Appendix 1

Form № 24

PROJECT

Development of an inelastic neutron scattering spectrometer in inverse geometry at the IBR 2 reactor.

Theme: « Studies of functional materials and nanosystems using neutron scattering»

Theme code: 04-4-xxxx-2021/2025

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DATE OF LABORATORY STC _________ DOCUMENT NUMBER _________

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Appendix 2

Form № 25

PROJECT ENDORSEMENT LIST

Development of an inelastic neutron scattering spectrometer in inverse geometry at the IBR 2 reactor.

Theme: « Studies of functional materials and nanosystems using neutron scattering »

Theme code: 04-4-xxxx-2021/2025

APPROVED BY JINR DIRECTOR	_________________	«____»_______2020

ENDORSED BY

JINR VICE-DIRECTOR	_________________	«____»_______2020
CHIEF SCIENTIFIC SECRETARY	_________________	«____»_______2020
CHIEF ENGINEER	_________________	«____»_______2020
HEAD OF SCIENCE ORGANIZATION DEPARTMENT	_________________	«____»_______2020
FLNP DIRECTOR	_________________	«____»_______2020
FLNP CHIEF ENGINEER	_________________	«____»_______2020
PROJECT LEADER	_________________	«____»_______2020

ENDORSED

PAC for CMP	_________________	«____»_______2020

ABSTRACT

The status of Inelastic Neutron Scattering Spectroscopy at FLNP has been under concern of both the user community and the Laboratory Management, instigating discussions within both parties. It became clear that the two spectrometers available at the moment, once competitive to European class instruments, have lost the race years ago, and they no longer satisfy the needs of the user community in the Eastern Europe region. It is therefore of the utmost importance to advance INS Spectroscopy in the direction set historically, and to support the maintaining the world renown scientific position of the FLNP JINR.

In order to regain competitiveness with the instrumentation of European neutron scattering facilities, it is necessary to build new INS spectrometers that would make use of up to date neutron optics and design solutions in order to deliver high resolution results, of excellent signal-to-background ratio over a broad range of energy transfer, from as small samples as possible in a time-wise highly efficient way. The first instrument to build will be an inverted geometry versatile spectrometer. The brightness of the IBR-2 source will be made use of to the maximum, and a very large surface or the reflecting energy analysers will assure the highest luminosity (largest acceptance) design.

Following the recommendations of the previous PAC meeting, we are going to open the new project “Development of an inelastic neutron scattering spectrometer in inverse geometry at the IBR-2 reactor” in the framework of the theme “Investigations of Functional Materials and Nanosystems by Neutron Scattering Methods”.

Contents

1. Scientific background

1.1. Introduction

Neutrons can excite atomic vibrations or activate stochastic motions via energy and momentum exchange during inelastic scattering (and the reverse of course holds as well). For inelastic incoherent scattering, the intensity is proportional to the space and time Fourier transforms of the self-correlation function, $G_s(\vec{r},t)$, the probability of finding a particle at position $\vec{r}$ at time t when the same particle was at $\vec{r}=\vec{0}$ at $t=0$. This makes it possible to develop neutron spectroscopy, both stochastic and vibrational.

Since neutrons are not in abundant supply, nor are they cheap to deliver to the instrument, two questions have to be answered:

- Are neutrons any better a probe to study vibrational properties of condensed matter than photons (e.g. IR or Raman)?
- Even if so, do they deliver “value for money”?

Such questions are often asked by financing bodies when it comes to spending considerable funds on the instrument development.

Neutrons as a probe to study atomic vibrations are neither better nor worse than photons. They are complementary. This complementarity means, among others:

- one method can better reach the goal where the others can’t or do it with difficulty,
- one method can provide specific information that the others can’t.

Vibrational spectroscopy can and does serve two main research fields:

- a big, and growing area of chemical compound detection and identification tasks. This includes the feasibility studies of application of vibrational spectroscopy in medical diagnosis, especially early, pre-clinical oncology. The research in this field involves conventional, laboratory spectrometers, but also synchrotron sources and confocal microscopy, high spatial resolution in-plane microspectroscopy. This is, in general, applied spectroscopy. It develops towards high speed, effectiveness, working with very large volumes of data (hyper-spectroscopic data sets), requiring “Big-data” approach.
- Study of specific properties of matter through the investigation of atomic vibrations therein, for fundamental research, which is now almost always application driven.

Therefore, complementarity means that advances in the use of one probe, e.g. photons, generates an increased demand for information that can only be provided by another probe, e.g. neutrons.

Therefore, in order to:

- maintain the quality of results obtained, meet world instrument’s standards, and complementarity, the existing instrument which is 35 years old and cannot compete with the world best spectrometers (TOSCA at ISIS and LAGRANGE at ILL) now urgently requires replacement;
- attract new users from JINR member states and beyond;
- broaden considerably the spectrum of research topics, thereby increasing competitiveness, effectively raise the level of scientific work performed at FLNP;

we put forward the present case for major upgrade of FLNP Inelastic Neutron Scattering instrumentation.

To continue with the introduction on the neutron and optical spectroscopy methods, let us shortly outline the most important features of both.

Neutrons

- "Directly" probe atomic (nuclear)vibrations via simple two-body interactions. The underlying theory is simple and straightforward.
- There are no selection rules.
- Neutron has a mass. Hence 2D or even 4D spectroscopy can be carried out: $S(Q,\omega)$ or $S(Q_x, Q_y, Q_y, \omega)$.
- Can measure e.g. soft acoustic modes (esp. close to phase transition).
- Can routinely access the THz region ($< 400cm^{-1}$).
- Exhibit very large sensitivity to Hydrogen vibrations (due to exceptional incoherent scattering cross section of ^{1}H nuclei), but also to almost identical masses: $m_n \cong m_H$.
- Have a magnetic moment, hence magnetic excitations are measured easily.
- Are a highly penetrating probe. Light-absorbing materials (such as supported catalysts) studied easily.
- Cause a nearly-zero energy deposition in the sample.
- Their scattering cross section is isotope-dependent, hence isotope substitution can and is routinely used to refine spectroscopic information.

Photons

- Probe vibrations "indirectly" through associated variation of electronic properties, such as electric dipole moment or polarizability. Theoretical description is difficult, and requires advanced, many-body approach.
- Operate close to the Γ-point only, but usually with high resolution.
- Have very high sensitivity.
- Can ensure very fast experiments.
- Micro-spectroscopy is gaining popularity, both on synchrotron sources and through confocal in-plane scanning techniques.
- Some light-absorbing materials cannot be studied at all (such as supported catalysts).
- High energy deposition, high-intensity synchrotron radiation often destroys the sample.
- The laboratory instrumentation is easily accessible, low-resolution portable spectrometers can be purchased "off the shelf" for e.g. forensic investigation.

The above arguments give just the most important and most frequently considered characteristics concerning the complementarity when it comes to vibrational spectroscopy. **Fig.1.** presents the scope of inelastic neutron scattering spectroscopy as compared to other spectroscopic methods commonly used nowadays.

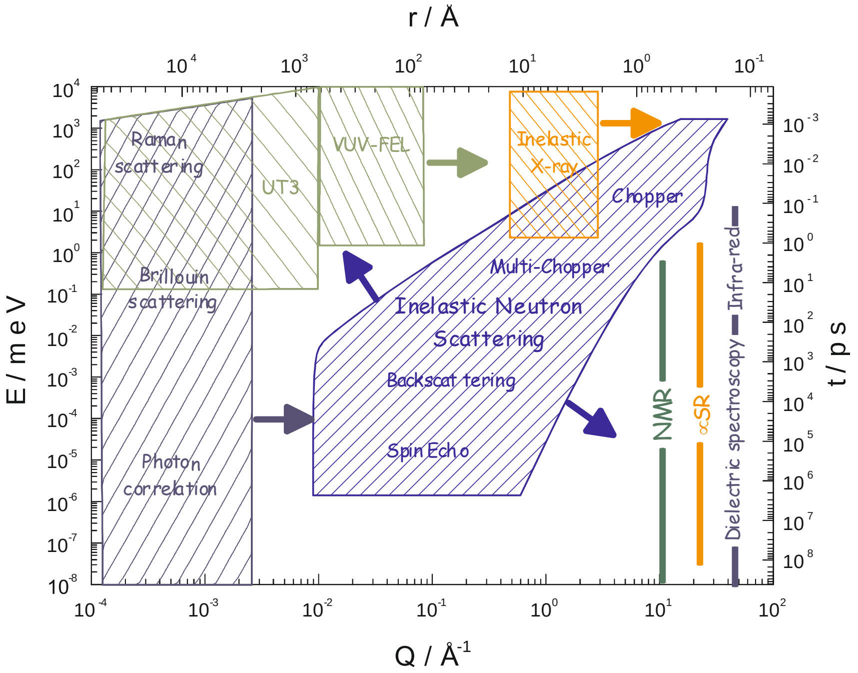


Fig. 1. Neutron spectroscopy on the map of various spectroscopic methods.

1.2. Published research examples of purposefully and fruitfully applied neutron spectroscopy

fac-Tris[2-phenylpyridinato-C^2, N]iridium(III) (tris-(2-phenylpyridine) iridium, Ir(ppy)$_3$ in short, was recently studied by S. Parker [1]. This is a metalloorganic compound, whose photo-emitting and photo-catalytic properties are of high importance to the industry. In particular, Ir(ppy)$_3$, as an emissive heavy metal complex, is the most frequently used precursor molecule for the synthesis of photonic materials. These are used in organic light emitting diodes (OLEDs). **Fig.2.** shows the comparison of neutron scattering, infrared and Raman spectra. For a complete vibrational characterisation, INS spectra are essential, especially if one needs to understand the low-energy metal-to-ligand modes. They are important in energy dissipation, which is an issue in these materials.

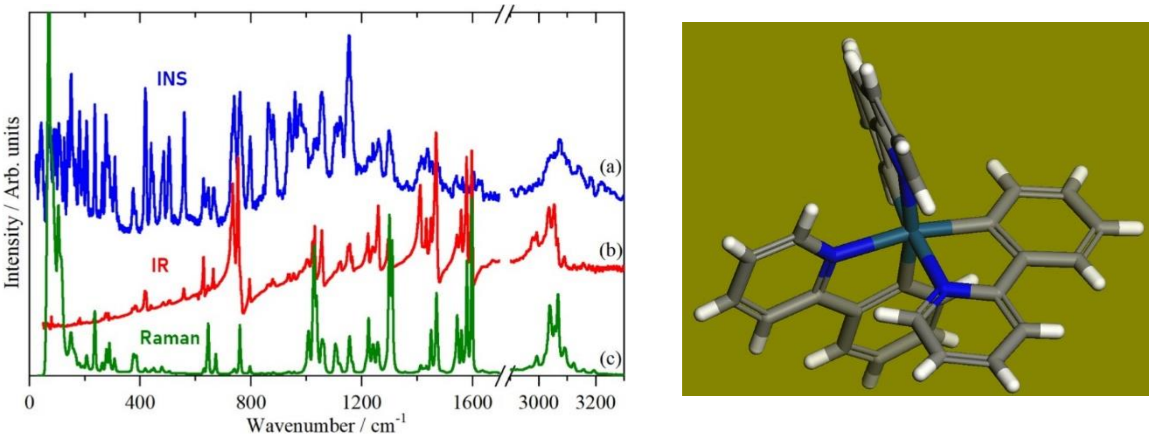


Fig. 2. Left: Vibrational spectra of fac-tris[2-phenylpyridinato-C^2, N]iridium(III). Right: The structure of the Ir(ppy)$_3$ molecule.

Another interesting compound of high applicational importance is N-phenylsuccinimide – an important compound in thermosetting resin industry. This compound has carbonyl group (C=O) in its structure. Vibrations of s carbonyl group are well-known to be a very sensitive probe of inter- and intramolecular interactions.

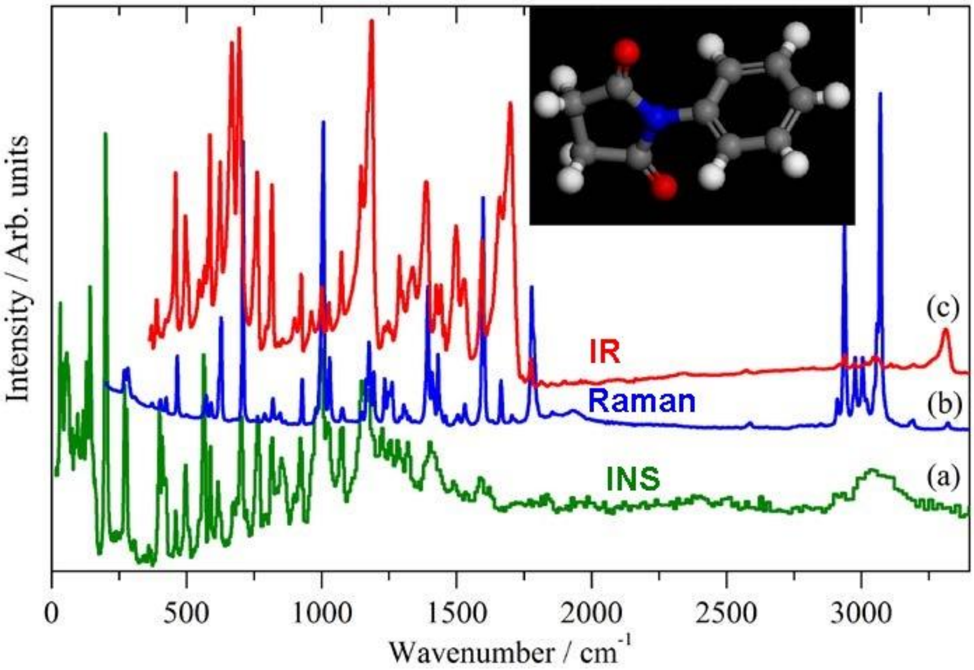


Fig. 3. Vibrational spectra of N-phenylsuccinimide.

Out-of-phase carbonyl stretch vibrations at 1700 cm^{-1} is very strong in the infrared absorption spectrum, weak in the Raman and absent in the inelastic neutron scattering.

On the other hand, the out-of-plane carbonyl bending mode at 407 cm^{-1} is strong in the INS, weak in Raman and absent in the IR [2].

The cited paper of S. Parker and co-workers (**Fig.3.**) illustrates the common limitation to laboratory mid-infrared absorption spectroscopy, and to Raman spectroscopy as well. This is the low-end cut-off (for mid-IR about 400 cm^{-1}). Raman is cut-off at lower wavenumbers, i.e. ca 200 cm^{-1}. Often IR can be extended towards 0 cm^{-1}, i.e. into the far IR region, however at a considerable cost and with experimental limitations. Inelastic neutron scattering routinely reaches towards 0 cm^{-1}, within a single experiment (no need to re-adjust optics, etc.).

There are cases, however, where light absorption spectroscopy is not feasible at all. Such are e.g. the supported metal catalysts where the absorption of light by the support results in a spectral cut-off that can be as high as 1400 cm^{-1}. For neutrons, most support materials e.g. alumina, silica, carbon, are transparent and the entire spectral range is accessible [2].

1.3. Examples of topics currently under the interest of the INS community at Frank Laboratory of Neutron Physics

Inelastic neutron scattering spectroscopy is based on the measurement of neutron energy and momentum transfers due to the interaction with the sample under study. The former can be achieved in two ways: either in the so-called direct geometry, whereby monochromatic beam is shined upon the sample, and the energy of the scattered neutrons measured, e.g. by time-of-flight method. Another way of measuring the energy transfer is called an inverted geometry, and it is easy to arrange on a pulsed neutron source, such as the pulsed reactor IBR-2. Here, pulses of "white" neutron beam are directed upon the sample, and incident neutron energy is measured by the time-of-flight method, while the final neutron energy is fixed by energy analyser. Note that the direct scattering geometry probes the whole (Q, ω) plane, while in the inverted geometry, the instrument measures along defined trajectories in the (Q, ω) plane.

On a pulsed source (IBR-2), especially with a long primary flight path, it is natural to build first an inverted geometry instrument. However, in order to complete the suite of INS spectrometers, a direct geometry one has to be planned for the future.

Before we proceed to the selected study cases, let us have a look at some challenges that Inelastic Neutron Spectroscopy meets worldwide.

- Study of small samples. This requires a well-designed focusing neutron optics. Most interesting materials for the investigation are usually available in very small amounts. Of special interest to the FLNP INS community are e.g.:
    - pharmaceuticals
    - matter under spatial confinement
- In highly correlated systems we need a 4D (Q_x, Q_y, Q_z, ω) spectroscopy. Example: topological Hall effect
- Access as large as possible range of timescales
- Study of magnetic multilayers
- Study of samples under extreme environments, e.g. very high pressures. This is being aimed at by the FLNP community for quite a long time
- Study of highly absorbing isotopes (as e.g. the performance of Li-ion batteries). Now this topic is of special interest to the FLNP community.
- Provide flexible resolution. Very useful, to be aimed at by the FLNP in future.

Nowadays the FLNP users of inelastic spectrometer are working on the following research topics. At the time the new instrument becomes operational, still "hotter" topics will certainly emerge.

These topics take into account the effective limitations to the existing neutron scattering instrumentation, both in terms of the accessible ω range (now up to 800 cm^{-1}, 1000 cm^{-1} at most) and too large incident neutron beam size $(5 \times 5$ cm).

- Molecular crystals and glass-formers at low temperatures in connection with
    - complementary studies and ab initio quantum chemical calculations.

    - Debye (‘crystal-like’) vs. non-Debye (‘glassy’) $G(\omega)$ at low ω at liquid He temperatures
    - dynamics in glasses of ODIC, CONDIS [3] and like phases
- Pharmaceuticals in bulk (native) state and as “micronized” or amorphized powders
- Matter under spatial confinement
    - “hard” nanomatrices (e.g. membranes)
    - “soft” confinement (e.g. microfibres)
- Materials for energy storage, e.g. plasticizer-SPE [4] systems for Li batteries
- Catalysts
- Photonic materials of industrial applications

Some of the challenges mentioned above cannot be addressed at FLNP in the nearest future, such as four-dimensional (Q_x, Q_y, Q_z, ω) spectroscopy due to the reasons which are outside the scope of our analysis.

Out of the above set, let us show two examples of science drivers that illustrate the elegance and power of inelastic neutron scattering spectroscopy if purposefully applied to particular research problems and carried out with top-quality instrumentation.

The study of soft and molecular matter at FLNP has a long history that once started with simple liquid crystals and molecular crystal complexes. Now it has evolved towards the physical phenomena behind phase transitions, and glass transitions in particular. The classical, in a sense, research by a Japanese group [5] is an excellent science driver for his type of work. It requires top-quality calorimetric studies plus a state-of-the art inelastic neutron scattering experiment to work out a scaling formula that brings together excess heat capacity measured by adiabatic calorimetry for a series of lithium borate glasses $x\,(\mathrm{Li_2O})\cdot\,(100-x)\,(\mathrm{B_2O_3})$ using the parameters of the excess(non-Debye) vibrational density of states measured INS. The scaling quality is shown in **Fig.4**. coming from [6].

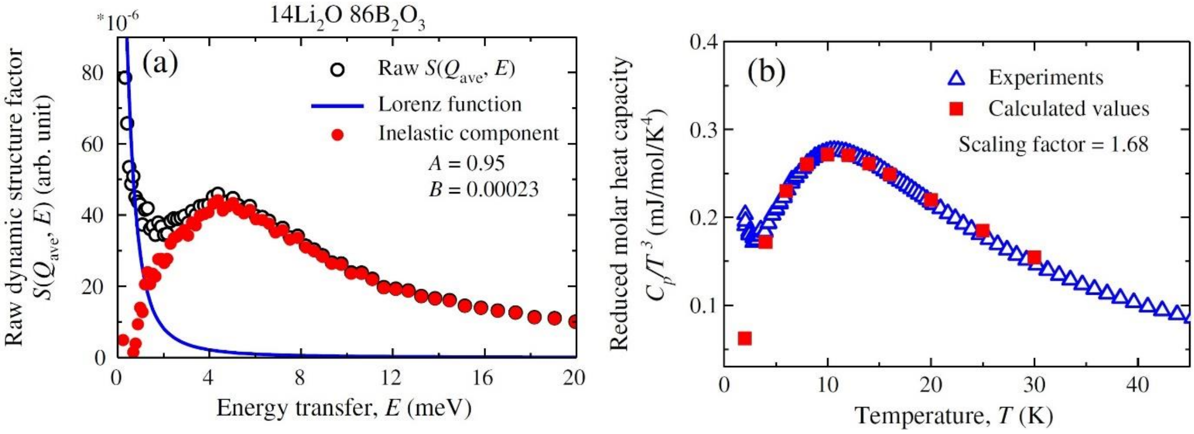


Fig.4. Scaling of adiabatic calorimetry excess heat capacity by low temperature excess density of states measured by INS.

Materials for lithium batteries are under extensive study by various experimental techniques. In order to build a high-performance battery, high internal currents have to be maintained, without side effects of e.g. generating excess heat that can result in unwanted accidents. Solvent-free solid polymer electrolytes (SPE) used therein, cannot meet the objective due to low ionic conductivity.

If PEO is taken as the basic polymer, then PEO-miscible electrolyte solvent can be used as the plasticizer to promote the segmental motion of PEO chains [6]. The amount of solvent is then increased. When the amount of the liquid plasticizer reaches a level where the ionic conduction is dominated by the liquid-in-polymer instead of the segmental motion of polymer chains, the SPE becomes a gel polymer electrolyte (GPE), and the internal mechanism of conductivity changes, as shown in **Fig.5**. The GPE then combines the advantages of the polymer electrolyte (high viscoelasticity) and liquid electrolyte (high ionic conductivity), and is of great significance in the Li-ion batteries.

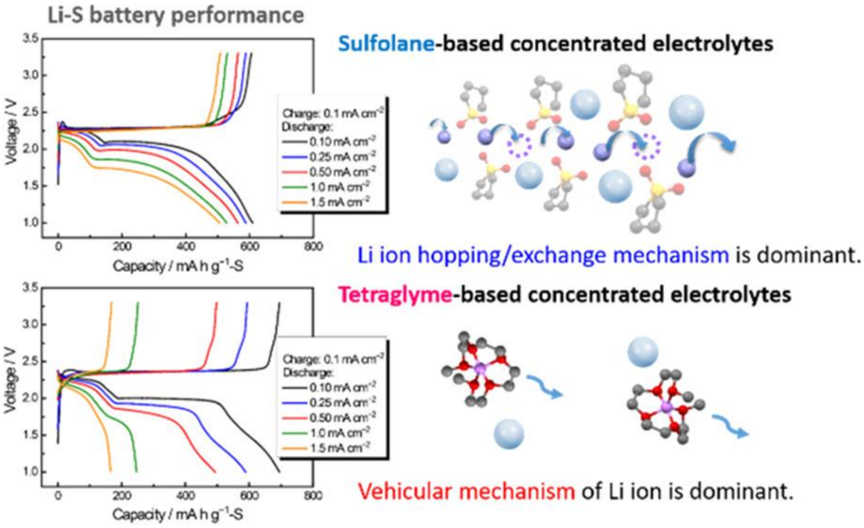


Fig. 5. The change of electric conductivity mechanism when solid polymer electrolyte (SPE) becomes gel polymer electrolyte (GPE) upon the addition of solvent-plasticizer.

In order to maintain the safety of batteries gel-polymer porous membranes are used to contain the electrolyte. This brings us to the research of soft matter under spatial confinement (currently underway), which is another science driver for the INS work in Frank Laboratory of Neutron Physics.

In-depth understanding of polymorphism of liquid crystals, as it is influenced by the chemical structure of molecules, by possible atomic substitution (e.g. fluorination) and many other is the crucial importance for both fundamental science and materials engineering. Low-level driver for the occurrence of structural transitions, and hence the polymorphism itself, is the inter- and intramolecular dynamics, influenced by the chemical structure of the molecules, and by all sorts of intermolecular interactions, such as hydrogen bonding, dipolar coupling, stacking interactions. Such dynamics is studied by means of a suite of spectroscopic techniques, and Inelastic Neutron Scattering is often of utmost importance, as it can easily access low-frequency modes via the vibration of hydrogen atoms, and other reasons mentioned earlier in this document.

Model DFT calculations were performed for two- or three-molecular clusters, as such systems are known to satisfactorily describe experimental vibrational spectra in the sense of band and frequency assignments (**Fig. 6**).

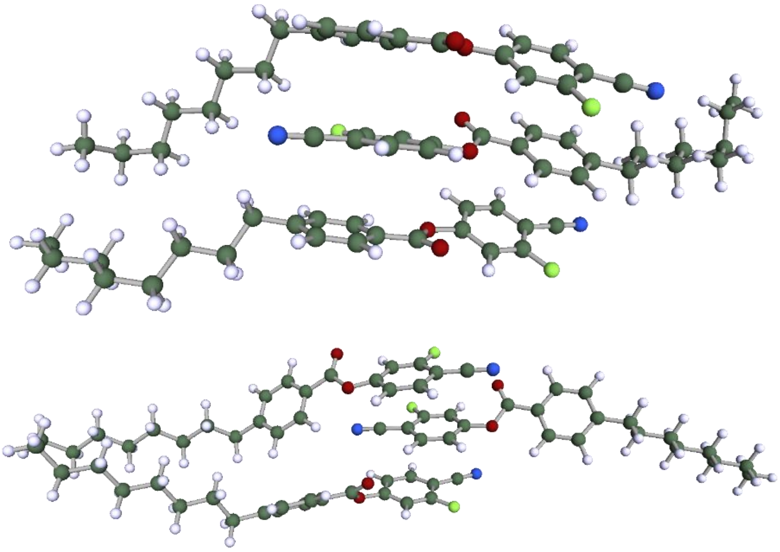


Fig. 6. Two of a few possible arrangements of the 8CFPB molecules in a cluster kept together by weak non-covalent interactions, yet quite stable, used for model calculations. After DFT optimization with triple-zeta basis functions and dispersion corrections.

Comprehensive study of the homologous series of 4-cyano-3-fluorophenyl 4′-n-benzoates (abbreviated nCFPB) [7] is an example of research along this line. **Fig. 7**. brings a comparison of INS spectra with model calculations for three members of this series.

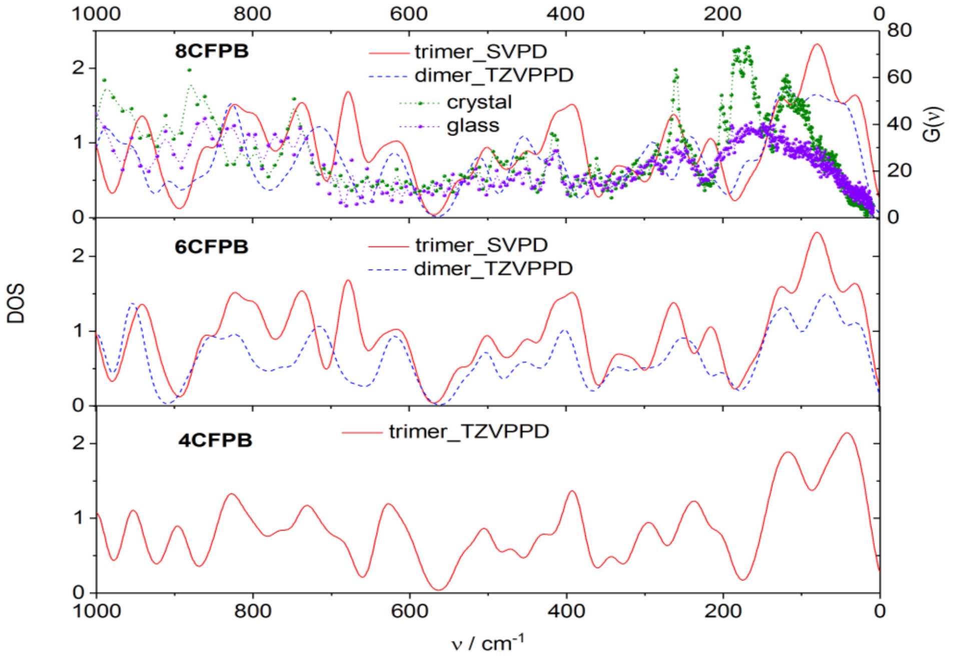


Fig. 7. INS spectra of three nCFPB homologues compared with model calculations

This work is still underway, and more results will be published in 2020. nCFPB molecules used to form model clusters can be arranged and energy-minimized in a number of ways, as well as a variety of molecular conformations have to be taken into account and more plausible solutions have already been obtained. Of special interest here is the nature of glassy state, and the low frequency part of the INS spectrum al liquid helium temperature in general. This region is not accessible to the mid-infrared FTIR spectroscopy.

It is well known that non-crystalline forms of pharmaceuticals are being sought as their biocompatibility is usually much higher than that of crystal phases. Single phenyl ring alcohols resemble the structure of several pharmaceuticals, and in-depth understanding of their properties is likely to facilitate the quest for pathways to obtain the desired forms of drugs or drug carriers. The aim of the work carried out, among other techniques, by means of Inelastic Neutron Spectroscopy at the NERA spectrometer of FLNP was to determine changes of vibrational dynamics of alcohols with globular shape of molecules containing a single phenyl ring, and their fluorinated isomers, i.e. 2-(trifluoromethyl)phenethyl alcohol (2TFMP), 3-(trifluoromethyl)phenethyl alcohol (3TFMP) and 4-(trifluoromethyl)phenethyl alcohol (4TFMP), with following structural formula: $CF_3C_6H_4CH_2CH_2OH$, see **Fig. 8**.

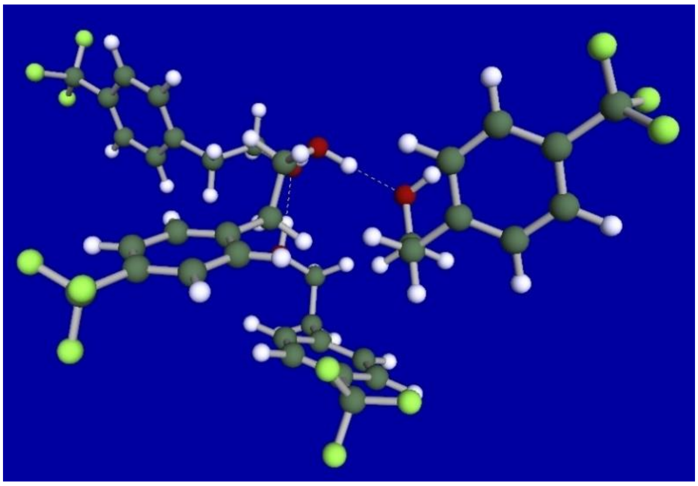


Fig. 8. A cluster of four hydrogen-bonded 2TFMP molecules

The compounds studied are glass-formers, and cold crystallization of metastable supercooled liquid state was evidenced by us. Inelastic neutron scattering was measured in a broad temperature range of 5 K – 255 K and the systematics of changes to the vibrational density of states $G(\omega)$ analysed. Vibrational dynamics seen by INS at the low temperature (5 K) reveal details of molecular properties in the following thermodynamic states:

- glass of supercooled liquid (fast cooling) and
- the crystal phase (slow cooling, cycles).

The INS and density of states $G(\omega)$ measured at 5 K illustrate well the difference between both states (**Fig. 9**).

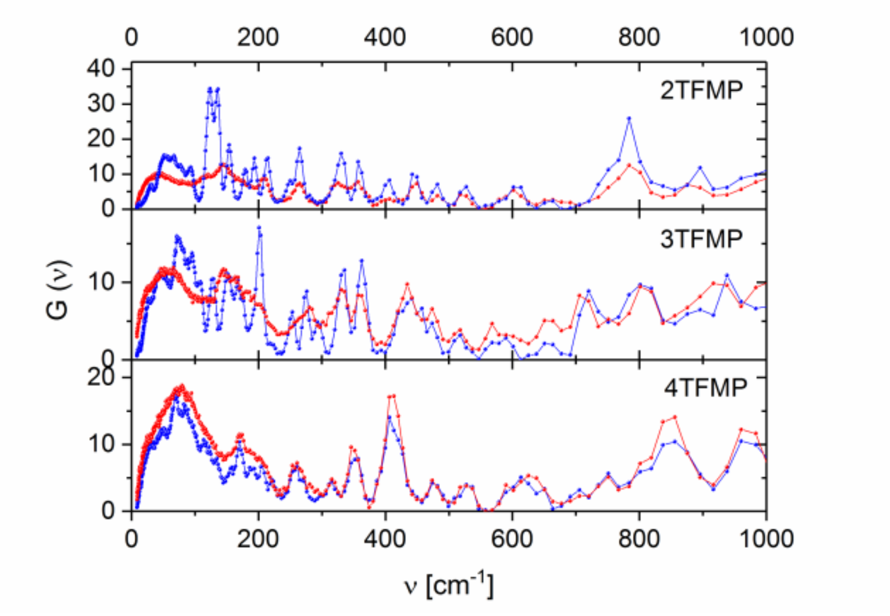


Fig. 9. Density of states spectra of solid phases, i.e., crystal (blue lines) and glass of isotropic liquid (red lines) measured at 5 K for 2TFMP, 3TFMP and 4TFMP.

Comparing low temperature spectra of ordered crystal phase with glass of isotropic liquid phase one can easily notice a broadening of vibrational modes caused by rotational and translational molecular disorder frozen-in in the glassy state. Below intramolecular vibrations (in a crystal these are phonon excitations), another significant difference appears, i.e., the boson peak. Torsional motions of -$(CH_2)_2$ (functional groups connected to phenyl ring) are visible on $G(\omega)$ spectra in the lattice vibrational region due to the low energy barrier. Some results of our investigation have been already published [8].

nTFMP forms a so-called fragile glass. This has been evidenced by the analysis of the boson peak, measured by INS at low temperatures (**Fig. 10**).

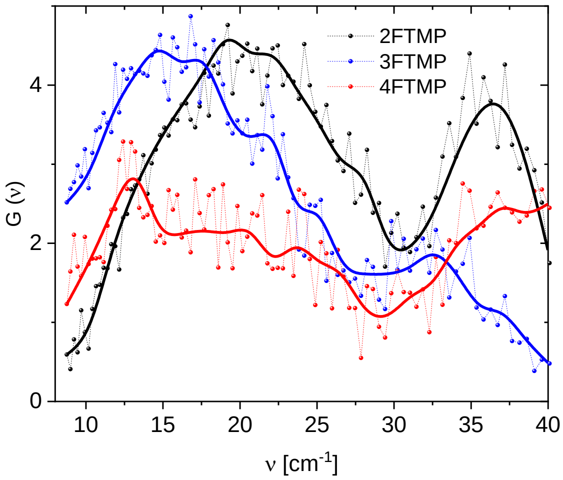


Fig. 10. Low intensity of the boson peak evidences a fragile glass.

Ethosuximide (ETX), **Fig. 11** is a medicine used to treat absence seizures. It is an example of a drug, whose physical form plays an important role in biocompatibility, and hence dose/efficacy ratio.

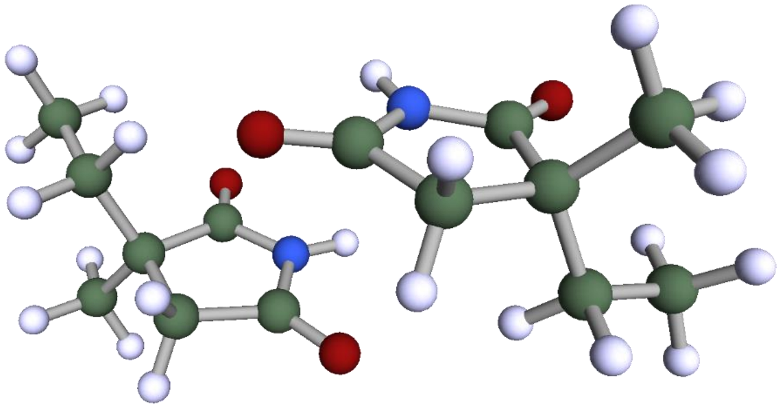


Fig. 11. Two Ethosuximide (ETX) molecules and their preferential arrangement (ring stacking), according to the DFT calculations.

The dynamic details obtained from density of states spectra $G(\omega)$ (**Fig. 12**) have been analysed against vibrational wavenumbers (harmonic frequency) obtained for isolated molecule of ETX following ab initio calculation. It was in the course of these calculations that three stables, almost isoenergetic conformers of ETX were found.

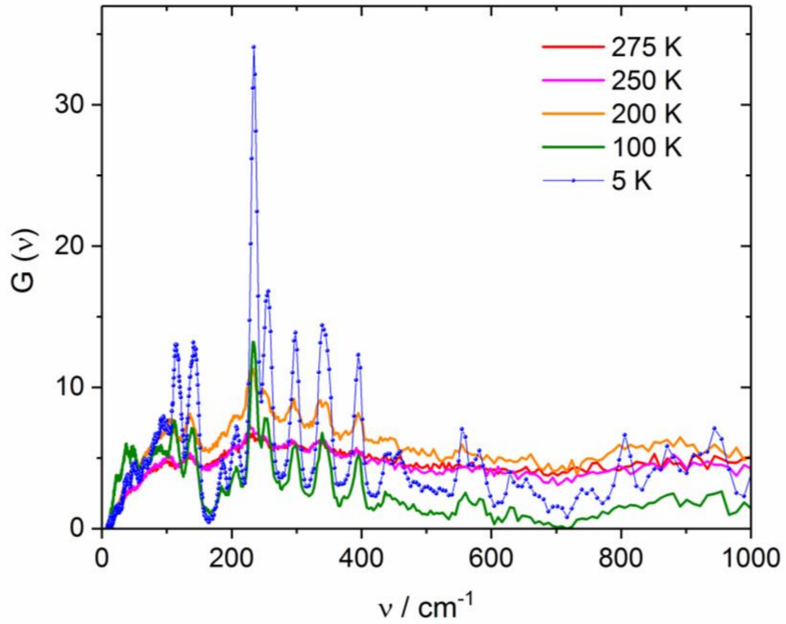


Fig. 12. Vibrational density of states G(ω) of Ethosuximide, as obtained from the INS spectra measured on the NERA instrument.

Torsional motions of protons from functional groups CH_2, CH_3, CH of the imide ring, and the N-H vibrations are visible on $G(\omega)$ spectra in the lattice vibrational (low frequency) region due to the low energy barrier. The motion involving the torsion angle made up by the ethyl chain (C10, C9), carbon atom from imide ring (C6) and methyl group (C7) of ETX is relevant for conformational flexibility within the molecule.

1.4. Scientific community at the FLNP JINR

The scientific community of FLNP mainly comes from the scientific members states of JINR. Within the past decade they have been carrying out studies of molecular dynamics using the existing spectrometers NERA and DIN-2PI. FLNP hosted both regular users as well as new user groups who applied for the beam time only recently.

Russia:
- Department of Nuclear Physics Research Methods, St Petersburg University, St.Petersburg
- Institute For Nuclear Research RAS, Moscow, Troisk, Russia
- the Institute of Solid State Physics of Russian Academia of Science, Chernogolovka
- RUDN University, Moscow
- Institute of Metallurgy UrD RAS, Ekaterinburg
- National Research Nuclear University Moscow Engineering Physics Institute, Moscow
- General Physics Institute, RAS, Moscow
- Nikolaev Institute of Inorganic Chemistry (NIIC) of the Siberian Branch of the Russian Academy of Sciences, Novosibirsk
- Institute of High-temperature Electrochemistry UB RAS Ekaterinburg

Poland:
- Faculty of Physics, Adam Mickiewicz University, Poznań
- Institute of Nuclear Physics Polish Academy of Sciences, Kraków
- Faculty of Chemistry, University of Wrocław, Wrocław
- West Pomeranian University of Technology, Faculty of Chemical Technology and Engineering, Department of Organic Chemistry and Physical Chemistry, Szczecin
- Department of Chemistry, University of Lodz, Łódź
- Department of Chemical Physics, Faculty of Chemistry, Jagiellonian University, Kraków
- Institute of Chemistry, University of Natural Sciences and Humanities, Siedlce
- Institute of Nuclear Chemistry and Technology, Warsaw

Germany:
- Forschungszentrum Jülich, Jülich
- Helmholtz Zentrum für Materialien, Berlin, Germany

Romania
- National Institute for Research & Development in Electrical Engineering ICPE - Advanced Research Bucharest

Slovak Republic
- Institute for Experimental Physics, SAS, Kosice

Ukraine
- Institute for Problems of Material Science, UAS, Kiev

Bulgaria
- Institute for Nuclear Research and Nuclear Energy, Sofia

1.5. Concluding Remarks

Presented in brief were just a few out of a broad range of state-of-the-art scientific areas that will benefit from the construction of the new INS Spectrometer. One of the advantages of the new spectrometer at IBR-2 reactor will be its high luminosity. This feature leads to a better, almost loss-less use of the neutron beam, resulting in a significant shortening of the experiment duration time and the possibility to work with a small mass of the sample. Altogether, the instrument proposed will be the best one as far as the neutron use efficiency is concerned. The results obtained with the new spectrometer will be at the level of those delivered by the leading neutron scattering instruments in Europe.

Having set these basic concepts, we now proceed with the instrument planning and optimization.

1.6. References

[1] S.F. Parker, J. Phys. Commun. **3** (2019) 065010

[2] S.F. Parker et al, Journal of Physics: Conference Series **554** (2014) 012004

[3] ODIC – orientationally disordered crystal; CONDIS – conformationally disordered

[4] SPE – solid polymer electrolyte

[5] Yu Matsuda, H. Kawaji, et al., J.Non-Cryst. Sol. **357** (2011) 534–537

[6] A. Nakanishi, K. Ueno, et al., J. Phys.Chem. C 2019, 123, 23, 14229–14238

[7] E. Juszyńska-Gałązka, W. Zając, Mesomorphic behaviour and vibrational dynamics of nCFPB liquid crystalline homologues, Phase Transit., 92 (2019) 1077-1088

[8] E. Juszyńska-Gałązka, W. Zając, Y. Yamamura, K. Saito, N. Juruś, Vibrational dynamics of glass forming: 2-phenylbutan-1-ol (BEP), 2-(trifluoromethyl)phenethyl alcohol (2TFMP) and 4-(trifluoromethyl)phenethyl alcohol (4TFMP) in their thermodynamic phases, Phase Transit., 2018 91 170-185.

2. Instrument Concept

2.1. Layout

The time of flight spectrometer in inverted geometry is a relatively inexpensive and very efficient instrument for neutron spectroscopic studies and practically the one ideally suited for pulsed neutron source.

The main efforts in the design project are focused on (Fig. 13):

- the primary spectrometer - neutron guides, choppers system, splitter;
- the secondary spectrometer - system of analyzers (cooled beryllium filters, HOPG), detector system.

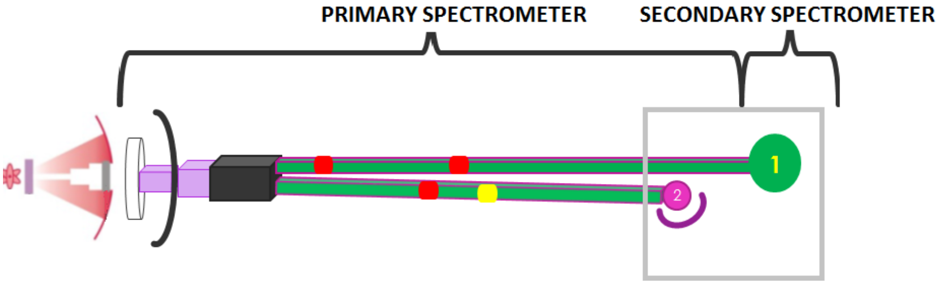
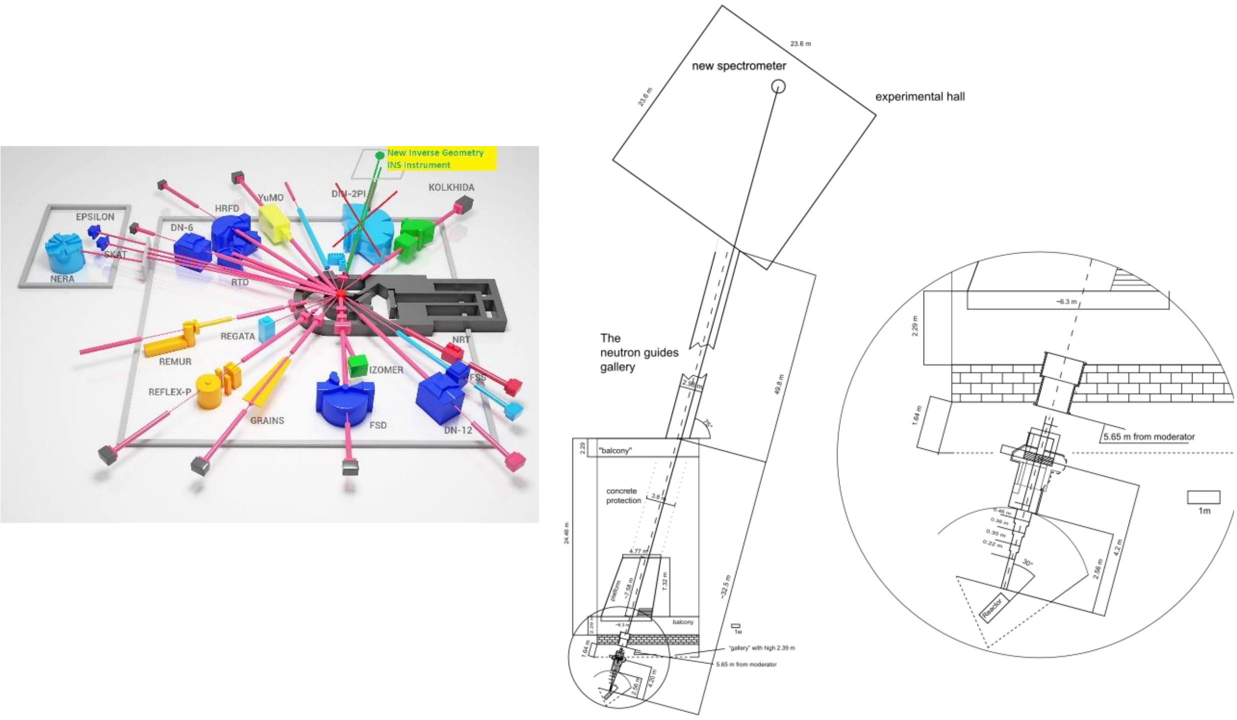


Fig. 13. The layout of the new spectrometer.

The new inelastic spectrometer will be installed at the channel 2 in the building 117/1 at the distance of 105 m from the moderator, replacing the DIN2PI direct geometry instrument which will be decommissioned. (**Fig. 14**).



a) *b)*

Fig. 14. a) and b) Planned position of the new inverse geometry.

The maximum possible distance between the source and the sample (~105 m) was chosen in order to have best possible resolution and energy range.

2.2. Primary spectrometer

The primary spectrometer comprises three main elements:

2.2.1. Splitter

The moderator has an area of 33.5 x 40.5 cm2 (WxH). Half of the area should be reserved for future installation of cold neutron source (CNS). In our simulation only the lower half of the moderator was taken into account i.e. the area of 33.5 x 20.25cm^2 was assumed for calculations. For simplicity was assume no neutrons are produced by the other half of the moderator as the CNS will suppress the thermal part of the neutron spectrum. In view of the perspective to build the second inelastic scattering instrument (direct geometry) on the same neutron channel, a splitter is foreseen so that another guide tube can be fed from the same moderator area.

2.2.2. Chopper systems

A cascade-arranged suite of choppers is foreseen to serve the following purposes:

- Pulse shaping. The desired pulse shape will be formed by a double-disk chopper located at the very beginning of the neutron guide, at a distance of 2.6 m from the moderator. Two counter-rotating disks with adjustable relative phase will produce a symmetrical pulse shape of tunable duration needed to vary the instrument resolution, especially in the high energy transfer part of the spectrum.
- Suppression of the delayed neutron background.
- Bandwidth definition (BW) and frame overlap (FO) prevention. Together with long-wavelength mirror filter the chopper system, located in the central part of the guide tube, will effectively cut out unwanted cold neutrons.

It is expected that disk of 700 mm in diameter will be used. The exact width of all disk windows will be defined after the final guide design is fixed.

2.2.3. Neutron guide with focusing end-part

The neutron optics will be optimized for the 0.5-1 Å wavelength band (thus for the large values of energy transfer 80-330 meV), and capable of fully illuminating samples two sizes: standard 3x3 cm^2 and small 1x1 cm^2 (as an option).
Based on the previous experience the constant cross-section guide equipped with a long focusing end-part has been chosen as the simplest solution. The model used during the optimization is depicted in **Fig. 15**. Shown in red is the focusing end-part. Two possible end-part geometries were considered: elliptic and parabolic ones. The full elliptic guide can be regarded as a particular case when the length of the focusing end-part is equal to the total guide length. Parameters to be defined

include: main guide height, focusing end-part length, positions of the foci (2 in elliptic case and 1 in parabolic) defining the shape of the focusing nose.

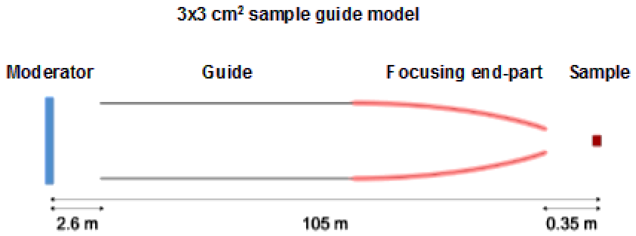
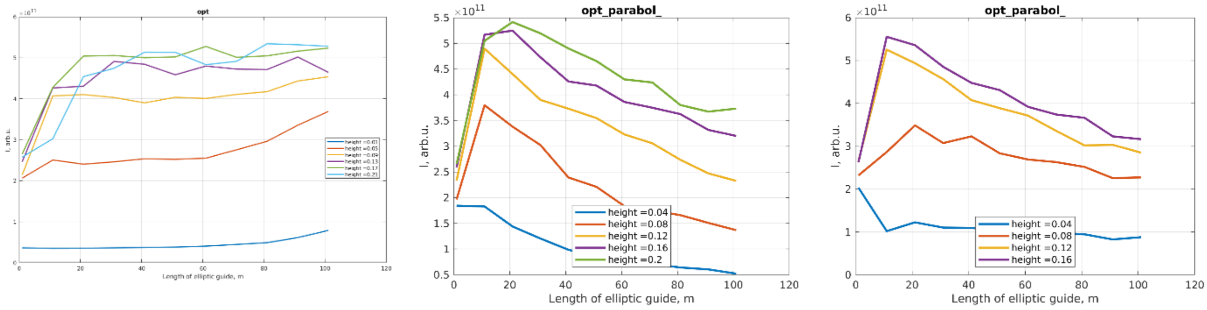


Fig. 15. Guide model used during the optimization.

The relative inefficiency of the ballistic geometry in our case is related to several facts:

- Large moderator volume of 15x20 cm^2
- Large distance between the source and the optics (4.2 m)
- Thermal-to-hot neutrons usage.

Fig.16 a). shows sample flux dependence upon the elliptic end-part length for several guide heights. For each point, a set of optimal ellipse parameters (left and right foci) has been found using particle swarm optimization. Two important conclusions can be made:
- There is no reason to expand constant cross-section guide dimensions above certain value. In this case guide height of 13-17 cm could be optimal;
- There is no reason to expand end-part length above certain value. In this case, the end-part length of 20 m could be optimal.
Figs. 16b) and 16c) show similar dependences calculated for a parabolic end-part, varying both the vertical and horizontal dimensions. Important feature of the parabolic end-part shape is a clearly visible maximum instead of plateau in the elliptic case. Both cases provide similar sample fluxes.



a) b) c)

Fig.16.a) Sample flux dependence upon the elliptic end-part length for several guide heights. b) Sample flux dependence upon the parabolic end-part length for several guide heights. c) Sample flux dependence upon the parabolic end-part length for several guide widths

Neutron optics starts at the distance of the 2.6 m from the moderator surface. At this position only the lower half-plane of the moderator surface was considered available to save space for the possible future second guide. Reflectivity R was approximated by the standard formula (1):

$$R = \begin{cases} R_0 & Q \le Q_c \\ \frac{1}{2} R_0 \left[1 - \tanh\left(\frac{Q - mQ_c}{W}\right)\right] \left[1 - \alpha(Q - Q_c)\right] & Q > Q_c \end{cases} \qquad (1)$$

where R_0 - reflectivity in the total reflection regime, W- width of the cutoff at mQ_c, α - slope of the reflectivity in the supermirror regime. Based on the data from SwissNeutronics [3] (see **Fig.17**) the following values were used in all cases: $R_0 = 0.99$, $Q_c = 0.0218$ Å^{-1}, $\alpha = 3.3$ Å and $W = 0.003$ Å^{-1}.

In all optimization simulations for simplicity we used m = 7 coating. The coating optimization is a subject of a further study. As long as we stay within the best possible performance, the guide geometry is independent from the choice of the m value (probably in the range of m = 3 - 7 for constant cross section part). Should a lower value of m be decided upon, providing a smaller flux (for any practical reason such as cost or production time) the geometry should be reoptimized.

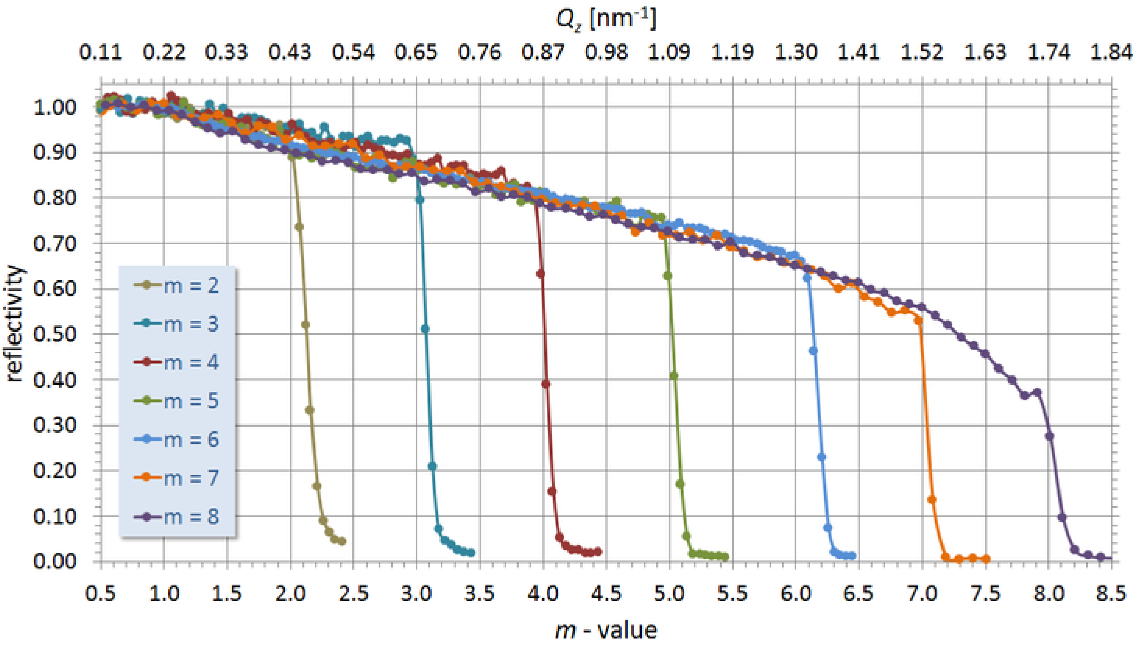


Fig.17. SwissNeutronics supermirrors reflectivity profiles [1].

2.3. Secondary spectrometer

We have started engineering design of the secondary spectrometer and now we have decided on the possible configuration of the instrument. The basic concept of the secondary spectrometer is outlined in **Fig. 18**.

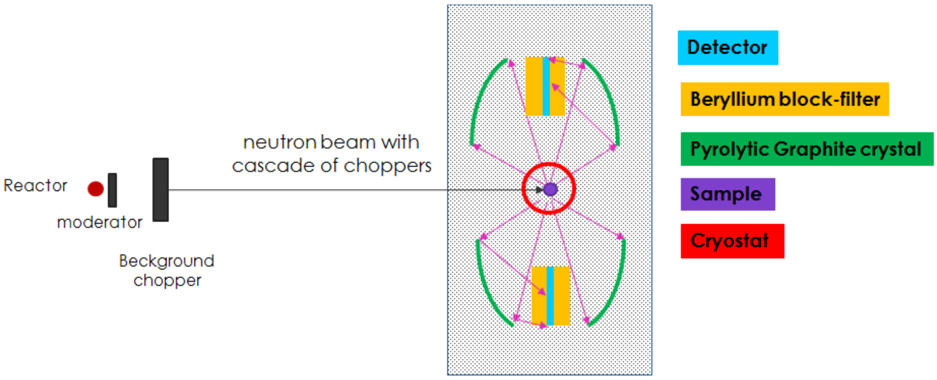


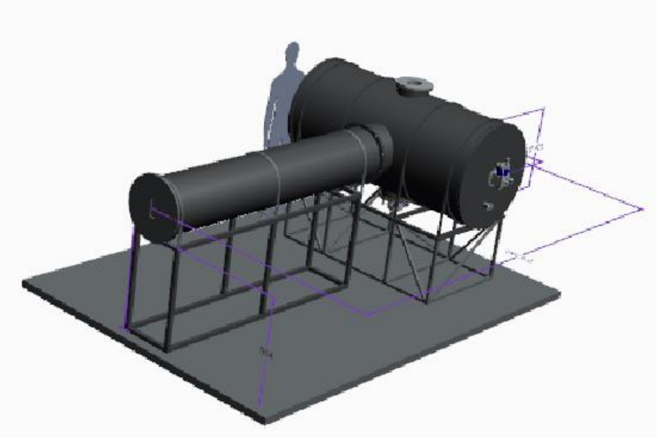
Fig.18. A schematic top view of the proposed secondary spectrometer.

The overall structure is composed of two bell-shaped sets of Pyrolytic Graphite Analysers, symmetrically placed with respect to the sample plane.

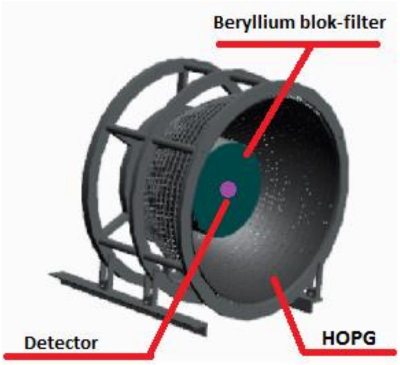
The optimal choice for the material for the reflecting surface turns out to be a broad-mosaic pyrolytic graphite, able to reflect a relatively wide neutron energy band around 4.5 meV with a reasonable (~70%) peak reflectivity. Cooled beryllium blocks dedicated to filtering out higher order reflections from graphite crystals will be installed around the detector inside the bells with HOPG crystals.

Conventional ^{3}He detector banks are planned to be used.

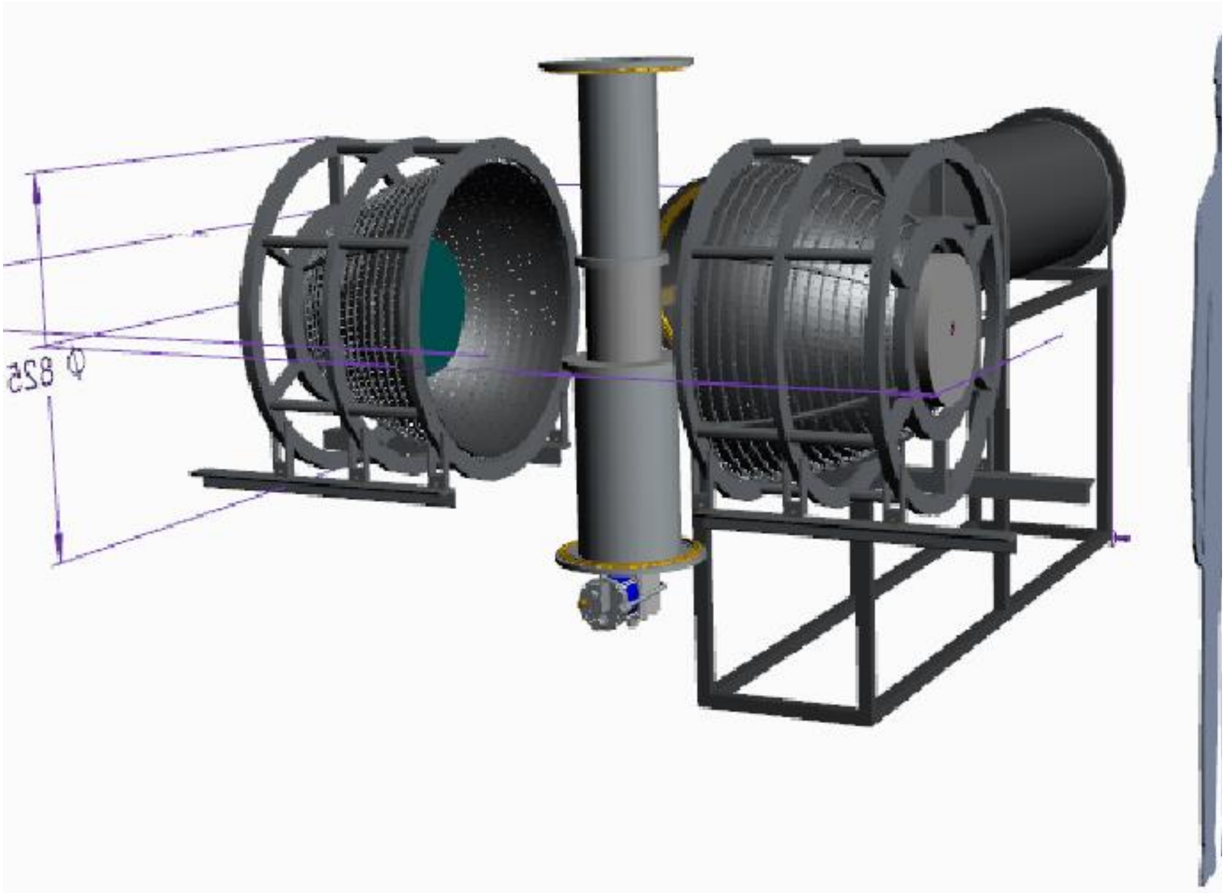
The conceptual design of a new inverse geometry INS spectrometer for FLNP JINR is depicted in **Fig. 19**.



a)



b)



c)

Fig.19. Schematic view of a new inverse geometry INS spectrometer: a) in vacuum container; b) one bell-shaped system of Pyrolytic Graphite Analysers; c) Main constituent parts without vacuum container.

In the nearest future the simulation of secondary spectrometer will be finished. Knowing the geometry of the new instrument, we will determine the size of analyser plates (Highly Orientated Pyrolytic Graphite) and details of their positioning. This part of the design work is of utmost importance, science further repositioning of a large number of rather expensive HOPG plates should be avoided at any cost.

Until now, just a conceptual design has been worked out. The next steps, consisting in further digital modelling and designing of construction details are to be taken without unnecessary delay.

The design of the supporting frame, construction of Pyrolytic Graphite holders and vacuum system as well as cryogenic system and detectors system will be performed after the project becomes approved.

[1] C Schanzer, M Schneider, and P Boni. Neutron optics: Towards applications for hot neutrons. In Journal of Physics: Conference Series, volume 746, page 012024. IOP Publishing, 2016.

2.4. Expected characteristics

The expected characteristics of the new INS spectrometer ware compared with this from NERA spectrometer.

	NERA	New INS Spectrometer	Comments
Analyzer area	15x3x25 1125cm^2	10000x2 20000см^2	
Ratio input/output to neutronguides	16x5cm^2/5x5cm^2 3.2	20x20cm^2 / 3x3cm^2 44.44	a gain in flux density (without taking into account the higher quality of the neutronguide) 44.44/3.2 = 14
Solid angle	~ 0.2 sr	~ 2 sr (1 pc of new spectrometer)	Solid angle gain 18
Ratio of luminosity of new spectrometer and NERA			18x14 = 250 times higher i.e. measurements of a sample with the mass of 10-20 mg will be possible.

3. Human resource assessment

Table 1.

Employees involved in the project.

№	Name of Division / Department / Group	Number of persons	Amount of employment
1	FLNP DCMRD DNICM	Chudoba D. +1	full-time
2	FLNP DCMRD DNICM	Goremychkin E.	full-time
3	FLNP DCMRD	Belushkin A.	part-time
4	FLNP DCM	Bodnarchuk V.	part-time
5	FLNP DCMRD DNICM	Kruglov A.+5	part-time
6	FLNP DCM Group № 1	Churakov A. + 1	part-time
7	FLNP DCM Group № 5	Chernikov A. +1	part-time
8	FLNP DCM Group № 4	Altynov A. + 1	part-time
9	FLNP DCM Group № 3	Petukhova T. + 1	part-time (by request)
10	FLNP DB	Kustov A. + 1	part-time (by request)
11	FLNP workshops	Kuznetsov A.	part-time (by request)

4. Partner companies and equipment suppliers

- FRAKO-TERM, Poland
- SwissNeutronics, Switzerland
- FP AMU, Poland
- INP PAN, Poland

5. Time schedule of activities

Form № 26

Proposed time schedule and required resources for realization of first part of the project «Creation of an inelastic neutron scattering spectrometer in inverse geometry at the IBR 2 reactor»

Description of units and systems, resources, funding sources	Cost of units (k$). Resource requirements for 1st part of the project	Proposals of the Laboratory for distribution of funds and resources		
		2021	**2022**	**2023**
Neutron guide	**2500**	600	900	1000
Construction design and technical specification	**250**	250	-	-
Highly Orientated Pyrolytic Graphite	**550**	150	250	150
^{3}He Detectors and electronics	**50**	-	-	50
Manufacture of vacuum, cryogenic systems and beryllium filters	**350**	250	50	50
Total	**3700**	**1250**	**1200**	**1250**

Project costs will be distributed by: new theme “Studies of functional materials and nanosystems using neutron scattering”, new theme “Scientific and methodological support for studies of condensed matter in extracted neutron beams”, theme 1105 and Grants of Polish Plenipotentiary at JINR.

The second part of the project is planned to cover rest costs for neutron quide system, manufacture of vacuum, cryogenic systems and beryllium filters (~2000k$).

Project leader D. Chudoba

6. Cost estimate

Form № 29

Cost estimate of the first part of the project « Creation of an inelastic neutron scattering spectrometer in inverse geometry at the IBR 2 reactor »

№	Description of cost items	Total cost	2021	2022	2023	
Direct expenses						
1	Design	k$	250	-	-	**250**
2	Materials	k$	150	250	200	**600**
3	Equipment	k$	850	950	1050	**2850**
4	Payment for research performed under contracts	k$	40	40	40	**120**
5	Travel expenses	k$	10	10	10	**30**
Total		k$	**1300**	**1250**	**1300**	**3850**

PROJECT LEADER

FLNP DIRECTOR

FLNP LEADING ENGINEER-ECONOMIST