MULTICRYSTAL INVERTED GEOMETRY SPECTROMETER NERA-PR AT THE IBR-2 PULSED REACTOR

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Abstract

The NERA-PR spectrometer is intended for inelastic (INS) and quasielastic (QNS) neutron scattering research with simultaneous check of sample structure by powder neutron diffraction (ND). The wavelength spectrum of incident neutrons is analyzed by time of fight on the 109 m reactor - sample flight path. Beryllium filters and pyrolitic graphite or Al, Cu and Zn monocrystals in front of the detectors are used as monochromators of the scattered neutrons. This permits wide variation of the spectrometer's resolution. The design and performance details of the spectrometer are reported. The variable configuration of the spectrometer setup allows simultaneous performance of INS, QNS and ND experiments which fact extremely suits the aims of investigation of substances exhibiting phase polymorphism.

1. Introduction

The inverted geometry spectrometers developed for the instrument suit of the JINR pulsed reactors in Dubna are the result of evolution of a simple energy loss inelastic spectrometer with a beryllium filter towards a difference Be-BeO filter [1] and, finally, a combination of a beryllium filter and crystal monochromator in front of the detector [2]. The idea of simultaneous investigation of the dynamics and structure of solids [3] that B. Buras suggested in 1966 was experimentally tested in the next two years [4] at the IBR pulsed reactor. However, wide demand for and high cost of electronics hindered construction of multipurpose spectrometers at that time. The obstacles were removed with appearance of semiconductor-based memories and mini-computers for data collection and instrument control in neutron scattering experiments. The new generation of multi-detectors inverted geometry spectrometers [5-7] made routine the simultaneous investigations of the structure and dynamics of complex molecular crystals [7,8], superionic compounds [9], metal hydrides [10] etc. at the IBR-2 high flux pulsed reactor.

The inverted geometry spectrometer KDSOG-M [6] on the 30 m flight path of the IBR-2 reactor has high luminosity, but limited resolving power of both inelastic and diffraction spectra because of rather large duration of the thermal neutron pulse (about 300 μsec). Considerable resolution improvement, especially for the INS investigations in the region of energy transfers above 20 meV, could be achieved by making essentially longer (by about a factor of 3) the flight path to be traveled by the incident on sample neutrons. The design of the NERA-PR spectrometer [11] exploits the idea. It was realized after mirror vacuum neutron guides have been manufactured and installed to guide thermal neutrons to this long distance without essential loss of the flux intensity. The arrangement of this spectrometer additionally allows investigation of stochastic motions of molecules or their fragments with the help of quasielastic scattering of neutrons. The beam of scattered by sample neutrons is reflected on monocrystal energy analyzers in the direction of the detector, i.e. "nearly backward", and allows one to reach the resolving power of about 40 μeV.

2. Description of the Spectrometer

The schematical lay-out of the spectrometer is shown in Fig.1. The neutron beam having been formed in the active core of the reactor and thermalized when passing through the moderator is guided by the vacuum, mirror neutron guide to a distance of 2.5 m from the sample (1) which is positioned at a distance of 109.05 m from the moderator. The aperture of the nickel mirror neutron guides is 50 mm wide by 160 mm high. On the path of the primary neutron beam at a distance of 5.5 m and 15.5 m from the moderator, respectively, two choppers are installed: the disk which rotation about the horizontal and the drum about the vertical axis is synchronized with reactor pulses. This arrangement allows effective reduction of the background of fast neutrons.

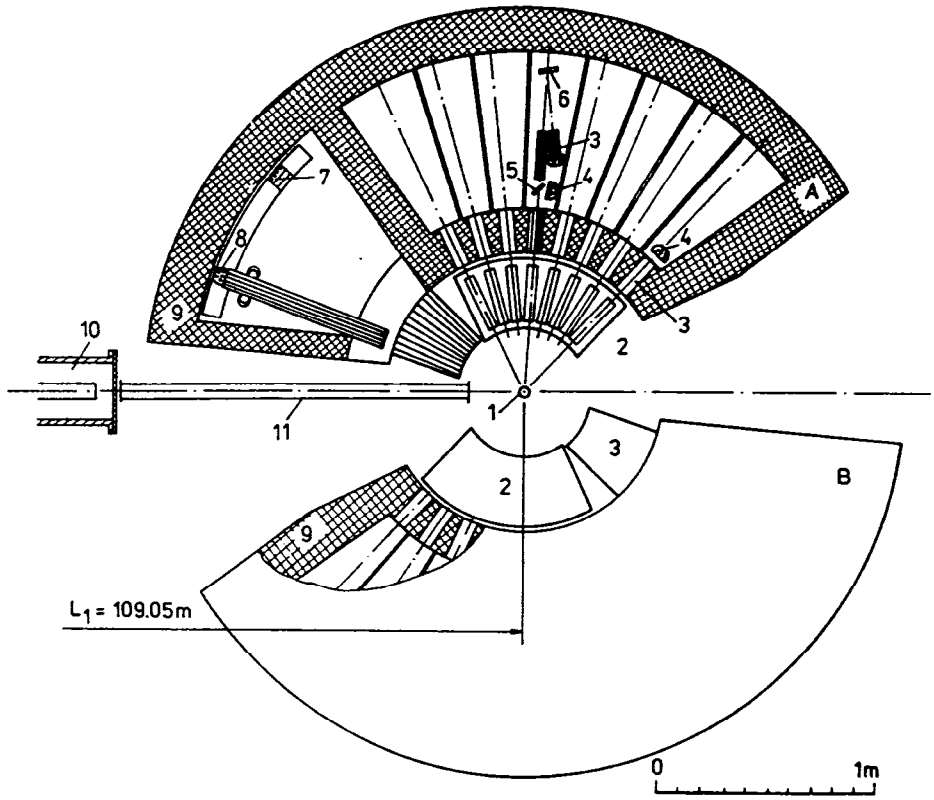


Fig.1. Sketch of the spectrometer NERA-PR:
1 - sample, 2 - Be-filters, 3 - collimators, 4 - ^{3}He detectors (INS and QNS), 5 - pyrolitic graphite analyzer, 6 - single crystal QNS analyzer, 7 - detector for high intensity diffraction, 8 - detector for high resolution diffraction, 9 - spectrometer shielding, 10 - Ni-coated mirror neutron guide in a vacuum tube, 11 - vacuum neutron guide.

The spectrometer has the two shoulders A and B of like built. Each has eight sections for neutron elastic and quasielastic scattering and one for diffraction measurements. Rails allow displacements of both shoulders about the spectrometer's (1) axis and variation of the angle between the neutrons incident and scattered on the sample. The diffraction sections of A and B for the scattering angles $170 \geq 2\theta \geq 110$ and $70 \geq 2\theta \geq 10$, respectively have neutron detectors which permit one to carry out the measurements with either high luminosity (7) or high resolution (8). The detectors (8) are the assemblies of five ^{3}He proportional counters (SNM-17) arranged in a chess order for the detector to have the area equal to the beam cross section (50 mm x 160 mm) and the thickness about 20 mm. Collimation of the scattered neutron beam is accomplished with a 750 mm length soller collimator with a minimum vertical width of slit of 5 mm giving an angular divergence of about 20'. The detectors (7) are the assemblies of four counters operating independently with an

angular shift of 1.2 degrees. In the latter case the collimation of the scattered beam depends on the diameter of a single counter and the distance between the radialy converging plates of the collimator (3) to make 45'.

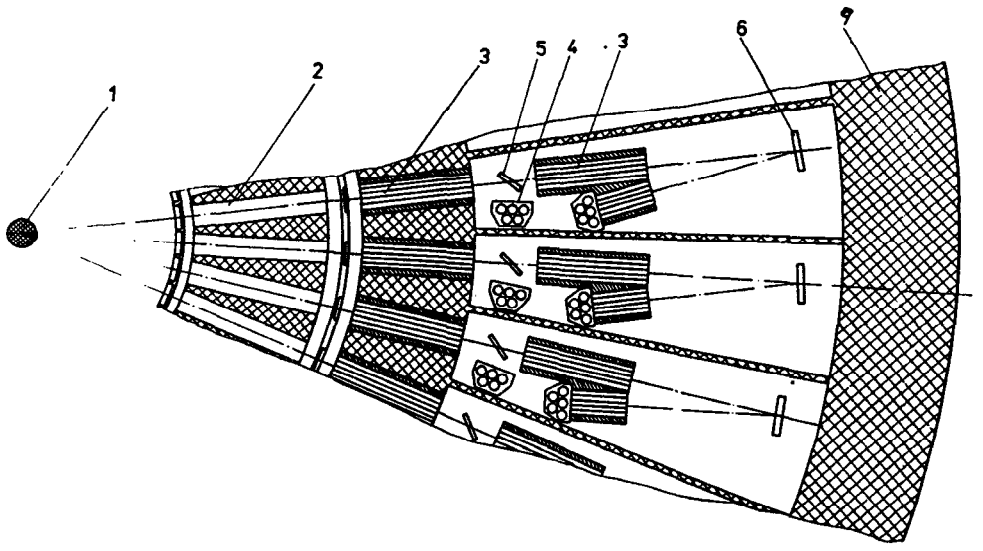


Fig.2. The INS and QNS neutron energy analyzers and detectors of the spectrometer NERA-PR. The numbers indicate the same options as in Fig.1.

The sections for neutron inelastic and quasielastic measurements are detailed in Fig.2. The neutrons scattered by sample (1) pass through a common for eight scattering angles beryllium filter-collimator (2) which is being cooled with liquid nitrogen. It is made of polycrystalline beryllium plates and cadmium sandwiched between every 20 mm. The total thickness of beryllium is 240 mm and the window is 40 mm x 160 mm. Behind the filter the soller collimators (3) with a minimum slit width of 5 mm are positioned which having the length of 200 mm ensure the horizontal collimation of 1.2°. The design of this sections allows analysis of scattered neutron energies to be carried out in three ways:

a) With the help of the beryllium filter, only. The neutron detector (4) having the analogous with (8) design (Fig.1) is installed immediately after the collimator (3). In this case the spectrometer has high luminosity at low energy resolution.

b) With the help of monocrystal analyzers (5) which exploit the Bragg reflection on a given crystallographic plane. The energy for analysis is usually selected in the immediate vicinity of the beryllium edge by rotating the crystal analyzer (5). The filter allows suppression of higher order reflections. Plates of pyrolytic graphite are conventionally used as analyzers, because of their high reflectivity. The analyzers are composed of four plates having the total size of 50 mm x 200 mm.

c) By making use of the Bragg reflection on a monocrystal (6) in the direction "nearly backscattering" ($2\theta = 172°$). This version can be carried out simultaneously with the second one because the positions of elastic lines for the monocrystals of Al(111) or Zn(002) are sufficiently different from those of neutron energies reflected on a pyrolytic graphite at the scattering angle of about 43°. The elastic peak width is determined by both the TOF resolution of the reflected neutron wavelength and the mosaicity of the used monocrystals. For tested Zn(002) and Cu(111) monochromators it reaches the value of 30 and 50 μeV, respectively. This method has rather limited luminosity and, therefore, is mainly applied to investigation of the quasielastic scattering of neutrons.

All spectra of scattered neutrons are measured with two-dimensional analyzers, "number of detector - TOF". The analyser's units (different for diffraction, quasielastic and inelastic scattering) are made in the CAMAC standard. Control of analyzers and data files accumulation are performed by a PC/AT-386 linked to the local ETHERNET network.

3. Luminosity and Resolution

The thermal neutron intensity at the exit of a mirror neutron guide measured with the neutron activation analysis method makes 4.6×10^6 n/cm^{2}sec and it is comparable with the neutron flux 6.6×10^6 n/cm^{2}sec in a 30 m vacuum neutron guide of the spectrometer KDSOG-M [6] in agreement with the estimates made when designing the mirror guide. The geometry of neutron distribution on a sample was investigated using neutron sensitive photo-films. It appears to be very homogeneous and the beam divergence from the mirror guide edge to the spectrometer axis does not exceed 5 mm.

The intensity of scattered neutrons $N(t_0)$ detected at the time t_0 after the start of the time analyser is described with good approximation by the formula [2]:

$$N(t_0) = \Delta t_0 \iiint de\, E_i\, dE_f\, dt\, F(E_i, E_f, t_0, t)\, \sigma(E_i, E_f, \phi, T), \tag{1}$$

where Δt_0 is the time interval corresponding to the width of the time channel of the analyser, $F(E_i, E_f, t_0, t)$ is the apparatus function describing the resolution of the spectrometer, $\sigma(E_i, E_f, \phi, T)$ describes the neutron scattering law, which in its general form depends on the energy, E_i and E_f, of incident and scattered neutrons, on the scattering angle, ϕ, and the temperature of the sample, T.

To calculate the resolution of the spectrometer we used the scattering law in the form $\delta(E_i - E_f - \omega)$ at a given value of energy transfer, ω. The apparatus function has the form:

$$F(E_i, E_f, t_0, t) = \rho(E_i, t)\ \Phi(E_i)\ n(E_f)\ \delta\left(t_0 - t - \frac{\alpha L_1}{\sqrt{E_i}} - \frac{\alpha L_2}{\sqrt{E_f}}\right), \tag{2}$$

where $\rho(E_i, t)$ describes the time distribution of incident neutrons with the energy E_i, $\phi(E_i)$ represents the energy distribution of the neutrons leaving the moderator; $n(E_f)$ is the distribution of neutrons after scattering. The function

$$\delta\left(t_0 - t - \frac{\alpha L_1}{\sqrt{E_i}} - \frac{\alpha L_2}{\sqrt{E_f}}\right),$$

gives the conditions of registration at the moment t_0 of a neutron leaving the source with the energy E_i at the moment t to have after being scattered by the sample the energy E_f (L_1 and L_2 denote the moderator-to-sample and sample-to-detector flight paths, respectively, α is the energy to velocity conversion coefficient).

The time distribution of incident neutrons $\rho(E_i, t)$ is calculated as convolution of an IBR-2 fast neutron pulse shape function (the Gauss function with the width 215 μsec) with the function describing the process of neutron moderation.

$$Z(t, E) \sim \frac{1}{\tau(E_i)} exp\left[-t/\tau(E_i)\right] U_+(t), \tag{3}$$

The value of

$$\tau(E_i) = 210.\left[1 - exp(-E_i/113.2)\right], \tag{4}$$

for the cock-shaped moderator of IBR-2 was taken from [12]. The energy distribution of neutrons leaving the moderator $\phi(E_i)$ was determined in accord with the results of fit to the experimental spectra of elastic scattering by vanadium. The energy distribution of detected neutrons $n(E_f)$ was

obtained with account of the transmission function of the beryllium filter [1] and the gaussian distribution of neutrons reflected on a crystal analyser. The width of this distribution was considered equal to the energy width at half maximum of an elastic peak. In this way the collimations of beams were taken into account, though not entering directly in the formulas (1) and (2).

The resolution functions calculated after these formula for different analysers of scattered neutron energy are shown in Fig.3. It is seen that at energy transfers above 100 meV the resolution analysing crystal are determined by the power pulse width from the IBR-2 reactor.

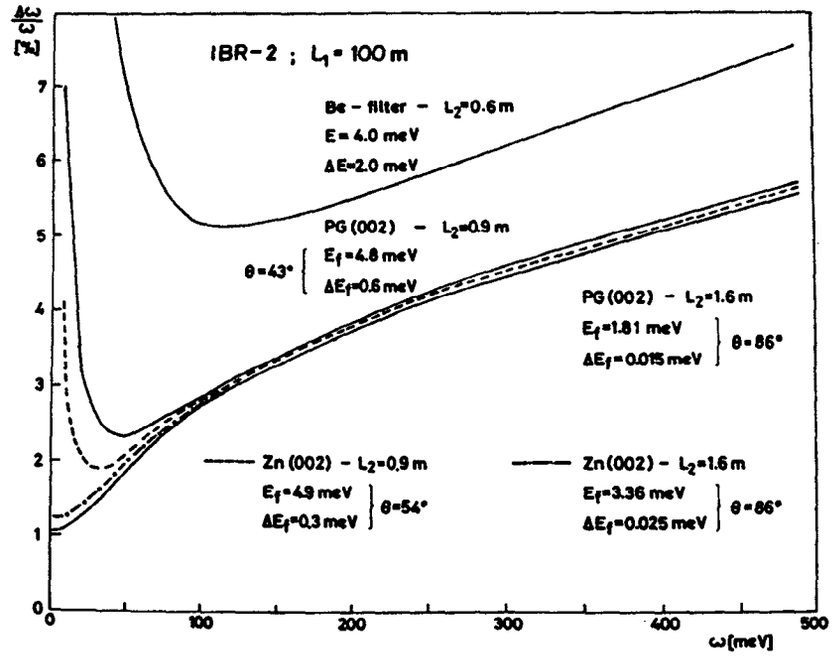


Fig.3. The resolution functions vs energy transfer for different analysers: upper solid curve — for Be filter; middle and lower solid curves — for pyrolytic graphite at different angles; dashed and dot-dashed lines — for zinc analyzer.

Figure 4 compares the INS resolutions of the NERA-PR and KDSOG-M spectrometers with pyrolytic graphite analysers. At energy transfers above 10 meV the resolution of the NERA-PR spectrometer becomes three times better and makes 3-5 % up to the energy transfer of the order of 500 meV.

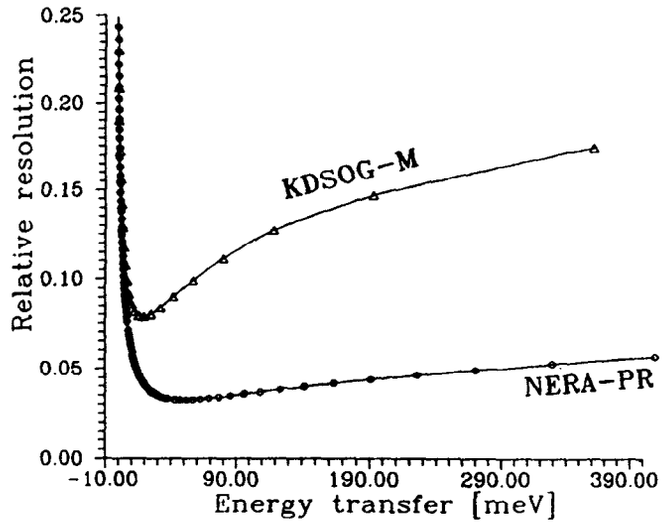


Fig.4. Relative INS resolutions $\Delta\omega/\omega$ of the KDSOG-M and NERA-PR inverted geometry spectrometers with pyrolytic graphite analysers at the IBR-2 pulsed reactor.

Calculated resolution functions are in good agreement with those measured with standard samples for all three types of spectra: inelastic and quasielastic scattering and diffraction of neutrons.

Figure 5 illustrates the spectrum of NH_4Cl measured on the KDSOG-M spectrometer with graphite analysers in comparison with that measured on the NERA-PR spectrometer with a beryllium filter. This experiment confirms the prediction that under these conditions both spectrometers are comparable in resolution and luminosity.

Figure 6 shows comparative high resolution spectra of the hydrazine sulfate sample measured on the NERA-PR spectrometer at IBR-2 and the TXFA [13] spectrometer at the neutron spallation source ISIS. In the region of energy transfer below 100 meV both spectrometers have practically coinciding resolution and luminosity. At higher energy transfers the TXFA luminosity is higher thanks to optimization of the moderator for epithermal neutrons.

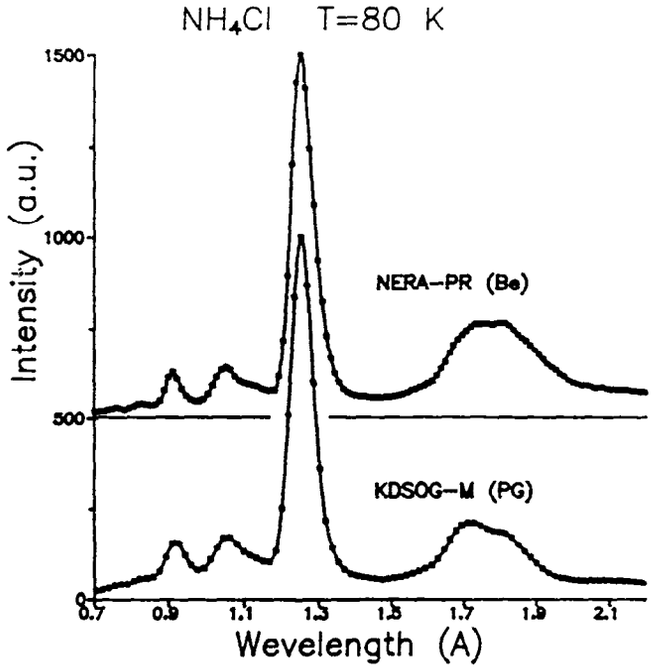


Fig.5.The INS spectra of ammonium chloride at 80 K measured on NERA-PR with beryllium filter analysers and on KDSOG-M with pyrolytic graphite analysers.

Figure 7 shows the neutron quasielastic scattering spectra of an ammonium chloride sample measured with a Cu(111) analyzer. At nitrogen temperature NH_4Cl is in the ordered phase and the elastic peak at the energy E_0 = 4.69 meV corresponds to the resolution of the spectrometer and its full width at half maximum is 48 μeV. One can see that this resolution is sufficient for observing diffusion jumps of ammonium groups in a disordered phase at room temperature. Investigation of QNS intensities in dependence on momentum transfer allows one to study the geometry of diffusion jumps and their frequency determines the broadening of the QNS peak. The spectrometer NERA-PR permits study of QNS in the range of scattering angles from 20° to 160° with the energy resolution of about 1% in characteristic energies of the first order reflections on a monocrystal used as analyser.

The ND spectra simultaneously measured at scattering angles ranging from 20° to 70° and from 110° to 170° allow one to study the phase state of an investigated sample together with the behaviour of structural parameters in dependence on temperature, in particular. Figure 8 illustrates the diffraction spectrum of a Ni powder measured with a high resolution detector at the scattering angle of 160°.

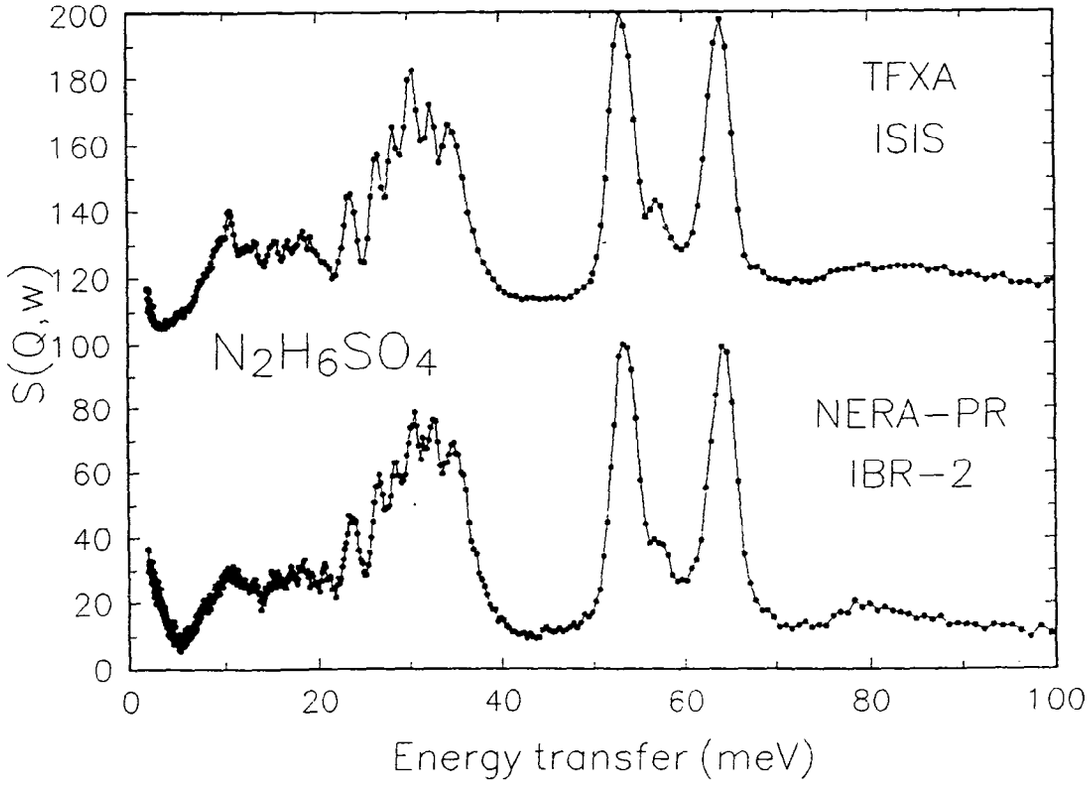
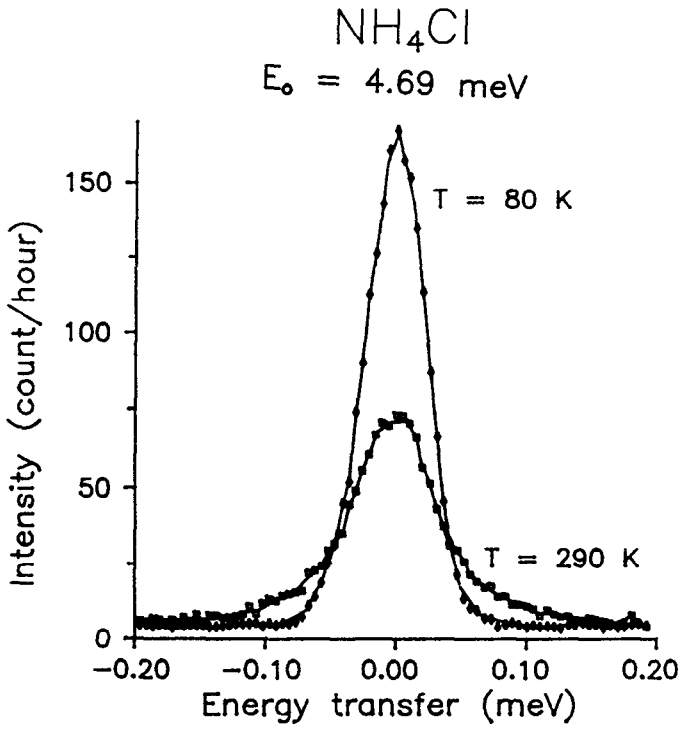


Fig.6. The high resolution INS spectra of hydrazine sulfate measured on the TFXA (ISIS) and NERA-PF (IBR-2) spectrometers with pyrolytic graphite analysers.



Under these conditions the resolution of the diffractogram is $\Delta d/d$ <0.5% for d_{hkl} <1Å. With such resolution of the NERA-PR spectrometer one can measure the interplane distances up to 4Å. By measuring ND spectra in the angular range from 20° to 70° one can study the interplane distances up to 15Å with the resolution of the order of 1 to 3%.

Fig.7. The QNS spectra of ammonium chloride measured with a Cu(111) analyser at the scattering angle 80°.

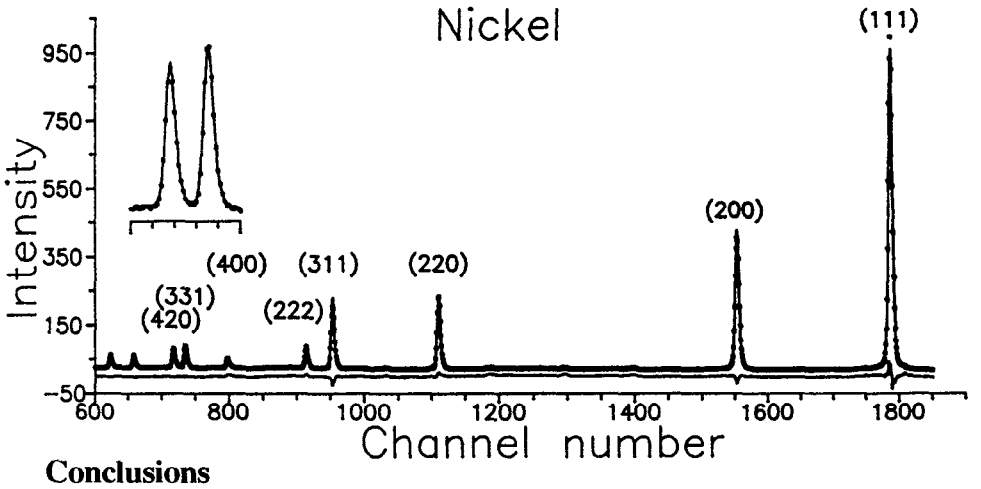


Fig.8.The Ni powder diffraction spectrum normalized to the incoming neutron spectrum measured at 160° scattering angle by a high resolution detector (8). Solid line represents a difference between the experimental and calculated by Rietveld method [14] spectra.

4. Conclusions

The NERA-PR design is optimized for operation at the high flux pulsed reactor IBR-2 producing neutron pulses of ca. 215 μ sec at a frequency of 5Hz. Of the period of 200 msec between the neutron pulses 95% of the time is allocated for neutron spectra analysis and about 5% i.e. about 10 μsec, for automatic control of the experimental conditions. Incoming neutrons in the wavelength range from 0.5 to 7 Å are effectively analysed.

The reported results of the test measurements of the physical parameters of the NERA-PR spectrometer show that this universal facility for measuring simultaneously INS, QNS and ND spectra of scattered neutrons belongs to the class of neutron spectrometers of relatively high resolution at good luminosity. These results confirm the calculated parameters laid in the basis of the spectrometer's design. Their comparison with the results obtained for other spectrometers show that all necessary experimental parameter are chosen correctly and the instrument can be transferred to the user's regime of operation. For instrument details see the updated edition of the FLNP User's guide [15].

Last year's experience of operation of the NERA-PR spectrometer has demonstrated usefulness of simultaneous investigation with the INS, QNS and ND methods of the dynamics and structure of

solids. It is especially suitable for investigation of substances, where phase transitions are followed by changes in the structure, lattice dynamics or stochastic motion behaviour of molecules or molecular groups.

Acknowledgements

The authors are sincerely grateful to Prof. J A Janik and the late Prof. Yu M Ostanevich for continuous interest and support in realization of this project; to Dr J Sciesinski and Dr S Urban for helpful discussions and comments in the time of preparation of the project; to eng. J Dec and the late eng. J Pawelczyk for the assistance in preparation of design documentation and manufacture of spectrometer parts; to technicians: R Pukala, H Zdziarski and the late T Pukala for the assistance in adjustment of the mirror neutron guides and assembly of the spectrometer; to engs. B Motyka, A P Sirotin, J Sokolowski and A I Zhuravlev for the assistance in manufacture and adjustment of detecting tracks and electronic systems for data storage and control of experimental conditions; to engs. W Iwanski and M L Tchelnokov for development and upgrading of the program for control of spectrometer operations; to Drs A V Belushkin, J Krawczyk. A Pawlukojc and W Zajac for the assistance in development of data procession programs and interest in the utmost use of this instrument's advantages in a physical experiment.

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