JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory of Neutron Physics

FINAL REPORT ON THE SUMMER STUDENT PROGRAM

Creating a database of inelastic and quasielastic neutron scattering spectra from NERA spectometer

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Abstract

In Frank Laboratory of Neutron Physics of the Joint Institute for Nuclear Research in Dubna is fast pulsed reactor IBR-2M. The reactor is surrounded by many experimental devices based on a neutron beam from this reactor. One of these devices is the NERA spectrometer. On this spectrometer scientists can measure inelastic and quasielastic neutron scattering and neutron diffraction. The results of the measurements are available on the website: neradb.jinr.ru . This report is about theoretical introduction to neutron scattering and about practical part of the project – the expansion of the database.

Contents

Spis treści

1. Description of the project

I was the participant of Summer Student Program in Joint Institute for Nuclear Research (JINR) in Dubna for six weeks. During the practise course, I studied opportunites of research in JINR. I visited the reactor and the following laboratories:

- Frank Laboratory of Neutron Physics – FLNP (where I was working),
- Flerov Laboratory of Nuclear Reactions – FLNR,
- Veksel and Baldin Laboratory of High Energy Physics,
- Dzelepov Laboratory of Nuclear Problems – DLNP.

The main purpose of my job was creating a database of inelastic and quasielastic neutron scattering spectra obtained from NERA spectrometer in Frank Laboratory of Neutron Physics. The project was a continuation of the work undertaken last year's summer practise. The task aimed to create a database which will display information such as: sample's name, experiment's author and author's affiliation, date of start and end of measurement, comments and information of publication of results. For some samples pictures of spectra are shown. It was supposed that the user of database will be able to search for sample giving: sample's name, experiment's author or/and year of the measurement. The database contains an administrator's panel where administrator can change data, add new samples and delete samples. Databse is available as the website: neradb.jinr.ru.

My report I started from about basic information about neutron spectroscopy.

2. Neutron spectroscopy

2.1 Neutron properties

The neutron is a particle without electric charge and with a mass about 1,0087 u ($1{,}675*10^{-27}$kg). Neutron consists of three quarks: two quarks *d* with the charge about – 1/3 and the one *u* with a charge +2/3 (Figure

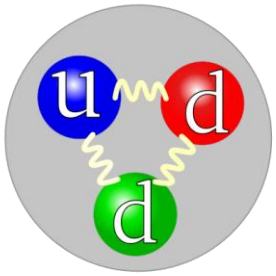


Fig. 1.: The scheme of neutron from: https://upload.wikimedia.org/wikipedia/commons/thumb/8/81/Quark_structure_neutron.svg/2000px-Quark_structure_neutron.svg.png

1.) Neutron has a spin $s = ½$.

Neutron was discovered by James Chadwick in 1932 (Nobel prize in 1935). The α-particles beam was fired on beryllium nucleus, which gave neutron and the carbon nucleus:

$$^{9}_{4}Be + \ ^{4}_{2}He \rightarrow \ ^{12}_{6}C + \ ^{1}_{0}n + 5{,}7\ MeV$$

The obtained neutrons collided with paraffin foil and ejected the protons from it. The protons were recorded by Geiger-Muller counter.

Neutrons are stable only in stable atomic nucleus. Free neutrons are labile and they decay with the mean lifetime τ = 889,1 s into a proton (p), an electron (e) and an antineutrino ($\tilde{\nu}_e$). This process is named β decay:

$$n \rightarrow \ p^{+} + \ e^{-} + \ \tilde{\nu}_e + 0{,}77\ MeV$$

Neutron has the wave-particle properties. The de Broigle relation describes the neutron wavelenghts:

$$\lambda_n = \frac{h}{m_n \vartheta_n} = \frac{h}{\sqrt{2 m_n E_n}}$$

According to their energy there are different categories of neutrons (Figures 2.):

- Epithermal neutrons
- Thermal neutrons
- Cold neutrons
- Ultracold neutrons

Quantity	Unit	Definition	Ultracold	Cold	Thermal	Epithermal
Energy E	meV[a]		0.00025	1	25	1000
Temperature T	K	E/k_B	0.0029	12	290	12,000
Wavelength λ[b]	Å	$h/(2mE)^{1/2}$	570	9.0	1.8	0.29
Wave vector $\mathbf{k}$[c]	$Å^{-1}$	$(2mE)^{1/2}/\hbar$	0.011	0.7	3.5	22
Velocity v[d]	m/s	$(2E/m)^{1/2}$	6.9	440	2200	14,000

[a] 1 meV = 1.6022×10^{-15} erg, the energy required to raise a proton up to a potential of 1 mV.
[b] λ (Å) = 9.0446 [E (meV)]$^{-1/2}$.
[c] k ($Å^{-1}$) = 0.69469 [E (meV)]$^{1/2}$.
[d] v (m/s) = 437.39 [E (meV)]$^{1/2}$.

Figure 2.: Characteristics of Neutrons t Selected Energies from *R. Celotta, J. Levine, Methods of Experimental Physics. Neutron Scattering, Academic Press, INC, London 1986.*

2.2 Neutron sources

There are different types of neutron sources:

- Small neutron sources
- Nuclear fission
- Neutron sources based on particle accelerators

The γ radiation is used in *small neutron sources.* The γ quantum incident on a beryllium nucleus giving its isotope and neutron:

$$^{9}_{4}Be + \gamma \rightarrow {}^{8}_{4}Be + {}^{1}_{0}n - 1{,}66\ MeV$$

In *nuclear fission* an ^{235}U absorbs thermal neutron and decays into two smaller nuclei and gives on average 2,5 neutrons (Figure 3.) with releasing 200MeV of energy:

$$^{1}_{0}n + {}^{235}_{92}U \rightarrow 2\ nuclei + 2{,}5\ {}^{1}_{0}n + 200\ MeV$$

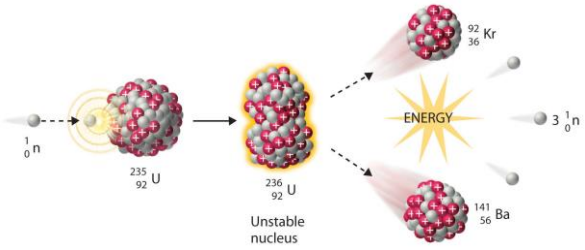


Figure 3.: An example of nuclear fission of ^{235}U which decays into ^{141}Ba and ^{92}Kr from *http://www.nucler-power.net/wp-content/uploads/2014/10/nucler_fission.jpg?32b4fc.*

The obtained neutrons have the energies around 1 MeV.

This reactions chain proceeds in neutron reactors. There are two types of reactors: stationary and pulsed. In a stationary reactor from 2,5 neutrons 1 is needed to maintain the chain reactions 0,5 is absorbed by the construction elements and there is one which can be used in research. The Figure 4. shows the scheme of stationary reactor. The nuclear uranium fuel is marked by 1. Between it there are regulating and emergency roots (2). The 3 is a cover and there is also a biological shield (4). The moderator (for example water) can also be used as a refrigerant (5). The obtained neutrons pass by neutron guide (6).

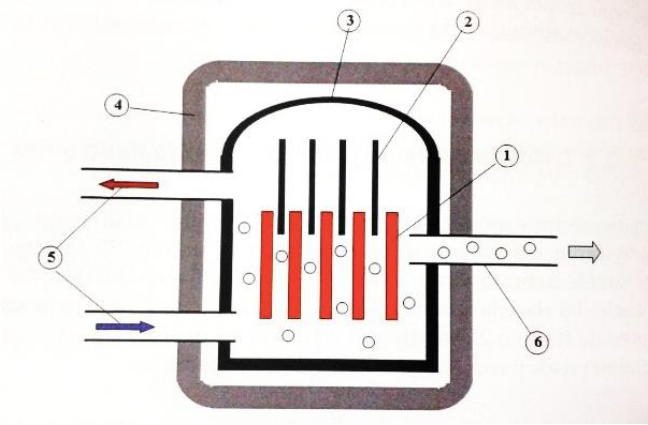


Figure 4.: The scheme of a stationary reactor. *V. Belushkin, J. W. Wąsicki, Wprowadzenie do neutronowych metod badania fazy skondensowanej materii, Wydawnictwo Naukowe UAM, Poznań 2013.*

The second type of reactor is a pulsed reactor, which was built in 1960 in Joint Institute for Nuclear Research in Dubna (Figure 5.) and named IBR-2. It is a fast reactor with periodic action. Its core is made by 69 fuel assemblies filled by PuO_2. Those assemblies are directly cooled by liquid sodium. The core is surrounded by 5 stationary reflectors and the sixth movable reflector. This movable reflector consists of: the min movable reflector and the auxiliary movable reflector. They rotate against each other with different rotating speed. While two reflectors are near the core the neutrons are reflexed to the core and it genertes a power pulse. When the reflectors move away the power of reactor decreases. The whole reactor is also cooled by liquid sodium and covered by two biological shields.

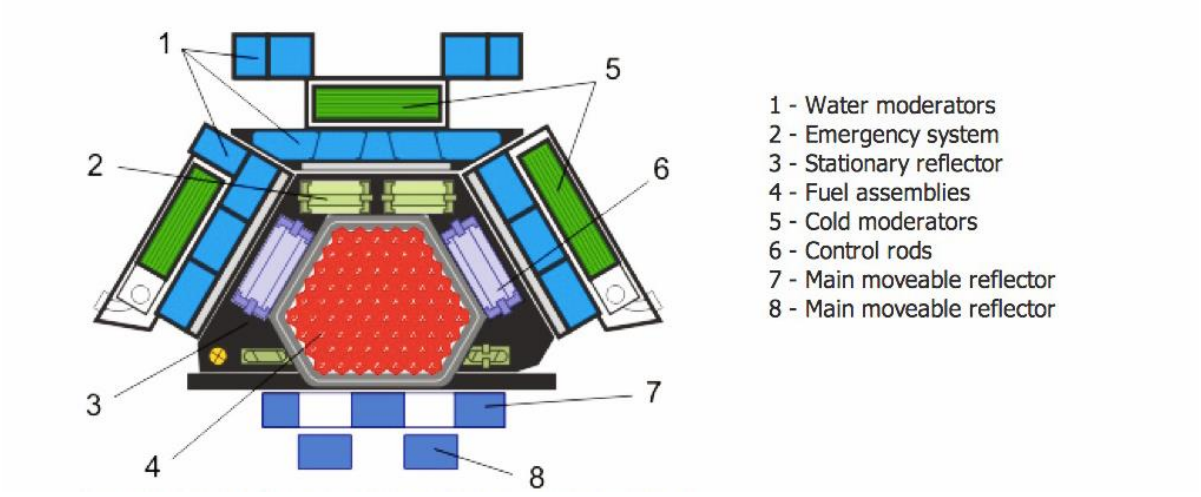


Figure 5.: The scheme of reactor core of IBR-2 and its description from: http://flnp.jinr.ru/251/.

There are also *particle accelerators*, which can be neutron sources. A particle beam is fired on heavy nucleus. For example, deuterons are accelerated to the energy of 100keV and they incident on a target of TiT_2. In this process we obtain the nuclei of helium and neutrons:

$$^{2}_{1}D + ^{3}_{1}T \rightarrow \ ^{4}_{2}He + ^{1}_{0}n + 17{,}6\ MeV$$

When there is a target built from heavy chemical elements like Pb, W, Ta and very fast electron beam is fired on it, it provides a photonuclear reaction. Inhibitory electrons produce the γ quantum fluxes. This kind of radiation is called Bremsstrahlung. When these γ quants interact with the nuclei of a target, the neutrons are formed (γ, n). This source gives short and pulsed neutron beams. The accelerator, which uses that type of neutron source, is IREN in Dubna.

And the last example of neutron source based on a particle accelerator is the *spallation process.* It takes place in the nuclear reaction between the high-energy particles and heavy elements. Firstly the intense beam of protons is focused onto a target (e.g. uranium). The nucleus of the target becomes excited and there are multiple intra-nuclear reactions taking place. As a result nucleus produces the very high-energy neutrons. Finally, the nucleus throws out neutrons, protons, deuterons, α particles, photons and neutrinos. So in those reactions one proton nd one heavy nucleus create many neutrons.

To sum up it is important to use the neutron source, which produces many neutrons but also does not release much energy so that it is needed to optimize the neutron production.

2.3 Interaction with matter

When a neutron is near the nucleus it could be:

- Absorbed by nucleus and excited it. After that nucleus could: emit γ quantum, undergo fission or throw away α particles, protons etc.
- Scattered by the nucleus that changes its energy and direction movement.

When the sample is under neutron beam $Ø_0$ and I_s is the number of scattering acts and I_a is the number of absorbing acts, the cross section for scattering σ_s and absorption σ_a are given by equations:

$$I_s = Ø_0\, \sigma_s$$
$$I_a = Ø_0 \sigma_a$$

The σ_s and σ_a are measured in barns (1 barn = 10^{-28} square meter).

The differential cross section for neutro scattering XXXX describes the probability that the scattered neutron can be detected at given angle OMEGA and will be found in the solid angle DOMEGA. A double differential cross section describes the probability that the neutron will leave the sample at a given angle OMEGA and will be found in the solid angle DOMEGA and its energy will be changed from XX to xx. That gives an equation:

$$\sigma_s = \int d\Omega \frac{d\sigma}{d\Omega} = \int d\omega \int d\Omega \frac{d^2\sigma}{d\Omega d\omega}$$

Neutrons interact with nuclei by nuclear and magnetic forces. For nuclear forces the potential impact between neutron and nucleus is given by the Fermi pseudopotential, ehich is given by equation:

$$V(\vec{r}) = \frac{2\pi\hbar^2}{m} \sum_i b_i \delta(\vec{r} - \overrightarrow{R_i})$$

where b_i is the scattering length, R_i is the position of nucleus *i* in a sample and $\vec{r}$ is the position of a neutron. The mass of a neutron is given by *m* and δ is the Dirac delta. This is summed over every nuclei taking part in scattering. The scattering length is the base parameter, which describes the interaction between neutron and nucleus and it depends on the isotope and the relative spin orientation of each other. The average value of $<b_i>$ for every isotopes and spin orientations is named coherent scattering length $b_i^{coh} = <b_i>$. The average square variation of b_i is the incoherent scattering length:

$$b_i^{inc} = \sqrt{< b_i^2 > - < b_i >^2}$$

The scattering lengths for chosen elements and their isotopes are shown in Figure 6. Opposite to x-ray scattering, the length scattering for neutrons depends irregularly to the Z number of an element and it depends of its isotopes. However, in the sample there is a mixture of isotopes, it means that it has both coherent and incoherent scattering length. The cross sections for the coherent and incoherent scattering will be:

$$\sigma_{coh} = 4\pi < b >^2$$
$$\sigma_{inc} = 4\pi(< b^2 > - < b >^2)$$

Relatively and the total cross section will be the sum of the coherent and incoherent cross section:

$$\sigma = \sigma_{coh} + \sigma_{inc}$$

The coherent and incoherent cross sections for neutron scattering allow recognizing various properties of condensed matter. Coherent scattering gives the information about the equilibrium structure, crystal structure excitation or magnetic subnet excitation. Incoherent scattering describes single atoms or molecules, for example its diffusion in the crystal.

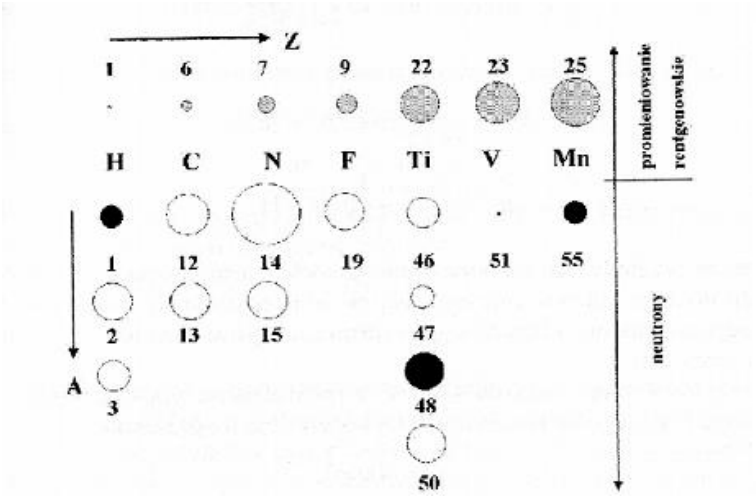


Fig. 6.: The comparison of the scattering lengths proportional to the diameter of cirles for x rays and neutrons. Tha black circle means the negative scattering lengths. *V.Beloshkin, J. W. Wąsicki, Wprowadzenie do neutronowych metod badania fazy skondensowanej materii, Wydawnictwo Naukowe UAM, Poznań 2013*

2.4 Inelastic incoherent neutron scattering

The investigation method being used in the scientific team in which I worked is inelastic *incoherent neutron scattering*. To study this method it is indispensable to know about different kinds of neutron scattering.

The first one is *elastic neutron scattering*. It is when a neutron beam incident on a fixed, point scatterer like nucleus and there is no change in the energy of the incident neutron. Although the direction of the wave vector of the neutron changes, its magnitude does not. The example of this kind of scattering is *diffraction*.

When neutrons are scattered by matter, the scattering from each of the individual nuclei are added. Nuclei are not fixed because atoms in matter undergo atomic vibrations. Nuclei can also recoil during a collision with a neutron. As nuclei are moving they can impart or absorb energy of neutrons. This kind of neutron scattering is named *inelastic neutron scattering* and in this process the direction and the magnitude of the neutron wave vector change.

These two types of neutron scattering provide different scattering effects. So we are talking about *coherent neutron scattering* when the neutron wave interacts with the whole sample as a unit and then the scattered waves from different nuclei interfere with each other. As this type of neutron scattering depends on the relative distances between the constituent atoms, it gives the information about the structure of materials.

In incoherent neutron scattering an incident neutron wave interacts independly with each nucleus so that the scattered waves have indeterminate relative phases and they do not interfere with each other. However, the intensities from each nucleus add up.

The combination of these kinds of neutron scattering gives four methods and its applications is presented in Table 1.

Type of neutron scattering	elastic	inelastic
coherent	Equilibrium structure, crystal structure	Collective motions of the atoms which produce vibrational waves in crystalline lattice
incoherent	(quasielastic): fast stochastic motions of molecules and molecules groups	Single atoms/molecules as atomic diffusion in crystalline lattice

Table 1.: Types of neutron scattering.

To investigate the neutron scattering it is indispensable to get to know about the *phonons* and *magnons*. As we know, in reality atoms are not frozen in fixed positions in a crystal. As they have thermal energy, they oscillate about their lattice site and move around inside a small volume with the lattice site at its centre. We can name this position in the centre the official position. As atoms are connected with each other by the binding forces, when one atom oscillate, neighboring atoms also start to oscillate with different frequencies and wavelength travelling in different directions. In effect they form the wave, which starts passing up and down. There are many waves like that which are formed by these thermal motions of atoms. We can describe the superposition of this waves mowing through the lattice. These fundamental vibrational waves in a crystal in which nuclei oscillate in a coordinated manner are named *phonons.* The energy of phonons is quantized and has a υ that is the frequency of atomic motion associated with the phonon.

Besides the neutron interaction with atomic nuclei, there is also an interaction between a neutron and the magnetic field from the fact that a neutron has a magnetic moment. The neutron experiences this magnetic force, when it passes near the electron. As most electrons in atoms are paired, they do not have the magnetic moment, but in some other atoms there are no paired electrons from valence shell. So neutrons could be also scattered by the resulting magnetic moments. It happens for example in ferromagnetic and antiferromagnetic materials, which have aligned magnetic moments of their electrons (in ferromagnetic they are pointed in the same direction and in antiferromagnetic in opposite directions). In the neutron scattering only the component of the sample's magnetization that is perpendicular to the scattering vector, is effective. As in magnetized materials the directions of the atomic moments can oscillate, they form a wave of magnetic oscillation that passes through the sample. These magnetic excitations are called *magnons.* Thus, they are magnetic analogues of the phonons and can be measured by the inelastic neutron scattering.

Returning to the inelastic incoherent neutron scattering the double differential cross section for incoherent neutron scattering will be:

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_1}{k_0}\sum_i (b_i^{inc})^2 S^{inc}(\vec{Q},\omega)$$

where the k_0 and k_1 are the wave vector of incident and scattered (relatively) neutron wave and $S^{inc}(\vec{Q},\omega)$ is the incoherent scattering law. Mathematically it is a spatial and time Fourier transformation of autocorrelation function, which describes the probability that the atom being at one point in time *t=0* will be found at a second definite point in time *t.* Thus, this method allows investigating monoparticlesdynamics effects like:

- local and quasilocal vibrations of admixtures,
- crystal field splitting,
- intramolecular vibrations,
- phonons and magnons density of states.

In spectroscopic studies of intreamolecular vibrations of molecules containing hydrogen atoms we can say that in the scattering law there are only the vibrations of hydrogen. It is because for hydrogen there is a big value of a scattering length. It gives an equation for the scattering law:

$$S(Q,n\omega) = \frac{1}{n!}(Q^2 < U^2 >)^n \exp(-Q^2 < U^2 >)$$

where $<U^2>$ is an average value of a square vibration amplitude and *n* is the principal quantum number. For *n=0* this equation describes the dependence of the intensity of neutron scattering from *Q*. For *n=1* it describes the first excited state of oscillator. The neutron scattering is analogic to the optical spectroscopy. But there are differences, which make this method an important complement for optical spectroscopy. Firstly there are no selection rules. Secondly, the signal intensity is proportional to *U* meanwhile for optic spectroscopy it is not. Furthermore, the large amplitude of hydrogen vibrations and the anomaly large cross section for neutron scattering makes the neutron spectroscopy an unusually sensitive method for investigating the samples containing hydrogen atoms. As we know, deuter does not have that large cross-section, so by changing the hydrogen atom for deuter, we can define the type of vibrations that corresponds to the signals in neutron spectrum. Finally, these materials, which are not transparent for visible and infrared spectrum, are transparent for neutrons (e.g. metals).

The inelastic incoherent neutron scattering is also used in the studies of phonons density of states - *g(ω)*. This function characterizes the probability thet the vibrational waves with the frequency *ω* will exist in a crystal. For compounds containing hydrogen atoms the cross section for incoherent scattering will be described by:

$$\left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{inc} = \frac{k_1}{k_0} N \frac{|Q^2|}{4\omega M}(b_H^{inc})^2 e^{-2W} G_H(\omega)\left(cth\frac{\hbar\omega}{2kT} \pm 1\right)$$

where *M* is the mass of crystal unit cell and $G_H(\omega)$ is the phonons density of states for hydrogen.

2.5. Impact of isotopic effect on inelastic neutron scattering spectra

An isotopic effect occurs when there are the differences of chemical and physical properties for different isotopes of the same chemical element. It also provides the differences of chemical compounds properties built from different isotopes of the same element.

The most common examples of compounds consisting of two isotopes are water and heavy water. They have different molecular weights, densities, melting and boiling points or permittivity. They also have different biological activities.

The difference of isotopes mass causes also differences in chemical activity because it changes the velocity of reactions. This effect is called kinetic isotopic effect.

Isotopic effect is clearly visible and useful in spectroscopic research. For example, inNMR technique as hydrogen nucleus has the spin s = ½ and deuterium nucleus has s = 1, it provides that in ^{1}H NMR spectrum we will not see any signals of deuterium. This effect can be used for example to detect another signal, which was covered by the one of hydrogen. Hydrogen-deuterium exchange provides the change of vibration frequency. For IR spectroscopy it will be the shift about 1,35 towards smaller wave numbers. So, recording two spectra – for hydrogenated and deuterated samples and comparing them – we can detect the signals of changed hydrogen atoms.

Finally, in neutron spectroscopy there is a huge difference between the cross sections of the hydrogenated and deuterated molecules. As the incoherent inelastic neutron scattering cross section is anomaly large for hydrogen, this technique is very sensitive for this isotope but it is not for deuterium. These modes with deuterium appear only weakly in the spectrum. Often the bands seem to disappear altogether from the spectrum. The advantage over the optical technique is that the spectrum is less congested.

The hydrogen-deuterium exchange also gives another important opportunity: it allows studying individual hydrogen atoms free from the interference of other hydrogen vibrations. This is why neutron spectroscopy is a unique method for the study of hydrogen bonds.

3. Information about NERA – group

3.1 Frank Laboratory of Neutron Physics

Joint Institute of Nuclear Physics consists of seven laboratories. One of them is Frank Laboratory of Neutron Physics (FLNP). Frank Laboratory of Neutron Physics was founded in 1957. FLNP has a lot of experience in creation and development of neutron sources. There are two neutron sources in FLNP: pulsed fast reactor IBR-2M and the Intense Resonance Neutron Source. There is also EG-5 – Van der Graaf accelerator. FLNP has also experience in using neutrons in fundamental and applied science. There are research about neutron as an elementary particle and also studies of other materials using neutrons. Many

experiments are performed by the researchers from many countries. In FLNP ultracold neutrons were discovered. Enhancement of the space parity violation effect in neutron resonances was found.

3.2 NERA spectrometer

The NERA Group is a part of Frank Laboratory of Neutron Physics of the Joint Institute for Nuclear Research in Dubna. The NERA spectrometer is aimed to study inelastic and quasielastic neutron scattering with simultaneous control of the phase of the sample by neutron diffraction. This spectrometer is used to investigate the dynamics and structure of condensed matter.

The NERA spectrometer is an inverse geometry time-of-flight spectrometer. In this type of spectrometer a pulsed neutron source produces the polychromatic neutron beams with the period *T*. They have continuous energy spectrum and their energy E_i is appointed from the time of flight measured between a source and a sample. The energy of scattered neutrons is monochromated by a monochromator placed after the sample. The detectors are placed at different angles that provides recording of spectra for different values of scattering vector $\vec{Q}$ and for the same value of ω. The time-of-flight will be given by the equation below:

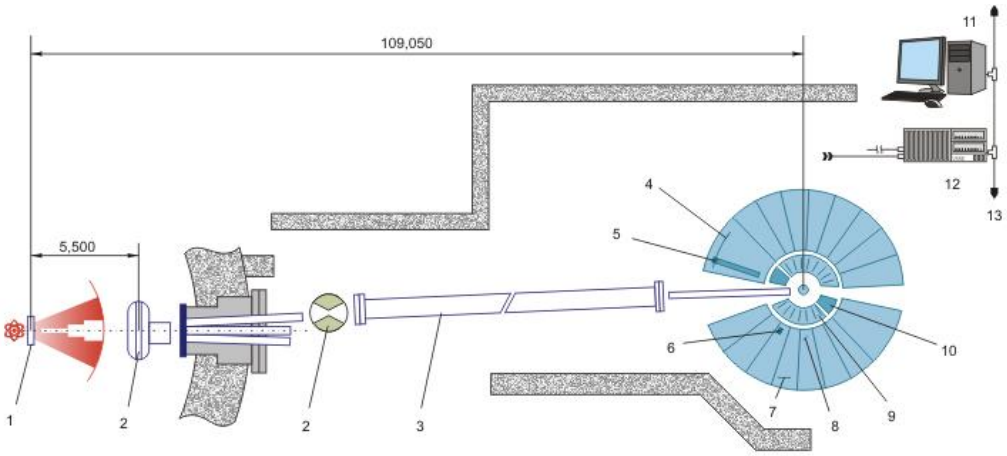
$$t_i = t - \frac{(L_{f1} + L_{f2})}{v_f}$$

where t is unrestricted time from the neutron beam generation, L_{f1} and L_{f2} are the distances between a sample and a monochromator and a monochromator and a detector relatively, $v_f = \sqrt{\frac{2E_f}{m}}$ is the velocity of scattered nautrons determined by crystal. Thus, the neutron energy will be:

$$E_i = \frac{m}{2}\left(\frac{L_i}{t - \frac{L_{f1} + L_{f2}}{v_f}}\right)^2$$

where L_i is the neutron time-of-flight from the source to the sample.

The spectrometer and its description is presented in Figure 7.



1 - Moderator
2 - Background Choppers
3 - Ni Guide Tube
4 - Detector for High Intensity Diffraction
5 - Detector for High Resolution Diffraction
6 - He^3 Detectors (INS and QNS)
7 - Single Crystal QNS Analyzer
8 - Pyrolytic Graphite INS Analyzer
9 - Be-Filters
10 - Collimators
11 - VME control and operative visualization/analysis
12 - VME Station (OS/9)Data Acquisition
13 - EtherNet Data Transfer

The produced neutron beam is slowed in the moderator (1) and background choppers (2) remove the background. After that, the neutron beam goes throw the neutron guide (3). The neutron beam incident on the sample (11). A spectrometer has 2 parts, each with 8 sections for measuring the inelastic scattering and one for the elastic scattering. The 4 and 5 are the diffraction detectors and (7) is a single crystal quasielastic neutron scattering analyser. For the inelastic incoherent neutron scattering the devices are used: ^{3}He Detectors (6) Pyrolitic Graphite INS Analyser (8), Be-filters (9) and collimators (10). At the same time 16 IINS spectra are registered at angles from 20^o to 160^o. Be-filters are cooled in liquid nitrogen.

4. Realization the project

The main point of my project was expansioning a database for spectra from NERA spectrometer. This database was written in MySQL language. The main website page consists menu: list of samples, search and administration (Figure 8.).

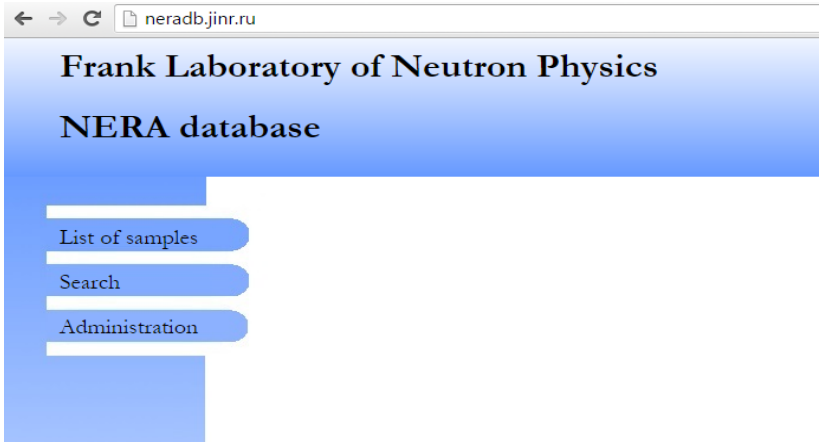


Figure 8.: Main page.

I worked mainly in tab 3. – Administartion:

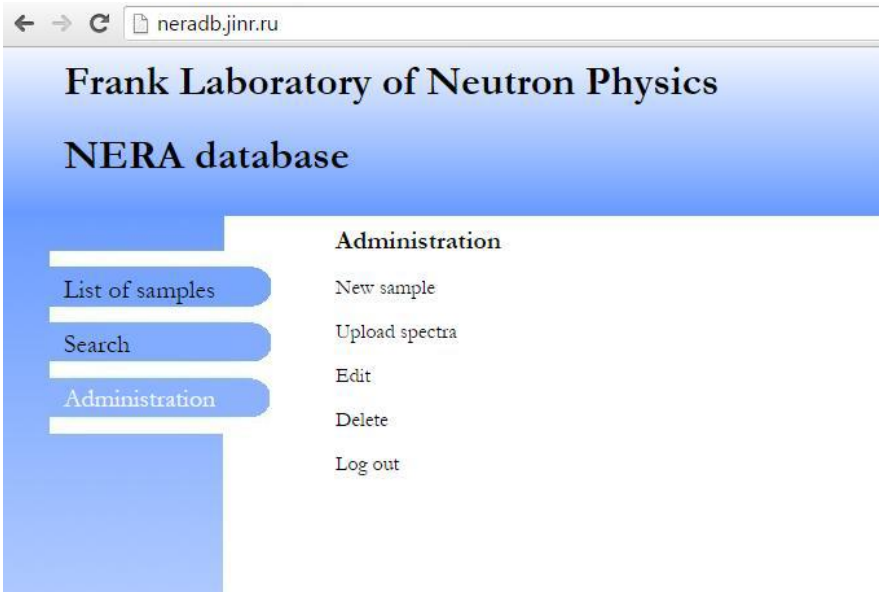


Figure 9.: Administration's menu.

During my work I completed the database for the years 2014 - 2017 and checked the year 2013. For this purpose, I used a lab log and user scripts of the NERA spectrometer. Result of my project are on the website: www.neradb.jinr.ru .

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