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NEUTRON SCATTERING CROSS SECTIONS OF CRYOGENIC MODERATOR MATERIALS: SOME RESULTS FROM THE SYNTHETIC MODEL

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Abstract

The Synthetic Scattering Function (SSF) allows a simple description of the incoherent interaction of slow neutrons with hydrogenous materials. Its original formulation, essentially aimed at describing molecular gases, has been extended to include the important case of molecular solids. In any case, the main advantages of the Synthetic Model reside in the analytical expressions that it produces for double-differential cross sections and energy-transfer kernels, which in turn permit the fast evaluation of neutron scattering and transport properties.

The SSF routines were integrated into the NJOY code, in such a way that the cross sections can be generated with the same format either from its standard library or from our model.

We discuss in this work some aspects of the formalism developed to include the case of molecular solids in a more detailed manner, and present applications of the original and extended Synthetic Model to recent experimental results on liquid Hydrogen, Mesitylene, and solid Methane.

1. Introduction

Neutronic design calculations involving thermal and subthermal neutron energies demand the knowledge of reliable cross section data relative to the materials which conform the system under consideration.

The relevant quantity to describe the interaction of thermal neutrons with condensed matter is the Van Hove scattering function $S(\boldsymbol{Q},\omega)$, as it embodies all the dynamical and structural information about a scattering system [1]. First-principles theories were developed in the past to evaluate the scattering function, but the resulting expressions are usually not appropriate for calculational procedures. Moreover, a detailed knowledge of the scattering function over a wide range of energy ($\hbar\omega$) and momentum ($\hbar\boldsymbol{Q}$) transfer is not required in many cases, and in fact only those interactions which are more operative under a given condition must be carefully accounted for.

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.

Those ideas were part of the main motivations for the development of a 'Synthetic Scattering Function' $T(Q,\omega;E_0)$, which incorporates the main dynamical characteristics of the molecular unit, still retaining a high degree of simplicity in its formulation [2]. The SSF uses the incident neutron energy E_0 as the variation parameter, to determine the values of effective translational masses, temperatures, and vibrational factors across the energy range.

The SSF has been applied to the evaluation of neutron cross sections and thermalization properties of several moderator materials [3,4]. We have included the SSF routines into the NJOY code, in such a way that the cross sections can be generated with the same format either from its standard library (ENDF/B-VI) or from our synthetic model.

In this work we will briefly review the characteristic features of the synthetic model, and its predicted results for the cross sections and other integral magnitudes for mesitylene, liquid hydrogen, and solid methane, and compare them with the data produced with the latest NJOY version and with experimental data when available.

2. The Synthetic Model

The synthetic scattering function has been developed to the extent of producing analytic forms for a variety of magnitudes [2,3], and the accuracy of them have been verified in a number of cases [4]. We will just summarize the main features of this formulation.

In terms of the synthetic model, the double-differential scattering cross section of a molecular unit is written as, (see Ref. 3 for details)

$$\frac{\partial^2\sigma}{\partial\Omega\,\partial E}=\sum_i^N n_i\frac{\sigma_b^i}{4\pi}T^i(\vec{Q},\omega,E_0)\ , \tag{1}$$

where N is the number of dynamically nonequivalent atomic species, and n_i represents the number of atoms of each equivalent atomic species with a bound scattering cross section σ_b. Finally, $T(\vec{Q},\omega,E_0)$ stands for the basic expression of the Synthetic Scattering Function (SSF):

$$T(\vec{Q},\omega,E_0)=\frac{k}{k_0}\left[S_{\mu_0,\tau_0,\Gamma}(\vec{Q}\ ,\omega)-\sum_{\lambda,\pm}^m C_{\lambda,\pm}\frac{\partial}{\partial\Gamma}S_{\mu_0,\tau_0,\Gamma}(\vec{Q}_{\lambda,\pm}\ ,\omega_{\lambda,\pm})\right]\ , \tag{2}$$

where k_0 and k denote the modulus of incident and scattered neutron wave vectors, respectively, $\vec{Q}=\vec{k}_0-\vec{k}$ and $\hbar\omega=E_0-E$ are the momentum and energy exchanged in the collision process. $S_{\mu_0,\tau_0,\Gamma}(\vec{Q},\omega)$ is the scattering law for the neutron interaction with a quasi-rigid molecule, and the second term on the right-hand side of Eq.(2) is a corrective one which accounts for processes where the neutron exchanges energy with the m internal modes of the atomic species, by creating or annihilating one phonon. The summation over inelastic processes is performed under the assumption that the internal modes are represented by Einstein oscillators, each with eigenfrequency ω_λ, and effective mass M_λ.

The quantities μ_0 , τ_0 and Γ represent the effective mass, temperature and vibrational factor, respectively, that the scattering nucleus would present in the interaction, and they depend explicitly on the incident neutron energy E_0.

Under those conditions, $S_{\mu_0,\tau_0,\Gamma}(\vec{Q},\omega)$ may be written as

$$S_{\mu_0,\tau_0,\Gamma}(\vec{Q},\omega) = \left[\frac{\mu_0}{2\pi\ \hbar^2 Q^2 k_B \tau_0}\right]^{0.5} \exp\left[-\left(\hbar\omega - \frac{\hbar^2 Q^2}{2\,\mu_0}\right)^2 \left(\frac{\mu_0}{2\hbar^2 Q^2 k_B \tau_0}\right) - \Gamma\frac{\hbar^2 Q^2}{2}\right] . \quad (3)$$

Analytic expressions are then obtained in a straightforward manner for the scattering kernels, and the total scattering cross section, while the usual expansion of the double-differential cross section in Legendre polynomials leads to the definition of the energy-transfer scattering kernels. These kernels also admit analytic expressions within the frame of the Synthetic Model [5-7].

3. The Case of Molecular Solids

In its original formulation, the SSF was devised to describe the interaction of slow-neutrons with molecular gases [2], and its applicability extended to liquid and even solid [8,9] systems as long as the energy of the incident neutrons is not too low as to detect departures from a free gas behaviour during the interaction time.

When the cross sections of a material have to be properly described at low neutron energies, as it is the case of solid cryogenic moderators, a different approach is imperative. A new formulation has been developed to tackle those situations, and it is at present under validation. Our new Synthetic Model for molecular solids (SMMS) uses a more realistic description of the dynamics at low energies, and switches over the traditional molecular gas formalism at higher neutron energies. Its basic hypotheses are just outlined in the following paragraphs, where we can think of solid methane as a test case.

Let us assume that the low frequency part of the system's density of states can be written as:

$$AZ(\omega) = \omega^2 e^{-(\omega-\omega_0)^2/2\sigma^2} + \frac{3\omega^2}{\omega_D^3} \quad (4)$$

where the first term on the r.h.s. represents very low energy rotational excitations ('1'), and the second term is the Debye approximation to the lattice modes ('2'), existing for $\omega \leq \omega_D$. The constant A reminds us that this is just a portion of the complete, normalized frequency spectrum that also includes the intramolecular vibrational modes.

Under the assumption of no coupling between those modes, the intermediate scattering function is the product of those associated to each mode. In addition, each of the factors is assumed to satisfy the Gaussian approximation

$$\chi_i(Q,t) = \exp\{-\gamma_i(t)Q^2\} \qquad ; \text{i=1,2} \quad (5)$$

where the time-dependent mean-square displacement $\gamma(t)$ is related to the frequency spectrum

$$\gamma(t) = \int_0^\infty \frac{Z(\omega)}{\omega}\left[\{n(\omega)+1\}e^{i\omega t} + n(\omega)e^{-i\omega t}\right] \quad (6)$$

and $n(\omega)$ is the occupation number.

The intermediate scattering functions admit a phonon expansion

$$\chi_i(Q,t) = e^{-2W_i}\left\{\sum_m \frac{(2W_i)^m}{m!}\left[\frac{\gamma_i(t)}{\gamma_i(0)}\right]^m\right\} \quad ; \text{where} \quad 2W_i = \frac{\hbar Q^2}{2M_i}\gamma_i(0) \tag{7}$$

and the associated scattering law is given by

$$S_i(Q,\omega) = e^{-2W_i}\sum_p \frac{(2W_i)^p}{p!}\frac{G_p^{(i)}(\omega)}{\hbar} \tag{8}$$

The scattering law due to the two low-lying dynamical modes under consideration is calculated as

$$S(Q,\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\,\chi_1(Q,t)\chi_2(Q,t)e^{-i\omega t} = S_1(Q,\omega)\otimes S_2(Q,\omega) \tag{9}$$

and then, to describe the low energy neutron cross sections in our Synthetic Model for molecular solids, we include up to three phonon terms in the inelastic scattering law:

$$\begin{aligned} S_{inel}(Q,\omega) \cong\; & e^{-2W_1}\sum_{p=1}^{3}\frac{(2W_1)^p}{p!}\frac{G_p^{(1)}(\omega)}{\hbar} + e^{-2W_2}\sum_{k=1}^{3}\frac{(2W_2)^k}{k!}\frac{G_k^{(2)}(\omega)}{\hbar} + \\ & + e^{-2(W_1+W_2)}\frac{4W_1W_2}{\hbar^2}\left\{G_1^{(1)}\otimes G_1^{(2)} + W_2 G_1^{(1)}\otimes G_2^{(2)} + W_1 G_2^{(1)}\otimes G_1^{(2)}\right\} \end{aligned} \tag{10}$$

The functions $G^{(1)}(\omega)$ related to the 'rotational band' in the frequency spectrum were calculated, as well as their convolution [10] with the $G^{(2)}(\omega)$ already obtained by Sjölander for a Debye model [11,12], producing an analytical expresion for $S(Q,\omega)$. Our final prescription for a SSF for molecular solids is then formulated as a combination of the last expression with the SSF for molecular gases, using a switching function determined by the elastic scattering component [10].

The NJOY code [13] was modified in order to include the SSF formalism as an optional way to calculate cross sections for moderator materials. The main modifications were introduced in the THERMR module where new subroutines were added, adapting the formalism of the synthetic model to the NJOY system.

4. Results for Some Cryogenic Moderators

Applications of the SSF to a variety of hydrogenous systems were presented in the past [14], where different magnitudes were predicted and compared with available experimental data or previous theoretical results. Here, we present NJOY results for thermal neutron scattering kernels and total cross sections of liquid hydrogen, solid methane, and mesitylene

obtained from both its original ENDF/B-VI.2 library (when available) and the Synthetic Model formalism integrated in our modified version of the code.

4.1 Liquid Hydrogen

Our model for H_2 was built on the basis of the Young-Koppel [15] formalism to describe the rotational elastic and one-phonon inelastic cross sections, together with the prescriptions of the general Synthetic Model for the vibrational and translational forms of the molecular scattering function [16]. The energies of the three Einstein oscillators that represent the actual (acoustic, rotational, vibrational) modes are 0.005, 0.0147 and 0.546 eV.

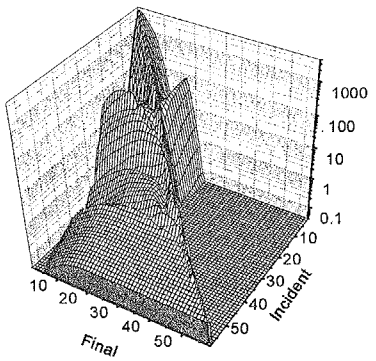

Fig.1(a) Scattering Kernel for para-H_2 at 20K, calculated by NJOY from ENDF/B-VI

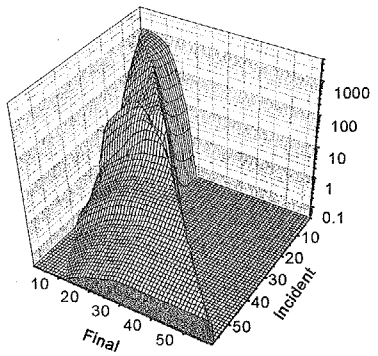

Fig.1(b) Scattering Kernel for para-H_2 at 20K, calculated by NJOY from SSF

In Figs. 1(a) and 1(b), we show the scattering kernels of liquid parahydrogen at 20K, calculated by our modified NJOY with its two options, ENDF/B-VI and SSF, respectively. The main differences between them are observed at the low energy groups, and they are related to the different treatment of the translational motion in both models.

The total cross sections of normal and para-hydrogen calculated at 20K are compared in Fig. 2 with the experimental results of Seiffert [17]. Once again, some discrepancies are apparent at the low energy side, which emphasize the need for new measurements over that energy range. Other SSF results for this system were presented previously [18].

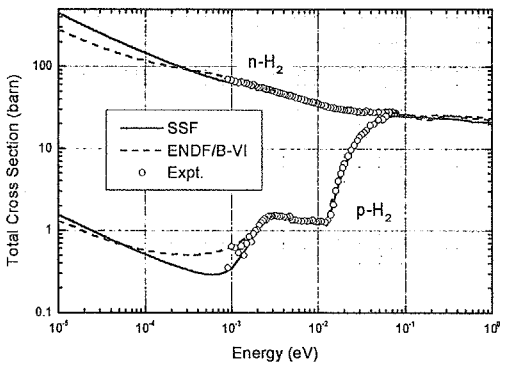

Fig.2: Total cross section of liquid Hydrogen.

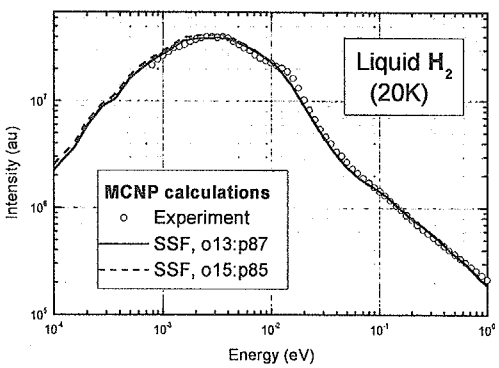

Fig.3: Observed and calculated neutron spectrum emerging from a LiH_2 moderator.

Spectra calculations were performed using our libraries in MCNP, to describe an experiment carried out at Hokkaido University [19]. In Fig.3 we compare the experimental results [20] with our calculations for two different ortho/para ratios in liquid H_2 at 20K, thus demonstrating the good agreement achieved through the use of the SSF scattering kernels and the sensitivity to the o/p concentration for this particular case.

The same set of input data for MCNP was used to predict the spectrum which should have been observed in the Hokkaido experiment at different temperatures. The result is shown in Fig.4, in the form of the logarithmic derivative of the neutron flux as a function of T, valid for temperatures between 19K and 25K, and at constant density. In a similar way, the logarithmic derivative of the neutron flux as a function of the ortho/para ratio has been calculated for the same temperature range. This kind of information should be of interest to evaluate the effect on instruments performance caused by variations of the cold moderator parameters.

Besides the question of the spectrum shape, referred to in the preceding paragraphs, the other important parameter characterizing the quality of a cold moderator at a pulsed neutron source is its time response. When the size of the moderator volume is large enough for a fundamental decay mode to be established after the injection of a fast neutron pulse, the time constant corresponding to that fundamental mode dominates the pulse width produced by the source as a function of neutron energy. At the lower energies, whenever neutron diffusion theory is valid for a particular geometry, the decay time becomes energy-independent and determined solely by the geometrical buckling of the moderator system. We show in Fig.5 values of that quantity predicted by the SSF for different ortho concentrations in liquid H_2 at 20K, together with an experimental result due to Würz [21]. The agreement in this case is good, but one must be aware that the very strong variation of the mean free-path with o/p concentration severely complicates the calculation of the actual buckling for a given situation (geometry, concentration). This means that any atempt to correlate measured decay times with concentration in a real cold source will depend on its geometry, but in any case it must rely very heavily on model calculations. Conversely, it is possible for some special geometries to optimize the neutron pulse shape in a specific case as a function of the o/p concentration.

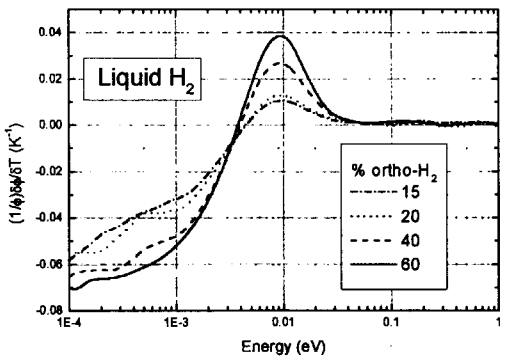

Fig.4: Spectrum derivative as a function of temperature, for different o/p concentration.

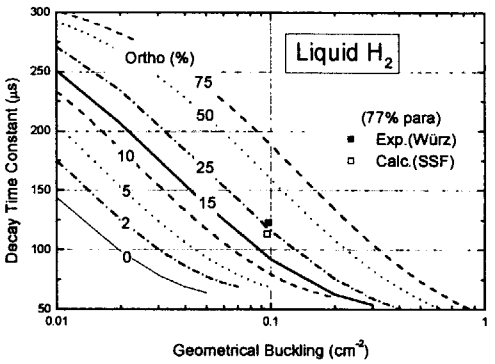

Fig.5: Decay constant of the fundamental mode.

4.2 Methane

The SSF for this molecule was described in [22]. Due to the very low energies associated to the rotational motion, these modes are usually excited by temperature and a quasi-classical treatment of them is applicable in the frame of the SSF formalism. The three parameters (μ_0 , τ_0 , Γ) of the model change as the neutron energy goes over the intramolecular vibrations'

energies (lumped into two Einstein oscillators at 0.17 eV and 0.38 eV) according to the model prescription. In fact, this set of input data for the SSF is appropriate to describe the neutron scattering properties of methane in the gaseous and liquid phases. Solid methane is treated in a different manner, in order to properly account for the lattice vibrations. As discussed in Section 3, the SSF for the solid system is built as a composition of a 'low' and a 'high' incident energy parts, represented by a phonon expansion of the scattering law for a molecular solid, and the classical form of the Synthetic Model, respectively.

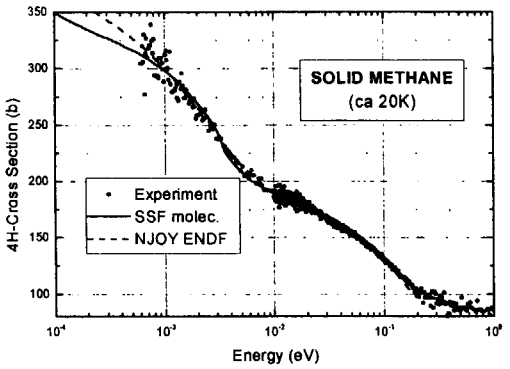

Fig.6: Total cross section of solid Methane.

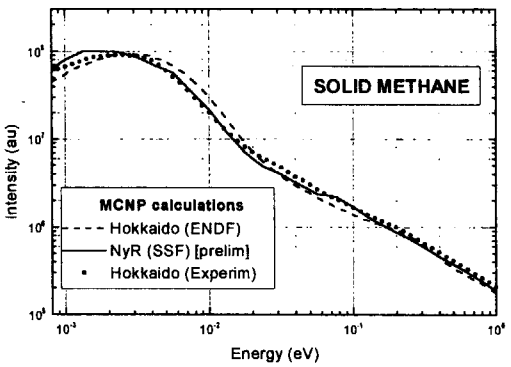

Fig.7: Observed and calculated neutron spectrum emerging from a solid methane moderator.

While the 'solid model' for methane is being developed according to the algorithm sketched in Sec.3, some preliminary results are included here to illustrate the expected quality of the model description. The total cross section of solid methane *ca* 20K is presented in Fig.6, where we compare the experimental data [23] with results obtained from NJOY using ENDF/B-VI and the traditional SSF, with Einstein oscillators at 0.0065eV to represent the onset of rotational excitations, and at 0.008eV as a characteristic excitation representing lattice modes described by a Debye spectrum with a temperature T_D = 140K [24]. The Hokkaido group has measured the neutron spectrum emerging from a solid methane system [19], and in Fig.7 we compare their experimental results with NJOY calculations based on its standard library and using our preliminary scattering kernel; once again the SSF seems to produce highly encouraging results. Finally, in Fig.8 we present one result concerning the SSF prediction for the time response of a solid methane slab (12x12x5 cm^3), compared with the decay times measured for that system [25]. Other neutronic properties of this moderator evaluated through the SSF have been presented previously [14,22].

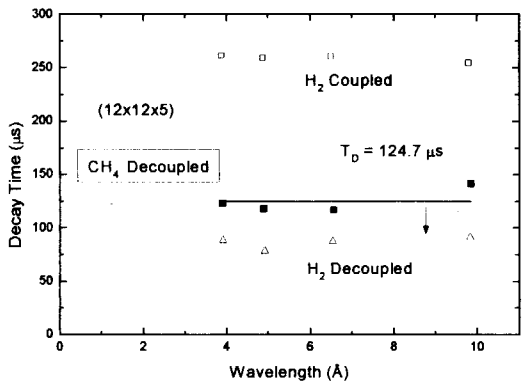

Fig.8: Experimental and calculated decay constant for a specific solid CH_4 (20K) moderator

4.3 Mesitylene

This material has been also proposed and used as a cryogenic moderator [26-28], and a few measurements were performed on it at low temperatures [29,30]. Mesitylene was modeled in the frame of the Synthetic Scattering Function [31], and we will show here some preliminary

results from this modeling. As in the case of solid methane, a more detailed description of solid mesitylene will include a phonon expansion as outlined in Sec.3 for low incident neutron energies, and its conversion into the traditional SSF at the other end of the energy scale.

Our model for mesitylene (1,3,5 trimethylbenzene) was based on our previous work on benzene [4] and methyl-group containing molecules [32], as well as on specific information about the dynamics of this material [29,30,33]. The actual description [31] involves four types of 'atoms': two elements (H, C), each of them composing either a methyl group or the benzene ring. The complete dynamics of the molecular system involves vibrations within the methyl units, their torsional motion respect to the ring, the rotations of the entire molecule and its translational motion. Using the low frequency part of the density of states measured for mesitylene at 18K [30] (weight = 0.2355) and independent oscillators at 0.12eV, 0.17eV and 0.37eV, we have also created an input for NJOY.

In Fig.9 we compare our calculated scattering cross section of H in mesitylene with the experimental points of Utsuro [29]. Although the theoretical curve seems to overestimate the cross section at low energies, it will be very convenient to reconfirm this situation at the light of new measurements. Finally, in Fig.10 we show our predicted total cross section and its elastic and inelastic components, from both the traditional SSF and NJOY.

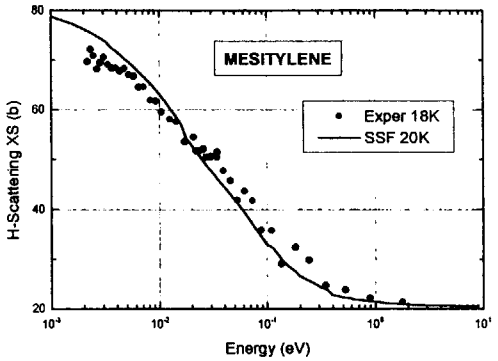

Fig.9: Total cross section of solid Mesitylene.

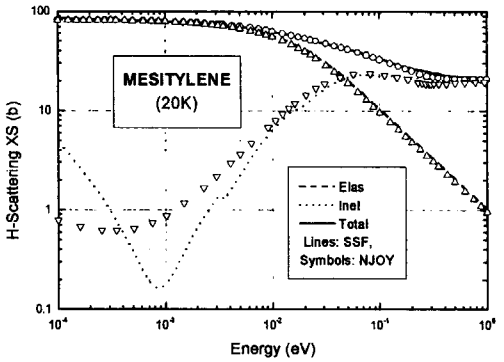

Fig.10: Calculated elastic, inelastic and total cross sections of solid Mesitylene.

5. Conclusions

The results presented in the previous section are typical of the SSF predictions for hydrogeneous materials, and serve to illustrate the flexibility of this function to model quite different molecular systems using a minimum set of input data with no adjustable parameters.

Some of those calculations have been performed using a modified version of the NJOY code, in which a package of subroutines corresponding to the Synthetic Scattering Function formalism has been integrated as a new option.

Although we continue benchmarking the results based on this new option, besides the extensive applications already performed using the Synthetic Model, we expect that the powerful capabilities of this model to describe complex systems will be very useful to tackle new ideas related to nonconventional moderator materials.

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