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Neutron diffraction on pulsed sources

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Abstract. The current capabilities of and major scientific problems solved by time-of-flight neutron diffraction are reviewed. The reasons for the rapid development of the method over the last two decades have been mainly the emergence of third-generation pulsed sources with a megawatt time-averaged power and advances in neutron optical devices and detector systems. The paper discusses some historical aspects of time-of-flight neutron diffraction and examines the contribution to this method from F L Shapiro, the centennial of whose birth was celebrated in 2015. The state of the art with respect to neutron sources for studies on extracted beams is reviewed in a special section.

Keywords: neutron diffraction, advanced neutron sources, pulsed neutron sources, time-of-flight technique, crystal structure

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Dedicated to the memory of F L Shapiro

1. Introduction

About 50 years ago, in 1963–1964, the first full-fledged diffraction experiments based on the time-of-flight method were performed at the Laboratory of Neutron Physics (LNP) of the Joint Institute for Nuclear Research (JINR) in Dubna, which served to catalyze the subsequent rapid development of research in this field. Soon after the experiments were carried out in Dubna, the time-of-flight method was applied simultaneously at several neutron centers around the world utilizing targets at electron accelerators, and it became clear that time-of-flight diffractometers at pulsed neutron sources are quite capable of competing with conventional installations at steady neutron flux reactors (steady state reactors). The main advantage of time-of-flight diffractometers involved the possibility of employing nearly the entire ('white') neutron spectrum from the source, whereas with conventional diffractometers with a monochromatic beam the fraction of 'useful' neutrons did not exceed 1%. As a result, it turned out to be possible to obtain quite interesting results even with the first relatively low-power pulsed sources (their power averaged over time was at a level of several kW) in Dubna, Harwell (the United Kingdom), and Tohoku (Japan).

As early as in the 1970s, work started on the construction of a new generation of pulsed sources. Two main lines of inquiry emerged. One was based on proton accelerators with heavy metal targets, which in the English-language literature were conventionally termed spallation neutron sources (SNSs), or proton neutron sources exhibiting a power up to 200 kW. Such sources were commissioned in the USA, the United Kingdom, and Japan. By the end of the 20th century, projects appeared which were conceived for pulsed sources of

Figure 1. Fedor L'vovich Shapiro (1915–1973) in his study at the JINR Laboratory of Neutron Physics. During the period of 1959–1973, F L Shapiro was deputy director of this Lab.

the subsequent, actually the third, generation with an average thermal power exceeding 1 MW. The first two of them started working relatively not so long ago in the USA and Japan, and research carried out with them plays a more and more significant role in the whole stream of neutron scattering experiments. The other line, essentially an original one, was concerned with successful operation of the world's first pulsed reactor IBR (Russian abbreviation for pulsed fast reactor) in Dubna [1]. Since the first such reactor was commissioned in 1960, reactors of this series have undergone much improvement and have played a significant part in the development of neutron studies. Suffice it to say that formulation of the concept of the new European Spallation Source (ESS), presently under construction in Sweden, was influenced by the IBR-2 reactor operating as a long-pulse source.

Twenty years ago, we have already considered neutron structural research carried out at pulsed sources until the middle of the 1990s [2]. Since then, the situation has undergone significant changes. First, the composition and quality of neutron sources has changed. Second, general technical progress, as well as new ideas in the construction of diffractometers, in the formation of neutron beams, and in the development of detector systems, have made it possible to achieve such capabilities in neutron diffractometry that two decades ago seemed incredible. Therefore, writing a new review became a necessity.

One more circumstance compelled us to deal with this issue once again: in the literature, notes and views are regularly published that are not quite correct in what concerns research development of diffraction at pulsed neutron sources with the aid of the time-of-flight method. As a rule, the opinion presented is the one formed on the basis of one or two publications that appeared at the beginning of the 1960s. Both the quite long prehistory and the relative importance of some event or another are totally ignored. We have endeavored to provide a more accurate analysis of the situation that preceded the first and subsequent diffraction experiments.

And, finally, 2015 was the 100th anniversary of the birth of Fedor L'vovich Shapiro, a physicist and teacher who influenced the destinies of very many people and founded the scientific school of neutron physics at pulsed sources in Dubna. During the period from 1959 up to his death in 1973, F L Shapiro was a deputy to I M Frank, director of the JINR Laboratory of Neutron Physics, and he significantly influenced the formation of scientific research plans, including those for the field discussed herein. We dedicate this review to the memory of F L Shapiro (Fig. 1).

In the present review, we consider the main scientific problems that are resolved with the aid of advanced time-of-flight diffractometers. For completeness of our presentation, a special section is devoted to the present-day state of affairs with neutron sources for research with extracted beams.

2. General situation concerning neutron diffraction by crystals

By the time J Chadwick discovered the neutron in 1932, X-ray structural analysis had already been a quite developed experimental method with the aid of which it became possible to analyze the structure of matter at the atomic level. By that time, owing to quantum mechanics, the wave properties of particles were no longer a puzzle; thus, for instance, successful experiments were performed in 1927 on the diffraction of electrons. However, the possibility of neutron diffraction was proposed and realized only four years after the discovery of the neutron (see original articles in the Jubilee collection [3]). The first real neutron diffraction experiments were performed about 10 years later [4], after the construction of nuclear reactors, i.e., the time delay relative to X-ray studies amounted to more than 30 years. Nevertheless, owing to the general principles of X-ray and neutron diffraction by crystals being practically identical, it turned out to be possible to rapidly adapt all the experience accumulated in X-ray structural analysis. Already by the end of the 1940s, Clifford G Shull had obtained outstanding results that were awarded a Nobel Prize in Physics 1994. Particularly, isotopic effects were observed [5] for the first time in neutron diffraction, and the hypothesized existence of antiferromagnetic ordering in crystals was confirmed [6]. These achievements and the construction of powerful nuclear reactors in the 1960s contributed to neutron diffraction becoming one of the most efficient methods for studying the atomic and magnetic structures of crystals.

The efficiency of structural neutron diffraction is primarily related to the differences between interactions with the matter of neutrons, on the one hand, and of X-rays and synchrotron radiation, on the other. A neutron penetrating into a medium undergoes interactions of several types, although the main ones are strong (nuclear) and electromagnetic. The first manifests itself when the neutron interacts with the atomic nucleus; magnetic dipole interaction arises between the magnetic moment of the neutron and the magnetic moment of the atom, which represents the sum of

the spin and orbital moments of the electrons. Low-energy neutron scattering by nearly all nuclei can be characterized by certain constants called scattering lengths that exhibit a specific peculiarity: their variation from element to element and from isotope to isotope is irregular. As a result, the scattering lengths for elements occupying different parts of the Periodic Table may happen to be comparable, and, contrariwise, neighboring elements or isotopes may have essentially different scattering lengths. X-rays do not distinguish between isotopes, and their scattering from elements only depends on the electron density. These differences determine those areas of structural studies on condensed matter in which the application of neutron diffraction is most efficient. Traditionally, these include structural analysis of compounds consisting of light and heavy atoms (hydrides, oxides), compounds of elements with close atomic numbers (alloys, intermetallides), biological compounds with the utilization of isotopic contrasting (mainly with deuterium substituted for hydrogen) of individual fragments of the structure, and the analysis of the magnetic structure of crystals, i.e., determination of the values and orientations of the atomic magnetic moments. In all the aforementioned cases, X-rays do not really 'see' any essential details.

One more important peculiarity of thermal neutrons consists in their linear absorption coefficients being in most substances several thousandths those of X-rays. Respectively, the high neutron penetrability permits the use of relatively complicated constructions in order to realize a certain external influence on substances, for example, in experiments with model electrochemical cells in studies of charge–discharge processes.

Presently, about 30 scientific centers exist in the world based on modern research neutron sources which are equipped with diverse neutron spectrometers. The latter necessarily include several dedicated diffractometers. As a rule, work in neutron centers proceeds within the framework of user programs, which implies submitting experiment applications, their estimation by experts, and the allocation of necessary time for applications having received high marks. An analysis of the state of and prospects for the development of neutron sources in the world is presented in Section 8.

3. Two types of neutron diffractometers: λ_0 and time-of-flight ones

Any stationary or pulsed research source produces a neutron flux with a close-to-Maxwellian neutron energy distribution. In the diffraction experiment with a crystal, the positions of peaks in reciprocal space are fixed — they only depend on the crystalline lattice, and for their registration it is necessary to choose the wavelength λ and scattering angle 2θ correctly, namely, so as to satisfy the Bragg–Wolf condition $2d\sin\theta = \lambda$, where d is the interplane distance. Respectively, there are two possible types of scanning the reciprocal space of a crystal: over the scattering angle at a fixed wavelength or over the wavelength for a fixed scattering angle, which results in the possibility of realizing two types of neutron diffractometers (Fig. 2).

Diffractometers traditionally used in steady state reactors involve a monochromatic neutron beam; these diffractometers actually represent enlarged-scale copies of X-ray devices, which is directly related to the large dimension of the reactor core. We shall further call them λ_0-diffractometers. At pulsed sources it is natural to make use of the entire Maxwellian ('white') neutron spectrum and of the time-of-flight method to determine their energy (wavelength). The relatively low velocity of thermal neutrons readily permits this process to be organized. Accordingly, devices at pulsed sources were termed TOF (Time-of-Flight) diffractometers. So-called energy-dispersive spectrometers, in which the continuous radiation spectrum is used, while the energy is analyzed by an energy-sensitive detector, serve as formal analogs of TOF diffractometers in the case of X-ray or synchrotron radiation.

The construction of TOF diffractometers turned out to be a significant event in the development of neutron diffractometry. Besides the aforementioned substantially more effective utilization of the neutrons produced than in the case of steady state reactors, TOF diffractometers provide several important methodical possibilities. First, the possibility of performing fixed-geometry experiments must be noted. Thus, for instance, this immediately permitted noticeably enhancing the range of achievable pressures in experiments with high external pressures [7]. Second, the TOF method readily permits the implementation of parallel three-dimensional (3D) scanning of the reciprocal space of a crystal, making use of a two-coordinate position-sensitive detector (3D PSD), while the neutron time of flight serves here as the third coordinate. It is important that the scanning can be extremely detailed (actually continuous), which renders TOF diffractometers irreplaceable for the analysis of diffuse scattering in the case of defective structures and incommensurably modified atomic or magnetic structures. Finally, the pulsed character of irradiation of the sample with neutrons from the source also provides the possibility of making the external influence on the sample pulsed. In this case, the influence can be made many times larger, which was already demonstrated back at the end of the 1960s in experiments with pulsed magnetic fields [8].

It should be stressed that in comparing the methods it is necessary to take many factors into account. Each one of the methods has its own advantages and drawbacks, and only a

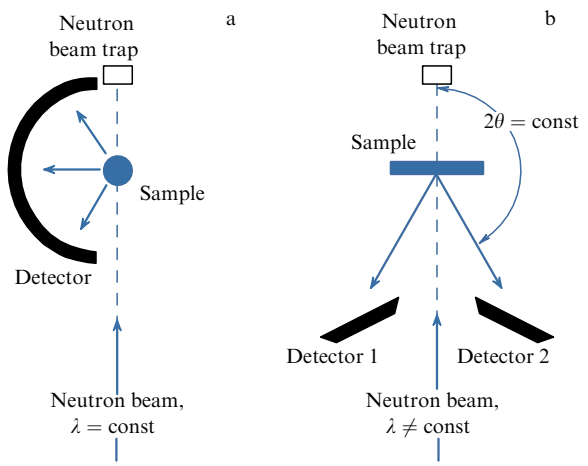

Figure 2. (a) Layout of a λ_0-diffractometer. A monochromatic neutron beam is utilized; scattered neutrons are registered by a single large position-sensitive detector with a scattering angle sweep. A cylindrical geometry of the sample is preferable. (b) Layout of a TOF diffractometer. A pulsed neutron beam with a continuous spectrum is used, as well as several individual detectors placed at fixed scattering angles. The diffraction pattern is analyzed by scanning the neutron wavelength; the geometry of the sample can be arbitrary.

comparison of the results of experiments can permit us to come to a conclusion concerning the preferability of one method over another. Further still, even a comparison of the rate with which diffraction data are obtained and of their accuracy in the case of one and the same sample, when the structure of a polycrystal is undergoing refinement in a standard manner, only makes it possible to draw qualitative conclusions, since, owing to the differences in the observed ranges of interplane distances d_{hkl} and in the behavior of the resolution function $R(d)$, the situations for the position and thermal parameters of atoms turn out to be different. Practice reveals that the quality of structural information obtained with TOF and λ_0-diffractometers of the same class, i.e., with approximately the same resolution and total flux falling on the sample, also turns out to be nearly the same, while certain details may differ quite significantly. This is related to several reasons: distinction in ranges of accessible interplane distances, in the influence of the microstructures of samples, in the dependences of microstructural parameters upon the momentum transfer, in the descriptions of the shapes of diffraction maxima, and so on. Therefore, the most complete and adequate structural information for composite objects can be obtained using diffractometers of both types in a sophisticated manner.

Notice that the time-of-flight method can also be realized in a neutron source with a continuous flux by periodically interrupting the neutron flux. Moreover, the method itself was established precisely in this way, while the very pulsed sources were then constructed for implementation of the method. Therefore, from the point of view of diffractometry, the significance of the term 'pulsed source' is more general than that of a 'type of device generating neutrons'.

4. Certain historical aspects of the origination of time-of-flight diffractometers

Historically, no unexpected issues concerning the origination of the neutron λ_0-diffractometer exist, so, accordingly, the priority of its construction is not disputed. On the contrary, the situation with the TOF diffractometer still remains somewhat unclear. For example, a relatively recent article [9] contains the assertion that the time-of-flight method was proposed by the Polish physicists B Buras and J Leciejewicz: "The Time-of-Flight (TOF) method for neutron diffraction was proposed half a century ago by Buras and Leciejewicz as a very efficient alternative to the crystal monochromator technique" [9]. It is not difficult, however, to verify that this assertion is, at least, not quite correct.

The actual TOF method in neutron spectroscopy has been known since the middle of the 1930s, when the first mechanical neutron flux chopper was constructed [10] for various purposes, including the monochromatization of neutron beams [11]. At the beginning of the 1950s, the idea of using the TOF method with a pulsed source for registering diffraction was put forward by P A Egelstaff, in particular, at the *Third Congress of the International Union of Crystallography* in 1954 [12]. Quite definite evidence concerning this issue may be found in the article by R D Lowde [13]. Moreover, not only is the idea presented in Ref. [13], but quite a complete theoretical justification is given for applying diffraction with the time-of-flight method, including formulae for the intensity of diffraction peaks. The next important event in the development of the TOF method in diffractometry turned out to be the long talk presented by P A Egelstaff in 1961 to the Saclay symposium [14], in which he discussed a concrete TOF diffractometer layout. The Polish physicist B Buras attended this symposium, and, apparently, the talk by P A Egelstaff subsequently encouraged him to attempt practical implementation of the ideas put forward in the talk.

Below follow several quotations, from which it is evident that at the beginning of the 1960s TOF diffraction was no longer something mysterious, and there was actually not much left to do — it was just necessary to arrange a concrete experiment. From the article by R D Lowde [13]: "A pulsed source of slow neutrons in conjunction with time-of-flight apparatus is ideally suited to such a technique [TOF diffraction]. Egelstaff has repeatedly emphasized that electron accelerators are capable of providing mean fluxes of thermal neutrons for these purposes equal or superior to the fluxes of modern reactors". From the article by P A Egelstaff [14]: "In the case of neutrons, one can consider the time-of-flight technique as an alternative to crystal diffraction. The pulsed neutron source may be an accelerator source or a chopper based on a reactor". From the article by B Buras [15]: "In the case of neutrons, one can consider the time-of-flight technique as an alternative to the above-mentioned conventional method. This has been pointed out independently by P A Egelstaff and in the course of a discussion on the program for the fast pulsed reactor at JINR in Dubna". From the article by I M Frank [16]: "Already from the very beginning, during the discussion of work planned for the IBR, it was obvious that neutron monochromatization was not necessary in the case of diffraction at a given angle of reflection, since different neutrons satisfying the Braggs condition differ in velocities and are separated by their time of flight."

Here, one must pay tribute to B Buras — precisely he initiated and carried out the first real TOF-diffraction experiments. How this was done is described in his detailed reminiscences published in the collection [3]. The very first experiments were conducted at the two-megawatt steady state reactor EWA (Experimental Water Atomic) of the National Center for Nuclear Research, Poland (Swierk, not far from Warsaw). A pulsed neutron beam (pulse duration ≈ 80 μs) was formed by a chopper, the total chopper–sample–detector flight path was about 5 m, and measurement of the first neutron diffraction patterns took up to 40 h for an Al sample of about 200 g in mass. After an increase in the solid angle of the detector and passage to BF_3 counters, the data acquisition time was successfully reduced to 4 h. The resolution also turned out to be not so good ($\Delta d/d \approx 0.025$), but the main goal was achieved — the method was experimentally shown to be capable of working. The results of these studies were published in Ref. [17].

However, the first real experiments based on the time-of-flight method, in which quantitative data were obtained on the structures of a series of powders, and the main relationships were checked concerning the relation between the intensity of diffraction peaks and their structural factors, were performed in Dubna [18] in 1963 and 1964 at the world's first pulsed reactor on extracted neutron beams.

Diffraction experiments started after B Buras arrived with his colleagues I Sosnowska and J Sosnovski in Dubna in 1963. A diffractometer was assembled with a total base of about 16 m, spectra were registered by a scintillation counter based on a $ZnS(Ag) + {}^{10}B_2O_3$ mixture 300 cm^2 in area, and the data acquisition time amounted to about 11 h. The results of these and some subsequent experiments are given in Ref. [18] and in review [15].

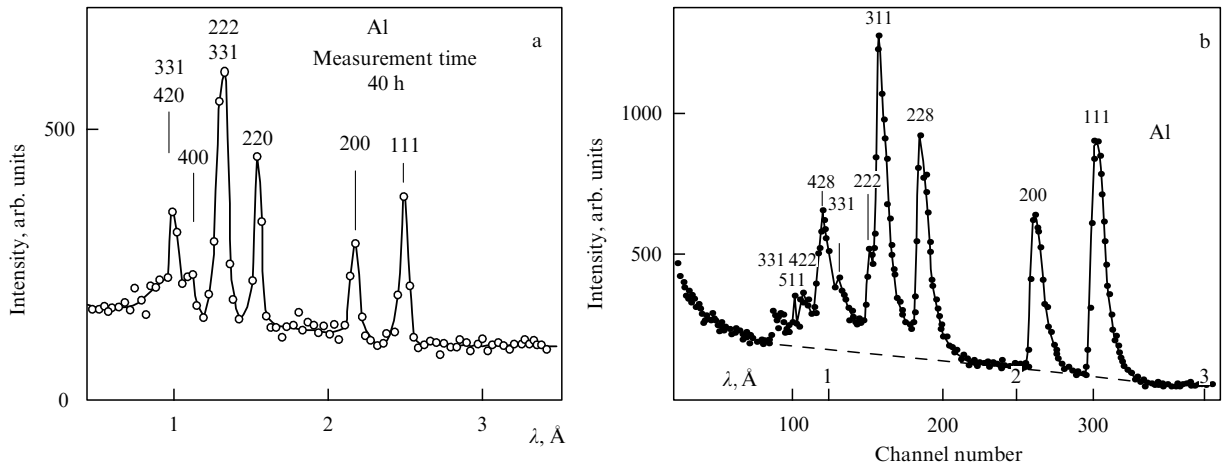

Figure 3. First neutron diffraction patterns of Al powder measured with TOF diffractometers. (a) Neutron diffraction pattern obtained at the steady state EWA reactor (measurement time of 40 h). The flight base was 4.76 m. (b) Neutron diffraction pattern obtained at the pulsed IBR-1 reactor (measurement time of 11 h). The flight path was 15.70 m. The wavelength dependence is plotted for the intensity of neutrons registered by the detector, and the Miller indices of diffraction peaks are shown. (Taken from Ref. [15].)

The first neutron diffraction patterns obtained in Swierk and Dubna are displayed in Fig. 3. The quality of the spectrum measured at IBR-1 is seen to be significantly better, but it was clear that the parameters achieved still did not permit performing systematic structural experiments. Suffice it to say that, although measurements were carried out with the most simple compounds exhibiting a maximum possible number of structural factors, the data acquisition time amounted to many hours, in spite of the mass of the sample being several hundred grams. But it became evident, at the same time, that this was only the beginning and that the prospects of the method were extremely impressive, including enhancement of the intensity and improvement of the resolution.

Thus, the key events in the development of the time-of-flight method in diffraction experiments were the presentations by P A Egelstaff [12, 14], the article by R D Lowde [13], experiments in Swierk and Dubna, and review talk [15] presented at Geneva conference in 1964. Only several years later, in 1968–1969, did communications appear concerning diffraction time-of-flight experiments in Harwell (United Kingdom) [7, 19], Troy (USA) [20], and Tohoku (Japan) [21].[1]

5. Work on neutron diffraction at the first pulsed reactor in Dubna

At the stage of planning construction of the first pulsed reactor, IBR-1, the latter was considered for use in resolving nuclear physics problems. But already at the construction stage, discussions also started concerning condensed matter physics (CMP). J Janik (Institute of Nuclear Physics, Polish Academy of Sciences, Krakow) recalls [22] that the first detailed discussions of experiments at the IBR took place back in 1958 in Dubna during a workshop of scientists from Member States participating in JINR. Subsequently, the initiative of carrying out studies on CMP in Dubna was mainly due to Polish physicists, who played a key role in the development of this line of inquiry at the reactors of the Laboratory of Neutron Physics.

During the first several years after the pulsed IBR-1 reactor was commissioned, all scientific work was actually performed under the leadership or F L Shapiro, and often with his personal participation. In the field of condensed matter physics, F L Shapiro was mainly interested in inelastic neutron scattering experiments. Thus, one of the most significant methodical achievements by F L Shapiro and his colleagues was the development of two principal methods for analyzing inelastic scattering, namely, the so-called direct and inverse geometry methods. In the former, energy analysis proceeds between the neutron source and the sample, while in the latter it is done between the sample and the detector. Experiments leaning upon these methods were actually the first scientific experiments on CMP carried out at a pulsed neutron source. A description of the methods and the results for inelastic neutron scattering in graphite, lithium fluoride (LiF), and vanadium were presented in 1961 at a workshop in Dubna [23]. A detailed analysis of the first work at IBR-1 with the recourse to inelastic neutron scattering can be found in the review by V V Golikov [24].

Of the publications by F L Shapiro concerning neutron diffraction, the most significant are review [15] and an original paper (by the way, without coauthors) [25]. In the first article, a detailed description is given to diffraction experiments performed during the preceding two years at the reactors in Swierke and Dubna, formulae are presented for the integral intensity of diffraction peaks and for the resolution of a TOF diffractometer, which are necessary for planning an experiment and for processing data obtained, and a comparison is made of measured and calculated structural factors for a series of simple substances (Al, Pb, Zn, ZnO, Si). In the concluding part of review [15], a detailed discussion is presented about the features of possible concrete implementations of the experiment and of tasks for the future steps. In considering the latter, attention is focused on the idea of a pulsed influence on the sample, which is synchronized with pulses extracted from the source.

[1] When our manuscript was already submitted for publication, we learned about the publication by I M Sosnowska, "The birth of time-of-flight (TOF) neutron powder diffraction at pulsed neutron source" [71], in which many interesting details are presented concerning the initial stage of development of the time-of-flight method at pulsed neutron sources. (*Note added in proofreading — Editor.*)

Original paper [25] describes a possible method for determining the structural factor phases in crystals with atoms having magnetic moments by varying the magnetization vector orientation. This idea is based on the possibility of controling the value of structural factors, i.e., it is close to the idea underlying the methods of anomalous scattering and of isomorphous substitution, which at present are extensively applied in the synchrotron radiation diffraction. The method is very difficult to implement, and has practically never been used, but the way it is presented reveals a profound understanding of the essence of the problem. It must be noted that in those years precisely the 'phase problem' was considered one of the most complicated problems in the structural analysis of crystals. It received enhanced attention, and to propose a new, original method for its resolving was anything but easy. With time, it turned out that the employment of advanced sources of X-ray and synchrotron radiation and of statistical methods for analyzing a large number of measured intensities permits us to obtain information on the phases of structural factors in a practically automatic mode. Whereas to perform neutron structural analysis, it is quite sufficient to take advantage of 'X-ray' phases, i.e., specific neutron methods for determining the phases of structural factors have, on the whole, totally lost their significance.

Actually, that F L Shapiro was coauthor of a relatively small number of publications on condensed matter physics should be qualified conditional. The point is that F L Shapiro was very reluctant to be coauthor of a publication if the text of the article was not written by him personally. A characteristic example is communication [26] on determining the Debye–Waller factor of tungsten, which was needed for calculation of the neutron interaction amplitude with an electron from diffraction data, obtained with the tungsten isotope ^{186}W [27]. F L Shapiro explained in detail that for such a calculation the idea must be entertained of the unambiguous relationship between the heat capacity of a one-atom substance and its Debye–Waller factor, since both of these characteristics depend on the phonon spectrum of the crystal. Moreover, F L Shapiro searched for and found detailed data in the literature on the heat capacity of tungsten and controlled the performance of calculations but flatly refused to be coauthor of the article presenting the results of calculations, so his name was only mentioned in the acknowledgments. As far as we know, a similar situation also occurred in the case of original work on the self-diffusion coefficient of ethane in the vicinity of the liquid–vapor critical point, and on the action of pulsed magnetic fields on antiferromagnetic crystals.

Before the end of the 1970s, after the first diffraction demonstrations performed in 1963–1964 at the IBR-1 reactor in the Laboratory of Neutron Physics, several experiments were carried out that were important from the scientific and methodical points of view. Already in 1965, measurements were performed of neutron diffraction patterns of the $BiFeO_3$ polycrystal, from which determination was successfully made of the orientation of the moments of iron ions relative to the crystallographic axes [28]. The success of the experiment was facilitated by an important feature of a TOF diffractometer — its resolution at a fixed scattering angle improves as the interplane distance increases. In 1967, I Sosnowska, a Polish physicist and one of the coauthors of this study, defended her Candidate of Sciences (PhD) thesis on this topic.

In 1967, A Holas et al. proposed and a year later implemented at the IBR-1 a method for focusing neutrons by time (or geometrical focusing) [29], which permits noticeably enhancing the luminosity of a TOF diffractometer without impairing its resolution. Independently, somewhat earlier, this method was proposed by J M Carpenter of the Argonne National Laboratory (ANL) (USA) [30], but the first focussed TOF diffractometer was constructed in Dubna. The method essentially leans upon the fact that in the case of diffraction, i.e., when the Bragg–Wolf condition is satisfied, there is total correlation between the scattering angle and the wavelength (velocity) of a neutron, and the time of flight of a neutron from the source to the detector can be rendered constant for neutrons of different wavelengths if they travel along different paths. For example, in the case of a moderator and a detector having the shape of plates, when certain conditions are fulfilled (see Ref. [2] for details), the geometrical contribution to the resolution function equals zero in the first approximation. In experiments with polycrystals, this method permits increasing the luminosity of the diffractometer by a factor of about five without noticeably impairing the resolution, and the method continues to be widely applied.

In 1967, the proposal was made in the Laboratory of Neutron Physics to perform diffraction experiments, which were subsequently realized (see book [27]), the aim of which reduced to determining the neutron–electron interaction amplitude related to the nonzero root-mean-square radius of the charge distribution in the neutron (n – e interaction). These experiments turned out to serve as an example of the application of the slow-neutron diffraction for determining nuclear-physical constants (coherent scattering lengths) and constants directly related to the quark structure of nucleons. The idea of the experiments was based on a relatively rare situation: preliminary estimates suggested that, owing to the interference of potential and resonance scatterings in the case of the isotope ^{186}W, its neutron coherent scattering length is close to zero. Accordingly, the relative contribution of the n – e interaction to the total scattering cross section undergoes a drastic increase.

As a first step, it was necessary to measure the scattering lengths of all the main isotopes of tungsten. To this end, four powder samples were prepared, one of which represented a natural mixture of tungsten isotopes, while the others were enriched with isotopes ^{182}W, ^{184}W, and ^{186}W up to about 80–90%. Measurements were performed at the IBR-1 reactor operating at an average power of 6 kW. Indeed, no Bragg peaks were observed to belong to the enriched ^{186}W-admixture, in spite of the very long measurement time (159 h) and the mass of the sample (77 g). The coherent scattering lengths of the isotopes ^{182}W, ^{183}W, ^{184}W, and ^{186}W were determined from the solution of a set of four equations for the measured intensities of diffraction peaks. The following coherent scattering lengths were obtained (in units of 10^{-12} cm): $b_{182} = 0.833(14)$, $b_{183} = 0.43(5)$, $b_{184} = 0.759(9)$, and $b_{186} = -0.119(5)$ [31]. These values were subsequently refined, but on the whole they turned out to be valid. Further experiments for determining the amplitudes of the n – e interaction were carried out with a ^{186}W single crystal at the steady state reactor in Obninsk.

The idea of a pulsed action on a sample, which is synchronized with pulses from the source, was realized at the IBR-1 already in 1968 and its results were presented in detail in review [15] Namely, for the first time in world

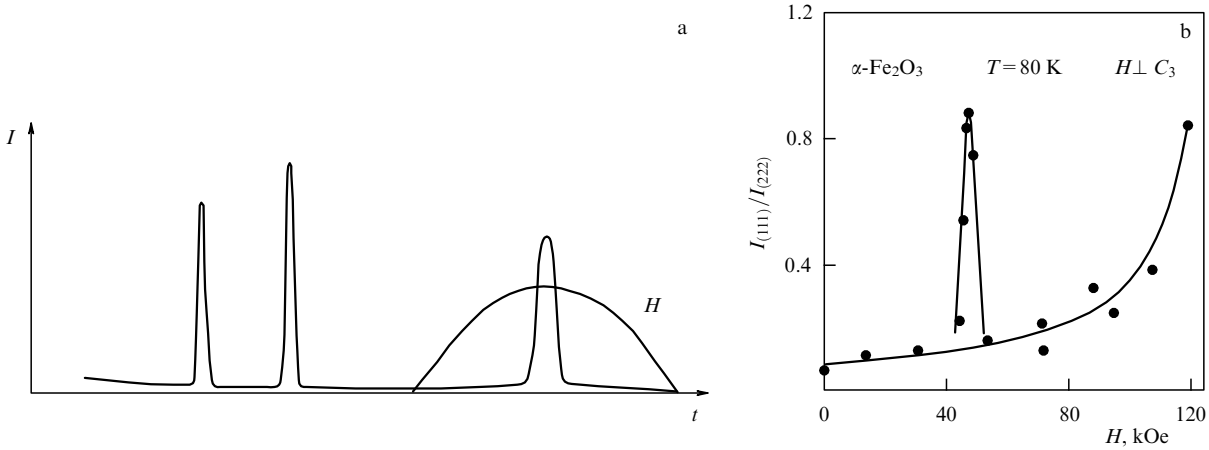

Figure 4. (a) Time diagram of the spectrum of neutrons scattered by a single crystal and of the pulse of the magnetic field H (segment of the sinusoid), the duration of which can vary within the range from 0.5 to 3 ms. (b) Dependence of the hematite (111) peak intensity, normalized to the intensity of the 'nuclear' (222) peak, on the amplitude of magnetic field pulses (taken from Ref. [32]). At $H \approx 4.95$ T, a peak of anomalous scattering is observed. C_3 is the rhombohedric axis.

practice, a pulsed magnetic device (PMD) was constructed and equipped with a TOF diffractometer, and it was utilized to perform a series of experiments with a hematite single crystal, α-Fe_2O_3, and a magnetic field amplitude up to 12 T [32]. Specially for these experiments, the reactor with a power of 6 kW was operated in the mode of 'rare pulses', namely at a frequency of 0.2 Hz, which was necessary in order to restore the initial PMD state between pulses. The maximum values of stationary magnetic fields used at the time in neutron experiments did not exceed 4.5 T. The time resolution that can be achieved in such experiments ranged between 2 and 10 μs, depending on collimation of the primary beam, which corresponds to a magnetic field amplitude resolution of about 2%. Figure 4 illustrates how the field pulse coincides with the diffraction peak, and one of the first results for hematite is also presented.

Subsequently, the construction of the PMD and of pulsed magnets repeatedly underwent improvement, and in 1988 at the IBR-2 reactor operation started of the spectrometer for neutron studies with a pulsed magnetic field (SNPM), with which numerous studies were performed on the kinetics of magnetic-orientation phase transitions of the 1st order, of phase rearrangement in the case of spin-flop transitions in collinear antiferromagnetics, of magnetic ordering induced by an external field, etc. A detailed discussion of the methodical basis of neutron studies in pulsed magnetic fields, and of the main physical results obtained at the LNP reactors is to be found in the review by V V Nietz [33].

6. Specialization of time-of-flight diffractometers

Besides the distinctions in their interactions with matter, exhibited by X-rays and neutrons, there is one more distinction that is essential. The neutron flux does not exceed 10^{15} cm^{-2} s^{-1} even at the most modern sources, while a laboratory X-ray diffractometer provides a flux of quanta $\approx 10^{18}$ cm^{-2} s^{-1}, and the flux at advanced sources of synchrotron radiation may be even 10 orders of magnitude higher. This leads to the necessity of dealing with samples of noticeably larger volumes and with longer times of acquiring the necessary statistics in neutron than in X-ray experiments. One more consequence of the insufficient luminosity of neutron diffractometers is the necessity of optimizing them when resolving certain concrete problems. Thus, neutron diffractometers differ not only in scanning modes of the reciprocal space of a crystal, but also in the types of the main experimental problem.

The schematic layout of a TOF duffractometer, dealt with in detail in review [2], has undergone no substantial changes in the 50 years since it appeared. Moreover, for a long time the initial layout of a TOF diffractometer (Fig. 2b) was considered to be universal, i.e., to permit resolving most of the problems that could arise. However, a clear tendency to the TOF-diffractometer specialization has been revealed during the past 20 years, namely, to the installation of specific constructions of certain components permitting certain structural problems to be optimally resolved.

Somewhat arbitrarily, it is possible to identify eight types of experiments, the conduction of which requires optimization of the diffractometer relative to its main characteristics (resolution and luminosity) and the construction of a detector system (Table 1). Certain additional requirements are indicated in the last column of this table. Detailed consideration of the construction versions of such diffractometers goes beyond the scope of the present review. Therefore, to illustrate modern tendencies, we shall only consider the powder high-resolution diffractometer that is always present in the suite of spectrometers at both stationary and pulsed neutron sources.

TOF diffractometer for structural studies of polycrystals. Starting from the first experiments carried out in 1948–1949 on nuclear reactors, structural studies of polycrystals have made up the main part of neutron diffraction studies. The fundamental possibility of analyzing one crystalline structure or another depends primarily on the resolution of the diffractometer.

As a rule, a neutron experiment is conducted in order to improve the information about a known structure or to analyze its variations under external influence on the substance studied. This problem is resolved by minimization of a functional that contains the measured and calculated intensities of diffraction peaks, with the formula for the calculation of intensities involving both the coordinates of atomic positions and their atomic occupancy factors as the parameters to be refined, as well as the atomic thermal factors. Correct mathematical setup of this problem assumes

Table 1. Main types of structural problems resolved with TOF diffractometers, and the requirements for the resolution, luminosity, and detector system of the diffractometer.

Problem	Resolution	Luminosity	Detector	Comment
Structure of a single crystal	Medium	Medium	Two-coordinate PSD	Position resolution of detector ≈ 2 mm
Structure of a polycrystal	Very high	Medium	Large area with $\Omega > 1$ sr	Large scattering angles required ($> 150°$)
Magnetic structure	Medium	Medium	Large area with $\Omega > 1$ sr	Measurement of large (> 10 Å) d_{hkl}
Real-time experiment	Medium	Very high	Several detectors at different, including small, angles 2θ	Large interval in d_{hkl}
High pressure	Medium	Very high	Several detectors at different angles 2θ	Large interval in d_{hkl}, low background
Macromolecular structures	Medium	High	Several detectors at different, including small, angles 2θ	Measurement of very large (≈ 60 Å) d_{hkl}
Local distortions of a structure	High	High	Large area with $\Omega > 1$ sr	Large momentum transfers required, $Q > 40$ Å^{-1}
Microstructure of materials	High	Very high	Large area at $2\theta \approx 90°$	Medium interval over d_{hkl}

that several (from three to seven) experimental points account for each individual parameter to be refined, with the role of these points attributed to the intensities of individual diffraction peaks.

Thus, the condition for successful resolution of a structural problem consists in measuring a necessary number of peak intensities. It can be shown that under the condition of a weak dependence of the resolution function $R = \Delta d/d$ on the interplane distance d, the number of peaks from a crystal measured individually on a TOF diffractometer amounts to $N \approx 1/(3\Delta d/d)$, i.e., an improvement in the resolution by several-fold permits increasing the number of parameters to be refined by the same factor. At present, several neutron centers have achieved resolution levels in the range from $R \approx 0.001$ to $R \approx 0.0005$; consequently, with these diffractometers it is possible to correct structures with up to several dozen independent parameters.

Special features of a high-resolution spectrometer with a monochromatic beam (λ_0-diffractometer) include a large angle of reflection ($\theta_M > 120°$) from the monochromator of low mosaicity and a good collimation of the beam incident upon the monochromator. At the Institut Laue–Langevin (ILL), Grenoble, the first high-resolution λ_0-diffractometer D1A [34], exhibiting $\Delta d/d \approx 0.002$ at the minimum of the resolution curve, was constructed precisely in accordance with the above principle.

High resolution of a TOF diffractometer can be achieved at neutron sources with short pulses if a relatively thin moderator is applied, and the flight path L exceeds 50 m. The best-known example is HRPD (High Resolution Powder Diffractometer) at the pulsed neutron and muon source ISIS[2] of the Rutherford Appleton Laboratory (RAL) [35] with $L \approx 100$ m, which, in principle, permits obtaining $\Delta d/d \approx 0.0004$.

The main development tendency of the TOF diffractometers at short-pulse sources consists in the use of complex constructions of mirror neutron guides (in particular, supermirror ballistic guides [36]) for transportation of the neutron beam over long distances, and of detectors exhibiting large solid angles (up to several steradians).

The following installations give examples of advanced TOF diffractometers with a resolution at a level of 0.001 or better: WISH (Wide angle In a Single Histogram) ($L = 50$ m) at ISIS, POWGEN (Powder Diffractometer Next Generation) ($L = 60$ m) at SNS in the Oak Ridge National Laboratory (ORNL), and SuperHRPD ($L = 94$ m) at J-PARC (Japan Proton Accelerator Research Complex). The high resolution of these diffractometers is due to the short pulse of the source ($\Delta t \approx 15-50$ μs) and long flight path, while the quite high luminosity level is provided by the large area of the detector system. For example, the POWGEN detector covers an area of 7 m^2, thus forming a solid angle $\Omega \approx 1.5$ sr.

As an illustration, Fig. 5 presents the layout of the TOF diffractometer WISH, which is mainly invoked in the joint analysis of crystalline and magnetic structures of polycrystals. The diffractometer is placed in a beam of cold neutrons with the maximum of its distribution at $\lambda \approx 4$ Å, the supermirror neutron guide with $m = 3$ provides a high transmission of cold neutrons, while the detector covering scattering angles from 10° up to 175° permits simultaneous registration of quite a large number of diffraction peaks within a very broad range of interplane distances. Precisely the last circumstance makes possible the simultaneous analysis of the crystalline and magnetic structures of the substance being studied.

In the case of pulsed sources with long pulses, it is necessary for obtaining a high resolution to reduce the pulse length with the aid of one type of mechanical chopper or another. In practice, either a standard single-slit chopper, with which arrangement of the experiment and the method of diffraction data accumulation are the same as in the case of a TOF spectrometer at a short-pulse neutron source, or a multislit chopper with a large number of regularly situated slits of the same width, known as a Fourier chopper, is used. To obtain the diffraction pattern, it is necessary in the latter case to apply correlation analysis of the signals from the source, the chopper, and the detector, which is done either online with the aid of special electronics, or by computer calculations after accumulation of the initial data.

[2] ISIS is not an abbreviation. The neutron source was named after the ancient Egyptian goddess Isis.

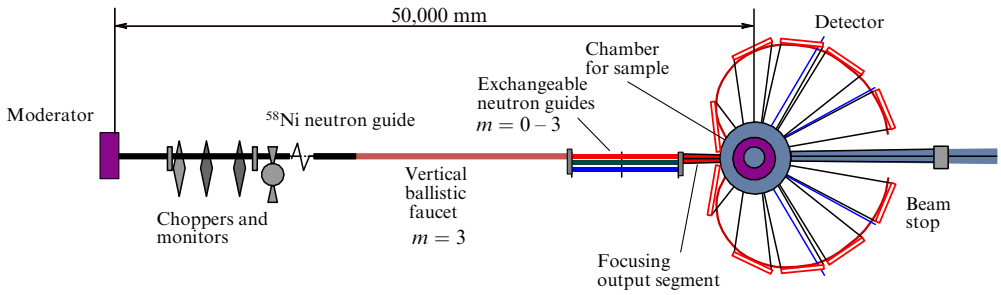

Figure 5. Layout of the TOF diffractometer WISH (ISIS), the scope of which is the joint analysis of crystalline and magnetic structures of polycrystals. Specific features of the facility include a relatively long distance between the source and the sample ($L = 50$ m) and the presence of a set of additional choppers, permitting choosing the required range of d_{hkl}, of a supermirror ballistic neutron guide with a coefficient characterizing enhancement of the critical angle of reflection from the neutron guide walls due to their multilayer structure, $m = 3$, and of a detector covering scattering angles from 10° up to 175°.

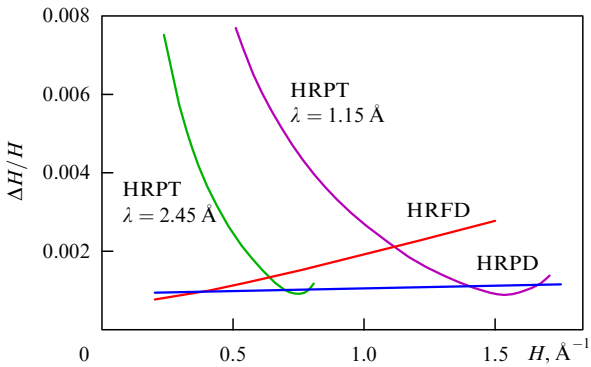

Figure 6. Resolution functions for polycrystals of two TOF diffractometers, HRFD (High Resolution Fourier Diffractometer) and HRPD, and the λ_0-diffractometer HRPT (High Resolution Powder diffractometer for Thermal neutrons) [SINQ (Schweizerische Intensive Neutronen-Quelle) of the Paul Scherrer Institute, PSI] for two wavelengths as functions of the vector length in reciprocal space H. While the resolution function for TOF diffractometers weakly depends on H, a λ_0-diffractometer is characterized by the presence of a deep minimum and by deterioration of the resolution at small H.

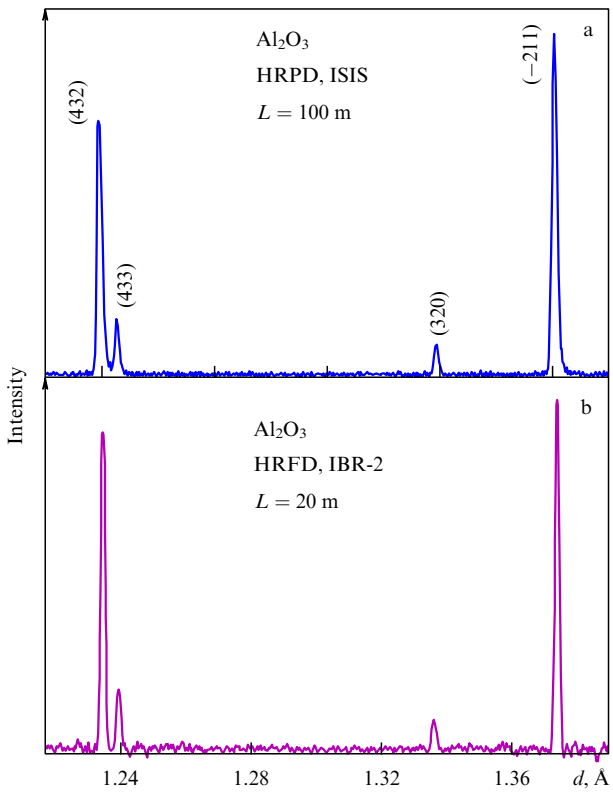

Figure 7. Comparison of segments of neutron diffraction patterns measured with the same Al_2O_3 polycrystal by TOF diffractometers HRPD (ISIS) (a) and HRFD (IBR-2) (b). In this region of the spectrum, $\Delta d/d \approx 0.0016$ for both diffractometers, although the flight path at HRFD is five times shorter than at HRPD, which is related to its correlation component. The Miller indices of diffraction peaks are indicated for a rhombohedral setting.

The functional dependences of the resolution on the interplane distance or the momentum transfer in the case of the three indicated types of high-resolution diffractometers differ noticeably. A common feature of λ_0-diffractometers consists in the presence in $R(H)$, where $H = 1/d$ is the vector length in reciprocal space, of quite a deep minimum, the position of which mainly depends on the angle of reflection of the primary beam from the monochromator. On the contrary, the resolution function in the case of TOF diffractometers depends weakly on H (Fig. 6). These peculiarities often happen to be extremely important, and they must absolutely be taken into account in planning experiments.

The high-resolution Fourier diffractometer (HRFD) at the pulsed IBR-2 reactor has been in operation since 1994, and essential information concerning the method of neutron Fourier diffractometry and the HRFD construction, as well as examples of the first structural experiments, were presented in our review [2]. Moreover, the development history of this method is considered in recent review [37], its technical and methodical peculiarities are examined in detail, and the scientific program implemented with HRFD is discussed. Therefore, we shall only briefly present here certain essential points in the application of a Fourier chopper at a long-pulse source, emphasizing its positive and negative features.

The Fourier chopper received its name owing to its periodic transmission function being close in shape to a sinusoid. Therefore, in the first approximation at a fixed frequency of the chopper rotation, the intensity of scattered neutrons, $I(\omega)$, represents a Fourier harmonic of the scattering cross section of neutrons by the sample. Consequently, it is possible to reconstruct the Fourier profile by measuring $I(\omega)$ within a broad range of frequencies, i.e., to obtain the scattering cross section $\sigma(t)$, where t is the time of flight related to the momentum transferred in scattering or to the interplane distance in the crystal.

The main advantage of the Fourier method lies in its high resolution at a relatively short flight path. The calculated value of $\Delta d/d$ for HRFD amounts to 0.0005 for a total flight path from the chopper to the sample of only 20 m. The same resolution at SNS with a short pulse can be achieved at $L \approx 100$ m (Fig. 7). In this case, a serious problem arises due

to overlapping the neutron spectra from adjacent source pulses. Thus, the range of d_{hkl} observed simultaneously by back scattering detectors at the HRPD (ISIS) with $L \approx 100$ m and the pulse repetition rate of 50 Hz for a neutron source is only 0.4 Å, and for widening it to at least 2 Å it is necessary to remove four of the five pulses with the aid of additional choppers, i.e., to work with an intensity that is five times lower.

Another important advantage of the Fourier method consists in the possibility of optimizing the resolution depending on the type of problem to be resolved — to this end, it is only necessary to change the rotation velocity of the Fourier chopper, since the time component of the resolution function is linearly related to the maximum modulation frequency of the neutron beam.

The main problems in applying Fourier choppers are due to the correlation nature of the accumulated information. As a result, an indispensable attribute of the measured diffraction patterns is the so-called correlation background, the appearance of which follows from the ambiguity in matching up a concrete event in the detector with one flight time or another. Moreover, the correlation nature of data and certain technical problems in modulation of the primary beam intensity lead to a relatively complicated profile of diffraction peaks, which cannot be described by any analytical functions with the required precision.

Among the possible methods for resolving these problems, the one presently considered as the most effective is the method of primary data accumulation in the list mode. With the aid of this method, instead of forming histograms (neutron diffraction patterns), accumulation touches upon individual events, each of which matches up its absolute registration time and certain parameters, in particular, the number of the triggered detector. The 'words' thus formed are recorded in quite a large memory. The advantages of such a way of recording information includes the possibility of registering data from a large number of detector elements and the fact that experimental data are retained in their initial form. Correspondingly, the possibility arises of processing experimental data applying code that involves necessary refinements, of which the most important is the refinement due to the discrepancy between the saw-shape transmission function of the chopper and a respective series of electronic signals.

To summarize, the Fourier diffractometer can be said to exhibit all the merits of a conventional time-of-flight diffractometer but, unlike the latter, it permits an extremely high resolution to be achieved with a relatively short flight path, which contributes to enhancement of the neutron flux on the sample and to reducing the cost of the device. An important circumstance is the weak dependence of the resolution function on the interplane distance within quite a broad interval. The problem of overlapping pulses from the source, which is quite unpleasant for conventional time-of-flight spectrometers, does not exist in the case of the Fourier diffractometer. A Fourier diffractometer can be most effectively applied at a neutron source with a long (exceeding 300 μs) pulse.

7. Scientific problems resolved with advanced time-of-flight diffractometers

Actually, each individual structural scientific problem of the ones listed in Table 1 can be resolved at any of the pulsed neutron sources presently in operation, since all of them are equipped with appropriate dedicated time-of-flight diffractometers. Therefore, in Sections 7.1–7.6, the solutions are considered of several of the most interesting problems, mainly taking advantage of studies carried out at the IBR-2 reactor, which, in addition, demonstrate how successful the development was of the technique and methodology that originated in the Laboratory of Neutron Physics 50 years ago.

7.1 Studies of single crystals

The most important advantage of a TOF diffractometer for single crystals is the possibility of three-dimensional analysis of large volumes of a crystal's reciprocal space without any rotations of a sample. This can be achieved, as was already mentioned in Section 3, by joint scanning over the neutron time of flight and over two coordinates of a 2D PSD. This possibility has been realized in its most complete form with the SXD (Single Crystal Diffractometer) (ISIS) [38], the detector system of which is composed of 11 two-coordinate PSDs, covering nearly half of the complete sphere (49.4%). Each detector contains 4096 sensitive elements with a total area of 192×192 mm^2. Such a geometry permits us to collect the necessary number of reflections from the crystal without any its rotations or reducing their number to a minimum. In the case of a typical crystal volume of 100 mm^3, sufficient statistics are accumulated in 1–2 h. Mainly, studies of organic substances, in order to establish the positions of hydrogen atoms, and of diffuse scattering, due to structural disorder or correlated thermal oscillations of atoms, are performed with SXD.

The detector system of the DN-2 diffractometer at the IBR-2 reactor also includes 2D PSD (a single one, so far) with linear dimensions of 250×250 mm^2 and a position resolution $\Delta x \approx 2$ mm, which permits detailed 3D scanning of reciprocal space. As an example, Fig. 8 demonstrates a small section of the diffraction pattern from an $La_2CuO_{4.04}$ single crystal, containing two orders of reflection. When this crystal is cooled, owing to transformational twinning upon phase transition to the low-symmetry (orthorhombic) phase, the nodes of the reciprocal lattice split into several components, two of which are well seen in the figure. Besides, bands of diffuse scattering are seen due to a certain level of disorder in the crystal's structure.

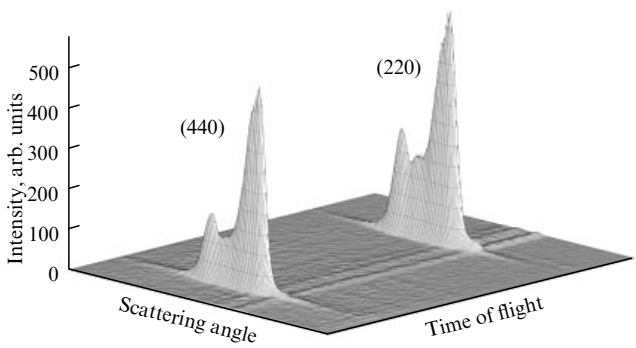

Figure 8. Two-dimensional intensity distribution measured with the DN-2 diffractometer within the ±5° range of scattering angles along the ($hh0$) direction in the $La_2CuO_{4.04}$ crystal. Two reflection orders are seen, (220) and (440), which are split into components. Sweeping of the spectrum over the flight time and the scattering angle corresponds to scanning along and across the radius vector of the reciprocal lattice. In addition to diffraction peaks, the bands of neutron diffuse scattering are also seen.

The intensity distributions measured by a TOF diffractometer with a 2D PSD contain up to 10^6 experimental points, and, accordingly, the time required for implementation of the experiment is enhanced by the same factor, as opposed to the relatively recent situation, when measurements were performed with the aid of a pointlike detector and a monochromatic neutron beam. The practically continuous scanning of the reciprocal space, achieved here, is especially important, when it is not known in advance within which region of space scattering peculiarities are to be found, for instance, in studies of incommensurate modulated structures.

7.2 Structural investigation of polycrystals

Structural studies of polycrystals at the IBR-2 reactor are mainly performed with the HRFD diffractometer [39]. In recent years, it has been used to carry out extensive work representing practically all the scientific lines of research presently of interest. Improvements have been achieved in the understanding of the structures of high-temperature superconductors (HTSCs) and compounds containing hydrogen, and of various oxides (perovskites, spinels, brownmillerites, etc.), and experiments have been performed to determine macro- and microstresses in bulky materials and products. Thus, for example, fundamental results were obtained on the structure of mercury HTSCs, and on the dependence of the critical temperature of the superconducting transition on the occupancy of positions by extrastoichiometric anions, oxygen and fluorine (see, for example, Refs [40–42]). Important results were obtained in a series of studies [43–45] on mesoscopic phase separation in complex manganese oxides and — based on it — the formation of the giant oxygen isotope effect manifested in the substitution of the insulating for the metallic state of the crystal, when the isotope ^{18}O is substituted for ^{16}O.

Much attention has recently been focused on studies of all kinds of structural anomalies [46, 47] and of the microstructure of crystals resided in a nanostructured state [48-50]. A key point of these studies lies in the high HRFD resolution, permitting the identification of microstrains in crystallites at a level of relative variation of interplane distances, due to their strained state, exceeding $\varepsilon \approx 0.0008$ and the average dimensions of coherently scattering domains at a level inferior to $L_{\mathrm{coh}} \approx 3500$ Å. Moreover, the effects of anisotropic broadening of peaks are being confidently determined. In these cases, an important circumstance is the weak dependence of the resolution on the interplane distance d_{hkl}, which provides the possibility of utilizing a large number of experimental points to analyze the widths of diffraction peaks within the framework of the Williamson–Hall approach. In this method, the dependence of diffraction peak widths is plotted versus some experimental variable — in the case of a TOF diffractometer, it is most convenient to invoke d_{hkl} as such a variable. The following relationship can be shown to hold valid for distribution functions approximated by a Gaussian:

$$(\Delta d)^2 = C_1 + (C_2 + C_3)d^2 + C_4 d^4 \,, \tag{1}$$

where Δd is the full peak width at half-height (or the integral peak width), constants C_1 and C_2, which can be determined in an experiment with standard samples, are related to the resolution function of the diffractometer: $C_3 \approx (2\varepsilon)^2$, $C_4 \approx (k/L_{\mathrm{coh}})^2$, and k is the Scherrer constant which is close to unity and takes into account possible anisotropy of the shape of coherently scattering domains.

By determining a certain number of peak widths and plotting the dependence of $(\Delta d)^2$ on d^2 within a sufficiently broad interval of d_{hkl}, it is possible to determine ε and L_{coh}. In the absence of the size effect (very large L_{coh}), dependence (1) will be linear; otherwise, it will be parabolic. Examples of Williamson–Hall plots are given in Fig. 9, where linear dependences on d^2 are shown of square peak widths for the resolution function and for composites with large crystallites, as are parabolic dependences, if the size effect exists. In the latter case, it was possible to show, with the aid of this plot, that the sizes of coherent scattering regions for NiO nuclear and magnetic structures differ significantly in the case of crystallites of average size, $\langle D \rangle = 100$ nm ($L_{\mathrm{nuc}}/L_{\mathrm{mag}} \approx 2.5$), i.e., the long-range magnetic order on the surface of a crystallite turns out to be, to a significant extent, destroyed.

In principle, studies of both crystalline and magnetic structures of polycrystals can be done with a TOF diffractometer. However, certain methodical peculiarities of the TOF diffractometer, primarily the dependence of diffraction peak intensities on the neutron wavelength distribution in the primary beam, hinder precise determination of the absolute value of atomic magnetic moments. Here, good results are achieved by joint analysis of data obtained with λ_0- and TOF diffractometers.

References [51, 52] present a recent example of such an analysis, where precision data were obtained with HRFD and the λ_0-diffractometer HRPT (SINQ, PSI) on the crystalline and magnetic structures of copper ferrite $CuFe_2O_4$, the temperatures of phase transitions, and the microstructural peculiarities of this compound. The data obtained with HRPT permitted us to determine with good precision the values of the magnetic moments of ions resided at the octahedral and tetrahedral sites of a unit cell. Remarkably, the difference between them turned out to remain constant within the entire range of the existence temperatures of an ordered magnetic structure and to correspond to the spin moment of the Cu^{2+} ion. Hence. it follows, in particular, that, contrary to wide-spread opinion, copper and iron ions do not exchange sites upon a structural transition from the tetragonal to the orthorhombic phase.

7.3 Studies of transition processes (real-time experiments)

Real-time studies of transition processes became possible when a drastic increase occurred in the luminosity of neutron diffractometers, which can be defined as the intensity registered by the detector per unit time (number of neutrons per second), which is tentatively

$$I = \Phi_0 S \frac{\Omega_{\mathrm{d}}}{4\pi} \delta \,, \tag{2}$$

where Φ_0 is the total neutron flux on the sample in the working range of wavelengths, Ω_{d} is the solid angle covered by the detector system, S is the area of the sample, and δ is its scattering capacity. Formula (2) permits estimating the feasible counting rate for known diffractometer parameters. For example, the total neutron flux at the DN-2 diffractometer amounts to $\sim 10^7$ cm^{-2} s^{-1}, while the solid angle of the existing 2D detector is close to 0.2 sr. For a sample area of 5 cm^2 and its scattering capacity (within a 4π solid angle) $\delta \approx 0.1$, we achieve a total neutron counting rate $I = 10^5$ s^{-1}. This is a large quantity that suffices for analyzing diffraction patterns of relatively simple structures in a measurement time of a single spectrum $t_{\mathrm{s}} \sim 1$ min with a reasonable statistical accuracy. The main reserve for enhancing the counting rate is

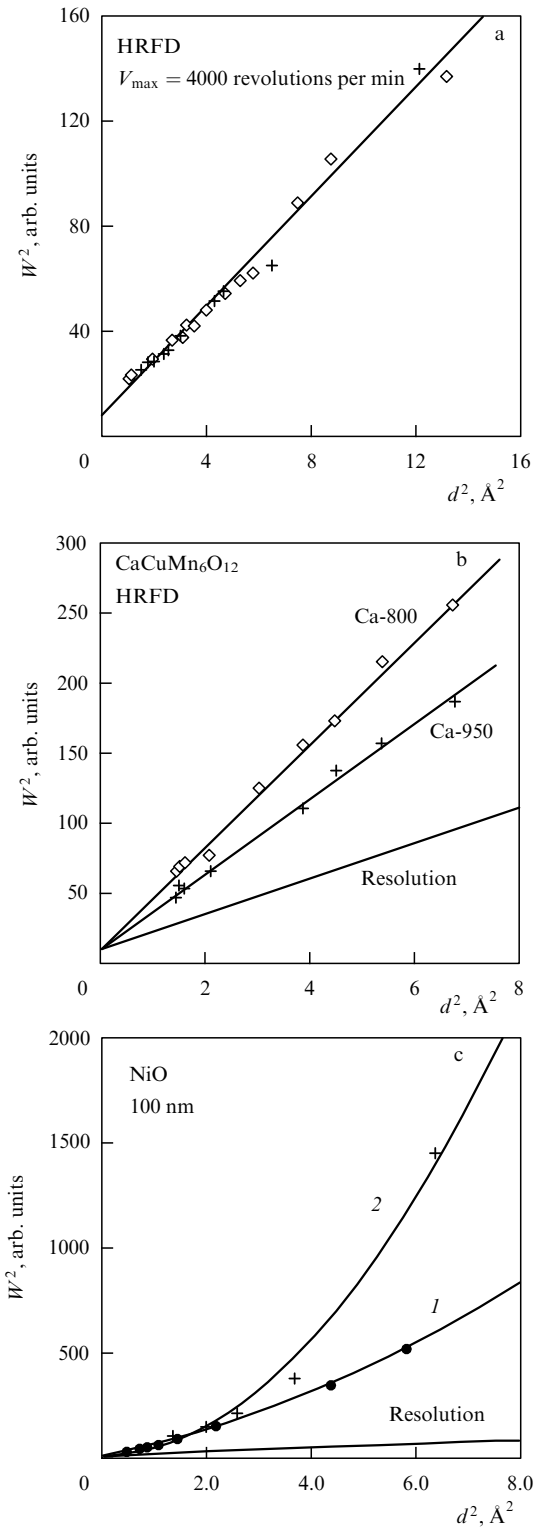

Fig. 9. Williamson–Hall plots for the HRFD resolution function, measured with standard samples (a) for Al_2O_3 (crosses) and $Na_2Al_2Ca_3F_{14}$ NAC (diamonds) at a maximum rotational speed of the Fourier chopper of 4000 rotations per min, (b) for two $CaCuMn_6O_{12}$ compositions annealed at temperatures 800 and 950 °C (the effect of microstresses is present), and (c) for NiO oxide with crystallites possessing an average size of 100 nm (the size effect is present). Two curves for NiO are shown, which correspond to the crystalline (curve *1*) and magnetic (curve *2*) regions of long-range order. The size of the magnetic regions in NiO is inferior to the those of crystalline regions, so the increase in the parabola is, respectively, more rapid.

to be found in the solid angle. Assuming the construction of a detector with $\Omega_d \approx 2$ sr to be realistic, we conclude that the neutron counting rate will amount to $\sim 10^6$ s^{-1}, which will make possible a reduction of t_s to several seconds.

The implementation of real-time experiments at the JINR Frank Laboratory of Neutron Physics has a long and rich history; in review [2], such examples are considered as solid-phase chemical synthesis, isotope exchange in lipid membranes, and phase transformations of high-pressure metastable ice (see also reviews [53, 54]). In these studies, the possibilities of high intensity λ_0- and TOF diffractometers are shown to be comparable, if processes are studied with characteristic times at the level of 1 min or more. The TOF diffractometer permits us, however, to register, besides diffraction data, small-angle neutron scattering, i.e., to follow the transformation of macrostructural inhomogeneities in a medium. Moreover, with a TOF diffractometer, the necessary statistics can be accumulated during a single power pulse, which is confirmed by the results of simulated experiments. In this case, in accordance with arguments presented in Ref. [55], investigation is feasible of irreversible processes with a time resolution at a level of 1 ms. This value is apparently close to the limit resolution in neutron diffraction of irreversible processes. It is comparable to the values achieved at synchrotron radiation sources and substantially inferior to the limits achievable at steady state reactors.

The long TOF diffractometer operational range of wavelengths permits successfully studying structures with repetition periods up to several dozen angströms, and also performing kinetic experiments with such structures. Thus, at the DN-2 diffractometer, studies are underway of hydration processes of multilayer oriented lipid membranes. A unique feature of these experiments lies in the simultaneous measurement of several reflection orders, which makes it possible to reconstruct structural variations in a lipid bilayer during its hydration–dehydration.

A demonstrative example is the experiment on hydration of an oriented multilayer membrane of dipalmitoylphosphatidylcholine (DPPC), in which the transition was studied from the state of low relative humidity of water vapor to a state of high relative water vapor humidity, and vice versa [56]. For this purpose, a dry sample that resided primarily in room conditions (relative humidity 46%, temperature 20 °C) was placed in an aluminum container inside of which a relative humidity of 98% was established with the aid of a saturated salt solution of K_2SO_4 in water with 8% of D_2O, and the temperature was kept at 20 °C. Neutron diffraction patterns were registered for ≈ 8 h, with the acquisition time of a single spectrum gradually increasing (from 3 to 60 min). Registration of the first spectrum started 2 min after the saturated salt solution was placed in the measurement chamber. When the sample reached an equilibrium state, the salt solution of K_2SO_4 was replaced by a saturated solution of NaBr, which assigned a relative humidity of 58% of water vapor. Registration of the membrane dehydration process went on for about 7 h, with the acquisition time of a single spectrum set from 2 up to 60 min. The sequence of neutron diffraction patterns measured in this process is displayed in Fig. 10, from which the intensities of diffraction peaks are seen to depend very strongly on the degree of membrane hydration, which corresponds to a significant rearrangement of the atomic structure.

The relatively large number of measured reflection orders and the standard procedure of inverse Fourier transforma-

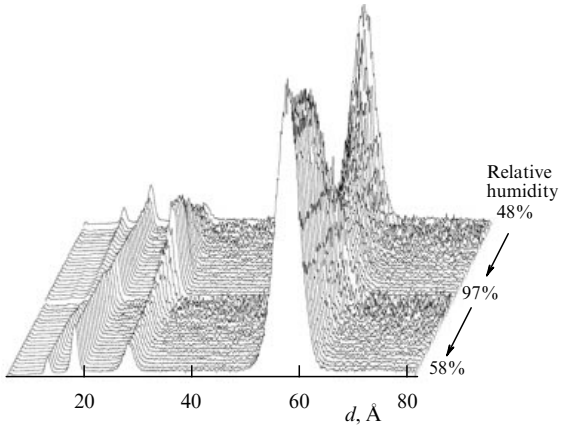

Figure 10. Sequence of real-time neutron diffraction patterns from a DPPC membrane measured during the process of membrane hydration and subsequent dehydration. The position of the first-order reflection varies within the limits from 58 to 60 Å. The pattern corresponding to the membrane in an equilibrium state in the case of a 58% relative humidity contains four visible diffraction peaks. A fifth diffraction peak is seen at larger humidity values.

tion (plotting the Fourier profile) permitted recovering of the lipid bilayer structure at all stages of the process with quite good accuracy. Information was obtained on changes in the water interlayer thickness between the polar heads, on the change in the inclination of hydrocarbon tails, and on characteristic time durations of different stages of the processes. Thus, for example, the hydration process turned out to be well described by a single exponential dependence with a characteristic time $\tau \approx 60$ min. On the contrary, the fast ($\tau_1 \approx 15$ min) stage is observed first in the course of dehydration, and then the slow ($\tau_2 \approx 100$ min) stage.

In recent years, neutron diffraction has been actively applied in studying transition processes in chemical sources of electric current, in particular, in Li-ion batteries. The potential of TOF diffractometers is perfectly consistent with experiments of this kind, since they make it possible to obtain detailed information on the evolution of structural phases during the battery charging/discharging process and on changes in the microstructure of the electrode material. As an example, Fig. 11 demonstrates the evolution of neutron diffraction patterns from an 18650 commercial cylindrical battery during several charging–discharging cycles, performed at different rates. From an analysis of the intensities and positions of diffraction peaks, information is obtained on the structural details of the lithium ion transitions from the cathode to the anode and vice versa, on the fraction of the electrode material involved in the process, on the extent of volumetric expansion or compression of the electrodes, etc. Examples of investigations of this type can be found in review [57].

7.4 Studies of atomic and magnetic structures at high pressures

As noted in Section 3, the TOF diffractometer was considered especially promising for implementation of experiments at high external pressure from the very beginning of its construction, which is related to the possibility of operation with a fixed scattering geometry. Another important circumstance for such experiments is the repeatedly aforementioned broad range of wavelengths permitting coverage of a large interval of interplane distances at a fixed scattering angle and,

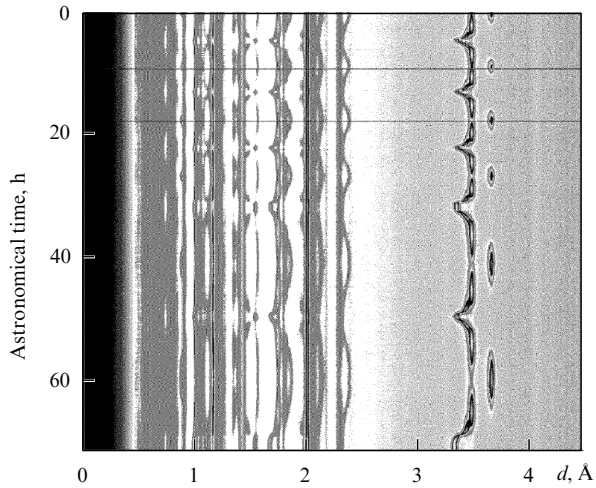

Figure 11. Evolution of neutron diffraction patterns from an 18650 battery in the course of several charging–discharging cycles performed at different rates. The vertical bands are the diffraction peaks corresponding to the respective materials of the anode and cathode, and they somewhat change their positions and intensities during the process running. In the region of the interplane distance $d \approx 3.5$ Å, there are diffraction peaks from the graphite anode, into which lithium is incorporated in the course of charging with the formation of several intermediate LiC_n phases and the final LiC_6 phase (the diffraction peak is situated separately at $d \approx 3.7$ Å), corresponding to the total charge of the battery.

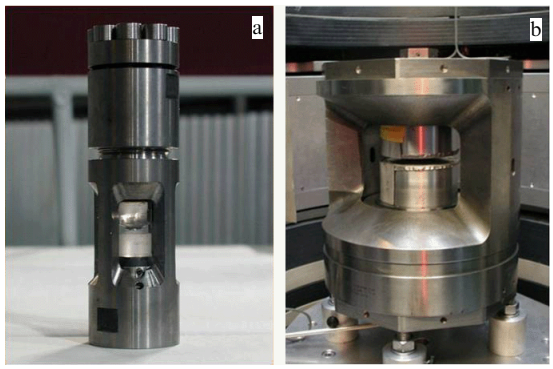

Figure 12. (a) High-pressure cell with single-crystalline anvils. Anvils of sapphire crystals permit working at pressures up to 10 GPa, and diamond anvils at pressures right up to ≈ 50 GPa. The volume of the sample is ≈ 5 mm^3 in the first case, and ≈ 0.1 mm^3 in the second. The cell with the sample can be cooled from room temperature to temperatures of several kelvins. (b) 'Paris–Edinburgh' press for operation within the range of pressures up to ≈ 30 GPa with a sample volume of 30–100 mm^3. The working range of temperatures extends from 90 to 1000 K. Cells of both types are used with the dedicated DN-6 and DN-12 diffractometers at the IBR-2 reactor.

accordingly, obtaining sufficient data set for the analysis of both atomic and magnetic structures.

For a long time the chambers (cells) applied in neutron studies at high pressures were of cylinder–piston types, which permitted reaching pressures of 2–3 GPa. However, in the 1980s, the cells that started to be used in Russia were with single-crystalline and then with toroidal anvils (Fig. 12). The first of them, together with a low-background multidetector registration system of diffraction patterns [58], permitted the study of samples of volumes 0.1–10 mm^3 at pressures up to ≈ 7 GPa (in sapphire anvils), and up to ≈ 50 GPa (in

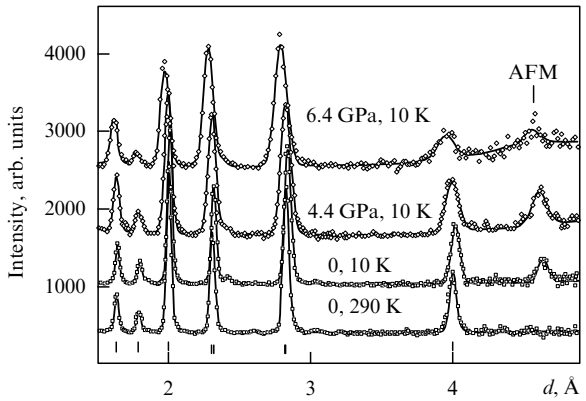

Figure 13. Neutron diffraction patterns for $PbFe_{0.5}Nb_{0.5}O_3$, measured with DN-12 at several values of the external pressure and temperature, and processed by the Rietveld method. Seen are the appearance of the antiferromagnetic (AFM) peak at low temperatures, the shift of peaks toward shorter interplane distances with an increase in pressure, and the change in the relationship between the intensities of magnetic and nuclear peaks.

diamond anvils). At the beginning of the 1990s, teams of physicists from Paris and Edinburgh developed a cell (that was termed the 'Paris–Edinburgh' press) based on anvils of tungsten carbide (WC) of toroidal geometry for neutron spectrometers [59]. The pressure achieved in this cell was ≈ 30 GPa, with a sample volume ranging 30–100 mm^3.

Cells of both types are applied at the IBR-2 reactor for experiments at high pressures, but work was particularly effective when cells with sapphire and diamond anvils were employed. For this purpose, two dedicated diffractometers (DN-6 and DN-12) were constructed with multidetector registration systems. Shown in Fig. 13 are typical neutron diffraction patterns that were measured using DN-12 in a cell with sapphire anvils with the $PbFe_{0.5}Nb_{0.5}O_3$ composition exhibiting relaxation and multiferroic properties [60]. Their quality is sufficiently good for analysis of atomic and magnetic structures with respect to relatively simple compounds, such as perovskite-like oxides. The resolution of the diffractometer is not so high, about 1%, but, as a rule, diffraction peaks are broadened owing to the pressure gradient in the cell, and this resolution is optimal.

Studies at high pressures are one of the most successful scientific topics at the IBR-2 reactor. In the relatively short time that such studies have been performed (DN-12 was commissioned in 1995 [61]), several large series of studies have been carried out with ammonium halogenides, mercury halogenides, manganese and cobalt oxides, multiferroics, etc. (see, for example, Refs [62–64]).

7.5 Investigation of local structural disorder

The construction of short-pulse SNS has permitted the achievement of substantial progress in diffraction studies of local disorder effects in the structure of crystals. The idea, termed PDF technique (Pair Distribution Function technique), consists in analyzing the total radiation–crystal scattering function, which, besides coherent Bragg peaks, involves a diffuse component. This component, an addition to the intensity of Bragg peaks, arises if the long-range order is violated in some manner and diffuse scattering appears in the form of a more or less smooth intensity distribution. For example, in the structure of the complex oxide $La_{1-x}Ca_xMnO_3$, the La and Ca ions are distributed randomly over the unit cells and, accordingly, incoherent scattering arises with an intensity proportional to $(b_{La} - b_{Ca})^2$, where b_{La} and b_{Ca} are the coherent scattering lengths manifested as a constant diffuse background. The diffuse scattering related to distortion of the shape of the oxygen octahedron around the manganese ions in this structure is more complex. At $x = 0$, all the manganese ions reside in the Mn^{3+} state, which leads to distortion of the MnO_6 octahedrons owing to the Jahn–Teller effect. When $x = 1$, on the contrary, all the manganese ions are in the Mn^{4+} state and the MnO_6 octahedrons have a regular shape. In the intermediate situation, the distorted and regular octahedrons are distributed randomly over the volume, the long-range order is violated, and diffuse scattering arises, the analysis of which permits us to obtain information on this local disorder. In this case, from an analysis of Bragg peaks, information can only be obtained on the structure averaged over the volume.

Mathematically, the PDF technique (see a detailed presentation in monograph [65]) is based on the Fourier transform of the total intensity of elastically scattered neutrons, $S(\mathbf{Q})$, where $\mathbf{Q}$ is the momentum transfer, $Q = 4\pi \sin\theta/\lambda = 2\pi/d$, which permits the pair correlation function to be obtained:

$$G(\mathbf{r}) = \frac{2}{\pi} \int \mathbf{Q}\big(S(\mathbf{Q}) - 1\big) \sin(\mathbf{Qr})\, \mathrm{d}\mathbf{Q}\,, \tag{3}$$

where the assumption is made that $S(\mathbf{Q})$ involves refinements taking into account the background and other methodical factors. The polycrystalline sample is also assumed to scatter neutrons isotropically, which permits reducing the three-dimensional Fourier transform $S(\mathbf{Q})$ in $G(\mathbf{r})$ to a one-dimensional integral over the $\mathbf{Q}$ modulus. In an ideal version, the integral in formula (3) should be taken within the limits from zero to infinity, but experimental restrictions both on the lowest and on the highest momentum transfers actually exist. These restrictions ('cut-off errors') lead to broadening of the peak-like distribution of interatomic distances, $G(\mathbf{r})$, and to the appearance of a modulated background.

From the formula for Q it follows that for its enhancement it is necessary to measure the spectrum at a scattering angle close to 180° and to use neutrons with short wavelengths. The application practice of the PDF technique has revealed that the effects in crystals, interesting from the point of view of local long-range order violations, are observed if $Q_{max} \gtrsim 25$ Å^{-1}. This means that a diffraction pattern must be measured for interplane distances down to $d \approx 0.2$ Å. Such small interplane distances can be measured with acceptable statistics and resolution only at short-pulse SNS neutron sources; accordingly, the PDF technique is one of the few whose implementation is practically impossible at steady state neutron sources.

Origination of the necessary experimental technology at neutron and synchrotron sources, as well as development of computational methods have already permitted the application of PDF-analysis for resolving many interesting problems related to the existence of a local structural disorder. Thus, for instance, numerous studies have been carried out on high-temperature superconductors and manganites with colossal magnetoresistance (CMR), in which local structural violations often play an important part in the formation of their physical properties (see monograph [65]).

7.6 Investigation into the microstructure of crystalline materials

From the point of view of diffraction, the microstructure of crystalline materials is conventionally considered to involve various kinds of structure imperfections violating long-range order. The microstructure of single crystals includes, in particular, the mosaicity value of single crystals, the characteristic size of coherently scattering blocks, the existence of twins, and defects of various kinds. In the case of polycrystals, besides the above, the crystallographic texture is added together with various internal strains, leading to diffraction peaks undergoing broadening and shifts. Sometimes, determination of microstructural characteristics only accompanies the analysis of atomic structure, and in other cases its investigation is a necessary stage preceding structural analysis. Most often, microstructure investigation is considered to be a so-called applied research, a typical example of which is the determination, with the aid of diffraction, of internal mechanical strains in bulky materials, products, or machine components, usually of interest to engineers in connection with the mechanical strength and serviceability of materials.

Diffraction methods of microstructural analysis can be divided, in the first approximation, into two types: the analysis of certain geometric characteristics of diffraction peaks (positions, widths), and the analysis of the actual peak profiles. Both approaches are realized in two versions:

— individual peaks are identified in the diffraction pattern, then their independent analysis is carried out at a level of widths or profiles;

— analysis is performed simultaneously of the entire diffraction pattern measured within quite a broad range of momentum transfers (interplane distances).

The classic Williamson–Hall approach is usually adopted in the first case, while in the second the Rietveld method [66] is applied together with a method that originated quite recently and was termed WPPM (Whole Powder Pattern Modeling) [67]. Examples of how the characteristic sizes of coherently scattering domains and microstrains in crystallites are determined with the aid of the Williamson–Hall method are considered in Section 7.2. In WPPM, contrary to the Rietveld method, the profiles of diffraction peaks are formed on the basis of the physical microstructure model of the crystalline medium, which, for instance, permits direct analysis of parameters responsible for the distribution of crystallite sizes. There have hitherto been few examples of the application of the WPPM method to analyze neutron diffraction patterns, but, apparently, precisely this method should be considered the most promising one for obtaining information on the microstructure of crystalline materials.

The WPPM method is based on a well-known property of Fourier transforms, namely, that if a function $\rho(x)$ is a convolution of two other functions $\rho_1(x)$ and $\rho_2(x)$, then its Fourier image is a product of the Fourier images of functions ρ_1 and ρ_2. Expressing the cross section $\sigma(Q)$ of neutron scattering by a crystal as the convolution of functions related to various kinds of effects distorting the spectrum profile, for example, as $\sigma(Q) = \delta(Q) \otimes f_S(Q) \otimes f_D(Q)$, where $\otimes$ denotes the convolution operation of functions, and $\delta(Q)$ is the scattering cross section on a crystal without defects, subscripts S and D indicate the distribution functions related to the description of the size and deformation effects, we obtain

$$F(\sigma) = F(\delta)\, F(f_S)\, F(f_D) \tag{4}$$

(here, F symbolically denotes the Fourier transform).

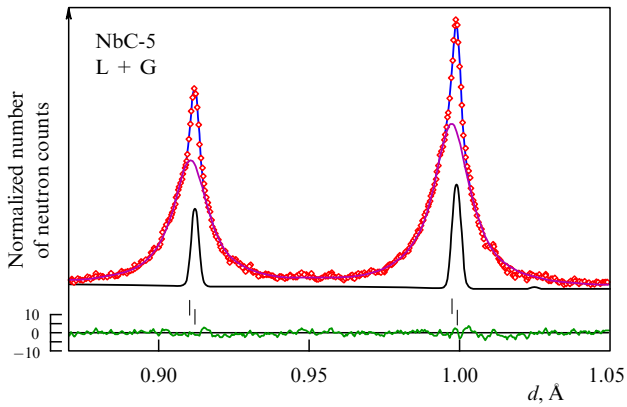

Figure 14. Peaks (422) and (420) from NbC-5 (5 h of milling) and their description by the sum of a Lorentz (L) and a Gaussian (G) functions, the positions of which are somewhat different. Shown are experimental points, the resulting curve, and contributions of the components (Lorentzian + Gaussian). The lower part of the figure shows the difference curve normalized to the error at an individual point. The vertical bars indicate the positions of components. The width of the Lorentzian is about four times broader than the width of the Gaussian, and the position of the Lorentzian is shifted toward smaller d. The χ^2 factor for this segment of the pattern equals 1.71. Attempts to describe the diffraction pattern by a pseudo-Voigt function (a linear combination of Lorentz and Gauss functions with the components having identical positions) results in $\chi^2 = 5.31$.

Relationship (4) is readily generalized by including factors related to the resolution function of the diffractometer, to the existence of antiphase domain boundaries, to variations in the elemental composition over the volume of the sample, etc. The distribution functions $f_S(Q)$, $f_D(Q)$ and others are calculated on the basis of the assumed microstructure model, then their Fourier transforms are computed and analysis is performed of experimental data on the basis of equation (4). The listed algorithms are realized in the code package PM2K [68].

Recent paper [69] presents an example of the application of the WPPM method to analyzing data obtained with high-resolution neutron diffractometers. In this work, four sets were studied of niobium carbide, $NbC_{0.93}$, powders with crystallites of different average sizes obtained by high-energy grinding of the initial large-grain powder for different milling times: NbC-n, where $n = 0$ (initial powder), 1, 5, 10, and 15 is the milling time in hours. The neutron diffraction patterns were measured with the TOF HRFD (IBR-2) diffractometer and the λ_0-diffractometer HRPT (PSI). Preliminarily, the experimental data applying the Rietveld and Williamson–Hall methods were processed. The results of this processing revealed that the ground powders contain two fractions, F1 and F2, with unit cells having several different parameters and essentially different peak widths of these fractions (Fig. 14). Moreover, the plotted dependences of $(\Delta d)^2$ on d^2 revealed a strong anisotropy in the peak widths, i.e., their dependence upon the concrete set of Miller indices, which did not permit obtaining reliable estimates of the size of coherent scattering domains applying the Williamson–Hall method.

Applying the WPPM method for processing the neutron diffraction patterns from NbC-10, measured with the aid of

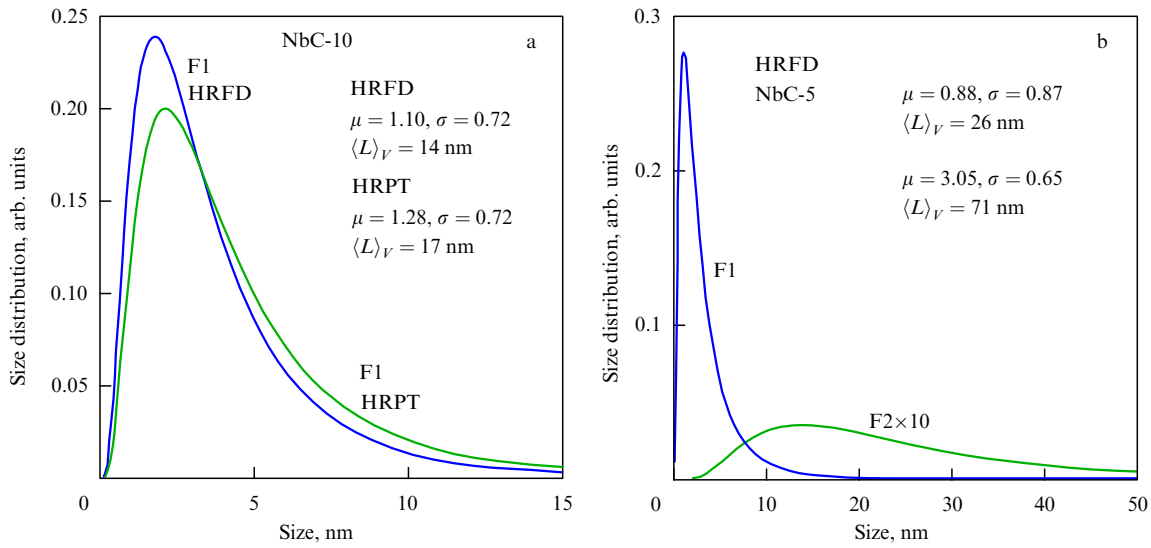

Figure 15. (a) Log-normal size distributions of the main crystallite phase for NbC-10, calculated from diffraction data obtained with HRFD and HRPT. The respective diffraction dimensions are 14 and 17 nm. (b) Log-normal size distributions of crystallites for F1 and F2 fractions in NbC-5, calculated from diffraction data obtained with HRFD. The parameters of distributions correspond to diffraction sizes of 26 and 71 nm.

HRFD and HRPT, yielded very close results for the distribution of crystallites over sizes, defined in the computer program as a log-normal distribution:

$$g(D) = \frac{1}{D\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\ln D - \mu}{\sigma}\right)^2\right], \tag{5}$$

where D is the crystallite size, μ and σ are fitted parameters, and the crystallite size averaged over the sample volume, which corresponds to the size determined from the widths of diffraction peaks by the Williamson–Hall method, was calculated by the formula

$$\langle L \rangle_V = \frac{3}{4} \exp\left(\mu + \frac{7\sigma^2}{2}\right). \tag{6}$$

The calculated size distributions are shown in Fig. 15a, from which the shapes of the distributions, as well as the average crystallite sizes (14 and 17 nm), are seen to be in quite a good agreement. From Fig. 15b, it is seen how strongly the size distributions of fractions F1 and F2 in the NbC-5 powder differ from each other.

The complex analysis of diffraction data presented has permitted us to reveal important details of the microstructure of niobium carbide powders that form when they are milled, and to obtain objective numerical characteristics for it. For their interpretation, high-energy grinding was assumed to lead to an inhomogeneous state of the resulting powder. The main part of the powder is milled effectively, but a certain part of it turns out to be difficult to grind. Here, a certain amount of carbon is removed from the main part, which leads to a decrease in the parameters of the unit cell.

8. Advanced neutron sources

The main points and tendencies in the development of neutron sources for research in extracted beams, expounded in our review [2], are still quite relevant. Therefore, we shall only note here developments that took place during the past two decades and that affect the possibilities of research today and in the future.

The type of device (stationary or pulsed) generating neutrons does not depend on the type of nuclear reaction. As a rule, nuclear reactors (fission) are stationary, but there do exist pulsed devices, too. Sources based on accelerators (photonuclear or 'spallation' reaction) are pulsed, as a rule, but there are also sources with a quasicontinuous flux. As we already mentioned, pulsed neutron beams for TOF diffractometry can also be produced at a stationary source. Therefore, we shall adhere in our presentation to the conventional division of sources in accordance with the method of neutron generation: steady-flux and pulsed.

8.1 Steady state sources

Most of the reactors producing continuous neutron fluxes (steady state reactors) were constructed between 1955 and 1960. These were first-generation reactors intended for producing isotopes, for irradiation runs, and for radiation research. After 1960, the construction of reactors of the second generation started, which were already intended precisely for radiation research and for research with neutron beams. The first third-generation reactor with a continuous neutron flux, i.e., a dedicated reactor for beam research, was constructed in 1965 in Brookhaven (closed down in 1996) five years after the construction of the IBR pulsed reactor in Dubna, which from the very beginning was intended only for beam research.

Until about 1960, the neutron flux increased together with the increase in reactor power, then further enhancement of the neutron flux started to leave the increase in reactor power behind. This advance became especially apparent at the beginning of the 1970s, when effective application started of third-generation reactors, such as HFR at the Institut Laue–Langevin (ILL) (Grenoble, France), Orphée at the Leon Brillouin Laboratory (LLB) (Saclay, France), WWP-M (water–water modernized reactor) at the B P Konstantinov Petersburg Institute of Nuclear Research of RAS (PINP, RAS) (Gatchina), and the research reactor IR-8 in the National Research Centre 'Kurchatov Institute' (NRC KI) (Moscow), JINR IBR-2 (Dubna), and others.

At present, about 30 research reactors used for beam studies are in operation around the world. Most of these reactors have been in operation for over 40 years, which is close to their natural lifetime, i.e., most reactors require modernization or replacement by new ones.

Ultimately, the operation efficiency of a research reactor is determined by the work of the experimental facilities (stations). Table 2 shows the characteristics of steady state reactors of the third and partly of the second generations, used for studies on extracted beams and mentioned most often in the literature. Experimental stations are conventionally divided into five classes: diffractometers for elastic Bragg and elastic diffuse scattering, devices for small-angle scattering, reflectometers, spectrometers for inelastic scattering, and other devices. The last ones include everything that is not involved in the first four categories. Facilities for irradiation and activation analysis are not considered here.

From Table 2 collating various neutron sources, HFRs can be seen as the most effective, from all points of view, among high-power reactors, while the FRM II is the most effective among intermediate-power reactors. HFRs exhibit the largest absolute and specific (reduced to power) neutron fluxes and the most developed infrastructures for implementing experiments. The FRM II construction is, apparently, optimal for intermediate-power reactors.

Besides technical parameters, cost is a quite significant parameter. To a great extent, the cost of a reactor depends on its average power; therefore, the ratio of average power to flux density can be considered a conventional characteristic determining the production cost of a neutron. Another factor is the number of experimental devices at a reactor; the higher it is, the higher the operation efficiency of the reactor; in other words, the lower the production cost of a single neutron. From Table 2, HFR and FRM II are seen to surpass all existing reactors.

From the point of view of further development aimed at obtaining higher fluxes of extracted neutrons, nuclear reactors are subject to restrictions due to technological reasons mainly related to heat removal. Thus, the HFR in Grenoble or the PIK reactor in Gatchina [70] have reached the limit for steady state reactors in the extracted neutron flux: $(4-5) \times 10^{15}$ cm^{-2} s^{-1}.

On the whole, in spite of the means for enhancing the application efficiency of reactors — the use of new types of moderators, the formation of beams with the aid of neutron guides, the installation of new detector systems — being quite developed, the number of advanced reactors in the world serving as neutron sources is clearly insufficient for physical research.

In order to achieve higher fluxes, projects for sources based on proton accelerators (spallation sources) and producing continuous neutron fluxes were proposed at the beginning of the 1960s. Here, the proton beam can be pulsed, but the pulse frequency is so high that the flux of neutrons traversing the moderator is practically continuous.

Such a source was made in Switzerland, where the isochronous cyclotron of the Paul Scherrer Institute (Villigen) with a proton current of 1.5 mA and energy of 600 MeV was utilized. The SINQ neutron source, projected to be similar to advanced reactors, has been in operation since the end of 1996 and provides a neutron flux of 10^{14} cm^{-2} s^{-1}, which is consistent with the fluxes from most advanced steady state reactors (see Table 2).

8.2 Pulsed sources

The history of pulsed neutron sources originates in 1945, at the time of the Manhattan Project, within the framework of which a self-quenching, or aperiodic, pulsed nuclear reactor was constructed at Los Alamos. Such superpowerful pulsed reactors, which were constructed for special purposes, are not used for physics research, but the idea underwent further development. In 1955, in the city of Obninsk at the Institute for Physics and Power Engineering (IPPE), work started under the leadership of D I Blokhintsev on the development of a fundamentally new pulsed reactor of periodic action — of a pulsating reactor. Previously, pulsed neutron fluxes for nuclear spectroscopy were achieved with the aid of beam choppers (Fermi choppers [11]) at steady state reactors. The reactor application efficiency, which was already not so high, was drastically reduced. A pulsating reactor could resolve this problem.

The IBR pulsating reactor, the construction of which started in Dubna in 1957, was commissioned in 1960. This was the first reactor in the world in which pulses were generated periodically with frequencies of 5 and 50 Hz owing to rotation of a part of the core. For an average reactor power of only 1 kW, the neutron flux was higher than at steady state reactors with a power of 10 MW and with a chopper.

Successful operation of the IBR reactor and its modifications stimulated further development in this direction. Several new projects were proposed in the middle of the 1960s. Of all the proposals for new high-flux pulsating reactors, only the project of the IBR-2 reactor was realized, which became possible owing to the experience of working with such systems in Dubna and Obninsk. The essential differences between IBR-2 and the series of the first IBR reactors were the modulation of the core reactivity and cooling of the core with liquid sodium.

The pulsating IBR-2 reactor commissioned in 1984 had the highest pulsed flux of thermal neutrons in the world: 10^{16} cm^{-2} s^{-1}. Moreover, it is an extremely economical installation — the operating time of the reactor core is about 20 years. A specific feature of the IBR-2 reactor is its relatively long duration of the neutron pulse, exceeding 300 μs for thermal neutrons.

The first pulsating source at an accelerator was constructed at the beginning of the 1950s in Harwell using a linear electron accelerator; here, in 1959, the idea was proposed and realized of a booster: the accelerator-multiplying target system. The multiplying target is that of heavy metal placed inside a subcritical assembly. Photonuclear neutrons initiate a chain reaction in the assembly, which leads to an increase in the neutron flux by a factor of 10–30. The chain reaction in the subcritical assembly takes place only when the accelerator is in operation, which is the main difference between such a system and a reactor. The IBR-30 booster was in operation in Dubna until 2006. At present, it is being replaced by a new photonuclear source IREN (Russ. abbr. for the source of resonance neutrons) (without multiplying), which is at the development stage.

Although construction of linear electron accelerators is relatively simple, at present they are not employed much (owing to their low efficiency compared with the efficiency of proton accelerators) and mainly for nuclear physics studies. Boosters have not become widespread either. The main problem is that society is against any systems containing fissile materials. However, the development logic of neutron

Table 2. Steady-flux neutron sources for scattering experiments mentioned most often in the literature.

Reactor *	Site	Commissioned/ modernized, year	Power, MW	Neutron flux, 10^{14} cm^{-2} s^{-1}	Neutron channels (horizontal)	Moderators (C — cold, H — hot)	Devices for neutron scattering					
							Diffraction	Small-angle	Reflector	Inelastic	Others	Total number
OPAL	Australia, Sydney	2007	20	2	10	1 C	4	3	1	4	1	13
WWR	Hungary, Budapest	1959/1993	10	2.1	8	1 C	2	3	3	3	3	14
BER II	Germany, Berlin	1973/1991	10	1.2	10	1 C	10	12	1	3	5	16
FRM II	Germany, Münich	2005	20	8	20	1 C, 1 H	6	4	3	8	2	23 + 7
Dhruva	India, Bombay	1985	100	2	13	1 C	3	1	1	8	1	14
NRU	Canada, Chalk River	1957	120	3	6	1 C	3	0	1	2	0	6
CARR	China, Beijing	2010	60	8	13	1 C	5	2	1	3	5	16
CMRR	China, Mianyang	2012	20	2.4	8	1 C	3	2	1	2	3	11
HFR	Netherlands, Petten	1961/1970	45	1–24	12	—	3	1	0	2	1	7
PIK	Russia, Gatchina	2019, planned	100	45	16	2 C, 2 H	5	2	2	—	3	up to 50
HFIR	USA, Oak Ridge	1966	85	30	4	—	4	1	0	5	0	10
NBSR-NIST	USA, Geithersburg	1969/1985	20	4	24	1 C	2	4	3	7	8	24
HFR ILL	France, Grenoble	1975/1995	58	15	26	2 C, 1 H	14	4	4	4	2	38
Orphée	France, Saclay	1980	14	3	20	2 C, 1 H	11	4	2	5	3	25
LVR-15	Czech Republic, Řež	1957/1965	15	1	6	—	1	1	1	1	1	6
SINQ	Switzerland, Villigen	1996	1	1	14	1 C, 1 H	5	3	3	5	6	22
JRR-3M	Japan, Tokai	1991	20	2	26	1 C	4	1	0	10	5	33

* Abbreviations stand for: OPAL — Open Pool Australian Light-water reactor, WWR — Water-Water Reactor, BER II — Berlin Experimental Reactor II, FRM II — Forschungsreaktor München II, NRU — National Research Universal, CARR — China Advanced Research Reactor, CMRR — China Mianyang Research Reactor, HFR — High Flux Reactor, PIK — Beam research vessel reactor, HFIR — High Flux Isotope Reactor, NBSR-NIST — Neutron Beam Split-core Reactor at the National Institute of Standards and Technology, HFR ILL — High Flux Reactor at ILL, LVR — Light water research reactor (from Czech Lehkovodni výzkumný reaktor), and JRR-3M — Japan Research Reactor 3M. The meanings of the remaining abbreviations are provided above in the text.

sources will apparently lead to widespread application of, namely, boosters.

Proton accelerators for pulsed neutron sources started to be used at the beginning of the 1970s. In 1973 and 1975, prototypes of ZING (Zero gradient synchrotron Intense Neutron Generator) and ZING-P′ were constructed at the Argonne National Laboratory and, subsequently, the IPNS (Intense Pulsed Neutron Source) device was also in operation there from 1981 up to 2008. Similar installations were made in Los Alamos in 1977, where the powerful source LANSCE (Los Alamos Neutron Science Center), recently given the new name LNSC (Luan Neutron Scattering Center), has been in operation since 1985. In 1980, the KENS source was commissioned (it was in operation till 2009) at the High Energy Accelerator Research Organization (KEK) in Japan. All these first-generation neutron sources were constructed at accelerators intended for nuclear physics studies.

The first pulsed second-generation proton source of neutrons, for which the main accelerator was especially constructed, ISIS, was put into operation in 1985 at the Rutherford–Appleton Laboratory (United Kingdom). At present, ISIS is an intense pulsed neutron source that is best equipped and adapted for usage. In 2006 and 2009, proton sources of neutrons were commissioned in the USA (SNS) and Japan (J-SNS), respectively. They are the most powerful and most intense (during the pulse) third-generation neutron sources, with which only IBR-2 can compete. Doubtless, precisely such sources have the best prospects for advancement.

The proton sources of neutrons listed above pertain to the class of pulsed sources with a neutron pulse of small duration (< 50 μs) (Short Pulse Source — SPS). Recently, the issue of creating sources with a long (> 300 μs) pulse (Long Pulse Source — LPS) has been quite actively discussed. The construction of proton storage rings, providing energies of several GeV required for enhancing the neutron flux, is quite expensive. It is much cheaper to construct a powerful linear proton accelerator, but the duration of the neutron pulse in this case increases. The pulse length determines the resolution of the experimental device: in the standard approach, the shorter the pulse, the better the resolution. However, development of the experimental technique at the first LPS — the IBR-2 reactor — reveals that in the case of a long pulse the application of modern electronics and mathematical software makes possible the formation of neutron pulses of the necessary duration, which permits obtaining a resolution at the level of the resolution of the best SPS both for elastic and inelastic scatterings. But the neutron flux increases here by an order of magnitude. This experience is used in constructing LPSs at proton accelerators. A source of this type started operating in 1999 at the linear accelerator of the Moscow meson factory.

At present, construction is under way in Sweden (Lund) of the most powerful LPS-proton source of neutrons — the European Spallation Source (ESS) of the next generation exhibiting a power of 5 MW, i.e., exceeding the power of ISIS by 10 times.

Table 3 presents high-intensity pulsed neutron sources used in studies of condensed media and having received international acknowledgment.

ESS will be the neutron source of highest intensity. Its intensity averaged over time (the neutron flux density) will be comparable to the intensity of HFR at ILL.

From the point of view of implementation of scattering experiments, the main characteristic of a source is the average neutron flux which determines not only the rate of performing experiments, but also the possibilities: of improving the measurement accuracy; of studying objects of small size, complex objects, and objects with small scattering cross sections, and of performing experiments involving analysis of the neutron polarization before and after scattering. As is seen from comparing Tables 2 and 3, according to this characteristic, existing pulsed neutron sources are inferior to steady state reactors.

However, as revealed by the analysis of conditions in which scattering experiments are implemented at steady-flux and pulsed sources, the flux at a pulsed source averaged over time is equivalent to the peak flux in the case of perfectly constructed devices requiring the neutron beam to be monochromatized. This means that even at existing pulsed sources the conditions for performing experiments may be better than at a steady state reactor.

8.3 European development strategy

In Table 4, ten neutron sources are presented that have been recognized by the European Neutron Scattering Association (ENSA) to be the most promising ones. The user system for these sources is the best developed, and they have produced the main scientific results.

In principle, decisions have been taken concerning decommissioning of two of these ten sources: LLB in 2019, and BER II in 2020. The issue of closing down the reactor at ILL is under discussion. The analysis carried out by ENSA shows that in Europe the number of days neutron sources are available to users will drop from 32,000 to 20,000 in the period between 2007 and 2027. At present, the main contributions are due (in decreasing order) to HFR-ILL, FRM II, Orphée, ISIS, BER-II, and SINQ. However, the demand for neutrons does not fall, signifying a rising need for such sources as ESS and PIK.

We are most interested in the stand of Germany as a strategic partner in creating the International Neutron Center on the basis of the high-flux PIK reactor [70] under construction at PINP NRC KI in Gatchina (Leningrad region).

The neutron strategy of the Germany is based on decommissioning old reactors, in spite of their being well equipped and successful, replacing them by a reactor, albeit a single one, but advanced with new experimental stations and participating in advanced European projects. This strategy, consistently pursued for the past 20 years, is aimed at regular technical reequipment, which results in constant technological development. Such a strategy adopted in all kinds of scientific results in it being integrated at a high level into the economy of a country.

At the beginning of the 2000s, the FRM I reactor in Garching (Germany), which had been in operation since 1957, was decommissioned. At the same site, operation of the reactor FRM II started in 2005, which at present is a well-equipped neutron center, a European leader and the only reactor in the Germany that will remain intact until the 2040s. In 2006, the FRJ II reactor at Jülich was closed down. Neutron research was transferred to Garching, where in 2006 at the FRM II reactor inauguration took place of the Jülich Neutron Research Center, which constructed over half of the neutron stations and for which separate dedicated housing was constructed. At present, an advanced neutron

Table 3. Pulsed neutron sources for scattering experiments that have received international acknowledgment.

Source, year of commissioning/ modernization	Country, site	Power of target, mW	Pulsed neutron flux, 10^{14} cm^{-2} s^{-1}	Pulse length of thermal neutrons, μs; frequency, s^{-1}	Neutron channels	Cold moderators	Stations for neutron scattering					
							Diffraction	Small-angle	Reflector	Inelastic	Other	Total number
Short-pulse sources												
ISIS I, 1985 ISIS II, 2009	United Kingdom, Chilton	0.2	10 45	20–30; 50 20–30; 5	16 13	2 1	10 6	2 4	3 5	7 2	1 2	21 20
MLNSC,* 1985 SNS, 2006	USA, Los Alamos, Oak Ridge	0.1 1	7 100	20–30; 20 20–30; 60	16 14	2 1	4 7	2 2	3 2	2 7	2 3	13 21
JSNS,** 2009	Japan, Ibaraki	1	100	20–30; 25	21	1	7	1	2	3	7	20
CSNS,*** 2018, planned	China, Dongguan	0.1	50	20–30; 25	20							20
Long-pulse sources												
IBR-2, 1984/2012	Russia, Dubna	2	200	360; 5	14	2	6	1	3	2	2	14
ESS, 2019, planned	Sweden, Lund	5	400	2000								20 after 2025

* Manuel Lujan Neutron Scattering Center
** Japan Spallation Neutron Source
*** China Spallation Neutron Source

Table 4. Promising neutron sources in European development strategy.

Source	Commissioned, year	Thermal energy, MW	Average neutron flux, $cm^{-2}\ s^{-1}$	Peak neutron flux, $cm^{-2}\ s^{-1}$	Operation, number of days per year	Number of stations	Possible number of stations	Number of users per year	Operating costs, 10^6 euros
FRM II, Münich	2005	20	8×10^{14}		240	23 in operation, 7 under construction	35	1000	55
BER II, Berlin	1991	10	1.2×10^{14}		220	16 in operation	20	400	25
ILL, Grenoble	1975/1995	58	1.3×10^{15}		200	27 + 10 CRG*	> 40	1400	80 + CRG
ESS, Lund	2019, planned	5, LP		4×10^{16}	200	20 after 2025	> 20		103
PIK, Gatchina	2019, planned	100	5×10^{15}		200	22 after 2022	> 40		30
LLB, Saclay	1985	14	3×10^{14}		200	22	25	600	25
SINQ, Villigen	1996	1	1.5×10^{14}		200	15	20	600	30
ISIS/ ISIS-II, Abingdon	1985/2009	0.2, SP		4.5×10^{15}	180	34	41	1500	55
IBR-2, Dubna	1984/2012	2, LP		2×10^{16}	108	14	14	200	1
WWR, Budapest	1959/1993	10	2.1×10^{14}		140	14	14	100	10

* CRG — abbr. for Collaborative Research Group instruments.

center (the Maier–Leibnitz Center), constructed by München Technical University and the Jülich center, is functioning at FRM II.

In Jülich, HBS (High Brilliance neutron Source), a compact neutron source based on a linear accelerator, is to be made for methodical research and for training the students. The absence of such university neutron sources is one of the reasons that the spread of neutron, as compared unlike synchrotron, methods are weak. There are very many photon, in particular, X-ray sources, which contribute considerably to the formation of a broad community of users of synchrotron radiation sources.

In 2010, the FRG I reactor in Geesthacht (Hamburg), which had been in operation since 1958, was closed down; neutron research was transferred to other sources (including the PIK reactor) and synchrotron research started to develop actively at external sources. Finally, the BER 2 reactor at the Helmholz scientific center in Berlin is to be closed in 2020. Although this neutron center is equipped perfectly and new experimental stations are being constructed, neutron research will be terminated there and it will be fully reoriented toward the synchrotron radiation source BESSY[3] II.

Germany is one of the founders (together with France) of the European Center of Neutron Research ILL, which has played an important part in the development of neutron research in Germany. It has also played an extremely important role for Russia during the period of its participation in ILL as a scientific member from 1997 up to 2007. The HFR ILL reactor will be in operation tentatively until 2023; therefore, Germany is actively participating in the construction of two new sources: ESS and PIK. Determining factors are the record high neutron fluxes and the possibility of developing international cooperation. The latter factor plays an important role not only owing to the high cost of work at megadevices, but also and even more owing to the ever increasing necessity of cooperation in science.

The ESS and PIK sources may become very good partners in the future. These two sources are absolutely mutually complementary. The neutron community expects experimental stations, systems for data processing, and systems for surrounding samples to be constructed in accordance with modern trends in the development of science. The PIK reactor occupies its own niche as a steady-flux source. Since it can essentially replace HFR, its experimental stations must already belong to the next generation. The entire path of neutrons from reactor to sample must be equipped for effective operation of the reactor: the best moderators, neutron guides, and other optical elements must be installed to reduce the background and to enhance the resolution, and the use of polarized neutrons must be extensive. The modern development of detector technologies and electronics permits significant expansion of the acquisition and improvement in the processing of experimental data.

The European community has accumulated significant experience in international cooperation, including preparation of the substantiation and development of the concept of the ESS device base, and is prepared to share it with Russia in creating the International Neutron Research Center on the base of the PIK reactor.

8.4 Neutron sources in Russia

The situation in Russia seems quite modest against the background of world development. No new neutron sources

[3] From the German: Berliner Elektronenspeicherring-Gesellschaft für Synchronstrahlung — Berlin electron-storage-ring community for synchrotron radiation.

Table 5. Characteristics of neutron sources in Russia for studies with extracted beams (characteristics of the IBR-2 reactor were presented in Tables 3 and 4).

Device	Organization	Commissioned, year	Power, MW	Neutron flux, 10^{14} cm^{-2} s^{-1}	Number of stations
IR-8 reactor	NRC KI, Moscow	1957/1981/2012	2/5/8	1	4 + 5
WWR-M reactor	PINP NRC KI, Gatchina	1959/1978 Prolonged shutdown since 2016	5/18	4.5	12
WWR-Ts reactor	Branch of RIPC, Obninsk	1964	13	1	3
IWW-2M reactor	IRM, Zarechnyi	1966/1983	15	2	5
IRT-T reactor	RI TPI, Tomsk	1967/1977	6	1.2	—
IRT reactor	NRU MEPhI, Moscow	1967/1975 Prolonged shutdown since 2013	2.5	0.3	4
GNEIS (pulsed) $\Delta t_0 = 10$ ns	PINP NRC KI, Gatchina	1973/1983	3×10^{-3}	1	3
IN-06 sources (pulsed) $\Delta t_0 = 100-200$ μs	INR RAS, Troitsk	1999	3×10^{-1}	1	7 + 2
IREN (pulsed) $\Delta t_0 = 30$ ns	JINR, Dubna	2010	4×10^{-3}	0.1	3
PIK reactor	PINP NRC KI, Gatchina	2019, planned	100	45	22 after 2022

for research with extracted beams have been constructed during the past 30 years. The only exception inside Russian territory is a sole advanced source — the pulsed IBR-2 reactor in the international intergovernmental organization, the Joint Institute for Nuclear Research (Dubna) with a flux of neutrons from the surface of the moderator equal to 2×10^{16} cm^{-2} s^{-1}.

There are several low- and intermediate-flux neutron sources at which matter studies are performed with extracted beams with various levels of activity. These are steady-flux reactors at NRC KI (Moscow), PINP NRC KI (Gatchina, Leningrad region), the Institute of Reactor Materials (IRM, Zarechny, Sverdlovsk region), the National Research Tomsk Polytechnical University (NR TPU), the National Nuclear Research University Moscow Engineering and Physics Institute (NNRU MEPhI), and the Obninsk Branch of the L Ya Karpov Research Institute of Physical Chemistry (RIPC, Obninsk), as well as the pulsed sources GNEIS (Gatchina Neutron Spectrometer) based on the proton synchrocyclotron (PINP NRC KI), IN-06 (thermal neutrons), and RADEX (*Rad*iation *ex*periment) (epithermal and thermal neutrons) based on the proton linear accelerator at the RAS Institute of Nuclear Research (INR RAS, Troitsk, Moscow region). Two pulsed sources, IBR-2 and IREN, are in operation at JINR. In Table 5, comparative characteristics are presented for neutron sources in Russia. The IBR-2 reactor was presented in Tables 3 and 4.

We shall consider in greater details sources at which studies are carried out with extracted neutron beams.

The IR-8 (NRC KI) reactor was commissioned in 1981 after reconstruction of the IRT reactor (Russian abbreviation for Standard Research Reactor) constructed in 1957. The design power was 8 MW; there are 12 horizontal and 18 vertical channels. Most of the stations are in a stage of reconstruction. Three diffractometers for structural research and five stations for nuclear physics research are in operation condition. A project exists for reconstruction and building of the neutron-guide hall. The irradiation base of vertical channels is actively applied, which permits carrying out a large volume of irradiation runs of construction materials, implementing studies of fuel elements for the water–water energy reactor, as well as research for the development of methods and technologies for producing radioisotopes for medical purposes.

At NRC KI, the concept was developed in 2007 for the construction of a Specialized neutron center based on the IR-8 reactor. In 2007–2012, upgrading was performed of certain systems of the reactor, and the reactor design power of 8 MW was achieved. The development and construction is foreseen concerning the source of cold neutrons, the supermirror neutron guides (a project and proposal exist for implementing the design and construction at PINP NRC KI), and of three experimental stations: a small-angle scattering station and a diffractometer for investigation of microsamples at high pressures (a project and proposal exist for implementing the design and construction at JINR, Dubna), and a reflectometer of polarized neutrons (a project and proposal exist for implementing the design and construction at PINP NRC KI, Gatchina).

Construction of a cold neutron source and a neutron-guide chamber is planned to be carried out in 2015–2017.

Construction of the *WWR-M reactor* (PINP NRC KI) was completed by the end of 1959 in accordance with a standard design prepared at the Institute of Atomic Energy (Moscow) to yield a power of 2 MW. The design was immediately revised by PINP physicists to obtain a power of 10 MW. The reactor was commissioned in 1960 with a power of 5 MW, and in 1961 with a power of 10 MW. Upgrading to a

power of 18 MW took place gradually and was completed in 1967.

The reactor is used for research in condensed matter physics, radiative studies of materials, research in radiobiology, nuclear physics, and adjacent lines of scientific research.

The reactor is equipped with 15 dedicated devices for studies of nanosystems and materials.

For economic reasons, starting in 2016 the reactor will be in a state of prolonged shutdown. The research program will be formed with account of the possibilities provided by the IBR-2 and IR-8 reactors.

The *WWR-Ts reactor* (Obninsk branch of RIPC) possesses 22 vertical and 9 horizontal channels, of which five are used. The vertical channels are actively used for nuclear doping, radiation-assisted material modification, and obtaining protons; a neutron-therapy complex is under construction for oncological patients. Three diffractometers were installed at the horizontal channels, which at present are not being used.

RWW-2M reactor (research water–water nuclear reactor) (Institute of Reactor Materials, ROSATOM). A neutron center for materials science possessing four diffractometers and one small-angle device is in operation on the basis the RWW-2M reactor equipped with a complex of five experimental stations constructed by physicists of the Institute of Metal Physics of the RAS Ural Branch (IMP UrB RAS).

The reactor is used for conducting research within a broad range of problems of radiative materials science, radiation physics, condensed matter physics, magnetism, and superconductivity. Unique possibilities of realizing low-temperature neutron irradiation permit performing advanced research into the influence of radiative disordering on the structural characteristics and physical–mechanical properties of construction materials — steels, alloys, etc. — applied in reactor engineering.

The RWW-2M reactor is the only one in Russia at which neutron scattering methods are applied in studies of highly radioactive materials, including functional materials utilized in industry.

The *IRT-T reactor* (NI TPU) is well-equipped for studies in radiation physics, producing isotopes, and activation analysis. It is used for preparing specialists in the field of reactors. There are no devices for extracted beam scattering.

The *IRT reactor* (NRU MEPhI). There are four devices in horizontal channels. Since 2013, it has been in a state of prolonged shutdown.

GNEIS is the Gatchina neutron time-of-flight spectrometer (PINP NRC KI), based on the SC-1000 proton synchrocyclotron. The accelerator was commissioned in 1971. GNEIS is a spallation pulsed neutron source made in 1973. The proton energy is 1 GeV, the internal beam current is less than 3 μA, the neutron target is a $40 \times 20 \times 5$ cm lead block cooled by water, there is a polyethylene moderator, the pulse length ≈ 10 ns, the pulse repetition rate is up to 50 Hz, and the mean fast neutron intensity is about 3×10^{14} s^{-1}. Five channels depart from the target (one 'looks' at the target, the others are oriented toward the moderator).

The scientific program is based on high-resolution time-of-flight spectrometry in neutron nuclear physics and in the physics of fission. Moreover, it has the only neutron beam available in Russia with an atmospheric spectrometer for testing aircraft electronics.

The *proton accelerator of INR RAS* (*IN-06 and RADEX sources*) can be considered the basis for constructing a future powerful spallation source (see Section 8.3). At present, there are seven devices at extracted beams. The operation parameters of the accelerator (proton energy of 209 MeV, mean current of 50 μA, and neutron complex operation time of less than 300 h per year) do not permit effective application of these devices yet.

Upgrading of the proton strong-current linear accelerator by partially replacing the accelerating structure with superconducting resonators, raising the final proton energy to ≈ 1 GeV, and substituting a liquid-metal neutron source IN-06 for the target could allow its parameters to approach those of neutron facilities in the USA, Japan, and Europe.

The *IBR-2 reactor* (JINR), commissioned in 1984, exhibits, after undergoing modernization between 2006 and 2012, the highest pulsed neutron flux in the world achievable at research neutron sources (2×10^{16} cm^{-2} s^{-1}), and is the only advanced Russian neutron source of a world class for carrying out research with extracted beams.

The modernization of IBR-2 actually resulted in a new reactor being constructed with a doubled thermal neutron flux, a longer-lived resource of the movable reflector (by 2.5 times), sleeve-type fuel elements with enhanced admissible burnup depth, an upgraded emergency protection system, and a new advanced moderator facility (thermal and cryogenic neutrons). The new complex of cold moderators at IBR-2 permits, in a number of experiments, us to make use of neutrons from extracted beams 20–30 times more effectively. This, in turn, opens possibilities for creating a powerful spectrometric base for performing research into condensed matter physics. The average cold neutron flux density on the surface of each of the cryogenic moderators amounts to at least 9×10^{12} cm^{-2} s^{-1} (about the same as provided by the cold moderator of the 'second' target at the ISIS source, which is publicized as record). All the above will permit the new IBR-2 reactor to maintain a leading position among world neutron sources until 2037.

Scientists from Russia and other countries carry out over 200 experiments per year at IBR-2. The user system has been organized according to the European standard. At present, 14 world-class spectrometers functioning at IBR-2 permit formulated problems to be resolved efficiently.

The experience acquired in the operation of IBR-2 reveals IBR-2 to be quite an effective neutron source, which in most areas of application is not inferior to the best sources based on proton accelerators. This experience is especially important at present, when rising interest is observed in pulsed neutron sources with long pulses.

The operation period of the upgraded IBR-2 reactor has been established up to 2032–2037, but already today versions are considered for its prolongation. We believe the project of a pulsed booster based on a linear proton accelerator to be quite promising. The construction of an accelerator with the required parameters — a proton energy of about 1 GeV, an average beam current of 1 mA, a pulse length on the order of 100 μs, an accelerator length up to 200 m — is at present a resolvable problem. As the multiplying target it is possible to employ a device such as IBR-2 installed in the existing building. Such a target will permit enhancing a neutron flux by 10–100 times compared with that of the flux at ESS.

The source of resonance neutrons (*IREN*) (JINR) is a new-generation base installation intended to resolve a broad range

of problems in fundamental and applied nuclear physics. The IREN device is intended for research in nuclear physics with the use of the time-of-flight method in the region of neutron energies up to several hundred keV required for studies of photonuclear reactions.

The full-scale IREN scientific research facility involves a linear accelerator of electrons with energies up to 200 MeV, with a beam power of 10 kW, a deeply subcritical multiplying target, a beam infrastructure with pavilions for measurements, as well as technological, control, safety, and supply systems. The characteristics of the full-scale IREN facility bring this device to the same level as the best neutron sources of this class (GELINA (Geel Electron LINear Accelerator) (Belgium) and ORELA (Oak Ridge Electron Linear Accelerator) (USA)).

The PIK reactor complex. The PIK research reactor is under construction in PINP NRC KI (Gatchina). Its construction and physical and technical characteristics make it unsurpassed in reactor engineering for experimental purposes, and upon being commissioned it will become a unique base for scientific research. A description of the work required and of the plan for completing construction of the PIK reactor are presented in Ref. [70].

The PIK reactor with a power of 100 MW and thermal neutron flux of 5×10^{15} cm^{-2} s^{-1} together with its experimental devices may become an international center of neutron research in Russia, the basis of which will be the high scientific professional level of the PINP NRC KI team and the existing experience in organizing the work of neutron beam users. There are more than 40 places for experimental beam-used devices in the reactor chambers.

9. Conclusions

During the past 50 years, neutron diffraction at pulsed neutron sources has covered an impressive path of development, initiated by the work of F L Shapiro and his pupils and colleagues. The average source power increased drastically from several kilowatts to several megawatts. The techniques of neutron beam production and the formation of detector systems also changed radically. Effective methods were developed for processing the diffraction data. The research area has expanded many-fold, and at present it includes all the lines of inquiry developed with λ_0-diffractometers, as well as certain additional ones.

As to the situation in the Frank Laboratory of Neutron Physics of JINR, where the history of TOF diffractometers began, it reflects the world situation. Half of the 14 neutron spectrometers belonging to the facility constructed at the IBR-2 reactor are diffractometers. Of the scientific problems listed in Table 1, only research on local structural distortions is not under development. Some diffractometers (HRFD, DN-6, RTD (Real-Time Diffractometer), CKAT (Russian abbreviation of Spectrometer for numerical Analysis of Texture)) exhibit characteristics at the level of the best ones in the world. Detailed information on all spectrometers presently in operation at IBR-2 can be found at the FLNP JINR site (http://www.flnp.jinr.ru). Their further upgrading will be related to improvement of the existing experimental technique: of neutron optical devices, detectors, and registration systems. The development of Fourier diffractometry, real-time neutron scattering, and diffraction on microsamples at high pressures have especially good prospects.

The total number of TOF diffractometers in the world is relatively small, and this is consistent with only five advanced pulsed sources being in operation at present; nevertheless, the research performed with their aid is quite visible in the general flow of neutron diffraction studies. This is primarily due to some of their specific capabilities, noted in Section 3, that permit implementing experiments which are difficult or even impossible to perform with λ_0-diffractometers. The most important ones have the possibility of nearly continuous three-dimensional scanning of reciprocal space and of operation at very short wavelengths. The latter is related to peculiarities of the neutron spectral distribution at proton sources of neutrons, in which there are large numbers of epithermal neutrons. For example, while at steady state reactors with thermal neutron beams the minimum neutron wavelength is ≈ 0.9 Å, and it can be reduced to ≈ 0.4 Å in the presence of a 'hot' source, at proton sources the admissible neutron wavelengths are ≈ 0.2 Å, i.e., momentum transfers up to $Q \approx 60$ Å^{-1} are quite accessible. Another important circumstance lies in the weak dependence of the resolution function of a TOF diffractometer on the interplane distance, which is very helpful in the analysis of small parameter variations and of a unit cell symmetry in the case of structural phase transitions.

The main characteristics of TOF diffractometers — resolution, luminosity, range of interplane distances — are not inferior to the respective characteristics of the best λ_0-diffractometers. Like steady state and pulsed sources, neutron diffractometers of two types should be considered complementary rather than alternative. Essentially different experimental conditions result in investigations of one and the same object with λ_0- and TOF diffractometers yielding somewhat different information. The main reasons for the differences, as a rule, are the differences in intervals of momentum transfers, different effect-to-background ratios within different segments of the spectrum, differences in refinements to the intensity introduced upon transition to structural factors, etc. For example, a diffractometer at a steady state source usually provides a greater precision in structural data: bond lengths, valence angles, values of magnetic moments, etc. However, accurate structure determination at a local level is only possible with a TOF diffractometer manifesting a large range of momentum transfers.

Modern practice has revealed that a routine experiment can be performed with approximately the same success at a source of any type. But precise or nonstandard data can be obtained only in the case of an adequate choice of the type of source and diffractometer. In special cases, moreover, it is quite expedient to perform experiments using diffractometers of both types.

Prospects of enhancing the luminosity of TOF diffractometers are mainly related to improvements in the technique of forming and guiding neutron beams and to increasing the solid angles of detector systems. Both lines of inquiry have recently been under radical development, and in both cases there is room for further progress. For example, the installation of a new supermirror neutron guide at the HRPD (ISIS) diffractometer has permitted achieving a gain of 5–10 times in the neutron flux within the main range of wavelengths.

A similar or even greater gain can be achieved owing to the use of advanced large-format scintillation detectors covering areas of several square meters. While quite recently the solid

angles of detectors for powder diffractometers were several tenths of a steradian, now they already amount to several steradians, i.e., enhancement factors of 10–20 in this parameter are also quite achievable.

The possibilities of improving the resolution of TOF spectrometers are somewhat lower. Plans exist for increasing the flight path to 200 m at the ESS source under construction, but this is related to the cost of the device significantly rising. The correlation technique is more promising. Simple estimates reveal that an increase in the rotation speed of the Fourier chopper at HRFD to 11 thousand rotations per minute and of the flight path between the Fourier chopper and the detector to 30 m will result in the time contribution to the resolution function amounting to 0.0002 at $d = 2$ Å. The geometric contribution in this case can be made approximately the same, i.e., $\Delta d/d \approx 0.0003$ at $d = 2$ Å, which is close to the resolution of powder diffractometers at sources of synchrotron radiation.

Thus, the application of correlation methods can ensure the next step (after HRFD at the IBR-2 reactor) toward enhancement of the resolution in neutron diffractometry in the case of a neutron flux density on the sample, which is not achievable with steady state sources.

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