Journal of the Korean Physical Society, Vol. 59, No. 2, August 2011, pp. 1076∼1079

Neutron Cross Sections Libraries for Methane in Phase II and Solid Deuterium

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(Received 26 April 2010)

We present new cross section libraries for two materials of interest as very cold and ultra cold neutron moderators: solid Methane in phase II and solid Deuterium. A simple model to describe the interaction of slow neutrons with solid methane in phase II (T < 20.4 K) was developed, including the main dynamical features of the system and the effect of spin correlations. Its predictions are in good agreement with a quantum mechanical calculation over the limited range where the latter was formulated, and with available experimental information over the complete thermal energy range. Also, we introduce cross section libraries based on a new scattering kernel recently developed to describe the interaction of slow neutrons with solid Deuterium (T < 18.7 K). The main characteristics of that system are contained in the model, including the lattice's density of states, the Young-Koppel quantum treatment of the rotations, and the internal molecular vibrations. The elastic processes involving coherent and incoherent contributions are fully described, as well as the spin-correlation effects. The results from the new model are compared with the best available experimental data, showing very good agreement.

PACS numbers: 25.40.Dn, 25.40.Fq, 28.20.Cz
Keywords: Cryogenic moderators, Methane II, Solid deuterium
DOI: 10.3938/jkps.59.1076

I. INTRODUCTION

The Van Hove scattering function S($\mathbf{Q}$,ω) is directly related to the double-differential cross section [1]:

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k}{k_0} S\left(\mathbf{Q},\omega\right), \tag{1}$$

where $\mathbf{k}$, $\mathbf{k_0}$ are the scattered and initial neutron wave vectors, $\hbar\omega$ is the neutron energy loss, and $\hbar\mathbf{Q} = \hbar(\mathbf{k_0} - \mathbf{k})$ is the momentum transferred to the system.

The scattering law of a molecular system can be written as the sum of inter ($l \neq l'$)- and intra ($l = l'$) molecular contributions, or in terms of its Fourier transform as

$$\chi(\mathbf{Q},t) = \left\langle \sum_{l\neq l'} \sum_{v,v'} a^*_{lv} a_{l'v'} \exp\{-i\mathbf{Q}\cdot\mathbf{R}_{lv}(0)\} \exp\{i\mathbf{Q}\cdot\mathbf{R}_{l'v'}(t)\} \right\rangle + \left\langle \sum_{l} \sum_{v,v'} \overline{a^*_{lv} a_{lv'}} \exp\{-i\mathbf{Q}.\mathbf{R}_{lv}(0)\} \exp\{i\mathbf{Q}\cdot\mathbf{R}_{lv'}(t)\} \right\rangle . \tag{2}$$

The scattering lengths a_{lv}appearing in the above equation are spin dependent quantities, and therefore must be in principle included within the expectation value brackets of the intermediate scattering function $\chi(\mathbf{Q},\omega)$. In terms of the usual coherent, b_c^v, and incoherent, b_i^v, scattering lengths for nuclei v, one obtains:

$$a_v = b_c^v + 2b_i^v \left(\mathbf{S}_v \cdot \mathbf{s}\right) \left[S_v\left(S_v+1\right)\right]^{-1/2}, \tag{3}$$

where $\mathbf{S}_v$ and $\mathbf{s}$ are the spin operators for nuclei v and the neutron, respectively.

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The probability for the occurrence of a scattering process with the exchange of certain energy and momentum between the neutron and the scatterer is controlled by the system's structural and dynamical properties, as contained in its scattering law S(**Q**,ω). In the frame of the Gaussian approximation [1] the dynamics of the material is enclosed in its generalized frequency spectrum. Under the assumption of no coupling between the molecular 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

$$\chi_i(Q,t) = \exp\{-\gamma_i(t)Q^2\}, \tag{4}$$

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

$$\gamma(t) = \int_0^\infty \frac{Z(\omega)}{\omega}\left[\{n(\omega)+1\}e^{i\omega t} + n(\omega)e^{-i\omega t}\right] \tag{5}$$

and $n(\omega)$ is the occupation number. We constructed frequency spectra for the different materials considered in this work, by combining experimental [2,3] and synthetic [4] contributions, together with a fitting parameter related to the weight of the translational and librational modes, and a normalization condition for the weights of all the dynamical modes.

Scattering law data files were generated by the LEAPR module of the NJOY code [5] using the adopted frequency spectra, whereas the cross section data libraries were produced by the modules THERMR and ACER, in the appropriate format for MCNP calculations.

II. APPLICATIONS

1. Solid Methane in Phase II

We developed a simple model to describe the interaction of slow neutrons with solid methane in phase II, including the main dynamical features of the system and the effect of spin correlations. This effect occurs in molecules containing identical nuclei whenever spin and rotational states are coupled, thus imposing symmetry requirements on the molecular wave function. The scattering lengths a_{lv} are spin dependent quantities. After performing the spin averages appropriate to an unpolarized neutron beam, the scattering law can be written as the sum of inter and intra molecular contributions and its Fourier transform as

$$\chi^{\text{inter}}(\mathbf{Q},t) = \left\langle \sum_{v,v'} b_c^v b_c^{v'} f'_{vv'} \right\rangle; \quad \chi^{\text{intra}}(\mathbf{Q},t) = \left\langle \sum_{v,v'} \left\{ b_c^v b_c^{v'} + b_i^v b_i^{v'} \frac{(\mathbf{S}_v \cdot \mathbf{S}_{v'})}{[S_v(S_v+1)\,S_{v'}(S_{v'}+1)]^{1/2}} \right\} f_{vv'} \right\rangle, \tag{6}$$

where we have called

$$f'_{vv'} = \exp\{-i\mathbf{Q}\cdot\mathbf{R}_{lv}(0)\}\exp\{i\mathbf{Q}\cdot\mathbf{R}_{l'v'}(t)\}$$
$$f_{vv'} = \exp\{-i\mathbf{Q}\cdot\mathbf{R}_{lv}(0)\}\exp\{i\mathbf{Q}\cdot\mathbf{R}_{lv'}(t)\}$$

and clearly f' refers to position operators for scattering centres in different molecules ($l \neq l'$).

Detailed quantum calculations were performed to describe the low-level energy states in CH_4 II (Ref. 6) and applied to the evaluation of neutron cross sections for that system [7,8]. Those formulations were then used to analyze neutron scattering and transmission experiments [9,10,11]. In our study we oriented our effort to the development of a simple model which should preserve the main dynamical features (as well as a proper description of spin correlation effects) of the system and therefore be able to make reliable predictions in terms of neutron fluxes emerging from such cold moderator material.

From the measurements performed by Harker and Brugger [12] at different temperatures in phase I and phase II of solid methane, we derived frequency spectra for free, hindered, and average (1:3) molecules in phase II, and they are displayed in Fig. 1 as a function of the excitation energy ε. It is observed that the spectrum corresponding to free molecules is richer at low energies, on the contrary, the spectrum for hindered molecules shows a prominent peak around 0.0065 eV where the librational states coalesce. The third curve, the phase II spectrum, is the 1:3 weighted average of the other two.

A comprehensive set of measurements on this system was performed by Grieger *et al.* [9]. at temperatures ranging from 19.5 K down to 0.3 K, and we compare in Fig. 2 our calculations with their experimental total cross sections at 0.3 K and 10 K, over the relevant energy range. The agreement is very good except at the lowest energies, where the scatter of data points does not allow to confirm such assessment.

The predictions of our model for methane in phase II are in good agreement with a full quantum mechanical calculation over the limited range where the latter was formulated, and with available experimental information over the complete thermal energy range.

2. Solid Deuterium

Very recently, a new scattering kernel to describe the interaction of slow neutrons with solid Deuterium was

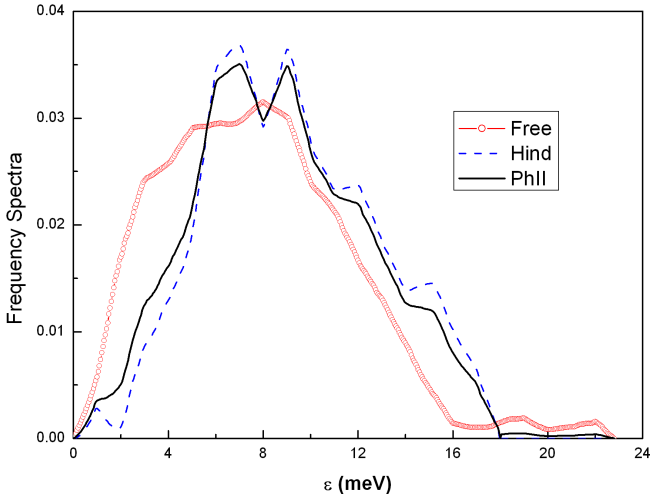

Fig. 1. (Color online) Frequency spectra derived from the experimental data of Harker and Brugger [12].

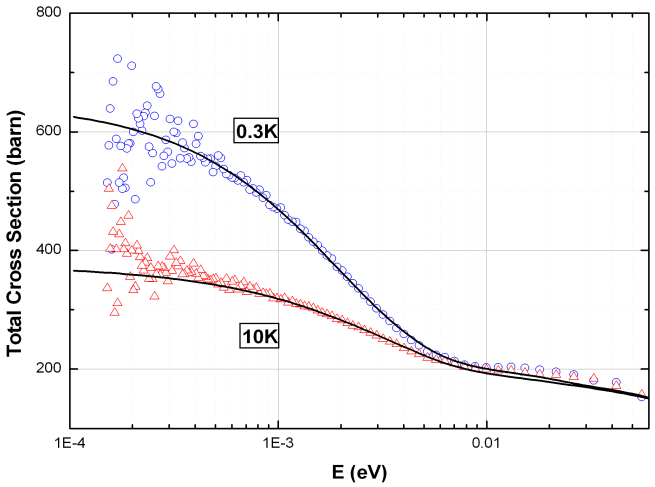

Fig. 2. (Color online) Comparison of the calculated total cross sections of methane at 0.3 K and 10 K, with measured points [9].

developed [13]. The main characteristics of that system are contained in the formalism, including the lattice's density of states, the Young-Koppel quantum treatment of the rotations, and the internal molecular vibrations. The elastic processes involving coherent and incoherent contributions are fully described, as well as the spin-correlation effects.

The deuterium molecule is formed by two bosons, and therefore its total wave-function must be symmetric under interchange of two identical nuclei [1]. Consequently, if the total nuclear spin **S** is even the spatial nuclear wave function must be symmetric, and antisymmetric if **S** is odd, which leads, respectively, to the existence of the *ortho* states, with S = 0, 2 coupled to J = 0, 2, 4, ... , and *para* states with S = 1 coupled to J = 1, 3, 5, ..., where J denotes the molecule's total angular momentum.

The neutron scattering laws S(**Q**,ω), energy-transfer kernels σ(E,E′), and cross sections σ(E) for inelastic

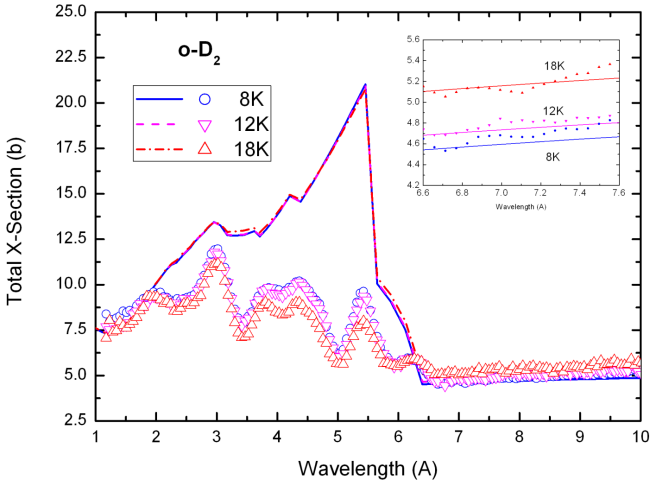

Fig. 3. (Color online) Calculated total cross section of solid o-D_2, compared with experimental data from [17,18]. The inset shows that comparison beyond the first Bragg peak.

scattering in solid ortho- and para-deuterium were calculated using the code NJOY [5], which is based on a phonon expansion for the lattice motion, and the Young-Koppel [14] formalism for the quantum rotational description. As part of the code's input data, the density of states (DOS) for solid deuterium derived by Schmidt *et al.* [15] was used. The elastic incoherent component produced by NJOY was modified in order to include spin correlation effects. The lattice structure factor was calculated using our code CRIPO [16] and then affected by the Debye-Waller factor $\chi^{\rm vib}$(**Q**,0) and the molecular structure factor to obtain the elastic coherent component.

The total cross section of ortho-deuterium is shown in Fig. 3 as calculated with the present model, compared with experimental data from Refs. 17 and 18 for a few temperatures over the thermal neutron wavelength range. The large elastic coherent contribution due to the *hcp* structure factor dominates the cross section at those energies, and the disagreement with the measured points is a clear indication of the lack of perfect polycrystallinity in the samples. However, and in spite of the significant difficulties to achieve a high precision normalization for the measured curves in those experiments, a very satisfactory agreement between both sets is observed beyond the first Bragg peak, a region dominated by total inelastic and incoherent (including spin correlation effects) elastic components of the total cross section (see inset in Fig. 3).

The comparison between the present model and existing cross section data for solid deuterium is further displayed through the curves presented in Fig. 4, where calculated values for solid ortho- and normal-deuterium are compared with the measurements from Refs. 18 and 19 at around 18 K.

At this temperature the inelastic cross section for ortho and para deuterium are very similar, and therefore the differences observed below the first Bragg peak for

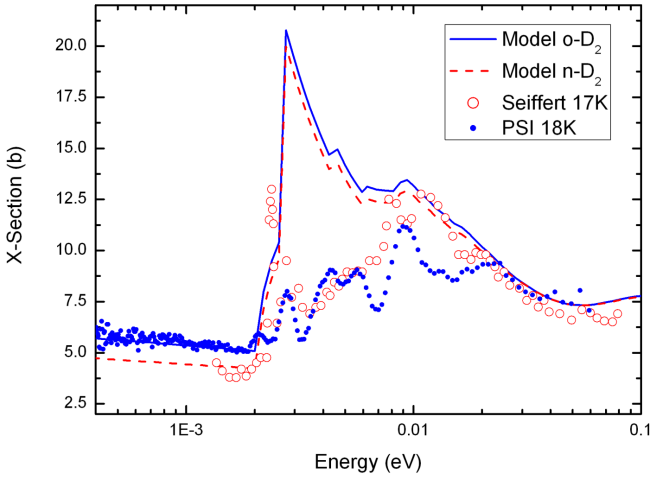

Fig. 4. (Color online) Calculated total cross sections of ortho- and normal-deuterium at 18 K, compared with the experimental data from Refs. 18 and 19.

the cross sections of ortho- and normal-deuterium are purely due to spin correlation effects on the elastic incoherent contribution.

III. CONCLUSIONS

We developed a new scattering kernel for solid methane in phase II, including the main dynamical features of the system and the effect of spin correlations. Good agreement with a quantum mechanical calculation over the limited range where the latter was formulated, and with available experimental information over the complete thermal energy range [20].

A new scattering kernel to describe the interaction of slow neutrons with solid Deuterium has been developed. Scattering functions and cross sections for both *ortho-* and *para-* Deuterium have been evaluated for temperatures ranging from the freezing point (18.7 K) down to 5 K. The new model has been compared with the best available experimental data, showing a highly satisfactory agreement.

ACKNOWLEDGMENTS

This work was partially supported by grant PICT 52963 from ANPCyT (Argentina) and IAEA RC N°14161 funds.

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