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NEUTRON SCATTERING: HISTORY, PRESENT STATE AND PERSPECTIVES

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I. How it all began

In 1932, J.Chadwick, the student and co-worker of E.Rutherford, published the results of his experiments on irradiation of beryllium with α-particles [1]. He interpreted the induced emission from beryllium as being due to neutral particles with a mass close to that of the proton. Thus the neutron, the particle whose existence was predicted by E.Rutherford in 1920, was discovered. This was of great importance for the nuclear theory and in 1935 J.Chadwick was awarded the Nobel Prize in Physics.

Already in 1936, the first experiments on the Bragg reflection of slow neutrons from MgO single crystals [2] and Fe [3] were conducted. As before, these were carried out with a compact laboratory neutron source.

Nuclear reactors allowed obtaining of much higher neutron fluxes and in 1946, E.Fermi and W.H.Zinn [4] demonstrated that neutrons can be totally reflected from polished surfaces if the angle of neutron beam incidence is sufficiently small. The experiments have formed the basis for the development of neutron guides, the neutron devices that operate like optical fiber in the case of light and make it possible to transport neutrons over long distances with minimum losses as well as deviate the neutron beam. This was also a milestone in the development of neutron reflectometry, the method for the study of surfaces and interfaces.

In 1949, Shull and Smart published the result of their study of antiferromagnetic ordering in MnO [5]. Below the magnetic phase transition temperature, they observed some magnetic diffraction peaks at the positions that are not allowed from the viewpoint of the chemical unit cell. This was the first direct observation of the antiferromagnetic structure and at that time, it could not have been realized using other experimental techniques.

The fact that the neutron possesses the magnetic moment makes possible the existence of polarized neutron beams with all neutron spins arranged so that they are parallel to one another. The practical realisation of neutron beam polarization was reported in the literature in 1951. To polarize the beam, one group of physicists used total reflection of the neutron beam from the magnetic mirror [6] and the other employed diffraction on a Fe or Fe_3O_4 magnetized single crystal [7]. These have created the basis for modern experimental techniques using polarized neutron beams.

Last but not least, a pioneer experiment of neutron scattering by phonons in an aluminum single crystal [8] gave rise to broad and very intensive studies of elementary excitations by the method of coherent neutron scattering in condensed matter.

II. Present status of condensed matter research with neutrons

Soon after the first neutron scattering experiments it was realized that the neutron is a valuable tool for the study of different properties of condensed matter. One can find numerous facts in favor of that in the literature, e.g. [9]. The wavelength of thermal neutrons lies within the range of interatomic distances in condensed matter. This allows the investigation of the crystalline structure, atomic arrangement in glasses and liquids, shape and volume of polymer or biologic macromolecules, etc. Neutrons are scattered on nuclei and the interaction does not only depend on the type of the element in the periodic table but it also depends on the type of the isotope of the particular element and relative orientation of neutron and nuclear spins. In contrast to X-rays, there is no regular dependence of the neutron scattering amplitude on the atomic number of the element. This often facilitates locating of light atoms in the structure by neutron scattering, which is not the case for X-rays. An additional advantage is the possibility of using isotope substitution to «highlight» or «hide» some structural details of the studied material. The neutron has no charge and it weakly interacts with matter. This makes it possible to study the bulk properties of materials and permits the use of complex sample environments to create the necessary experimental conditions. Also, this is why the neutron is practically a non-destructive probe for investigations of objects, including delicate biological objects. Moreover, weak interaction allows one to consider the scattering in the first Born approximation and makes the data analysis rather straightforward.

The existence of the magnetic moment of the neutron opens a unique possibility for the study of very complicated magnetic structures and magnetic fluctuations. Also, the fact that thermal neutron energies are equal to typical energies of atomic motion creates the basis for probing of different dynamic properties of condensed matter.

Neutron scattering techniques were first applied to the study of different physical phenomena in condensed matter. This is still a lively activity but it is not the only one. At present, neutron scattering methods are applied in almost all fields of natural sciences: physics, chemistry, materials science, life sciences, engineering science and Earth science. The survey conducted by the European Neutron Scattering Association (ENSA) shows that the neutron scattering science is as diverse as the condensed matter itself. One can go further and assert that neutron scattering affects even our everyday life, although it is not obvious to most people yet [10]. It helps an understanding of how to design best artificial crystals to obtain advanced materials with the necessary properties ranging from high-T_c superconductors and computer chips to cookware and prescription drugs. Neutron scattering provides unique information about the microstructure of chemical products, which, in particular, aids creation of new cosmetic goods, paints, shampoos, detergents, etc.

Complex fluids like microemulsions and solutions of macromolecules exhibit self-organization in addition to other very important properties. In particular, they can form different structures like micelles, membranes, vesicules. Membranes play an essential role in biological processes because all living cells are covered with them. Vesicules can deliver drugs to specified parts of the human body where the drug is released in precise doses. Study of such systems is a rapidly developing activity in today's neutron scattering.

Enzymes and proteins are the hormones that control the activity of the human body. Normally, they protect humans from diseases but when hormones transform into mutant forms they may cause cancer or AIDS. One of the key properties that defines the functioning of hormones is the shape and packing conformation of macromolecules forming the biological system. Such molecules contain a lot of hydrogen which is much better seen with neutrons than other methods of structural studies.

Amorphous materials often exhibit much better mechanical properties, such as elasticity, durability, resistance to corrosion, than crystalline materials. Since neutrons can penetrate deeply into matter they appear to be the best tool to study short range structures of metallic glasses and their dependence on the external conditions even during the process of synthesis. New methods of neutron data analysis allow, for example, to trace the diffusion process of mobile ions in conducting glasses.

Investigation of magnetism still remains one of the most fascinating applications of neutron scattering. Unique information obtained with neutrons makes it possible to improve high - density recording media. Permanent magnets with an extremely high magnetic strength in a small volume are of importance for the creation of motors and vehicles. The properties of such magnets on a microscopic level can be studied only with neutrons.

In polymer science, the possibility of using isotopic labeling of some molecules or their parts allows an increase of contrast and this enables a more detailed study of polymer structures, which is of importance not only from a pure scientific point of view. They may lead the researcher on his way to producing materials with the necessary properties, e.g. environmentally friendly products, including clothes, shopping bags, materials used for manufacturing cars and airplanes, etc.

In engineering science, it is worth to mention the application of neutron scattering to the study of residual stresses, textures of technologically important materials, corrosion and embrittlement investigations. This is of great importance as soon as it ensures that rails will not crack, trains will not crash, the wings of planes will not be destroyed in the flight, construction materials of nuclear plants will prevent radioactive contamination, pipelines will not let gas or oil leak, etc.

Last but not least is the use of neutrons themselves for applied purposes, such as control of the fissile materials traffic, neutron tomography of jet engines, rockets, refrigerators, detection of explosives and drugs in luggage, cancer treatment, etc.

Of course, all above said is just a small part of today's applications of neutron scattering. It was, therefore, not surprising that a long awaited event of awarding the Nobel Prize in Physics for a pioneering contribution to the development of neutron scattering techniques for condensed matter studies by the Royal Swedish Academy of Sciences finally took place in 1994. One half went to Professor Bertram N. Brockhouse, McMaster University, Hamilton, Ontario, Canada, for neutron spectroscopy and the other half went to Professor Clifford G.Shull, MIT, Cambridge, Massachusetts, USA, for neutron diffraction. In other words, the Nobel Prize is awarded for the invention of new methods which allow us to learn i) how atoms are arranged in condensed matter and ii) how they behave in time.

III. Present situation with neutron sources in the world

To do condensed matter research, one needs bright neutron sources. At present, two nuclear reactions are used for the purpose:

- fission reaction used in reactor sources
- spallation reaction where heavy metal atoms in the target are bombarded with high energy (about 1 GeV) protons from the accelerator.

At present, there are about 50 nuclear reactors in the world used for scientific research in condensed matter.

The main parameters of some of the reactors are shown in Table 1. More detailed information can be found in [11]. The best reactor sources produce the neutron flux of the order of 10^{15}n/cm^{2}s and this is close to the presently available or possible to be obtained in near future technological level. Thus, one can hardly expect any serious advances in research reactor parameters. An additional obstacle that hinders reactor development is the negative attitude to it, often purely emotional rather than rational, of a reasonable part of the society. As it can be seen from Table 1 the major part of the existing reactor sources was built in the '60s or '70s. The new project of the construction in the USA of a $ 2.8 billion Advanced Neutron Source - the 330 MW reactor with a thermal neutron flux of 7×10^{15}n/cm^{2}s, was canceled because of a high cost and plans to use weapons-grade uranium as fuel [12,13]. At present, there are only 2 new research reactors under construction in the world - PIK in Gatchina, Russia and FRM-II in München, Germany. Just two more research reactor projects may also be realized - HIFAR1 in Lucas Heights, Australia and IRF in Canada [14].

Table 1

Selected high flux research reactors in the world

Country	Reactor	Site	Start up/ modernization	Reactor power MW	Neutron flux 10^{14} n/cm^{2}s
Russia	VVR-M	Gatchina	1959/79	14	1.4
Russia	IR-8	Moscow	1957/81	8	1.0
Russia	IVV-2M	Ekaterinsburg	1966/83	15	0.1
Russia	IBR-2 (pulsed)	Dubna	1984	2 (average) 1500 (pulsed)	0.08 (average) 50 (pulsed)
France	HFR	Grenoble	1971/94	58	15
France	ORPHEE	Saclay	1980	14	2.5
Germany	BER-II	Berlin	1973/91	10	1.0
Japan	JRR-3M	Ibaraki	1990	20	2.0
India	Dhruva	Bombay	1985	100	2.0
Canada	NRU	Chalk River	1957	125	3.0
USA	HFBR	Brookhaven	1965	60	9.0
USA	HFIR	Oak Ridge	1966	100	30
USA	NBSR	Gaithersburg	1969	20	4.0

Also, upgrading of the existing reactor HFIR in Oak Ridge, USA and modernization of the pulsed reactor IBR-2 in Dubna, Russia are under way.

Construction of spallation neutron sources is currently considered as most perspective in the field. Today, only five such sources exist and some of their parameters are presented in Table 2. The advantages of the spallation over fission reaction are as follows.

Table 2

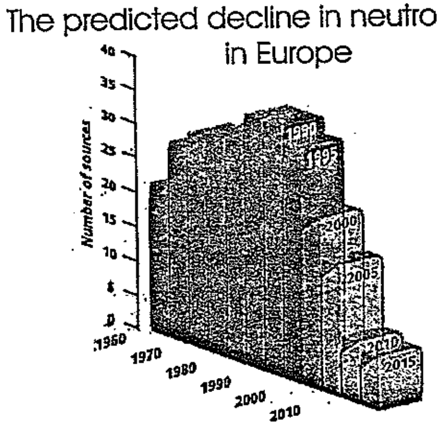
Spallation neutron sources in the world

Country	Source	Site	Start up	Proton beam power, kW	Repetition rate, Hz	Target	Protons per pulse	Proton current	Proton energy, MeV
UK	ISIS	Chilton	1985	160	50	Ta	2.5×10^{13}	200 µA	800
USA	LANSCE	Los Alamos	1985	56	20	W	2.5×10^{13}	80 µA	800
USA	IPNS	Argonne	1981	7	30	U^{238}	3×10^{12}	15 µA	450
Japan	KENS	Tsukuba	1980	3	20	U^{238}	1.5×10^{12}	5 µA	500
Switzer-land	SINQ	Villigen	1997	400	Steady state	Zircaloy	-	1500µA	590

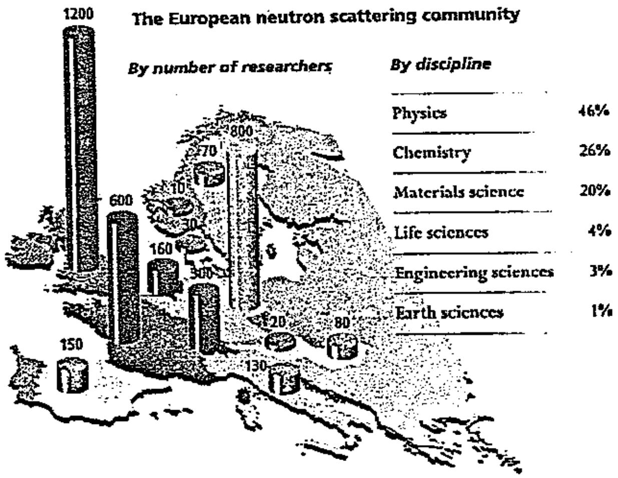
In the reactor source only 1 neutron per fission reaction can be used in neutron scattering research. The other 1.5 neutrons (in average) are necessary to sustain the chain reaction. The energy released in fission is 190 MeV per one generated neutron. Therefore, the higher neutron flux is needed the larger heat energy dissipates in the core and is removed by cooling. This is one of the major factors limiting the development of steady state reactors. In contrast, in the spallation process about 30 neutrons are produced per one 1 GeV proton that hits the target. The energy release is about 30 MeV per one produced neutron. In an ideal case, all generated neutrons are used in the experiment.

In addition, spallation sources are not a hazard to the environment because they produce neutrons only when the proton accelerator operates and there is no risk of uncontrolled chain reaction development.

However the majority of today's neutron sources are nuclear reactors. Since the average life time of the research reactor is about 20 years and most of the existing reactors are very close to that age while the number of new projects is quite limited, one may expect a rapid decline in the number of research neutron sources in near future. This is illustrated in Fig.1 taken from [14] and it reflects the predicted situation in Europe.



The decline will affect mainly national rather than international sources. This may concern approximately 7000 scientists in different scientific fields who conduct research by neutron scattering. In Fig.2 [14] there is illustrated the distribution of the neutron scattering community over Western Europe.



In addition, there are about 350 users in Russia, about 100 users in East European countries, 1000 on the American continent and others are in Japan, China, India, Australia and the Pacific region. It is important that the number of neutron users is steadily growing and at present, the demanded to available beam time ratio is almost a factor of two.

A specific feature of modern neutron facilities, like any large scale facilities today, is a high cost of construction and running. For illustration, in Table 3 there are summarized estimated costs of some new projects. It is seen that even neutron sources based on the existing technologies, like FRM-II and AUSTRON, cost several hundred million US dollars. The cost of the new generation sources, like ESS and SNS, exceeds one billion US dollars.

Table 3

Estimated cost of new neutron source projects

Country	Project	Capital cost, M$
Germany	FRM-II	440
Austria	AUSTRON	380
Europe	ESS	1050
USA	SNS	1000

Table 4 expresses the cost of running in the units of cost per instrument per day. It ranges from 900 $ for possibly the cheapest modern neutron source IBR-2 in Dubna up to 15000 $ for spallation sources based on the existing technologies.

Table 4

Estimated running cost of neutron sources

Country	Source	Running cost, instr./day, $
Russia	IBR-2	900
France	ORPHEE	3500
UK	ISIS	14500
Austria	AUSTRON	15000

So it seems that neutron scattering is an expensive technique used by a limited number of researchers, in the context of the total number of natural scientists in the world. However, an analysis of the situation in the United Kingdom and G7 countries [15] shows that the number of research papers coming from major neutron sources per invested million US dollars is about 10.5. According to [15] this proves that «the cost-effectiveness of neutron scattering research is thus comparable to that of UK civil research programs on average and is substantially greater than that of the national averages for other G7 countries». Therefore the efficiency of neutron research is very high. One of the explanations is that around the neutron source there can be simultaneously operated many experimental instruments. This permits hundreds and thousands of researchers to perform experiments with the source every year. Success of neutron experiments is based on the user program, which is a standard for any modern facility at present. User program implies a peer review of proposed experiments to estimate the quality of proposed science, feasibility and timeliness of the experiment. The OECD report [16] states: «The more accessible and easier a source is to use, and the fewer impediments are in the way of actually performing experiments, the more useful and productive a source will be». The same report concludes that «user organisations may be the only vehicle for promoting their common interest as regards sources, instrumentation and experimental techniques». For illustration, let us compare the HFR reactor of the Institute of Laue-Langevin in Grenoble, France and the HFIR reactor of the Oak Ridge Laboratory in the USA. The HFIR reactor has a higher thermal power and an almost two times larger thermal neutron flux than the HFR reactor (see Table 1). At the same time, the reactor in Grenoble is strongly oriented to the user while the Oak Ridge reactor has been mainly used for home research. As a result, in ILL there are 32 top level instruments compared to only 10 in Oak Ridge. ILL serves the needs of about 1400 scientists every year and the scientific output and international reputation of this neutron centre are on the top level. Americans have learned the lesson and are radically changing their policy in neutron scattering research as it is discussed in the next section.

IV. The future of neutron scattering

At present, it is generally accepted that the development of neutron sources demands a long-term strategy and convincing initiatives. The construction and operation cost of a modern neutron source is generally too high to be raised by a single country. In this connection, even facilities based on the existing technologies seek international cooperation. Below is a brief discussion of today's state of some projects for the construction of new neutron sources.

As it is mentioned in the previous section there are only two ongoing projects of research reactors in the world. One is the PIK reactor in Gatchina, Russia. Some of its parameters are presented in Table 5 [17]:

Table 5

Parameters of the PIK reactor

Power	100 MW
Max.thermal flux, D_2O reflector	1.2×10^{15}n/cm^{2}s
Active core volume	50 litres
Fuel	HEU (90%)
Coolant	H_2O
Moderators	1 hot 1 thermal 2 cold 1 ultracold
Neutron beams	10 horizontal 6 inclined 6 vertical
Max.number of instruments	50

The parameters of the PIK reactor are very similar to the parameters of the HFR reactor in ILL, Grenoble. The estimated date of the PIK startup is the year 2005. Since the

ILL reactor will be most probably shut down in the year 2013, the PIK reactor may be the real candidate for a steady state source to continue the international program of neutron scattering with.

The other reactor under construction is to start operation in the year 2001. This will be a German national source open to a wide international community. Some parameters of the reactor are presented in Table 6 [18]:

Table 6

Parameters of the FRM-II reactor in Münich

Reactor power	20 MW
Max.thermal flux, moderator	8×10^{14}n/cm^{2}s
Active core volume	17.6 litres
Fuel	HEU
Coolant	H_2O
Moderators	1 hot (2000°C graphite) 1 thermal (D_2O) 1 cold (liquid D_2)
Neutron beams	10 horizontal 2 inclined 1 vertical
Max.number of instruments	35

The FRM-II reactor is based on latest reactor technologies and, as it is seen from a comparison of the data summarized in Tables 5 and 6, in spite of a moderate power its rated neutron flux is comparable to the flux from more powerful reactors. This is due to a very compact reactor core consisting of a single fuel element cooled with light water. In addition, the design assumes the use of a new fuel of high-density uranium silicide in combination with highly enriched uranium.

The number of proposals to construct new spallation sources based on powerful proton accelerators is much larger than for reactor sources. At present in the world there exist five new projects of the spallation source of which three most advanced are discussed in this report.

In Japan, the construction of an accelerator complex of a 200 MeV Linac, 3-GeV Ring for 200 μA proton beams and a 50 GeV, 10 μA Ring, is under way. The complex will serve the needs of hadron physics (K-arena), muon (pion, neutrinos) physics (M-arena), nuclear physics with radioactive beams (E-arena) and neutron physics (N-arena) [19]. The neutron source is to begin operation in the year 2003 on the basis of a 200 MeV Linac and a 3 GeV Ring. Some parameters of the source are illustrated in Table 7, including future upgrades.

Table 7

Some parameters of the neutron source at the Japanese Hadron Facility

Linac length	122.3 m	
Linac final beam energy	200 MeV	
Booster ring circumference	340 m	
Final proton energy on target	3 GeV	
	1st stage	Upgrade
Beam power	0.6 MW	1.2 MW
Proton current	200 μA	400 μA
Repetition rate	25 Hz	50 Hz
Target	W or Ta Water cooled	Liquid Hg
Moderators	Room temp. H_2O Liquid H_2	– –
Average thermal neutron flux	~ 10^{14} n/cm^{2}s	
Peak thermal neutron flux	~ 10^{17} n/cm^{2}s	

The JHF N-arena will be the most powerful spallation neutron source on the basis of the existing technology and its construction (1st stage) will not require extensive R & D. The solid metal target will be replaced by a liquid metal target, most probably mercury, to upgrade the proton beam power to 1.2 MW. This will necessitate serious experimental work which will be very likely done in cooperation with other two projects discussed below.

On January 21, 1998 in Oak Ridge, the Vice-President of the United States A.Gore announced that the White House would request $ 157 million for the 1999 fiscal year to begin the construction of the Spallation Neutron Source (SNS). Thus, work that started in the USA in the early 1970s to develop a strategy to meet the future need for neutron sources completed with a very positive result. The DOE report on SNS recommends, in particular, that the source be highly reliable, that it has high availability and that it possesses the inherent design flexibility to be upgradable to provide future capabilities for the neutron user community [10]. SNS is the first of the next generation sources whose construction was approved and the work started. At the same time, even in the US, a single Laboratory or an Institute cannot carry out such a project. In this connection, DOE has selected the Oak Ridge National Laboratory (ORNL) to be the site for the neutron source and a truly collaborating team has been formed on the basis of Argonne National Laboratory, Brookhaven National Laboratory, Lawrence Berkeley and Los Alamos. Each Laboratory holds responsibility for a particular part of the whole project in which it has the best expertise. So, the Ion Source will be constructed by Lawrence Berkeley, LINAC will be built by Los Alamos, Accumulator Ring will be created by Brookhaven, Target will be produced by Oak Ridge, and the Experimental Facilities will be the products of Argonne and Oak Ridge. In the future, the Joint Institute for Neutron Science will be organized to serve the needs of the American national user community in the first place and also, it will be open to international users. Some parameters of the SNS are presented in Table 8.

Table 8

Some SNS parameters

Linac length	493 m	
Output Linac beam energy	1.0 GeV	
Accumulator ring circumference	220.7 m	
Pulse repetition rate	60 Hz	
Target	Liquid Hg	
	Initial parameters	Upgrade
Beam power	1 MW	2 MW
Proton current	1000 μA	2000 μA
Peak thermal flux	2×10^{16} n/cm^{2}s	4×10^{16} n/cm^{2}s
Moderators	2 thermal 2 cold (liquid H_2)	

As it can be seen for SNS, a liquid mercury target is necessary to cope with a high thermal load caused by the proton beam. There has been no experience in the construction of such targets yet and, therefore, expensive R&D work is currently being done in cooperation with the team on the ESS project.

ESS, the European Spallation Source, is the most ambitions project of today. It exploits most recent achievements in accelerator technology. The source will be operated according to a traditional scheme established in European neutron scattering centers. It will provide new opportunities for studies in physics, chemistry, biology, materials science, earth science, pharmacology, engineering, etc. The new source is needed by the European scientific community to escape the risk of a serious shortage in neutron beams in near future. In the last 20 years Europe played a leading role in condensed matter research with neutrons. At present, as discussed above, the United States and Japan are constructing next generation neutron sources and in Europe, the ESS project is still under consideration. Full information about the source, including the scientific program and technical studies of its components, such as the ion source, linac, accumulator ring, beam transport, target etc. can be found in [9]. Selected parameters of the source are presented in Table 9.

Table 9

European Spallation Source parameters

Linac length	710 m	
Output Linac beam energy	1.334 GeV	
Accumulator ring circumference	163.4 m	
	Target 1	Target 2
Beam power	4 or 5 MW	1 MW
Pulse repetition rate	50 Hz	10 Hz
Peak thermal flux	2×10^{17} n/cm^{2}s	
Average thermal flux	7×10^{14} n/cm^{2}s	

It should be noted that not only the peak thermal neutron flux of the source will be higher than from any existing neutron sources in the world but also the average thermal flux will be close to 10^{15} n/cm^{2}s, i.e. be about equal to fluxes available from modern research reactors (c.f. Table 6). Thus, the source will make it possible to conduct many experiments that at present, are only feasible with steady state sources. The project is, of course, developed internationally. The following European countries enter the ESS Council: Austria, Denmark, Germany, Italy, Netherlands, Sweden, Switzerland, United Kingdom, France, Spain, USA, together with some international organizations involved as observers. Best experts in accelerator physics, design and construction from CERN, DESY, GSI, RAL, Los Alamos etc. work on the project. Recent neutron target developments as well as a unique know how from Jülich, PSI, RAL, Riso, HMI are utilized. To develop the instrumentation base for the new neutron sources three working groups are formed:

- Upgrading of the existing instrumentation at pulsed sources;
- Development of good reactor instruments for pulsed sources;
- New types of instrumentation for pulsed neutron sources.

Today's state of the project makes it possible to start the next stage of its realization immediately. The only factor that limits the speed of project realization today is the absence of the final political decision of the financing bodies.

V. Conclusion

Above, an interesting factor is not discussed. All considered spallation sources produce short neutron pulses with a typical pulse width of about 30 μs for thermal neutrons. Therefore, expertise in the construction of instruments for spallation sources lies mainly in the field of short pulse sources. For many years, there has dominated the opinion that the short pulse is a must for efficient use of a pulsed neutron source. The experience of operation of the IBR-2 pulsed reactor in JINR has, however, shown that the pulse width is not a limiting factor [11]. A broad and very successful scientific program being conducted at that long pulse neutron source with the pulse width about 300 μs for thermal neutrons proves that a proper instrument design can make the long pulse be even an advantage [20]. This has stimulated activity in analysis of possible designs and the construction of next generation long pulse spallation neutron sources [21,22]. Today, the idea of building such a source based on a linear proton accelerator and a neutron target to produce neutron pulses of several milliseconds is actively developed in Los Alamos Laboratory in USA. There is a real hope that appropriate instrumentation for such neutron source will allow experimentalists to cover and even exceed the existing capabilities of steady state reactors.

The generally accepted opinion in the world neutron scattering community is that new facilities alone cannot satisfy all scientific needs. Continued use of the existing reactors and pulsed sources is desirable in foreseeable future. However, this is only possible if the sources are modernized and their instrumentation is seriously upgraded.

In this connection it should be emphasized that the IBR-2 reactor of JINR is at present, the only modern neutron source with advanced instrumentation that belongs to the member states of the Institute. Its modernization and further instrumentation development to ensure the prolongation of IBR-2 operation until the year 2030 is a necessity if we want to preserve the high level of neutron scattering research in JINR member states and in Eastern Europe in general.

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