II-2. Modeling and measurements of neutronic properties of new cryogenic neutron moderators

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Summary

With the advent of new projects to build intense neutron sources, and the requirement of copious fluxes of long wavelength neutrons, a revived interest in the development of a new generation of cold neutron sources has prompted an intense wave of efforts all around the world. Obviously, that means improving our present knowledge on existing materials and the development of new ones, in order to achieve higher levels of performance. Most likely, some of those new cryogenic moderators will consist of molecular systems in solid form. At the Neutron Physics Division of Centro Atomico Bariloche (Argentina), we have been working on both theoretical and experimental aspects in relation to the production of neutron cross-section data for molecular liquids and solids. In the context of this CRP we plan to apply that expertise to explore new materials that can be used as cold or very cold neutron moderators with improved performances respect to the existing ones. Our central activity will focus on the development of new physical models to describe those materials, and from that the production of scattering kernels and the generation of the cross section data in a proper format for MCNP calculations. On the experimental side, we wish to build and put into operation a new cold neutron moderator at our pulsed neutron source based on the Bariloche LINAC, to extend towards the cold neutron energy range our present experimental capabilities. We expect to collaborate with other participants of this CRP especially in connection with experimental validations of the new scattering kernels through neutron total cross-sections and spectra measurements.

Previous related work at NyR

The relevant quantity to describe the interaction of thermal neutrons with condensed matter is the Van Hove scattering function \( S(Q,\omega) \), as it embodies all the dynamical and structural information about a scattering system. Although first principles theories to evaluate the scattering function exist, the resultant expressions are usually not appropriate for calculational procedures and, moreover, a detailed knowledge of the scattering function over a wide range of energy \( (\hbar\omega) \) and momentum \( (\hbar Q) \) transfer is not required in many cases.

Those ideas were part of our main motivations for the development of a 'Synthetic Scattering Function', which incorporates the main dynamical characteristics of the molecular unit, still retaining a high degree of simplicity in its formulation. The main advantages of this model reside in the analytical expressions that produce for several magnitudes of interest in the field of Neutron and Reactor Physics. These include double-differential cross sections, energy-transfer kernels, and total cross sections, which in turn allow the evaluation of neutron scattering and transport properties.
More recently, we have introduced the concept of a Synthetic Frequency Spectrum (SFS) to represent in a simple manner the density of states of a molecular solid. Using this approximation together with the standard NJOY code, we were able to obtain very good agreement with available experimental results, for example for solid Methane in Phase I, and make reliable predictions for the cases of water ice and Methane Hydrate.

In addition, we have performed total cross section measurements using our pulsed neutron source based on the Electron LINAC facility of Centro Atómico Bariloche, on samples at room and low temperatures, in order to compare the experimental data with the calculated total cross sections from the nuclear data libraries generated at the same temperatures.

Much of those activities of the Neutron Physics Group were motivated by our participation in the International Collaboration on Advanced Cold Moderators (ACoM), which gathered a dozen of the leading groups in the world concerning cold neutron moderator development.

As it is reflected through our recent contributions in matters related to the present project (see ANNEX), activities have been in progress at Neutron Physics Division in theoretical, calculational and experimental aspects involving the study of hydrogenous materials of interest as cold moderators. The majority of results so far are in the form of new cross section libraries generated with the use of the NJOY code, starting from the physical models developed in our group and partially validated by comparison with total cross sections measured by us.

**Scientific Scope and overall Work Plan of the Project**

Cold neutrons are widely used in different fields of research such as the study of the structure and dynamics of solids and liquids, the investigation of magnetic materials, biological systems, polymer science, and a rapidly growing area of industrial applications. Besides the already classic reactor-based cold neutron sources, accelerator-based cold sources have also proved to be very successful in sustaining high class neutron scattering studies on the above mentioned fields.

The development and optimization of advanced cold neutron sources require neutronic calculations involving thermal and subthermal neutron energies, which in turn demand the knowledge of reliable cross section data relative to the materials which conform the system under consideration. The compromise solution adopted in standard Nuclear Data Libraries involves the inclusion of scattering cross sections for a few common moderators at some selected temperatures, and data for any different material or physical condition must be ‘constructed’ from pieces of information actually corresponding to those few cases found in the existing files.

Within this project we will complete the development of scattering kernels for a number of molecular systems of interest as cold moderator materials, with particular emphasis in methane and hydrogenous methylated aromatics such as toluene, mesitylene, xylene, and mixtures of those, including for some special cases consideration of their different solid phases. In order to validate those new libraries, we expect to compare predicted total cross sections and neutron spectra with experimental data obtained in our laboratory and at other facilities, like LENS at Indiana University and the Hokkaido LINAC. The Japanese source is well equipped for spectra measurements and indeed that group has a large experience in that sort of experiments. On the other hand, LENS seems to be an exciting new medium pulsed
neutron source, and no doubt it will be offering unsurpassed capabilities to study properties revealed by cold and very cold neutrons.

During a second phase of this project, we will tackle the problem of describing the interaction of slow-neutrons with deuterated molecular solids, typically d-methane and d-mesitylene at very low temperatures. Those materials are of primary interest as moderators for very- and ultra-cold neutrons, on account of their very small absorption cross sections and the expected shift in the excitation energies due to isotopic effects. Although interference phenomena revealed by the large coherent scattering length of Deuterium play no major role in the thermalization process, it will be useful to take it into account in order to compare model predictions with (single) scattering experiments.

Finally, we will construct the new cold source at our electron LINAC based neutron source, which has been recently designed and optimized according to a modular concept that offers great flexibility in terms of neutron intensity and pulse width. The design is based on solid mesitylene as moderator material.

**Work Plan for the first year**

During the first year of the project we will complete the validation of the scattering kernels that we have developed in recent years, through comparisons of their predictions with experimental data. The hydrogenous materials involved are: methane, liquid hydrogen, ice, methane hydrate, benzene, toluene, mesitylene, and mixtures of the two later, and the libraries will be produced at different temperatures starting from 4K. When phase transitions occur within the range of temperatures considered, the changes in the dynamics of the scattering system will be accounted for in the models.

Concerning experimental activities, measurements on some of the above materials will be done to determine their total cross section over an extended energy range. Part of those measurements can be performed at home, but at present with neutrons of energy not less than about 1 meV, and sample temperatures down to 30K only. It will be necessary to complement those experiments with data obtained at other facilities, like LENS of Hokkaido Linac, extending down both the neutron energies and sample temperatures.

Also during the first year, we will start construction of the new cold source at our electron LINAC based neutron source.

**Results obtained under the CRP**

As it is reflected through our recent contributions in matters related to the present proposal (ANNEX), activities have been in progress at Neutron Physics Division in theoretical, calculational and experimental aspects involving the study of hydrogenous materials of interest as cold moderators.

The majority of results so far are in the form of new cross section libraries generated with the use of the NJOY code, starting from the physical models developed in our group. In this context, we have just introduced a new model to describe the interaction of slow neutrons with solid methane in phase II (that is the stable phase below T=20.4K and atmospheric pressure), that produces cross sections in excellent agreement with measurements after taking proper account of spin correlation effects.
Recently, a large effort has been done in the implementation of a sample cooling system, which at present allows us to cool the sample down to about 30K. The new cold moderator system for our electron LINAC based pulsed neutron source has been designed and optimized, and we are now starting its construction.

**Recent publications related to the project (within the past 2 - 3 years):**

► Journals:

“*A New Thermal Neutron Scattering Kernel for Liquid Hydrogen*”
  J.R. Granada and V.H. Gillette  

“*Neutron Cross Sections of cold moderators: An approximate algorithm for the description of Synthetic Kernels for molecular solids*”
  S.N. Petriw, F. Cantargi, J.R. Granada, M.M. Sbaffoni and O. Lovotti  

“*New Scattering Kernels for some materials of interest as Advanced Cold Neutron Moderators*”
  F. Cantargi, J.R. Granada, S.N. Petriw, M.M. Sbaffoni  

“*Design of a cold neutron source for the Bariloche LINAC with solid Mesitylene as moderator material*”
  L. Torres, J.R. Granada  

“*Analysis of mean kinetic energies in H2O/D2O mixtures by neutron total cross section measurements*”
  J.J. Blostein, J. Dawidowski y J.R. Granada  

“*Thermal Neutron Cross Section of Liquid and Solid Mesitylene*”
  F. Cantargi, J.J. Blostein, L. Torres and J.R. Granada  

“*Total Cross Sections of Benzene at 90K and Light Water Ice at 115K*”
  L. Torres and J.R. Granada  

“*Neutron Cross Sections of cold moderators: A Synthetic Kernels for molecular solids*”
  S.N. Petriw, F. Cantargi, J.R. Granada  

"*Experimental Neutron Data: Sigma(E) of Mesitylene in the thermal range*".
  Nuclear Data Section, IAEA, EXFOR 31578 (2006).

"*Experimental Neutron Data: Sigma(E) of Solid Benzene and Ice in the thermal range*".
  L. Torres J.R.Granada and J.J. Blostein.  
  Nuclear Data Section, IAEA, EXFOR 31588 (2007).

“*Synthetic Scattering Kernel for solid methane in phase II*”
J.R. Granada

►Conferences:

“Problems with Low Energy Neutron Scattering Kernels for Moderator Materials”
J.R. Granada, V.H. Gillette, M.M. Sbaffoni, F. Cantargi, S. Petriw and L. Torres
Workshop on Long Wavelength Target Stations, Rutherford Appleton Lab., England
(Febr.2003).

“Neutron Scattering Cross Sections of Cryogenic Materials: Preliminary results for Mesitylene”, J.R.
Granada, F. Cantargi, L. Torres, M.M. Sbaffoni and S. Petriw
XIIIth. International Conference on Selected Problems of Modern Physics, Dubna, Russia
(Jun 2003).

“Some research and nuclear engineering teaching at a small accelerator driven neutron source”, R.E.
Mayer, J.R. Granada,
J. Dawidowski, J.R. Santisteban, J.J. Blostein, L. Torres, A. Tartaglione, A.I. Lazarte, A.J.Villanueva,
L.A.Rodriguez Palomino,
Proceedings del Technical Meeting on the Use of High Energy Accelerator for Research, Therapy and
Applications of Spallation Sources, IAEA Headquarters, Vienna, 7-9 (Dec. 2005).

“35 years of Pulsed Neutrons in Bariloche: Cases from the Past and some ideas for the Future”, J.R.
Granada et al.
International Workshop on "Neutron Science using Accelerator based Small Neutron Source",
Hokkaido University, Sapporo, Japan (Jan. 2006)

“Scattering Kernels for some materials of interest as cold neutron moderators: Recent Activities”
J.R. Granada, F. Cantargi, S. Petriw and M.M. Sbaffoni.
International Workshop on Advanced Cold Moderators, Bariloche, Argentina (Apr.
2006).

International Collaboration on Advanced Neutron Sources, ICANS.XVIII, Dongguan,
Guangdong, China (April 2007).

“Neutron Cross Section Libraries for Aromatic Systems of Interest as Cold Neutron
International Collaboration on Advanced Neutron Sources, ICANS.XVIII, Dongguan,
Guangdong, China (April 2007).