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Cross-Section Models of Liquid H_2, Solid and Liquid CH_4 and Light Water for a Pulsed Cold-Neutron Source

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A set of cross-section models for neutron scattering in hydrogenous moderators is developed, such as liquid para- and ortho-H_2 between melting and boiling points, solid and liquid CH_4 for temperatures down to 20 K, and light water around 300 K in a wide range of neutron incident energies from 0.1 μeV to 10 eV. Neutron scattering property, both up and down, is studied quantitatively by comparison with many experimental cross-section results. Typical features of each moderators are clarified: for instance, cold-neutron production due to para-to-ortho transition at lower temperatures, an increase in upscattering of lower-energy neutrons with an ortho-H_2 concentration, neutron slowing-down by rotational excitation of a CH_4 molecule and some energy exchanges inherent to hydrogen-bonded water molecules. It is shown that major intermolecular motions as well as intramolecular ones for these moderators are adequately described in cold and thermal neutron region. This permits us to use the models for neutronic considerations of a pulsed neutron source in terms of energy-dependent intensity and pulse width. A basic data library as a group-constant set is now being generated. Part of the results is given.

KEYWORDS: cold neutron, neutron source, liquid hydrogen, solid methane, light water

§1. Introduction

Low-energy neutrons such as cold(~1 meV) and thermal (~25 meV) neutrons are very useful for a broad range of condensed-matter studies. Hence more intense beams of such neutrons are being required. For this, a large number of fast neutrons emerged from a pulsed spallation source must be slowed down as efficiently as possible. And it is necessary to maximize a neutronic performance of ambient temperature(H_2O) and cryogenic temperature(H_2 and CH_4) moderators.

Among many moderating materials, these hydrogenous ones have many advantages: large scattering cross section leading to rapid moderation and small source volume; good removal of neutron kinetic energy largely by excitation of molecular rotation; favorable refrigeration requirements due to low melting and boiling points for H_2 and CH_4 ; and much less technical problems of liquid moderators as compared with solid ones suffering from radiation damage.

In the present paper, cross-section models for these hydrogenous materials are presented. It is shown that the models are able to cover a wide range of neutron incident energies from 0.1 μeV to 10 eV and of many different temperatures for liquid H_2 between 14.0 and 20.4 K and for solid and liquid CH_4 from 20 to 111 K. Preliminary results on group-constant sets for liquid para- and ortho-H_2 are also given.

§2. Liquid Hydrogen

A cross-section model for liquid H_2 has been developed,[1] in which intra- and inter-molecular motions are properly taken into account. The former is followed from the Young-Koppel model[2] describing the nuclear-spin correlations of two atoms in a molecule, free rotations of a molecule and harmonic stretching vibrations of an atomic bond. On the other hand, an intermolecular motion is expressed in terms of the product of a static structure factor and a self-scattering function. The following molecular motions are included: very short-time free-gas like translation, short-lived vibration and long-time diffusion. Note that coherent scattering from para-H_2 is described by a convolution approximation.

Good performance of the present models is found by a satisfactory agreement with many experimental cross-section results, both double-differential[3,4] and total,[5] for widely different temperatures and ortho-para ratios. An extensive calculation shows that the models are able to predict scattering processes occurring in the energy range from 0.1 μeV to 10 eV at any temperature between

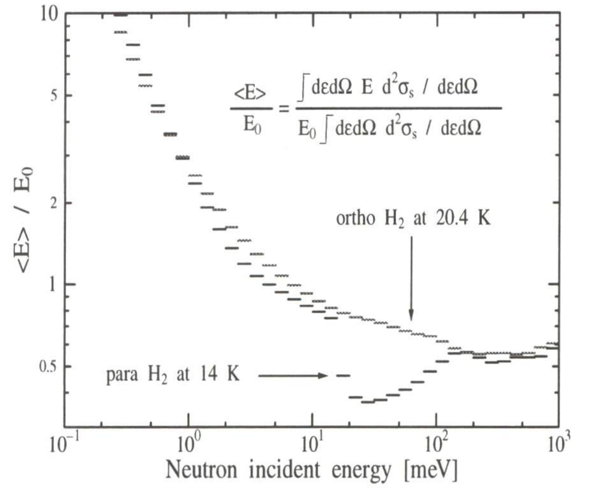

Fig. 1. Ratios of an average final energy to an incident neutron energy for liquid para- and ortho-H_2.

14.0 and 20.4 K for any ortho-para ratios.[6)]

From these studies, a slowing-down property of a low-energy neutron is clarified. It is summarized as follows.

- A para-to-ortho transition is very significant for incident energies around 19.4 meV, especially at low temperatures down to 14.0 K. For para-H_2 at 14.0 K, about two-thirds of the magnitude of total scattering cross-section is occupied with cold neutron production.[13)] This is shown in Fig.1 in terms of the ratio of an average scattered-neutron energy to an incident neutron energy.
- An ortho-H_2 as spin-incoherent scatter has a scattering cross-section being about 25 times larger than a para-H_2 in the cold neutron region. A thermal neutron is down-scattered largely by excitation of intermolecular vibration with a characteristic energy of about 5.3 meV, while a cold neutron is quasi-elastically scattered by molecular diffusion.
- With increasing energy of a neutron above several tens meV, a recoil scattering with a free molecule becomes dominant. At more higher energies, a free-atom scattering appears.

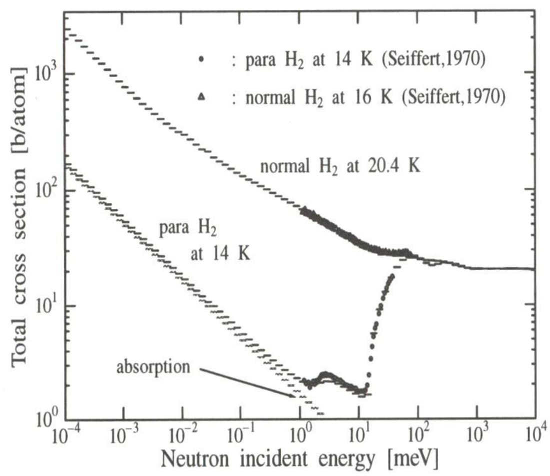

Fig.2. Total cross sections of liquid H_2, together with the experimental ones.[5)]

The present models are then utilized to generate a set of group-constants for para- and ortho-H_2 at 14.0 and 20.4 K: 80 energy groups from 0.1 μeV to 10 eV at an equidistant interval on logarithmic scale by P_3 approximation. The resultant total cross sections for para-H_2 at 14 K and normal-H_2 at 20.4 K are shown in Fig.2, in which experimental results on para-H_2 at 14 K and normal H_2 at 16 K[5)] are also presented. Hence it is ready for a neutron transport analysis on a pulsed cold-neutron source.

§3. Solid and Liquid Methane

A differential cross-section model for solid and liquid CH_4 has recently been developed. It is based on incoherent approximation adequate for scattering in four protons against a carbon. Use is made of Gaussian approximation of an intermediate scattering function, so that some molecular motions are expressed in terms of a width function defined by a frequency distribution function $g(\omega)$. The following molecular dynamics are included.

- Translational motions of a molecule by very short-time free-gas like behavior, short-lived vibration with a characteristic energy of about 6 meV and, in liquid state, long-time diffusion with a temperature-dependent coefficient $D(T)$.
- Rotational motions of a molecule by short-time free-rotator behavior and long-time diffusion. These depend largely on temperature through respective time constants and contribute to small-energy transfer scattering, i.e. down scattering of a thermal neutron and quasi-elastic scattering of a cold neutron.
- Intramolecular motions with two representative vibration energies of 170 and 387 meV, instead of 9 original ones.

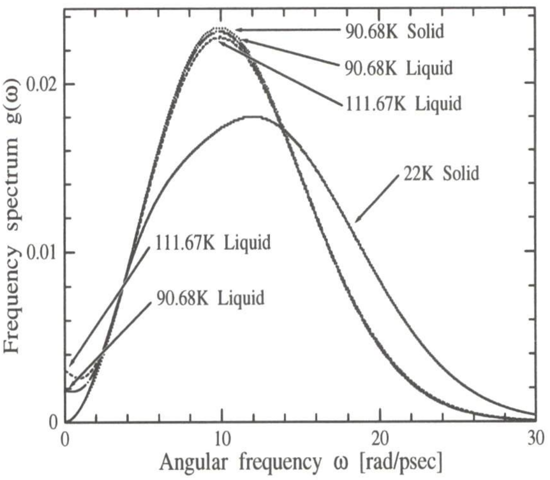

Fig.3. $g(\omega)$ for liquid and solid CH_4.

The resulting $g(\omega)$ for liquids at the melting and boiling points and for solids at the melting point and 22 K are shown in Fig.3. It is clear that there is significant variation in the form of $g(\omega)$ between liquid and solid states and at very low frequencies for liquid. The latter is due largely to $D(T)$. Total area of $g(\omega)$ is adjusted to a constant value of 0.28, while each component of molecular diffusion, vibration and rotation has a temperature-dependent magnitude.

Figure 4 shows the differential cross sections at four temperatures for an incident neutron energy of 4.87 meV at a scattering angle of 90°. In the liquid state, a quasi-elastic scattering peak due to translational and rotational diffusion is present, together with a broad peak of a intermolecular vibration. On the other hand, in the solid state, a very large elastic peak appears on the quasi-elastic component due to rotational diffusion. Although a free-rotation with small energy transfer is associated, it is included in the bottom of the quasi-elastic peak. Consequently, even for a neutron with several meV, this rotational motion is still effective for down scattering.

The present models can be compared with experimental data on total and double-differential cross sections.

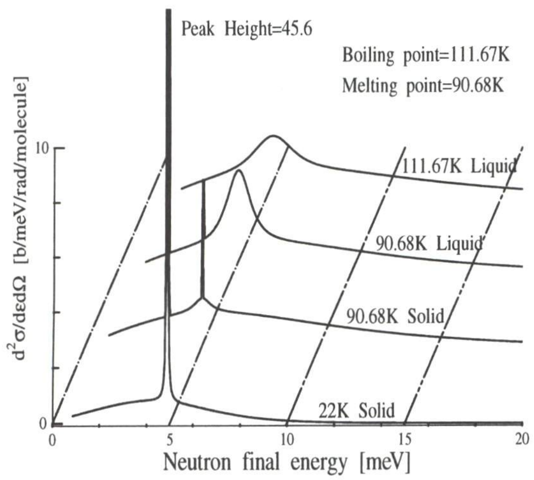

Fig. 4. Double differential cross sections of solid and liquid CH_4.

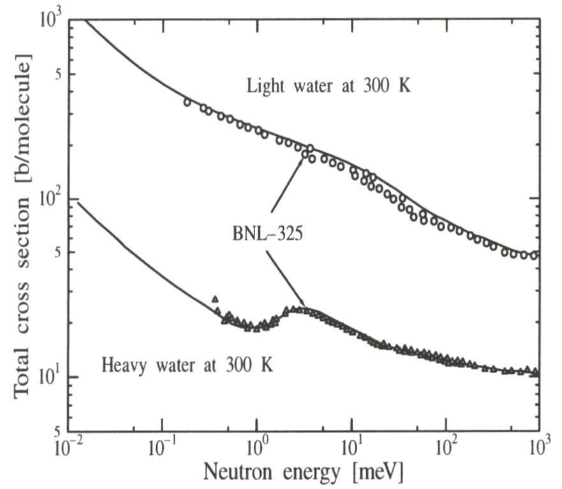

Fig. 6. Total cross sections of light water, together with the experimental ones from BNL-325.[12]

As an example, Figure 5 shows the energy spectra of scattered neutrons for solid CH_4 at 22.1 K at an incident energy of 34.7 meV and constant scattering angles of 30.2°, 68.0° and 90°. There is a satisfactory agreement between the calculated and experimental[7] results, except for the magnitudes of an elastic peak.

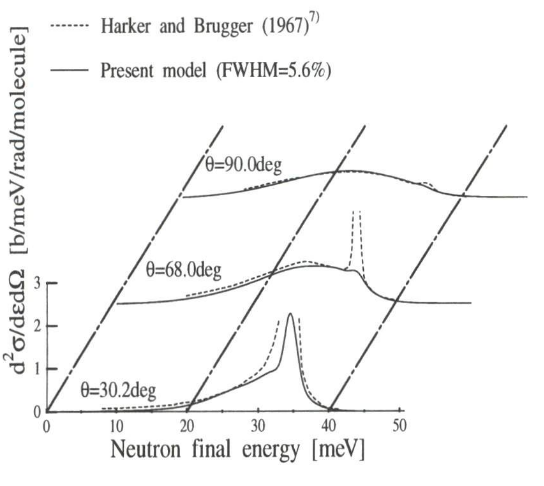

Fig. 5. Double differential cross sections of solid CH_4.

§4. Light Water

Cold and thermal neutron scattering from light water is studied theoretically by developing a differential cross-section model.[8] Molecular motions in water are expressed in terms of long-time diffusion and short-lived intermolecular vibration in a highly-bonded molecular cluster. Hindered rotations of a molecule and intramolecular vibrations, both bending and stretching, are also included. An approach to a free-atom cross section for high incident energies above about 1 eV is taken into account.

Based on the Gaussian description of a space-time density correlation function, the model is described basically by $g(\omega)$ and calculated numerically for double-differential and total cross sections. Good agreement with some typical experimental results[9–11] is found. Such an example is shown in Fig.6 in which total cross sections for H_2O and D_2O at 300 K are compared with the experimental ones from BNL-325.[12] The other integral properties such as an average cosine of scattering angle and an average final energy of a scattered neutron are also checked successfully. Hence the model is now being utilized to generate a group-constant set.

§5. Concluding Remarks

A set of cross-section models for the hydrogenous moderators on H_2, CH_4 and H_2O has been developed successfully. Hence they are now being utilized for generation of group constant sets and then a neutron transport analysis. This makes quantitatively clear the performance of a pulsed cold-neutron source in terms of energy-dependent intensity and other characteristic.

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