NEUTRON SCATTERING KERNELS
FOR COLD MODERATOR MATERIALS

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ABSTRACT

In this work we present scattering kernels and cross sections for a few cryogenic materials of interest as cold neutron moderators. These calculations are based on a Synthetic Scattering Function (SSF), developed to describe the incoherent interaction of slow neutrons with hydrogeneous materials in a simple way. The main advantages of this model reside in the analytical expressions that it produces for several physics parameters of specific interest to the field of Neutron and Reactor Physics. These parameters include double-differential cross sections, energy-transfer kernels, and total cross sections, which in turn allow the evaluation of neutron scattering and transport properties. The possibility of calculating those quantities in a fast and accurate way, permits the production of group constants for any specific material, at any temperature, any degree in the Legendre expansion, and over any required energy mesh.

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 review the SSF results for the cross sections and scattering kernels for water, liquid hydrogen, and liquid and solid methane, and compare them with the data produced with the latest NJOY version and ENDF/B-VI library, and with experimental data when available.

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1. INTRODUCTION

It has became evident in recent years the increasing demand of long wavelength neutrons from both pure and applied sciences, to explore the structure and behaviour of matter by exploiting the unique capabilities of neutron scattering techniques. As a consequence, projects to install new cold neutron sources at research reactors, upgrade existing ones, and to develop a new generation of cold sources for new (pulsed) neutron sources, have been launched in many places around the world.

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. Although fuel and structural components are of great importance, moderators play a central role in the thermalization process.

The relevant quantity to describe the interaction of thermal neutrons with condensed matter is the Van Hove scattering function $S(\boldsymbol{Q}, w)$, 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 ($\hbar w$) 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' $T(Q, w, E_0)$, which incorporates the main dynamical characteristics of the molecular unit, still retaining a high degree of simplicity in its formulation [2]. In the frame of the Synthetic Scattering Function (SSF), the incoherent interaction of slow neutrons with hydrogeneous materials can be described in a simple way. 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. In this manner, a kind of envelope represents the combined effect of the quantum excitations of the system's internal modes, thus avoiding the complexities involved in a complete description of them.

The main advantages of this model reside in the analytical expressions that produces 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. The possibility of calculating those quantities in a fast and accurate way, permits the production of group constants for any specific material, at any temperature, any degree in the Legendre expansion, and over any required energy mesh.

The SSF has been applied to the evaluation of neutron cross sections and thermalization properties of several moderator materials [3,4]. A very good agreement was achieved between our model predictions and the most recent experimental data for those examples, while the analytic character of the derived expressions showed its power in drastically reducing the computing time as compared with conventional codes.

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 water, liquid

hydrogen, and liquid and solid methane, and compare them with the data produced with the latest NJOY version and ENDF/B-VI library, and with experimental data when available.

2. THE 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)

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

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):

$$T(\vec{Q},\omega,E_0) = \frac{k}{k_0}\left[S_{m_0,t_0,\Gamma}(\vec{Q},\omega) - \sum_{l,\pm}^m C_{l,\pm} \frac{\partial}{\partial\Gamma} S_{m_0,t_0,\Gamma}(\vec{Q}_{l,\pm},\omega_{l,\pm}) \right] \quad , \tag{2}$$

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. $S_{m_0,t_0,\Gamma}(\vec{Q},\omega)$ 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_l, and effective mass M_l. These quantities are obtained from a realistic frequency spectrum and, in particular, the effective masses associated to each of those motions are taken from the areas of the corresponding part of it, and constrained to satisfy a proper normalization condition [2].

The quantities m_0, t_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. In this way, a simple prescription is obtained to handle situations ranging from cases in which the incident neutron cannot excite any of the l-oscillator levels, through those cases when that mode becomes fully excited in the collision and a quasi-classical treatment of it is applicable.

Under those conditions, $S_{m_0,t_0,\Gamma}(\vec{Q},\omega)$ may be written as

$$S_{m_0,t_0,G}(\vec{Q},\omega) = \left[\frac{m_0}{2\pi\,\hbar^2 Q^2 k_B t_0} \right]^{0.5} \exp\left[-\left(\hbar\omega - \frac{\hbar^2 Q^2}{2 m_0} \right)^2 \left(\frac{m_0}{2\hbar^2 Q^2 k_B t_0} \right) - G\,\frac{\hbar^2 Q^2}{2} \right] . \tag{3}$$

The reduced number of input parameters required for the SSF and the analytic character of the derived expressions, make this formalism a powerful and practical tool for describing the slow neutron - molecule interaction. In particular, it provides in a straightforward manner analytic expressions for the scattering kernels,

$$S(E_0, E) = \int_{4\pi} \frac{\partial^2 S}{\partial \Omega \, \partial E} \, d\Omega \tag{4}$$

and the total scattering cross section

$$S(E_0) = \int_0^\infty \int_{4\pi} \frac{\partial^2 S}{\partial \Omega \, \partial E} \, d\Omega \, dE \quad . \tag{5}$$

The usual expansion of the double-differential cross section in Legendre polynomials, leads to the definition of the energy-transfer scattering kernels. These are the coefficients of the Legendre polynomials which appear when the Boltzmann equation is solved to simplify its geometrical complexity. To evaluate these kernels, the calculation of the following integrals is required:

$$S_n(E_0, E) = \int_{4\pi} \frac{\partial^2 S}{\partial \Omega \, \partial E} \cos^n(\theta) \, d\Omega \quad , \tag{6}$$

but it has been shown that the P_N kernels also admit analytic expressions within the frame of the Synthetic Model [5-7].

III. MODIFICATIONS TO THE *NJOY* CODE

We briefly describe here the modifications to NJOY in order to include the SSF formalism as an optional way to calculate cross sections for moderator materials. The NJOY code 94.105 version [8] was installed and run on different platforms (PC, Sun Ultra with r-8 option, and Silicon Graphics).

NJOY calculates the cross sections for moderators through two ways:
- Free-gas theory
- Calculation of the scattering matrix from the $S(\alpha,\beta)$ matrixes (calculated previously with LEAPR module) supplied in the ENDF/B-VI library.

The SSF formalism was written as a set of computer routines, which were tested through a range of application examples that involved the evaluation of neutron cross sections and thermalization properties of several moderator materials. Up to now, SSF input data sets are available for light and heavy water, liquid hydrogen (ortho and para), methane, ethane, glycerol, polyethylene, Plexiglas, dodecane, TBP, benzene, dowtherm, metallic hydrides, but the data base will be extended in the future to other materials.

The main modifications were introduced in the THERMR module (subroutines CALCEM and SIGL, and function SIG), where new subroutines were added, adapting the formalism of the synthetic model to the NJOY system.

When NJOY calculates scattering kernels from the scattering function (iinc = 4) the initial energy grid is internally fixed (58 groups), and the final energy grid is calculated in an adaptively way, from the data provided in the $S(\alpha,\beta)$ tape. If the SSF is used (iinc = 5), the same grid of initial energies is adopted, and the same adaptive process is used for final energies determination, but from a fixed and extended grid of energy transfer values (β).

Input modifications for the THERMR module:

→ In card 2, a new option for IINC was implemented. If IINC is equal to 5, the program will use the "Synthetic Scattering Module" (MODEL Subroutine) to compute the total cross section and the energy-transfer kernels.

→ In this case, new data are needed, which are given in card 5.

The first parameter (**I0**) indicates which material will be treated. The meaning of the second and third parameters **(I1 and I2)**, depends on the material, because different routines may be used for different cases. For example, in the liquid hydrogen case, I1 can take the values 1, 2, or 3 if it is ortho-hydrogen, para-hydrogen or a mix of both materials, and I2 is read only in the case of I1.eq.3 giving the percentage of ortho-hydrogen in the mix. In any other case I1 indicates the type of cross section to be calculated, and I2 specify if the data will be for a molecule or for a single atom or atom family.

IV. A FEW APPLICATION EXAMPLES

Applications of the SSF to a variety of hydrogenous systems were presented in the past [9], where different magnitudes were predicted and compared with available experimental data or previous theoretical results.

In this work we present NJOY results for thermal neutron scattering kernels and total cross sections of water, liquid hydrogen and methane, obtained from both its original ENDF/B-VI.2 library and the Synthetic Model formalism integrated in our modified version of the code.

1. Water

As far as the SSF is concerned, its predictions for thermal cross sections and diffusion parameters of room temperature water have been comprehensively discussed and compared with existing information [10].

Water is represented by three modes. This set was adopted after a thorough analysis of measured frequency spectra with particular consideration of the internal modes' weights, especially for the low frequency part where the Sachs-Teller mass concept was used for normalization purposes [11]. Besides the two vibrational modes at 0.205 eV and 0.481 eV (that are almost the same as in the LEAPR input), the model uses a single Einstein oscillator at 0.07 eV to represent the actual broad rotational band, with a weight of 0.41667 (=7.5/18) rather than 0.44444 (=8/18).

2. Methane

The SSF is particularly appropriate to describe a gas of spherical molecules, as CH_4 at room temperature. In this case, due to the very low energies associated to the molecular rotational motion, these modes are thermally excited and a quasi-classical treatment of them is applicable in the frame of the SSF formalism. Therefore, at all incident energies below the lower vibrational energy, the neutron will 'see' the hydrogen atoms as having a mass equal to their Sachs-Teller mass (3.4 amu); the effective temperature and vibrational factor over this region correspond to the system's temperature and the zero-point motion of the vibrational modes. Those three parameters (μ_0 , τ_0 , Γ) of the model change as the neutron energy goes over the 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. The SSF for the latter system is built as a composition of a ´low´ and a ´high´ incident energy parts, represented by a three-phonon expansion of the scattering law for a molecular solid, and

the classical form of the Synthetic Model, respectively. A change-over function between both forms is introduced in connection with the behaviour of the elastic cross section as a function of energy.

3. Liquid Hydrogen

After the early total cross-section measurements performed by Squires and Stewart [12] on liquid H2 and the theoretical work of Sarma [13], many calculational techniques were developed, most of them based on the gas model of Young and Koppel [14]. As a consequence, the differentiating feature of those bound atom models has been in the treatment of the translational motion of the molecular unit.

The transformation properties of the total wavefunction in a homonuclear diatomic molecule causes the total nuclear spin **I** and the total angular momentum **J** to be correlated, thus producing a well-defined "selection rule" that governs the transitions between states corresponding to those quantum numbers. Those correlations are especially important in the case of H2 at low temperatures, where not many rotational levels are excited and, consequently significant interference effects show up in the neutron cross sections for the ortho and para forms.

Our model for H2 is built on the basis of the Young-Koppel 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. Concerning this latter mode, an additional Einstein oscillator was introduced to represent the low-energy collective excitations, as it is well known that a simple gas model is not adequate to predict some scattering properties at low neutron energies. The energies of the three modes are 0.005, 0.0147 and 0.546 eV.

Intermolecular interference effects show up in the measured total cross section of liquid H2 at low energies (<0.003 eV), where they partially cancel the intramolecular interference contribution for neutron wavelengths longer than the average distance between molecular centers. We have taken this behavior into account in a crude manner by imposing a partial cancellation of the coherent elastic scattering, whereas a more rigorous treatment of these effects should be based on the consideration of the static structure factor of the liquid.

V. RESULTS

Figure 1 shows the isotropic scattering kernels for water produced by NJOY using (a) its ENDF/B-VI.2 library and (b) the integrated SSF option. The great similarity between both sets is evident, besides some sharper features in the model-based result related to the crude approximation involved in representing the rotational band by a single oscillator, plus additional small discrepancies (groups # 30-40) in regions where the value of the scattering kernel is not significant.

The scattering cross section of hydrogen in room temperature water is shown in Fig.2, where again a very good agreement is observed between the NJOY results based on ENDF/B-VI.2 and SSF over the full thermal energy range. There is however a small systematic discrepancy between both sets below 0.1 eV, that we cannot attribute to the Synthetic Model in view of its excellent agreement with experimental data on the total cross section of water [4].

In Fig.3 we present the results obtained for the scattering kernel of liquid methane at 100 K, from the two options implemented in NJOY: (a) ENDF/B-VI.2 and (b) SSF. Some differences between both sets are apparent at the lower groups, reflecting the appropriate treatment of rotational and diffusive modes on which ENDF/B-VI.2 is based. However, those discrepancies are quite localized and with relatively small amplitudes.

The total scattering cross section of hydrogen in methane at 20K and 100 K is given in Fig.4 for the two evaluations. The agreement between them is not very good in this case, particularly at 100K, although the trend is similar. One may expect that at very low energies, the ENDF based calculation should give more reliable results, on account of its better description of the molecule's translation and rotations.

The scattering kernels of liquid para and ortho hydrogen at 20K are shown in Figs.5 and 6, respectively, in each case as calculated by our modified NJOY with its two options: (a) ENDF/B-VI.2 and (b) SSF. The main differences between both sets are observed at low energies, due to the different treatment of the molecular translational motion. In the case (a) a cluster of 20 molecules is considered, sustaining solid-like internal vibrations and diffusing according to the Egelstaff-Schofield [15] prescription. Instead, the SSF considers a much smaller effective mass (=2 molecular masses) associated to the hydrogen in the H2 molecule at neutron energies below 0.005 eV. Consequently, the latter produces a smoother behaviour of the scattering kernels over the low-energy groups.

We show in Fig.7 the total cross section of para- and normal-hydrogen as calculated by our modified NJOY with its two options, together with the experimental data of Seiffert [16]. Both calculations describe the experimental measurements quite well over the region where the latter exist. However, the discrepancies observed at the low energy side reflect again the different treatment of the translational motion mentioned above. New measurements at those very low energies are therefore required for a proper modeling of this system.

VI. CONCLUSIONS

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

The calculations have been performed using a modified version of the NJOY (94.105) code, in which a package of subroutines corresponding to the Synthetic Scattering Function formalism has been integrated as a new option. Those results illustrate the similarities and differences between the ENDF/B-VI.2 and SSF formalisms.

Although we are still 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 become a useful reality after its integration into NJOY.

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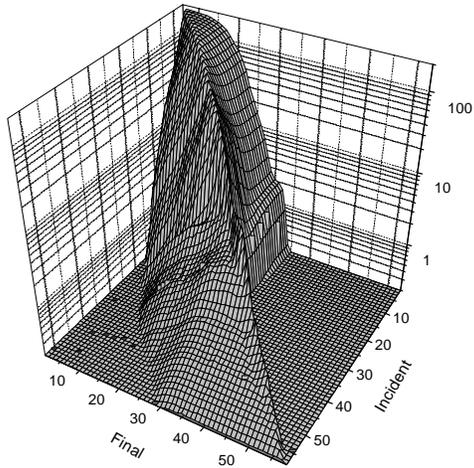[15] Egelstaff, P.A. and Schofield, P. *Nucl. Sci. Eng.* **12**, 260 (1962).
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Fig.1(a): Scattering Kernel for H in H_2O at 300K,
calculated by NJOY with option (a) ENDF/B-VI.2
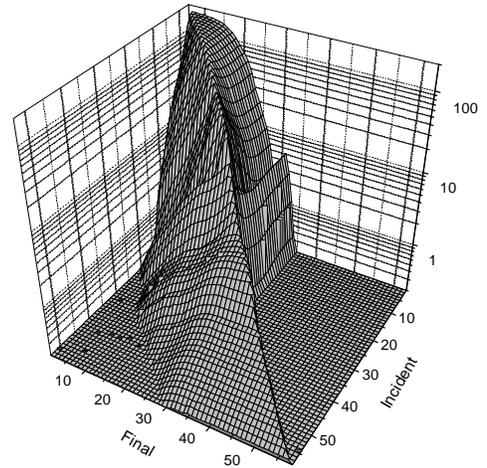
Fig.1(b): Scattering Kernel for H in H_2O at 300K,
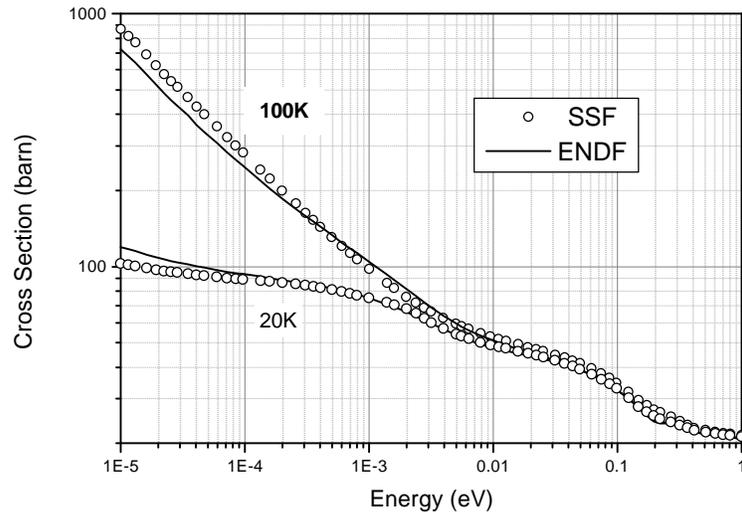calculated by NJOY with option (b) SSF
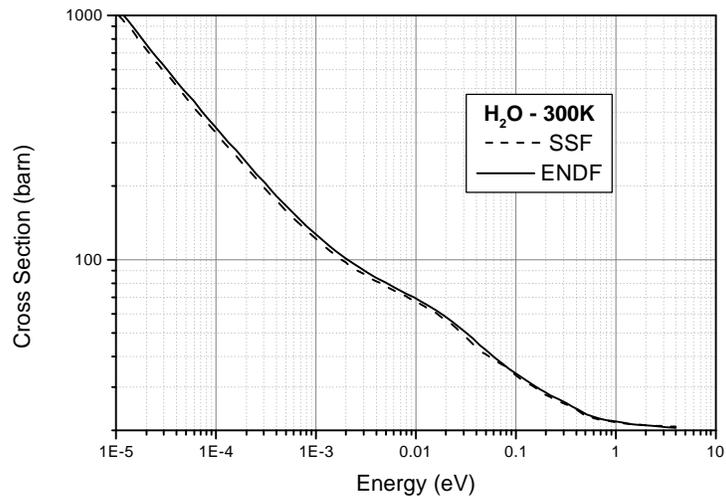
Fig.2: Cross section of H in H_2O at 300K calculated by our
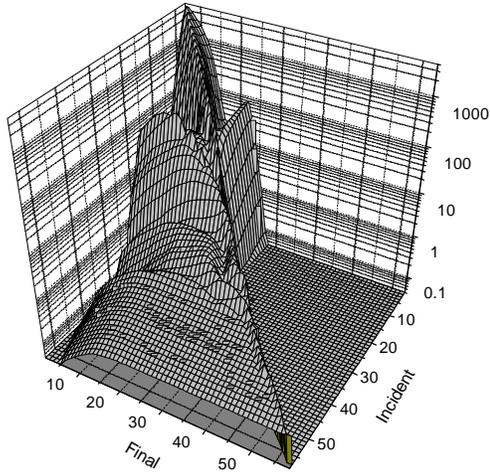NJOY with its two options, over the full thermal energy range.
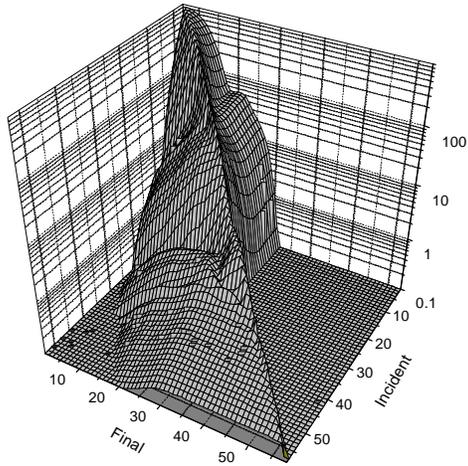
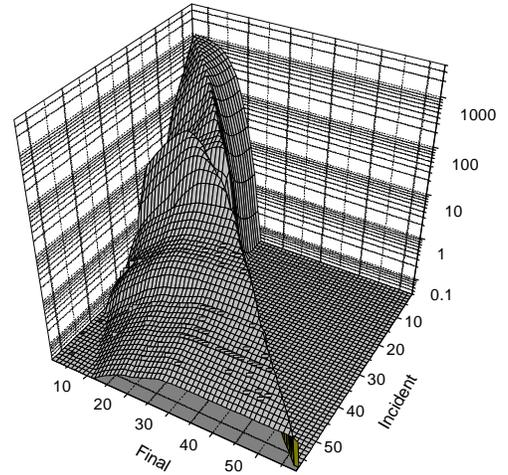**Fig.3(a):** Scattering Kernel for H in CH$_4$ at 100K, calculated by NJOY with option (a) ENDF/B-VI.2
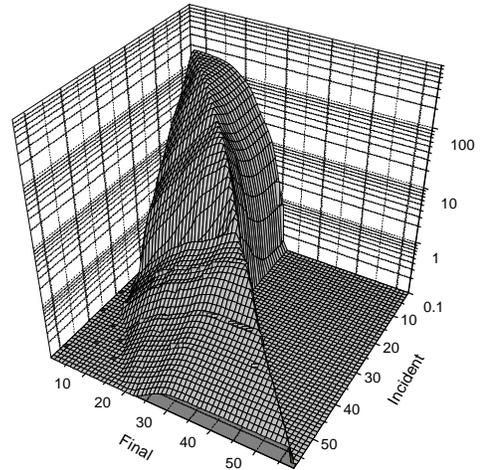
Fig.3(b): Scattering Kernel for H in CH$_4$ at 100K, calculated by NJOY with option (b) SSF

Fig.4: Cross section of H in CH$_4$ at 20K and 100K calculated by our NJOY with its two options, over the full thermal energy range.

Fig.5 (a) Scattering Kernel for H in Para H$_2$ at 20K, calculated by Njoy with option (a) ENDF/B-VI.2

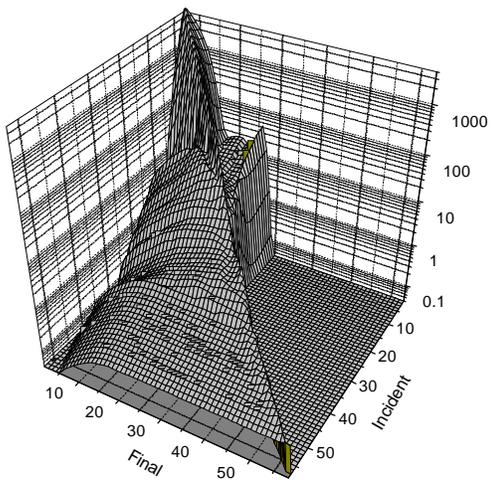**Fig.5 (b)** Scattering Kernel for H in Para H$_2$ at 20K, calculated by Njoy with option (b) SSF

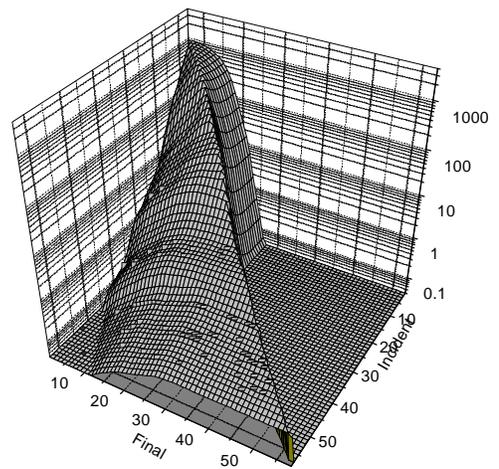**Fig.6 (a)** Scattering Kernel for H in Ortho H$_2$ at 20K, calculated by Njoy with option (a) ENDF/B-VI.2

Fig.6 (b) Scattering Kernel for H in Ortho H$_2$ at 20K, calculated by Njoy with option (b) SSF
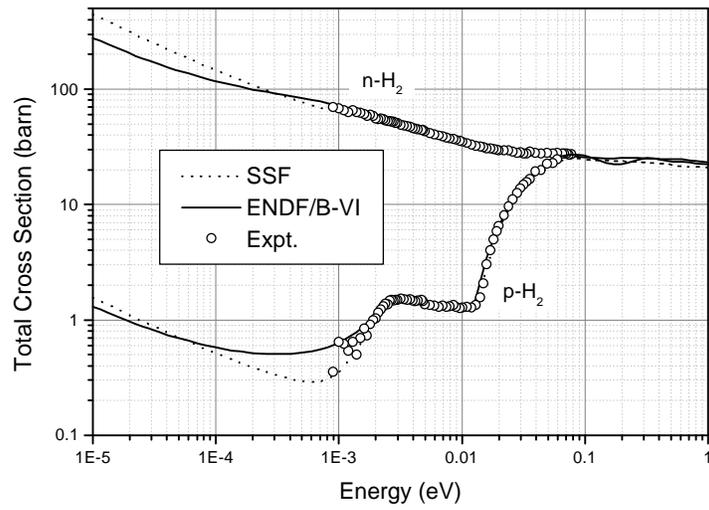
Fig 7 Cross Section for H in Para- and Normal H$_2$ at 20K, calculated by our Njoy with its two options, together with experimental data of Seiffert.[26]