The first studies of condensed media using a pulsed neutron source

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The first studies of condensed media using the IBR pulsed reactor at the Laboratory of Neutron Physics, JINR are reviewed. The foundations of the method of studying condensed media at pulsed neutron reactors were laid under the direction of F. L. Shapiro. The first experimental facilities are described, and the various research areas which have benefitted from the use of a pulsed neutron source and the time-of-flight method are discussed. © *1995 American Institute of Physics.*

1. INTRODUCTION

In the late 1950s at reactor centers around the world a new nuclear-physics method of studying condensed media became popular: low-energy neutron scattering. Neutron diffraction and inelastic scattering is valuable, owing to the following characteristics of the neutron, which make it a unique probe for studying the properties of condensed media:

1. The wavelength of a thermal neutron is comparable to the interatomic separations in condensed media, so that thermal neutrons undergo clearly expressed interference effects in scattering on the atoms of such systems. In addition, the neutron energy is lower than that usually required to knock an atom out of matter, so that the samples are not damaged in the experiment.

2. The change of the energy of a thermal neutron in an inelastic scattering process associated with the creation and absorption of excitations in solids and liquids is often of the same order of magnitude as the initial neutron energy. It is therefore relatively easy to measure it.

3. The strength of the interaction between a neutron and a nucleus varies irregularly with atomic number, so that information about light atoms can usually be obtained just as easily as information about heavy ones. In addition, since the strength of the neutron–nucleus interaction for different isotopes of the same element is different, it is sometimes possible to change the scattering properties of the sample by changing the isotopic content.

4. The neutron possesses a magnetic moment and therefore undergoes magnetic scattering on atoms which possess an atomic magnetic moment. This scattering process, which usually can be completely separated from nuclear scattering, gives information about the structural and dynamical properties of magnetic systems.

5. The neutron does not possess an electric charge and can deeply penetrate matter, which makes it possible to obtain information about the bulk properties of the sample and not only about the surface properties, as usually is the case in electron and x-ray scattering.

The new method of studying the properties of condensed media quickly became widespread largely because at the start of this research physicists at continuously operating reactors used the experimental technique and experience gained from working with x rays. For example, the first studies of neutron diffraction begun in 1945 at the Oak Ridge National Laboratory in the USA were carried out using a biaxial diffractometer whose second axis was a standard device usually used in x-ray spectroscopy. Most of the pioneering studies of Wollan and Shull were carried out using this setup.[1] The same is also true of the first experiments on inelastic neutron scattering performed by Brockhouse[2] in Chalk River, Canada, using a triaxial crystal spectrometer.

The first pulsed reactor IBR-1 was designed to solve a comparatively narrow range of problems, primarily, to determine neutron cross sections and study nuclear states.[3] The foresight, intuition, and talent of F. L. Shapiro are therefore all the more remarkable, as he initiated and directly participated in neutron research in condensed-matter physics at the Laboratory of Neutron Physics which began practically at the same time that the IBR-1 reactor came on-line in 1960. Here the problem was considerably more complicated methodologically, since at that time there was no experience at all in using pulsed neutron sources for such research, and direct comparison of the average power of the IBR reactor (1 kW) with that of a typical stationary reactor (10–20 MW) suggested that the pulsed reactor had no hope of competing in this area. Owing to the essential talent and enthusiasm of F. L. Shapiro, in 2–3 years a nearly complete arsenal of techniques for neutron studies of condensed media was created with the participation of his colleagues at the Laboratory of Neutron Physics. Already in May of 1964 at the Third United Nations International Conference on the Peaceful Uses of Atomic Energy,[4] it was reported that one of the main areas of research at the IBR-1 pulsed neutron reactor was the study of liquids and solids by means of neutron scattering. The use of a pulsed neutron source in such studies greatly simplified the design of an experiment, since it was no longer necessary to use a monochromator in the chopper of the incident neutron beam. Of course, the duration of a thermal neutron pulse turns out to be quite large; it is determined by the average lifetime of the neutron in the moderator, which is 200 μsec for an "unpoisoned" thick water moderator. Consequently, to obtain good resolution it is necessary to use large flight paths of order 10 m and above. It was emphasized that the IBR, as shown by the experiments, offers excellent possibilities for the development of solid-state research. At that time five installations were used in these studies: one for measurements using cold neutrons, one for studying liquids and solids by the "reverse-geometry" method, one for diffraction measurements, one for measuring

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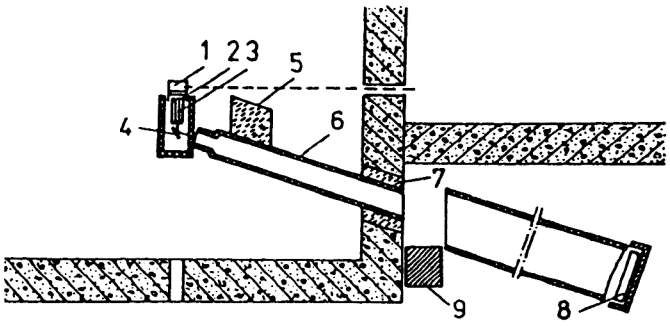

FIG. 1. Schematic view of a setup for measurements using cold neutrons. (1) Active zone of the reactor; (2) moderator; (3) beryllium filter; (4) sample; (5) and (7) shielding; (6) vacuum neutron pipe; (8) detector; (9) slide valve.

doubly differential scattering cross sections, and one for measuring the phonon spectra of crystals.

The successes of the studies performed at the IBR-1 reactor in condensed-matter physics were convincing. They led to the appearance of several projects using pulsed-neutron sources. At the International Conference on the Perspectives of Neutron Spectroscopy (October 1970) organized by the JINR with the support of the International Atomic Energy Agency, the reports of foreign scientists stressed the fact that interest in pulsed neutron sources had been stimulated by the series of pioneering experiments first performed at Dubna using the unique IBR reactor.[5] The competition between pulsed and stationary neutron sources for condensed-matter physics led to understanding of their differences and advantages. The experience gained in using the IBR-1 showed that pulsed sources were promising for further progress in condensed-matter physics and led to the creation of the IBR-2, still the most powerful pulsed reactor, and pulsed neutron sources based on proton accelerators.

We shall begin this review of the research in which use of a pulsed neutron source has proved fruitful by listing the results obtained using the first spectrometer built at the IBR-1 for research in condensed-matter physics: a setup for studying the quasielastic scattering of cold neutrons.

2. THE SETUP FOR STUDYING COLD-NEUTRON SCATTERING

The construction of this setup was begun in 1960 concurrently with the initial reactor studies, and the work was mainly carried out by V. V. Golikov and A. Shkatula under the direction of F. L. Shapiro. Experiments using cold neutrons were begun right after the IBR reactor came on-line at the end of 1960 (Ref. 6).

The design of the setup is shown schematically in Fig. 1. The IBR reactor is located at the center of a room of dimensions 10×10 m, whose walls serve as the reactor shielding. The active zone of the reactor together with the reflector and controls had dimensions of about 25×25 cm. The reactor produced neutrons in periodic pulses with a half-width of 36 μsec at a repetition rate of 8.3 times per second. The average heat output of the reactor was 1 kW.

A key feature of the setup is the cold-neutron source. According to the original design, fast neutrons from the reactor were moderated in a layer of paraffin 5 cm thick, located 5 cm from the reflector of the reactor. This version of the moderator is not the optimal way of obtaining an intense beam of cold neutrons at a pulsed source. Two new ideas were used to construct the cold-neutron source. First, a block of polycrystalline beryllium of length 24 cm acting as a cold-neutron filter was placed right behind the paraffin moderator. The placement of the beryllium right next to the moderator increased the cold-neutron flux by a factor of 3.5 compared to the case where the beryllium was located at some distance from the moderator. This intensity gain was achieved because the beryllium acted as a reflector of thermal and fast neutrons. Cold neutrons, to which the beryllium is transparent, are extracted from inside the moderator–beryllium combination, where the density of thermal and, accordingly, cold neutrons is considerably higher than at the edge of the bare moderator when the beryllium is some distance from it. In later years this experimental fact served as the basis for a series of studies on optimization of moderators for pulsed-neutron sources. Studies were made on the choice of materials for the moderator and its temperature, on the optimization of the shape and size, and on the use of reflectors. Since these studies were performed at other IBR setups, we shall describe the results later on.

Second, the moderator and the beryllium were initially placed in a foam plastic cryostat and cooled to liquid-nitrogen temperatures. The 6-cm thick walls of the cryostat were glued with epoxy. Cold neutrons were extracted from the cryostat via a window consisting of 15 layers of 10-μm thick aluminum foil with a gap of 5 mm between layers. This construction kept the window from sweating. The cryostat was filled with liquid nitrogen remotely from the experimental chamber; a single filling took 4 working hours. Cooling of the moderator by a factor of $\simeq 3.5$ increased the flux of cold neutrons (the cooling of the 24 cm of beryllium increased the transmission of cold neutrons by an additional factor of 3). In this way a version of a "cold moderator" was first realized at a pulsed source of cold neutrons.

The filtered beam of cold neutrons with average energy $\simeq 0.0035$ eV and a sharp boundary at 0.0052 eV fell on the sample, located 25 cm from the beryllium. One of two samples could be inserted remotely into the beam of cold neutrons. The energy of neutrons scattered on the sample at an angle of $75°\pm 4°$ was measured from the time of traveling the "sample–detector" distance. The flight path from the sample to the detector could be varied from 10 to 45 m. Neutrons were detected by a scintillation detector based on the mixture $ZnS(Ag)+B_2^{10}O_3$ of area 2000 cm^2 (Ref. 7).

In general, for a fixed value of the energy transfer in a scattering event the time t for detecting the scattered neutron (measured from the center of the reactor pulse) has some spread Δt due mainly to the spread in the lifetime of the cold neutrons in the moderator τ and the width of their energy spectrum. However, as F. L. Shapiro noted, the presence of the sharp drop in the cold-neutron spectrum at $E=0.0052$ eV in some cases leads to experimental results which could be interpreted as coming from a monochromatic line at $E=0.0052$ eV. In particular, when studying small energy transfers (quasielastic scattering in liquids), the energy trans-

fer and its spread can be determined from the location of the leading front of the spectrum of neutrons filtered by the beryllium and the change of its slope owing to the spread in the energy transfer. In this case the resolution of the setup is of order $\Delta t/t \approx \tau/t$. The spectrum of neutrons incident on the sample was measured in this setup using elastic scattering on vanadium. The time half-width of the slope of the beryllium edge was ~ 120 μsec, which for a flight path of 45 m corresponds to an energy resolution of 3×10^{-5} eV for neutrons with an energy of about 5.2×10^{-3} eV.

At the suggestion of F. L. Shapiro, the first object of research was the quasielastic scattering of cold neutrons on water. This was stimulated by the appearance a short time before of a study by the Brookhaven group[8] with the sensational conclusion that there is a completely elastic component in the scattering of cold neutrons on water. This result was extremely interesting to physicists studying the dynamics of molecules in liquids, and a large number of studies devoted to the scattering spectra of cold neutrons in water were performed at various laboratories around the world.[9,10] In analyzing the quasielastic scattering of cold neutrons on water, various authors arrived at inconsistent conclusions: values of the self-diffusion coefficient D of water molecules in the range from $D<0.1D_0$ (Ref. 11) to $D=D_0$ (Refs. 12 and 13) were obtained, where D_0 is the coefficient of self-diffusion measured by classical methods such as the nuclear paramagnetic resonance (at room temperature $D_0=1.85\times10^{-5}$ cm^{2}sec^{-1}). The quasielastic scattering of cold neutrons on water was a leading topic at the three IAEA Symposia on neutron inelastic scattering at Vienna,[9] Chalk River,[10] and Bombay.[14]

In order to resolve the contradictions mentioned above, a group at Dubna set out to study quasielastic scattering in water[15] with a resolution considerably better than that used in earlier studies. As mentioned above, for a flight path of 45 m we had an energy resolution of 3×10^{-5} eV for neutrons with an energy of about 5.2×10^{-3} eV. This value can be compared with the maximum resolution of 2×10^{-4} eV used in the earlier studies.[12]

Comparison of the spectra of neutrons scattered by a layer of water at room temperature and by vanadium showed that the 5.2×10^{-3}-eV line is certainly broadened in scattering on water. To obtain a quantitative estimate it is necessary to start from the expression for the dependence of the counting rate $N(t)$ of scattered neutrons on the time of flight t:

$$N(t)dt=dt\frac{dE}{dt}\int_0^{E_0}F(E')\sigma(E')W(E',E)dE', \qquad (1)$$

where $F(E')dE'$ is the spectrum of the flux incident on the sample, $\sigma(E')$ is the cross section for scattering on the sample, and $W(E',E)dE'$ is the spectrum of scattered neutrons for primary energy E'.

In the theory of quasielastic scattering[16,17] $W(E',E)$ has a Lorentz shape with width at half-max Γ. In the model of continuous diffusion[16]

$$\Gamma=2h\kappa^2D, \qquad (2)$$

where $h\kappa$ is the change of momentum in the scattering.

In the model of jump-like diffusion[17] the quasielastic peak also has a Lorentz shape under the condition that the jump time τ_1 is much smaller than the time for the oscillatory motion τ_0. In this case, if the average squared displacement in time τ_1 is much larger than the average squared deviation of a molecule from its equilibrium position, then

$$\Gamma=\frac{2h}{\tau_0}\left(1-\frac{e^{-2W}}{1+\kappa^2D\tau_0}\right), \qquad (3)$$

where W is the Debye–Waller factor.

When the incident neutron spectrum has a step shape [$F(E')=0$ for $E'>E_0$, $F(E')dE'=E'dE'$ for $E'\leq E_0$], quasielastic scattering leads to smearing of the spectrum boundary. The quantity Γ can be determined by using the distance from the center of the beryllium edge for vanadium to the maximum of the distribution for water. This distance has a complicated dependence on Γ and on the form of $F(E')$ and $\sigma(E')$. Since the latter were not studied, this method was shown by F. L. Shapiro to be ambiguous, and at his suggestion the authors of Ref. 15 chose to use the value of the logarithmic derivative $N'(t)/N(t)$ at the center of the beryllium edge. The value of the derivative depends weakly on the form of $F(E')\sigma(E')$; setting $F(E')\sigma(E')\simeq E'$, the following expression was obtained as a good approximation for Γ:

$$\Gamma=\frac{8E_0}{\pi}\frac{1}{t_0/\Delta t+5}. \qquad (4)$$

Here E_0 and t_0 are the energy and time of flight for the beryllium edge and Δt is the half-width of the boundary on the time-of-flight scale. Taking into account the instrumental resolution, Δt was calculated from the expression

$$\Delta t=\sqrt{(\Delta t_1)^2-(\Delta t_2)^2},$$

where Δt_1 and Δt_2 are, respectively, the half-widths for the liquid and for vanadium.

The value $\Gamma=(5\pm1)\times10^{-4}$ eV obtained for water at room temperature at a scattering angle of 75° ($\kappa^2=3.7A^{-2}$) corresponded to the value $D=0.5D_0$.

In later work[18] the quasielastic scattering of cold neutrons was studied for other hydrogen-containing liquids differing from each other in the nature of the intermolecular interaction. An improvement of the setup[19] made it possible to extend the measurements to large energy transfers (inelastic scattering) and to perform them at various temperatures, including below the melting point. These studies gave information about the parameters and mechanism of molecular self-diffusion in liquids, both when intermolecular hydrogen bonds are formed (water, ethylene glycol, acetic acid) and when they are not formed (benzene, naphthalene, and dioxane).

Analysis of the results showed that the measured values of Γ are lower than the calculated ones, and the difference is especially large for liquids in which there are intermolecular hydrogen bonds (Fig. 2). The decrease of Γ in relation to the value given in (2) can be attributed to the jump-like diffusion mechanism. It suggests that jump-like diffusion is manifested more strongly in liquids with hydrogen bonds.

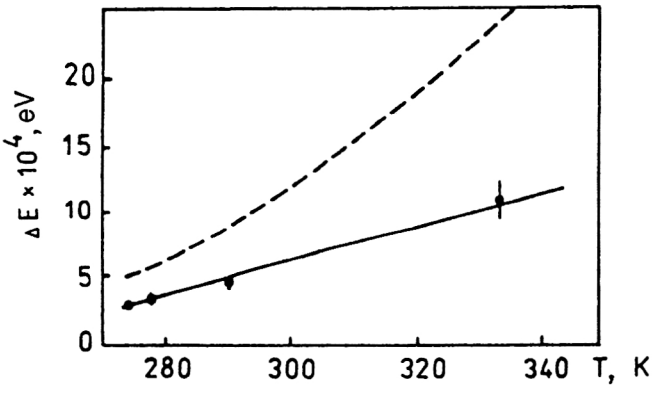

FIG. 2. Temperature dependence of the half-width of the quasielastic peak $\Gamma \equiv \Delta E$ for water. The solid line shows the experimental data, and the dashed line shows the calculation using the expression for continuous diffusion based on the experimental values of D.

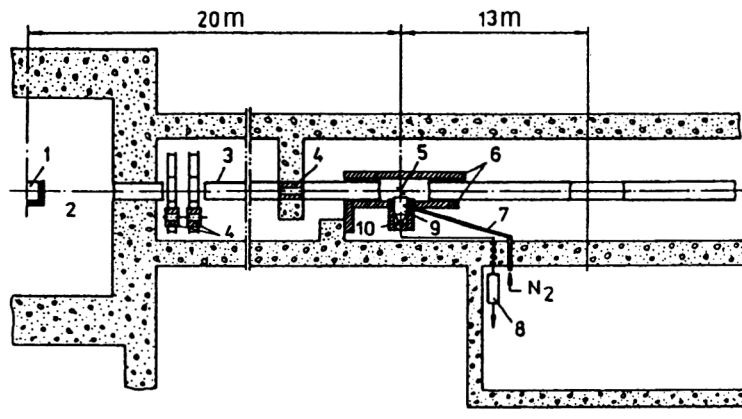

FIG. 3. Schematic design of a setup for studying liquids and solids by the reverse-geometry method. (1) Active zone of the reactor; (2) moderator; (3) vacuum neutron pipe; (4) collimators; (5) sample; (6) shielding; (7) nitrogen pipe; (8) detector electronics; (9) beryllium filter; (10) detector.

The setup described here was designed for measuring the elastic and quasielastic scattering of cold neutrons. For such measurements it was characterized by high resolution and high intensity. However, since the sample was located right by the active zone of the reactor, it was difficult to access. In addition, the scattered beam had to be extracted via a channel in the reactor shielding, so that it was not easy to change the scattering angle. It was therefore natural to adopt the method of "reverse geometry," which subsequently completely displaced the original technique.

3. THE TIME-OF-FLIGHT SPECTROMETER WITH A FILTER IN FRONT OF THE DETECTOR

In the reverse-geometry method the energy $\hbar\omega$ transferred by the neutrons to the sample in the scattering process, i.e., $\hbar\omega = E_0 - E'$, is measured. The energy E_0 of a neutron incident on the material under study is arbitrary and is determined according to the time of flight from the reactor to the detector. The neutron energy after scattering E' is determined by the transmission band of a filter located in front of the detector. In order to ensure a sufficiently long flight path, the sample is located at a distance of 20–30 m from the reactor. Therefore, it is not difficult for the experimentalist to change the experimental conditions (remove the samples, change their temperature, change the scattering angles, and so on). The method has the additional advantage that the cross section for energy transfer from the neutron to the atoms of the sample is measured. This cross section remains sizable even at low temperatures, in contrast to the cross section of the reverse process measured in the "direct geometry," which falls off as the sample is cooled.

At the suggestion of F. L. Shapiro, the reverse-geometry method was first used at the Laboratory of Neutron Physics in 1961 (Ref. 20). Here F. L. Shapiro together with T. A. Machekhina and Z. Ogzheval'skiĭ studied the possibility of using the resonance neutrons of a pulsed source to extract certain integrated characteristics of the frequency spectrum of a solid. For this purpose a filter (a gold foil) possessing an isolated narrow resonance with an energy of about 5 eV was placed in front of the detector. After two years F. L. Shapiro again returned to the reverse-geometry method, but now using thermal and cold neutrons. Under his direction, A. Bajorek, T. A. Machekhina, and K. Parlinski at the Laboratory of Neutron Physics created the first version of a time-of-flight spectrometer with a beryllium filter in front of the detector. It is shown schematically in Fig. 3 (Ref. 21). Slow neutrons leaving the moderator are scattered on the sample located 20–30 m from the active zone of the reactor. A cryostat with a beryllium filter cooled to liquid-nitrogen temperatures was placed next to the sample at a certain angle relative to the axis of the incident beam. The neutrons scattered on the sample were detected by a scintillation detector after passing through the filter. The cryostat together with the detector could be moved, allowing the scattering angle to be varied within the range 30–120°.

To select the best experimental conditions the authors of the study performed an extensive series of experiments comparing various moderators at the IBR reactor in terms of the average neutron lifetime and the neutron yield. The lifetime τ was determined from the slope of the leading front of the spectrum of neutrons filtered by a 16-cm thick beryllium layer. The shape of the beryllium boundary as a function of the time of flight T is described quite well by the expression

$$N(T) = \left(\frac{T_0}{T}\right)^5 - \exp\left(-\frac{T - T_0}{\tau}\right),$$

where T_0 is the time of flight corresponding to the start of the sharp drop of the beryllium boundary. Planar layers of water of thickness 36 and 56 mm and of ethyl alcohol of thickness 36 mm, in addition to more complicated configurations in the form of a hole moderator and a moderator with a beryllium reflector, were studied as sources of slow neutrons. The hole moderator was a continuous layer of alcohol of thickness 36 mm adjoining a 60-mm thick plexiglass block. Throughout the entire thickness of the plexiglass there were holes of diameter 5 mm which occupied ~50% of the area of the moderator. The moderator with a beryllium reflector was a planar layer of alcohol of thickness 36 mm next to an 80-mm thick block of beryllium.

The results of the measurements showed that the hole moderator and the moderator with a beryllium reflector cooled to liquid-nitrogen temperatures had the best characteristics as cold neutron sources, both in the neutron yield J and in the quality parameter J/t^2 (where t is the effective duration of the neutron pulse). Here the cooling to liquid-nitrogen temperatures raised the cold-neutron flux by a factor of 3 in both cases. For thermal neutrons the highest yield was obtained by using a hole moderator at room temperature, and the best in terms of the parameter J/t^2 was a water moderator of thickness 36 mm.

In the following years the idea of F. L. Shapiro about obtaining intense beams of slow neutrons at pulsed sources by extracting them from inside the moderator was developed by N. A. Gundorin and V. M. Nazarov.[22] That study describes a crested water moderator consisting of a continuous layer of water of thickness $\simeq 3$ cm. The height of the crest $h=10$ cm was chosen to ensure that the overall average thickness of the moderator in water was at least 8 cm. For such a water layer the maximum density of the neutron flux is obtained at a depth of 3 cm. Studies of the crested moderator showed that it is more efficient than the hole moderator and the moderator with a beryllium reflector. At present the crested moderator is widely used at the pulsed neutron sources of the Laboratory of Neutron Physics: the IBR-30 and the IBR-2 reactors.

The reverse-geometry spectrometer created with the direct participation of F. L. Shapiro possessed satisfactory characteristics regarding the resolution, the intensity, and the effect-to-background ratio in studying processes of elastic or quasielastic scattering of cold neutrons and the inelastic scattering of neutrons with energy transfer of up to 130 MeV. This spectrometer was developed further by a group of physicists from Poland under the direction of J. Janik and included the use of a single-crystal energy analyzer after the beryllium filter,[32] increase of the number of simultaneously operating detectors, and simultaneous recording of the diffraction spectra and the inelastic-scattering spectra.[33,34]

The main topic of study using this reverse-geometry spectrometer in the first years was the dynamics of the excitations of molecular crystals. The dynamics of excitations of double alloys, in particular, the effect of light impurity atoms on the oscillation spectrum, were also studied.[34]

Of the many experiments carried out using the reverse-geometry method, let us briefly discuss the work done by V. V. Golikov and Zh. A. Kozlov under the direction of F. L. Shapiro.[23] They studied the scattering of slow neutrons at small angles by liquid lead in order to investigate the neutron analog of Mandel'shtam–Brillouin light scattering at frequencies of order 10^{12} inaccessible by optical methods.

As is well known,[24] three lines are observed in the spectrum of Rayleigh scattering of light by liquids: an unshifted peak and the two components of the Mandel'shtam–Brillouin doublet symmetrically located on either side of the unshifted peak at a distance $\Delta\omega$ proportional to the speed of sound u:

$$\Delta\omega = \pm u|q|,$$

where q is the change of the photon momentum in the scattering. A similar spectral distribution of the intensity should also be observed in coherent neutron scattering if it occurs with sufficiently small momentum transfer κ.

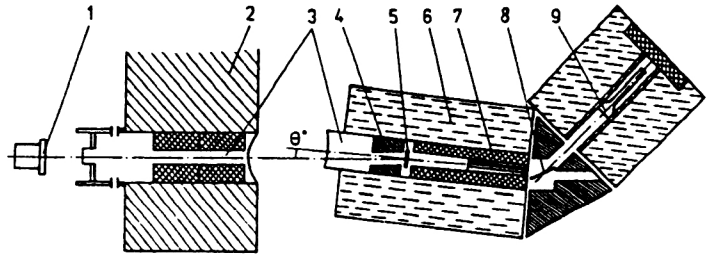

FIG. 4. Schematic view of the setup for small-angle neutron scattering. (1) Active zone with moderator; (2), (4), and (6) shielding; (3) vacuum neutron pipes; (5) sample; (7) Soller collimator; (8) crystal–monochromator (or beryllium filter with detector); (9) detector.

A setup for small-angle neutron scattering is shown schematically in Fig. 4. The energy of the neutrons incident on the sample was determined from the time of flight between the moderator and the sample (a distance of 10 m) and between the sample and the detector (2.4 m). Here the energy of the scattered neutrons recorded by the detector was in some experiments obtained using a crystalline monochromator, and in others using a cooled beryllium filter. The setup allowed the neutron scattering angle to be smoothly varied in the range from 3 to 90°. In order to study the effect of multiple scattering, a series of measurements was performed on a sample consisting of cells of liquid lead of dimensions $5\times5\times5$ mm^3, separated by barriers made of strongly absorbing material (cadmium covered with aluminum).

Analysis of the data obtained using liquid lead established the presence of two types of high-frequency collective excitation characterized by dispersion relations analogous to the dispersion laws of longitudinal and transverse oscillations in solid lead. The experimental results were compared with the polycrystalline model of a liquid and with the hydrodynamical model taking into account the frequency dependence of the kinetic coefficients, and their range of applicability was determined. The authors of the study concluded that for liquid lead the transition to the purely hydrodynamical description can be expected to occur at $\kappa \leqslant 0.1$ Å^{-1}.

4. THE TIME-OF-FLIGHT DIFFRACTOMETER

Before the appearance of pulsed neutron sources, the structural analysis of polycrystals consisted of measuring the angular distribution of monoenergetic neutrons scattered on the sample. The main reason why this method was so widespread was that it had been used for many years in x-ray diffraction studies. As we noted in the Introduction, the biaxial neutron crystal spectrometer is very similar to the spectrometer used in x-ray structural studies, and essentially the same expressions for the integrated intensity are valid for a quantitative analysis. However, with the appearance of pulsed neutron sources it became possible to view the time-of-flight method as an alternative. Already at the very beginning of the planning of the program of research at the pulsed

neutron reactor at the Laboratory of Neutron Physics it was obvious that there is no need for neutron monochromatization in diffraction at a given angle of reflection, because different neutrons satisfying the Bragg condition have different velocity and different times of flight.

The method of neutron diffraction using the time of flight was experimentally and theoretically justified by the work of F. L. Shapiro, V. V. Nitc, I. Sosnowska, J. Sosnowski, and a group of physicists from Warsaw under the direction of B. Buras.[25–29]

In the time-of-flight method, a pulsed neutron beam is scattered on a polycrystal and the neutrons scattered at a certain angle 2θ are recorded by a neutron detector. As a result, the dependence of the intensity on the wavelength is obtained. Peaks are identified in the usual way, but to obtain the structure factor from a neutron-diffraction pattern it is necessary to know the expression for the integrated intensity J. When thermal vibrations, absorption, and extinction are neglected, we have

$$J=\frac{i(\lambda)j(F_{hkl})^2\lambda^4}{4\sin\theta_0}\cdot\frac{\delta V}{V^2}\cot\theta_0\Delta\theta_0, \tag{5}$$

where $i(\lambda)$ is the neutron intensity per unit wavelength interval; j is the multiplicity factor for the plane (hkl); F_{hkl} is the structure factor; V and δV are the volumes of the unit cell and the crystal, respectively; θ_0 is the scattering angle and $\Delta\theta_0$ is the angular spread of the neutron beam. Equation (5) gives the integrated intensity over the entire Debye–Sherrer ring.

Taking into account the fact that certain quantities in (5) are constant (for each experiment), we easily find

$$j(F_{hkl})^2=\text{const}\,\frac{J}{i(\lambda)\lambda^4}, \tag{6}$$

from which it follows that to obtain the relative values of the structure factors from the neutron-diffraction patterns it is necessary to measure the areas of the peaks, normalize them to the neutron spectrum $i(\lambda)$, and divide them by λ^4. To avoid introducing corrections for the detector efficiency, the spectrum $i(\lambda)$ and the scattered beam should be measured by the same detector.

In Fig. 5 we show the design of one of the first experiments to understand the possibilities offered by a pulsed neutron source in structural neutron-diffraction studies. A beam of neutrons from a moderator passed through a collimator of the Soller type and scattered on a sample. The scattered beam passed through a second collimator, identical to the first and located at an angle of 90° to the direction of the direct beam, and was recorded by a detector. The flight path was 18.0 m.

Studies were carried out on Al, Zn, ZnO, and Si samples having different structures. As an example, in Fig. 6 we show the neutron-diffraction pattern of silicon. The experimental data were processed using Eq. (6). Good agreement between the theoretical values jF^2_{calc} and the experimental values jF^2_{exp} was obtained for all the samples. The reliability factor characterizing the accuracy of the results,

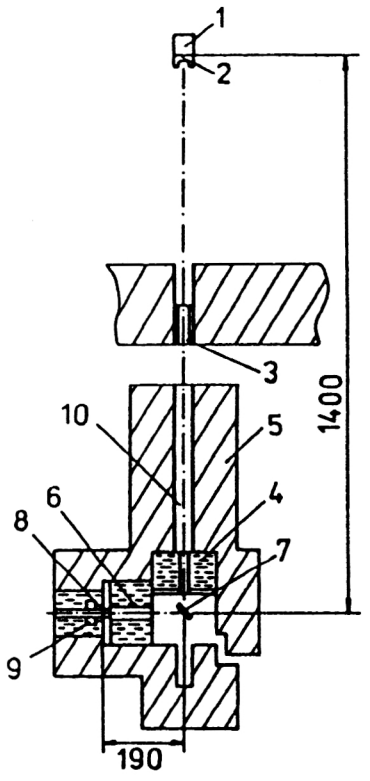

FIG. 5. Schematic diagram of the setup for diffraction measurements. (1) Active zone of the reactor; (2) moderator; (3) and (6) slit collimators; (4), (5), and (9) shielding; (7) sample; (8) detector; (10) vacuum neutron pipe.

$$R=\frac{\Sigma(jF^2_{\text{exp}}-jF^2_{\text{calc}})}{\Sigma jF^2_{\text{exp}}},$$

was about 2% in the case of silicon.

In the first studies the dependence of the resolution and the intensity on the shape and content of the moderator and also on the neutron scattering angle was studied. The following slow-neutron sources were used: a thick water moderator which was deeper in the middle, and also planar moderators (of thickness 40 mm) filled with pure water and water with

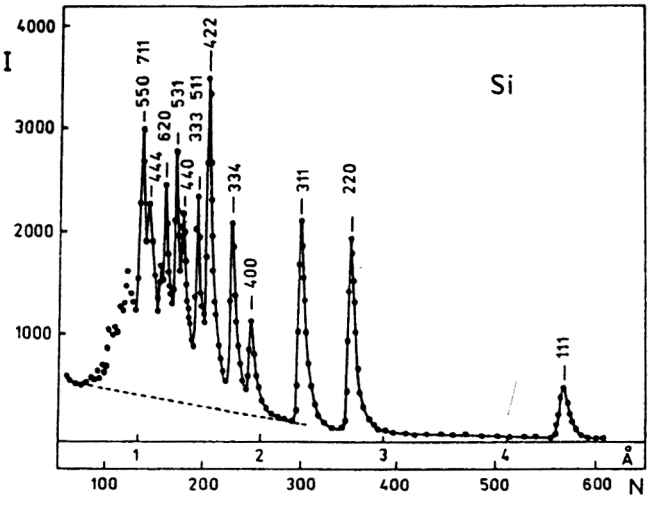

FIG. 6. Neutron-diffraction pattern of silicon for the angle $2\theta=90°$.

an admixture of 0.3 and 0.6% boric acid (by weight). The best resolution was obtained using a planar moderator filled with an aqueous solution of boric acid.

The time-of-flight method has a number of important advantages over the usual method. In the latter case the neutron wavelength used cannot significantly exceed 2 Å, owing to the admixture of higher-order reflections. In the time-of-flight method there are no limits on the scattering angle, so that neutrons of longer wavelength can be used. Because of the factor of λ^4 in (5) even the long-wavelength tail of the Maxwell spectrum can ensure sufficient intensity of the diffraction peaks. The possibility of using the long-wavelength part of the spectrum is particularly desirable when studying crystals with a large elementary cell, for example, in studying magnetic structures.

The time-of-flight technique possesses the advantage that all the results can be obtained directly at a given scattering angle. This is a great advantage when working with samples at high pressure, in which for reasons of stability only small windows can be left for the neutrons to pass through.

A drawback of the time-of-flight method is the appearance of an energy dependence of the neutron absorption cross section if the absorption is significant. In this case it is necessary to introduce corrections.

Among the subsequent methodological developments leading to the improvement of the time-of-flight method for structural studies, special note should be made of the work of A. Holas.[30] He was the first to show under what conditions focusing in the time of flight can be obtained in diffraction. It followed from the general expressions obtained by Holas that there was no need for strict collimation of the incident and scattered neutron beams. The Holas calculations have been verified experimentally. The absence of the need for strict collimation of the beam typical of the ordinary diffraction method makes the luminosity in the time-of-flight method very high.

In the first of their studies, the scientists who developed the time-of-flight method of neutron diffraction suggested that it could be used effectively for research beyond the classical neutron-diffraction studies. The time-of-flight method can be used in combination with a pulsed (electric or magnetic) field acting on a sample with frequency equal to that of the neutron-beam pulses. The effect of a field pulse simultaneous with neutron scattering can be used to study the change of structure induced by a very strong field, which is not possible in the stationary mode. Depending on the duration of the field pulse relative to the duration of the neutron pulse and on the time for the neutrons to travel to the sample, it is possible to observe the effect of the field on the entire spectrum, on several peaks, or even on a single diffraction peak.

By shifting the field pulse in time relative to the neutron pulse it is possible to study transient effects (relaxation processes). By using a high-intensity pulsed neutron source allowing a complete neutron-diffraction pattern to be obtained in a few minutes, it is possible to study processes with a relaxation time longer than several minutes.

The first study in this area[31] focused on the behavior of two diffraction peaks of neutron magnetic scattering in hematite placed in a large pulsed magnetic field (up to 120 kOe).

5. A SETUP FOR MEASURING DOUBLY DIFFERENTIAL SCATTERING CROSS SECTIONS

When studying neutron inelastic interactions it is useful to be able to expose matter to a monoenergetic neutron beam and to determine the entire spectrum of inelastically scattered neutrons for various scattering angles. A doubly differential slow-neutron spectrometer was built at the IBR for this purpose. The work was done by a group of physicists from the Physics and Power Institute in Obninsk under the direction of V. A. Parfenov.[35] The setup is based on a mechanical neutron-beam chopper rotating synchronously and in phase with the IBR disk and located at a distance of 10 m from it. By changing the phase of rotation of the chopper it is possible to change the energy of the neutrons passing through the chopper in the form of a pulse of a certain duration. Therefore, the selector acts as a monochromator. The spectrum formed as a result of the scattering of a monoenergetic neutron pulse on the sample is measured from the time of flight directly at eleven angles with flight path ranging from 5.5 to 10.5 m.

In the early years this setup was used to carry out extensive studies of the inelastic scattering of slow neutrons by zirconium hydride, water at various temperatures, and a series of organic compounds.

In the late 1960s at the suggestion of F. L. Shapiro, experiments were begun at this setup to study a fundamental object: the quantum liquid He II. The first stage of these studies was performed by Zh. A. Kozlov and V. A. Parfenov. The initial focus was the study of the quantum effect of Bose condensation in superfluid helium.[35] In these investigations, which made it possible to find the momentum distribution of the helium atoms experimentally, it was shown that there exists a Bose-condensation temperature which coincides with the temperature of the transition of liquid helium to the superfluid state. Below this temperature the number of helium atoms with zero momentum (the Bose condensate) is observed to grow as the temperature is lowered. The temperature dependence of the density of the Bose condensate coincides with the analogous dependence for the superfluid component of helium. In modern theoretical descriptions of superfluidity the Bose condensate is introduced *ab initio*. This causes the spectrum of single-particle excitations in liquid ^{4}He to acquire a complicated structure. In experiments performed recently at the Laboratory of Neutron Physics[36] it has been shown that in the transition to the superfluid state a new excitation branch observed only at a temperature below the λ transition is added to the already observed complicated structure of the excitation spectrum in He I. The intensity of this branch increases rapidly with decreasing temperature, and it is this branch which is related to the existence of the Bose condensate in superfluid ^{4}He.

6. CONCLUSION

The initiation and development of research on condensed-matter physics at the Laboratory of Neutron Physics at the JINR is inseparably associated with the name of F. L. Shapiro. The foundations of the methods of studying condensed media using pulsed neutron sources were actually laid in the 1960s under his direction. The first experience gained in the research in the early years of operation of the IBR showed that experimentalists had acquired a new effective neutron source which could be used to successfully solve various scientific problems.

As with any new technique, the first experiments amounted largely to the investigation of the experimental possibilities offered by this technique. At the same time, in several research areas in which the use of a pulsed neutron source and the time-of-flight technique proved fruitful, results were obtained which were either completely new or much more accurate than the data available earlier.

Excellent new possibilities for neutron studies in condensed-matter physics appeared after the powerful pulsed reactor IBR-2 was built. The IBR-2 not only extended the possibilities of existing areas of research, but also opened up new ones. At the present time at the Laboratory of Neutron Physics there are 12 time-of-flight spectrometers for studying the properties of condensed media by the methods of neutron diffraction and inelastic scattering. The scientific work performed at the Laboratory of Neutron Physics is in many cases closely related to the research interests of F. L. Shapiro or to his ideas.

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