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EVALUATION OF NEUTRON CROSS-SECTIONS FOR LIQUID HYDROGEN AND DEUTERIUM FOR THE DESIGN OF COLD NEUTRON SOURCES

W. Bernnat, D. Emendörfer, J. Keinert, M. Mattes

Institut für Kernenergetik und Energiesysteme, University of Stuttgart,
Pfaffenwaldring 31, D-7000 Stuttgart 80, West Germany

Abstract: Cross-sections for slow neutron scattering from H_2 and D_2 have been calculated taking into account the liquid state. The ability of the model is demonstrated by comparison with experimental results for differential and total cross-sections. For applications the scattering law $S(\alpha,\beta)$ has been prepared in the ENDF-5-format for different temperatures as a basis for the generation of scattering matrices. From calculations of neutron spectra with different scattering models it turns out that the gain factor for cold neutrons is sensitiv to the liquid state. Below 1 meV the frequently used Young-Koppel model for a molecular H_2-gas overestimates the gain factor by about 50 %.

(cold neutrons, liquid hydrogen, liquid deuterium, intermolecular interference, double differential cross-sections, total cross-sections, gain factors)

Scope of the Work

Liquid hydrogen and deuterium are favoured as moderator materials for cold neutron sources in research reactors. To improve design and optimization calculations, realistic scattering models should be applied for neutron-thermalization and -transport in liquid hydrogen and deuterium.

The well-known scattering kernel by Young and Koppel for H_2- and D_2-gas [1] takes into account the spin-dependence of the neutron-proton (deuteron) interactions, free rotations of the molecules, harmonic H-H (D-D) vibrations and free translations of the molecular units:

$$\frac{d^2\sigma}{d\Omega d\epsilon} = a^2 \frac{k}{k_0} S_{Mol.}(\kappa,\epsilon) \tag{1}$$

$$S_{Mol.}(\kappa,\epsilon) = \frac{1}{2\pi} \cdot \int_{t=-\infty}^{+\infty} dt e^{i\epsilon t} \chi_{Mol.}(\kappa,t) \tag{2}$$

$$\chi_{Mol.}(\kappa,t) = \chi_{Free\ Transl.}(\kappa,t) \cdot \chi_{Spin,Rot.,Vib.}(\kappa,t) \tag{3}$$

For an adequate description of the liquid state the scattering function for free translations in (3) has to be substituted in general by a scattering function for the superposition of several translational modes:

i = 1: harmonic lattice vibration
i = 2: diffusive motions
i = 3: hindered translations (with effective mass)

$$\chi_{Transl.}(\kappa,t) = e^{\frac{\kappa^2}{2M_{Mol.}T}\sum_{i=1}^{3} w_i G_i(t)} \tag{4}$$

The corresponding scattering law for translations of the molecules in the liquid state we can derive from the scattering function (4) by a numerical Fourier-transformation with the code GASKET [2]:

$$S_{Transl.}(\kappa,\epsilon) = \frac{1}{2\pi} \int_{t=-\infty}^{+\infty} e^{i\epsilon t} \chi_{Transl.}(\kappa,t)\, dt \tag{5}$$

Scattering Kernel for Liquid Hydrogen

By combining the molecular dynamics with the translational one, we can set up the scattering kernel for the molecular liquid. One way to derive the "self"-term of the double differential cross-section for the liquid state is the numerical convolution of the respective scattering laws [3]

$$S_{Liquid}(\kappa,\epsilon) = \int_{\epsilon'=-\infty}^{+\infty} S_{Transl.}(\kappa,\epsilon-\epsilon')\, S_{Spin,Rot.,Vib.}(\kappa,\epsilon')\, d\epsilon' , \tag{6}$$

$$\frac{d^2\sigma_{Liquid}}{d\Omega d\epsilon} = a^2 \frac{k}{k_0} S_{Liquid}(\kappa,\epsilon) . \tag{7}$$

An equivalent direct evaluation takes into account the effect of typical translational modes (i = 1, 2, 3) for the liquid state already in the multiple summations of the Young-Koppel kernel over the possible initial and final states of the system, compatible with conservation of energy and momentum. Splitting the scattering cross-section into coherent and incoherent contributions, we obtain [4] in extension of the Young-Koppel model

for parahydrogen

$$\frac{d^2\sigma_{coh.}}{d\Omega d\epsilon} = \frac{k}{k_0} a^2_{coh.} S_{even\,even}(\kappa,\epsilon) \tag{8}$$

$$\frac{d^2\sigma_{incoh.}}{d\Omega d\epsilon} = \frac{k}{k_0} a^2_{incoh.} S_{even\,odd}(\kappa,\epsilon) \tag{9}$$

and for orthohydrogen

$$\frac{d^2\sigma_{incoh.}}{d\Omega d\epsilon} = \frac{k}{k_0} a^2_{incoh.} \left[\frac{1}{3} S_{odd\,even}(\kappa,\epsilon) + \frac{2}{3} S_{odd\,odd}(\kappa,\epsilon)\right] \tag{10}$$

$$\frac{d^2\sigma_{coh.}}{d\Omega d\epsilon} = \frac{k}{k_0} a^2_{coh.} S_{odd\,odd}(\kappa,\epsilon) , \tag{11}$$

with the modified partial scattering laws

$$S_{even\,odd}(\kappa,\epsilon) = \sum_n \left(\frac{\kappa^2}{2\,M_{Mol.}\,\omega}\right)^n \cdot \sum_{J=even} P_J \sum_{J'=odd} (2J'+1) \cdot \tag{12}$$

$$\cdot \sum_{l=|J'-J|}^{J'+J} |\,A_{nl}\,|^2\; C^2(J\,J'\,l;0\,0)\;\cdot\; S_{Transl.}(\kappa,\epsilon_{nJJ'})\;,$$

$$\epsilon_{nJJ'} = \epsilon + n\omega + E_{J'} - E_J\;. \tag{13}$$

The matrix elements A_{nl} and the Clebsch-Gordan coefficients $C(JJ'l;00)$ can be taken from [1]. $S_{Trans.}(\kappa,\epsilon_{nJJ'})$ is determined by (5).

In the same way we get
for orthodeuterium

$$\frac{d^2\sigma_{coh.}}{d\Omega d\epsilon} = \frac{k}{k_0}\,a^2_{coh.}\;S_{even\,even}(\kappa,\epsilon) \tag{14}$$

$$\frac{d^2\sigma_{incoh.}}{d\Omega d\epsilon} = \frac{k}{k_0}\,a^2_{incoh.}\,[\,\frac{5}{8}\,S_{even\,even}(\kappa,\epsilon) + \frac{3}{8}\,S_{even\,odd}(\kappa,\epsilon)\,] \tag{15}$$

and for paradeuterium

$$\frac{d^2\sigma_{incoh.}}{d\Omega d\epsilon} = \frac{k}{k_0}\,a^2_{incoh.}\,[\,\frac{3}{4}\,S_{odd\,even}(\kappa,\epsilon) + \frac{1}{4}\,S_{odd\,odd}(\kappa,\epsilon)\,] \tag{16}$$

$$\frac{d^2\sigma_{coh.}}{d\Omega d\epsilon} = \frac{k}{k_0}\,a^2_{coh.}\;S_{odd\,odd}\;, \tag{17}$$

with $S_{even\,odd}(\kappa,\epsilon)$ according to (12).

To include intermolecular interference we use the approach by Vineyard [5], with the final result

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{d^2\sigma_{coh.}}{d\Omega d\epsilon}\;S(\kappa) + \frac{d^2\sigma_{incoh.}}{d\Omega d\epsilon}\;, \tag{18}$$

where $S(\kappa)$ is the static structure factor.

Comparison between Theory and Experiment

For translational modes of type i = 1,3 differential and total cross-sections for liquid hydrogen have been calculated [3] and compared with experiments. The frequency spectrum for harmonic lattice vibrations was chosen in accordance with data derived from experimental and theoretical investigations [6], [7], see Fig. 1. The comparison of calculated double differential cross-sections with measured data in Fig. 2 and Fig. 3 shows that our model for liquid hydrogen is able to represent the characteristic features in better agreement with the experimental results than the Young-Koppel kernel. The same conclusion can be drawn from comparisons of calculated and measured total cross-sections. Fig. 4 shows for example the total cross-section of parahydrogen. The liquid kernel reproduces the characteristic decrease between 50 and 10 meV much better than the gas kernel. The same tendency has been observed for a liquid kernel with translational modes i = 1,2, where the intermolecular vibrations are described by a single frequency parameter [8].

The second characteristic decrease below 5 meV is due to the intermolecular interference and can be represented according to (18). Our static structure factor (see Fig. 5) has been approximated for low κ-values by a solution of the Percus-Yevick equation for hard spheres [12], and for higher κ-values by a Fourier-Transformation of the low density pair distribution function with the Lenard-Jones Potential for the interaction of the molecules.

Fig. 6 shows our calculated total neutron cross-section for liquid deuterium and results from Utsuro [9] compared with experimental data. Apparantly for deuterium the agreement between theory and experiment is not as good as in the case of hydrogen.

Scattering Law Representation

As a data basis for nuclear engineering applications the scattering law for proton and deuteron in liquid hydrogen and deuterium,

$$S(\alpha,\beta) = e^{\frac{\beta}{2}}\;S(\kappa,\epsilon)\;, \tag{19}$$

$$\alpha = \frac{\kappa^2}{2\,MT} \qquad \beta = \frac{\epsilon}{T}\;, \tag{20}$$

has been generated in the ENDF-5 format for a fine (α,β)-grid and different temperatures. Since the principle of detailed balance is violated as a consequence of the spin dependence of the nuclear reaction, the scattering law data in ENDF-5 format are to be extended to negative β-values. This implies that the cross-section generation code NJOY [10], which carries out integrations over α and β to produce group cross-sections from the scattering law $S(\alpha,\beta)$, has to be modified [11].

Neutron Spectra

With scattering matrices generated from different scattering kernels for hydrogen, we have solved the neutron transport equation in B_N-approximation for the conditions of a cold neutron source, see Fig. 7. The intermolecular interaction in the liquid state contributes to the reduction of the slowing down power in addition to the effect of the chemical binding in the hydrogen molecule. Therefore, realistic neutron spectra calculated with the scattering kernel for liquid hydrogen are hardened relativ to spectra calculated with the Young-Koppel kernel. This means, that the number of cold neutrons predicted by the Young-Koppel kernel is too high. The gain factors for cold neutrons in liquid hydrogen at 20 K (related to water at room temperature) are overestimated by the Young-Koppel model in the energy range below 1 meV by about 50 %.

Acknowledgement

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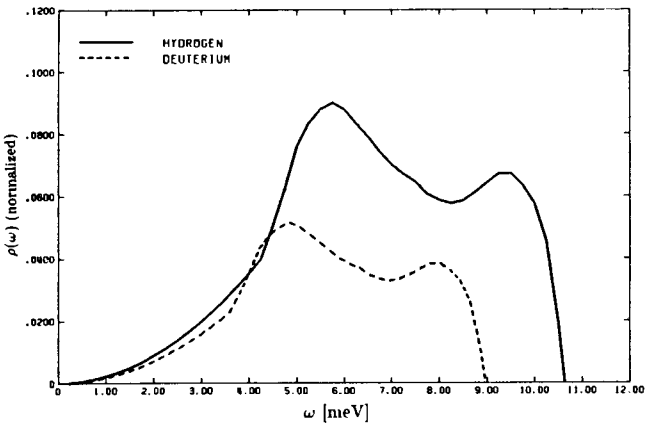

Fig. 1: Frequency distributions of the translational modes in liquid hydrogen (H_2) and deuterium (D_2)

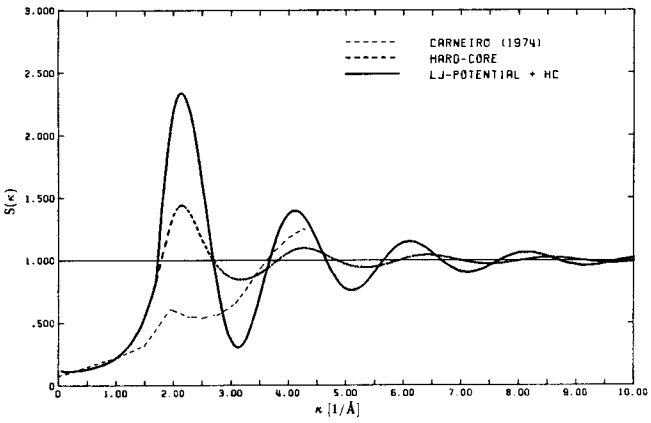

Fig. 5: Static structure factor S(κ) in liquid hydrogen (H_2)

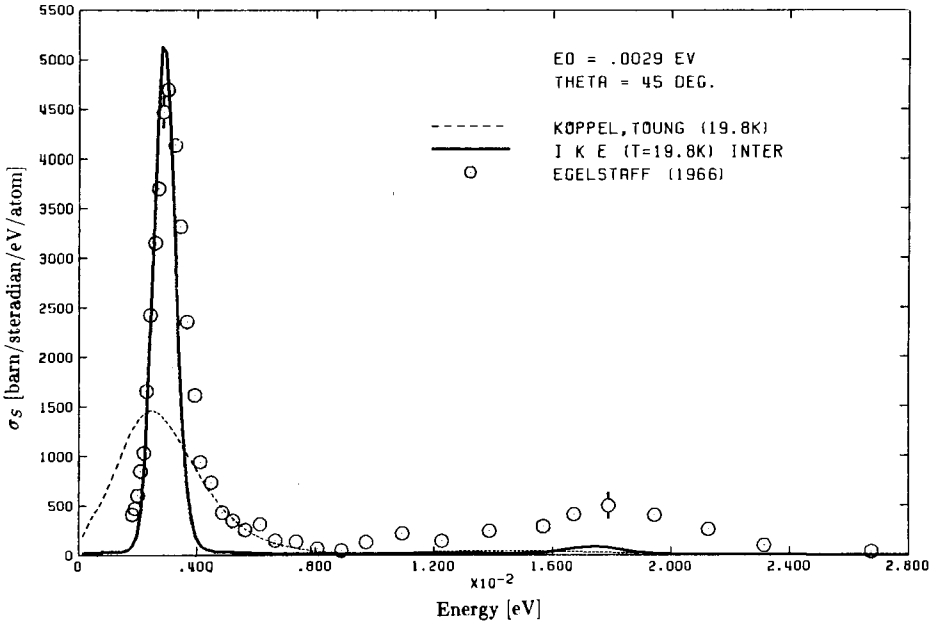

Fig. 2: Double differential neutron scattering cross-section for normal hydrogen (n – H_2) at T = 18 K

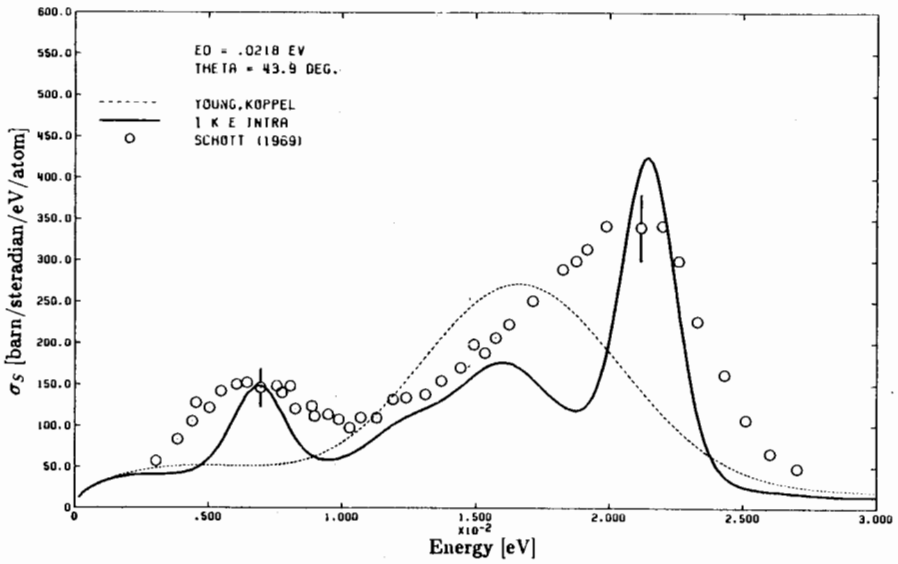

Fig. 3: Double differential neutron scattering cross-section for hydrogen (52 % p - H_2) at T = 19.8 K

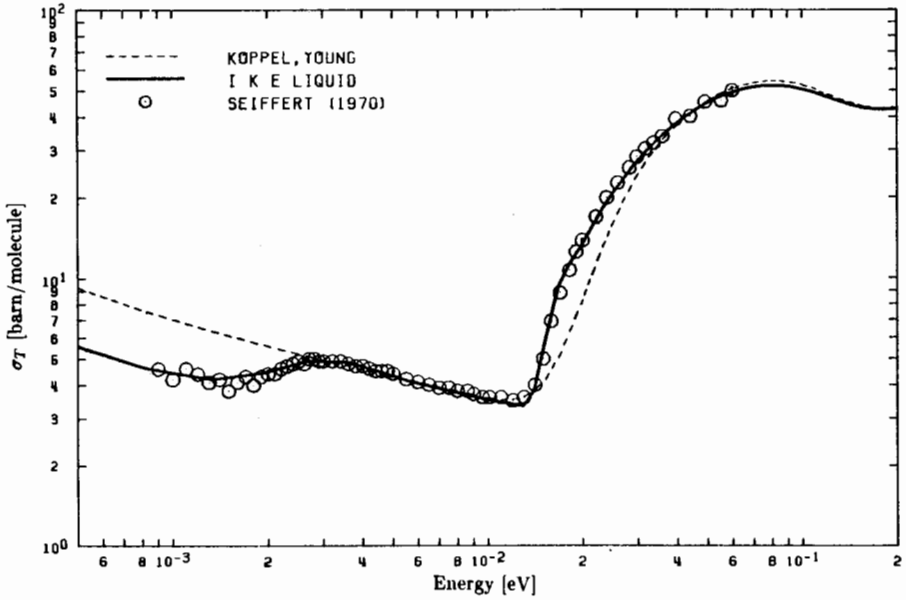

Fig. 4: Total neutron cross-sections for parahydrogen at T = 14 K

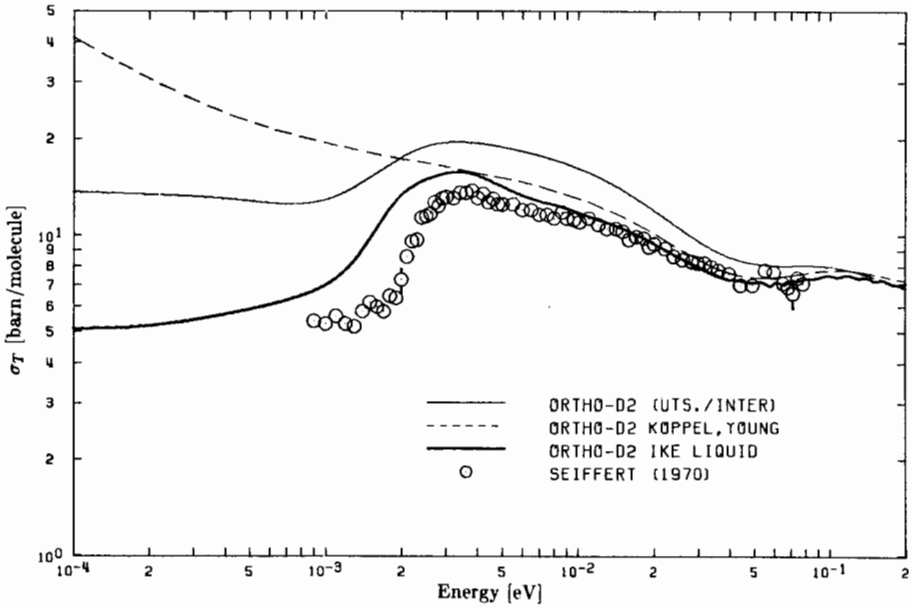

Fig. 6: Total neutron cross-sections for deuterium at T = 19 K

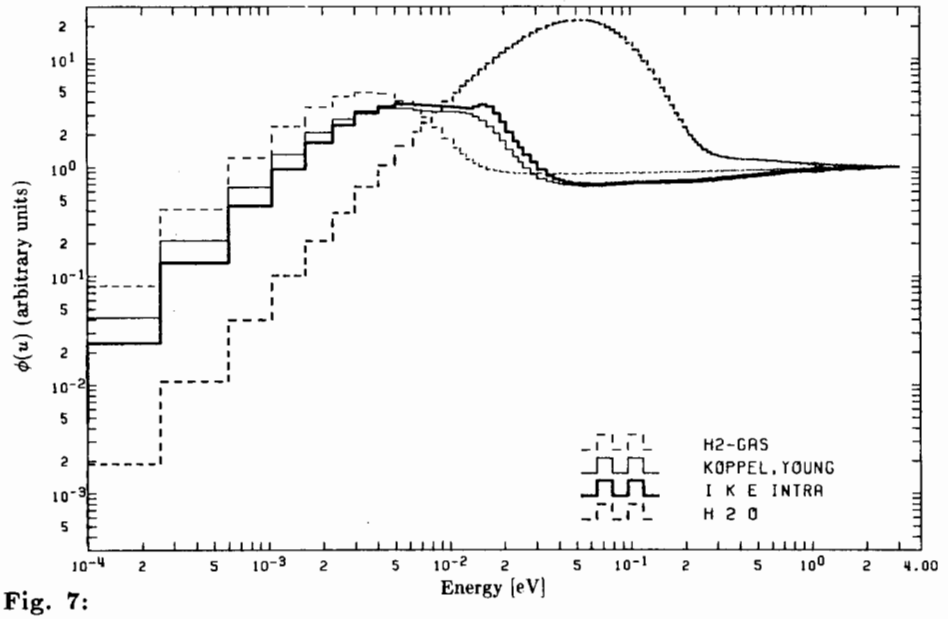

Fig. 7:
Neutron flux spectra in a liquid hydrogen (n - H_2) cold neutron source at T = 20 K