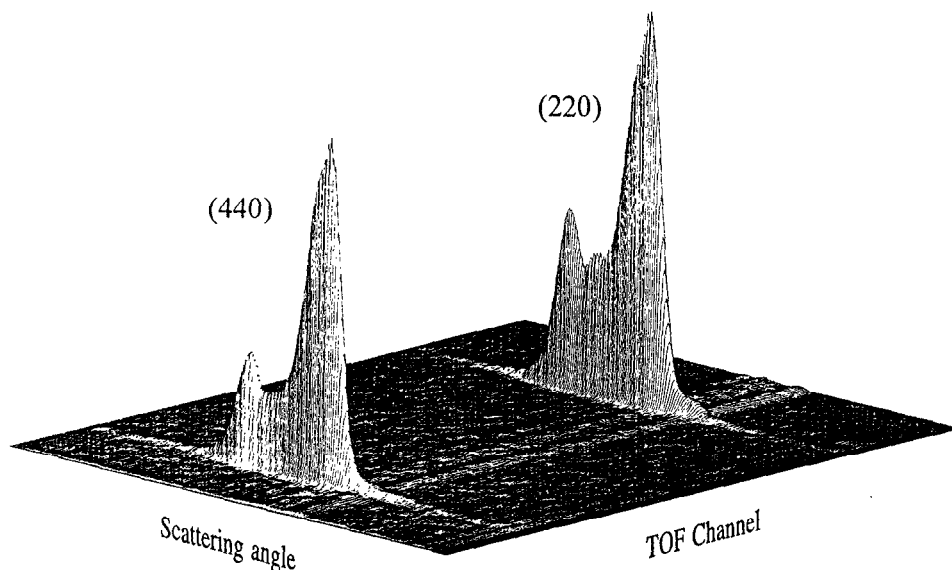


Fig.9. The diffraction pattern along the  $(hh0)$  direction of a  $\text{La}_2\text{CuO}_4$  single crystal (in the ferroelastic phase) measured with a two-dimensional PSD and summed over the vertical coordinate of the detector.

The problem of the coexistence of superconductivity and an ordered magnetic state in two different kinds of these  $\text{La}_2\text{CuO}_{4+\delta}$  single crystals – macroscopically homogeneous and phase separated – have been studied intensively with both HRFD and DN-2 diffractometers [8,14]. To check whether the observed by  $\mu\text{SR}$  technique magnetic transitions in  $\text{La}_2\text{CuO}_{4.02}$  and  $\text{La}_2\text{CuO}_{4.04}$  samples lead to the true long-range AFM order, the neutron diffraction spectra along the  $[100]$  direction were measured on DN-2. According to the  $\mu\text{SR}$  data, one could expect to find the (100) magnetic peak below  $T_N$ . Indeed, in the  $\text{La}_2\text{CuO}_{4.04}$  sample, this peak is well pronounced (Fig.10), whereas in the  $\text{La}_2\text{CuO}_{4.02}$  sample, neutron diffraction reveals no traces of such reflection (insert in Fig.10).

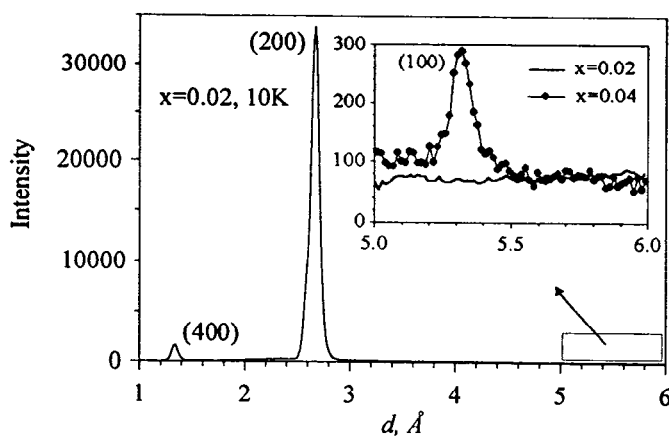


Fig.10. The diffraction pattern from the (h00) plane for  $\text{La}_2\text{CuO}_{4.02}$  measured at  $T=10\text{ K}$ . The insert shows fragments of (h00) diffraction patterns for  $\text{La}_2\text{CuO}_{4.02}$  and  $\text{La}_2\text{CuO}_{4.04}$  near the (100) AFM peak.

In conclusion, it can be confidently forecasted that the joint operation of improved HRFD, DN-12, and DN-2 diffractometers at the IBR-2 pulsed reactor will make a visible scientific impact in powder precision structural, powder high pressure, and single crystal neutron diffraction studies.

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