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NEUTRON SCATTERING AND TRANSPORT PROPERTIES IN LIQUID H_2 AND D_2 USING A SYNTHETIC SCATTERING FUNCTION

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Abstract: A synthetic model has been used to describe the interaction of slow neutrons with H_2 and D_2 molecules. The model yields analytic expressions for the double-differential scattering cross section, the P_0 and P_1 energy-transfer kernels and the total cross section. From these, neutron scattering and transport properties in liquid H_2 and D_2 are calculated and compared with available experimental results, showing very good agreement. These evaluations are relevant for cold neutron source design.

(cold neutrons, liquid H_2 and D_2, bound atom model, scattering and transport properties)

Introduction

Molecular hydrogen and deuterium represent a simple system from a theoretical point of view, although quantum effects are apparent in the liquid state. On the other hand, the unique neutronic properties of H_2 and D_2, have make them a natural choice as moderating materials for the production of cold neutrons.

From the total cross section measurements on liquid H_2 performed by Squires and Steward /1/ and the theoretical calculation of Sarma /2/, up to the recent work of Keinert and Sax /3/, a large body of calculational techniques has developed, most of them based on the gas model due to Young and Koppel /4/. Unfortunately, no much experimental information on the neutron field behavior in those system is yet available/5/.

In a homonuclear diatomic molecule, the transformation properties of the total wavefunction causes the total nuclear spin I and the total angular momentum J to be correlated, thus producing a well defined "selection rule" which governs the transitions between states corresponding to those quantum numbers /6/. Such restrictions are especially important in the case of H_2 and D_2 at low temperatures, where not many rotational levels are excited. Liquid hydrogen for example, exist in two forms: para (I = 0) and ortho (I = 1), with no rotational states with odd J in the case of antiparallel spins (para) and with states of odd J only in the case of parallel spins (ortho). As a consequence of the nuclear spins correlation, the neutron cross sections for the two forms show significant interference effects.

Equivalent rules hold for liquid deuterium, with the difference that having the deuteron spin 1, the possible total (nuclear) spins of the molecule are I = 0,1,2. Therefore, the symmetric (ortho) spin arrangements have I = 0,2 and these correspond to even J values while the antisymmetric (para) states have spin 1, corresponding to odd J values.

In general, a knowledge of the full molecular dynamics is required in order to describe the cross sections for the neutron-molecule interaction. However, many neutron properties can be evaluated through a simplified scheme to represent the actual frequency spectrum, and this has been the central idea behind the development of the Synthetic Scattering Function /7/. This formalism has been successfully applied to the description of neutron scattering and transport properties of several hydrogeneous systems /8,9/. In this work a synthetic model for H_2 and D_2 is presented, and some preliminary results are given and compared with available information.

Synthetic model for H_2 and D_2

In the frame of the synthetic scattering function hypothesis, the actual frequency spectrum of the system is lumped into discrete harmonic oscillator modes, each of these having an occupation number for the excited levels controlled by the system's temperature. Furthermore, collision-induced transitions between those levels are allowed in the neutron-molecule interaction, which in turn depends on the incident neutron energy E_0. In this way, a synthetic function $T(Q,\omega ; E_0)$ is obtained, where the effects of the molecular dynamics on the scattering cross sections is represented through the variation of an effective mass μ, temperature τ and vibrational factor Γ /7/.

Our model for liquid H_2 and D_2 is constructed on the basis of the Young-Koppel model /4/ to describe the rotational elastic and one-phonon inelastic cross sections, together with the usual prescriptions of the general synthetic model for the molecular translation and vibrational forms of the scattering function. 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 /10,12/. Its eigenfrequency was derived from accepted values of the Debye temperature, with a weight (effective mass) consistent with the assumption of two molecular units moving together as a description of the translational motion "seen" by very low energy neutrons. This latter idea was already suggested by Whittemore /13/ many years ago.

The experimentally observed drop in the total cross section of liquid parahydrogen

(and deuterium) below ~0.003 eV is caused by intermolecular interference effects. We have taken this behavior into account by imposing a complete cancellation of the coherent elastic scattering, as the result of the composition of intra-and intermolecular interference contributions. A more rigorous treatment of these effects should be based on the consideration of the static structure factor of the liquid system /10/.

The synthetic model provides analytical expressions for several magnitudes of interest in neutron physics calculations (i. e. total scattering cross sections, P_0 and P_1 energy-transfer kernels) as well as neutron transport magnitudes by a simple integration. In this way, evaluations involving changes of parameters (ortho - para concentration, density, temperature, weighting spectrum) can be easily performed in a fast and accurate manner. As an example, a complete calculation involving total scattering cross section and average cosine of the scattering angle at 76 incident neutron energies, P_0 and P_1 energy-transfer kernels (each a 76 x 150 matrix) and neutron diffusion parameters, takes about 100 sec of CPU time on a VAX 11.

The input data for all the calculations presented here are summarised in Table 1. Absorption and bound atom scattering cross sections were taken from Ref.14.

Table 1.Input data for the model calculations

Qty	H_2	D_2
σ_c (b)[1]	1.7583	5.592
σ_i (b)[1]	79.91	2.04
σ_a (b)[2]	0.105	1.65×10^{-4}
$\hbar\omega_D$(eV)	0.005	0.003
M_D(amu)	4	8
$\hbar\omega_r$(eV)	0.0147	0.0073
M_r(amu)	3	3
$\hbar\omega_v$(eV)	0.546	0.386
M_v(amu)	6	12

1: bound atom cross section.
2: molecular absorption constant; $\sigma_a(E)=\sigma_a/\sqrt{E}$

Results

Although the merits of a bound atom model cannot be assessed through its ability to predict a total cross section, a satisfactory description of this quantity is a necessary condition. Figure 1 shows the results of the present model, together with the experimental data of Seiffert /15/ for the total cross section of para-and normal-hydrogen. Small discrepancies are apparent at the low energy side, and these are of different origin for the upper (n-H_2) and lower (p-H_2) curves. In the case of normal hydrogen, it is caused by the crudeness in the treatment of the translational motion borned into the model, while all scattering components have been suppressed for incident neutron energies lower than ~ 0.001 eV in the case of para hydrogen. However, the overall agreement is very good and comparable to that obtained from more elaborated models /10,11,15/.

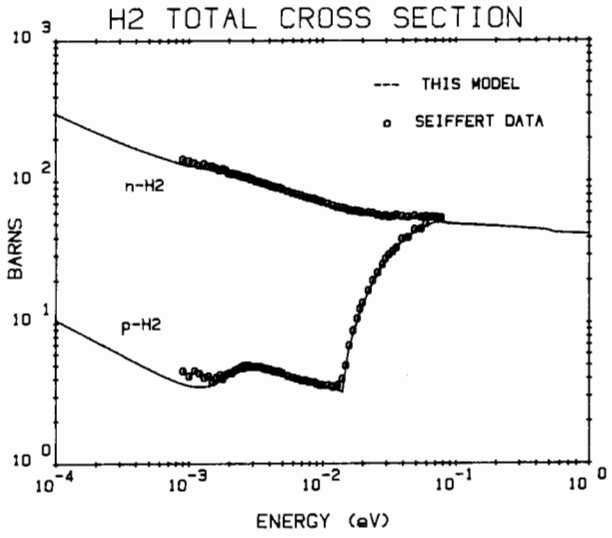

Fig.1 Total cross section of normal- and ortho-hydrogen.

In Fig.2 the total cross section data of Seiffert for liquid D_2 are compared with the synthetic model results for ortho and normal deuterium. The agreement between experiment and calculation in this case is not as good as for H_2 (due to coherence effects), but still quite satisfactory.

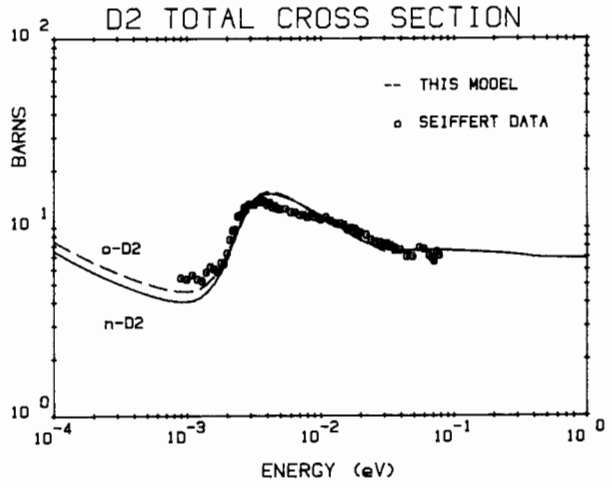

Fig.2 Total cross section of normal-and ortho-deuterium

Energy-transfer kernels at selected incident neutron energies are shown in Figs.3 and 4 for liquid H_2 and D_2, respectively. In the first of them, o-H_2 presents an almost gas-like behavior at E_0 = 0.018 eV, while p-H_2 shows the effect of the J = 0→1 flipping transition through a pronounced down-scattering contribution. Similar behavior is observed in the case of D_2 (Fig.4), but it is the ortho-deuterium which presents now a distinct downscattering component for an incident neutron energy E_0 = 0.01 eV

The analytically generated scattering kernels were used to evaluate some other integral properties. As an example, Fig.5 shows the slowing down power $\xi\sigma$ (per atom) of

ortho-and para-hydrogen which turn out to be very similar to those based on the Young-Koppel model /16/, although the J = 0 → 1 transition peak (~ 0.055 eV) for p-H_2 is not as high in our calculation. The large negative values of $\xi\sigma$ for o-H_2 at low energy reflect the importance of upscattering processes through the transition J = 1 → 0.

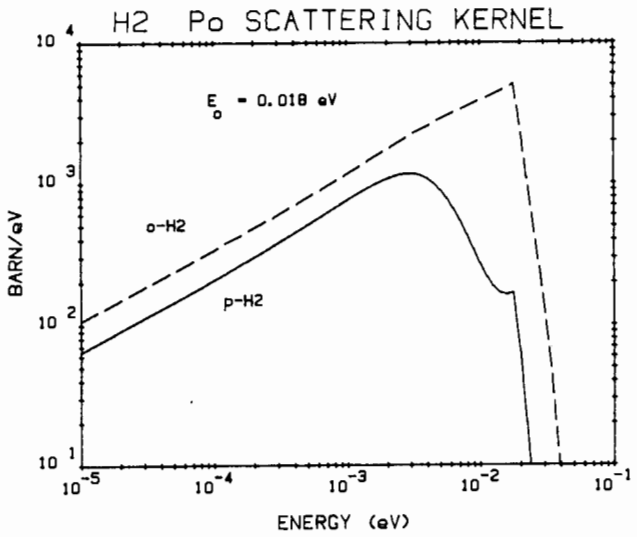

Fig.3 Energy-transfer kernels for H_2 at E_0 = 0.018 eV

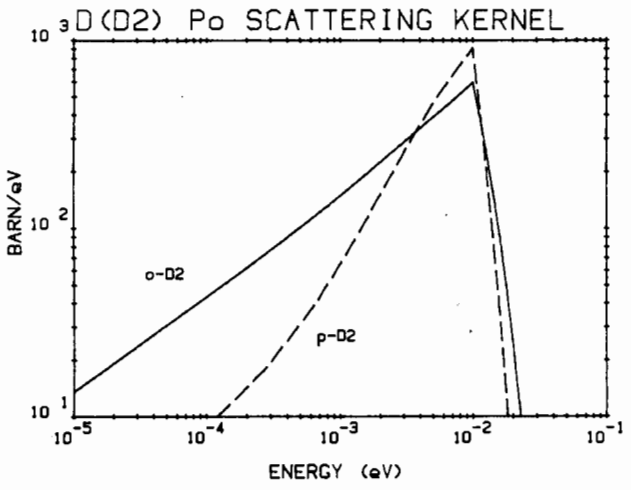

Fig.4 Energy-transfer kernels for D_2 at E_0 = 0.01 eV

Fig.6 shows our calculated curves for the average cosine of the scattering angle, $\bar{\mu}$, in ortho-deuterium.

It is obvious that neutron transport parameters will strongly depend on the para to ortho concentration, as well as on the neutron spectrum. Concerning this latter point, a Maxwellian distribution has been adopted for the sake of simplicity, with different temperatures T_M to simulate the spectrum hardening due to leakage in the smaller geometries.

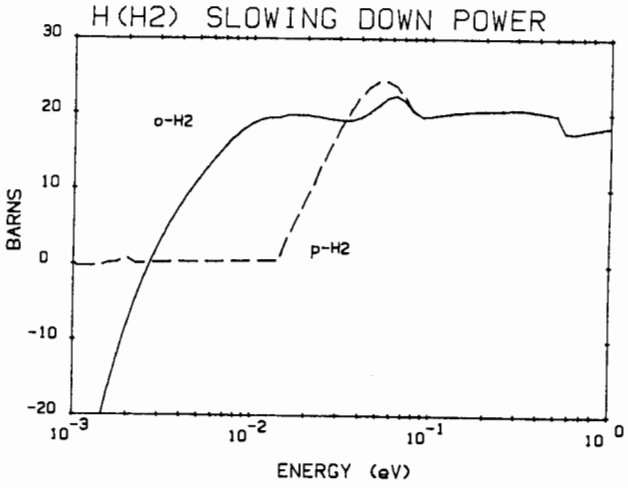

Fig.5 Slowing down power of H_2

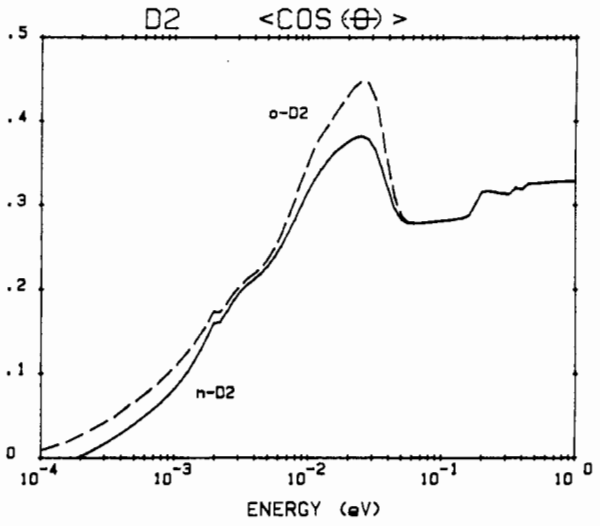

Fig.6 Average cosine of scattering angle for D_2

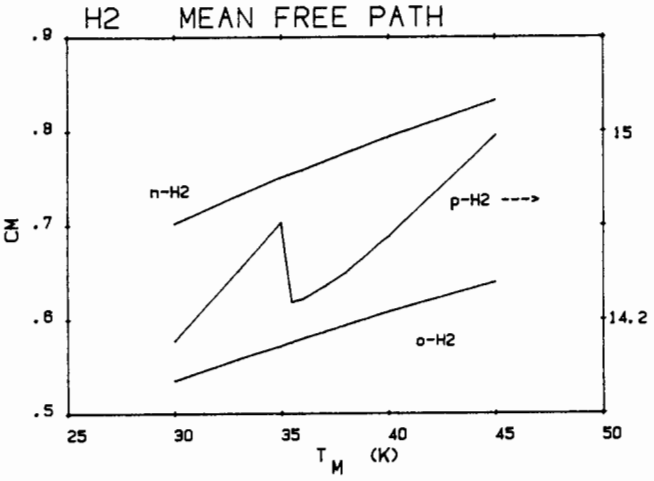

Fig.7 Mean free path in H_2 vs. spectrum temperature

In Fig.7 we present the results of the synthetic model for the neutron transport mean free path λ_{tr} , over a temperature range centred around the accepted equivalent Maxwellian temperature of 37 K /5,11,17/ for liquid hydrogen. A monotonic increase with T_M is observed for both ortho-and normal-hydrogen, whereas in the case of p-H_2 a large step occurs in λ_{tr} (note the different scale). Similar calculations for liquid D_2 are shown in Fig.8

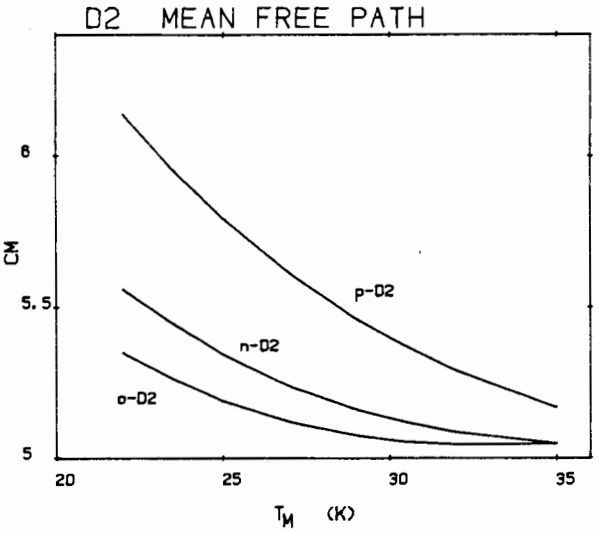

Fig.8 Idem Fig.7, but for D_2

The dependence of λ_{tr} for liquid H_2 on the ortho: para concentration is shown in Fig.9, together with the results of Wurz calculations /11/. There is good agreement between both evaluations over most of the range, except in the high para-concentration limit, where our model gives slightly larger values. Finally, Fig.10 shows our prediction for the neutron diffusion length L in H_2 as a function of the o - p concentration; a much smaller variation is obtained for these magnitudes in the case of liquid D_2.

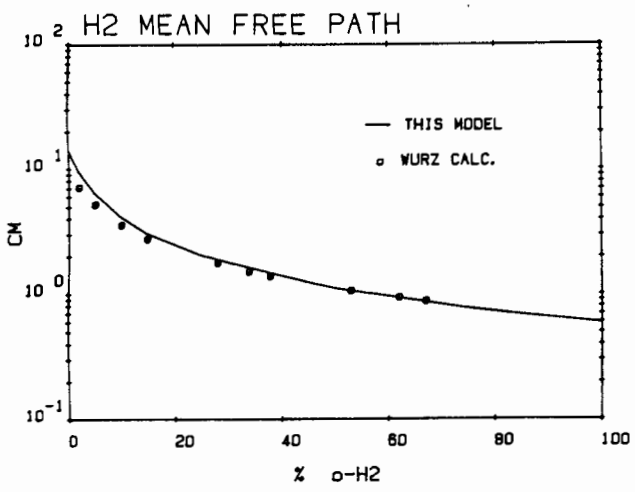

Fig.9 Mean free path in H_2 vs.ortho: para concentration.

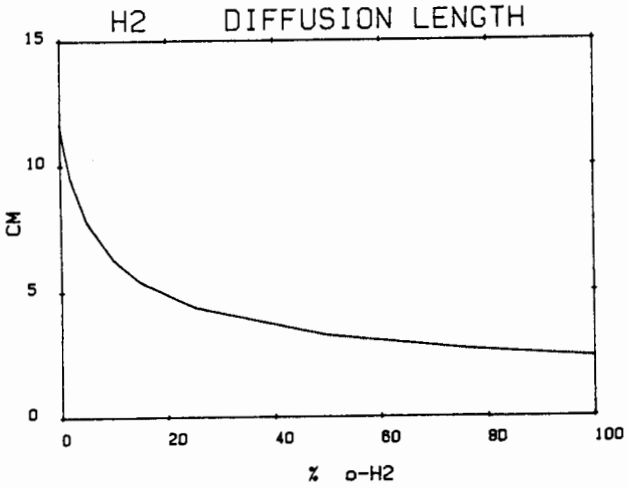

Fig.10 Idem Fig.9, but for the neutron diffusion length.

Conclusions

In the frame of the synthetic scattering function formalism, a model to describe neutron scattering and transport in liquid H_2 and D_2 has been developed. We have used the analytic expressions supplied for the model to evaluate a number of magnitudes of interest in cold neutron source design. A more comprehensive body of results, including spectra calculations and gain factors for different geometries and concentrations, will be presented in a forthcoming publication.

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