SCATTERING OF SLOW NEUTRONS BY WATER MOLECULES

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The double differential cross sections for the scattering of neutrons by molecules were measured by a method based on determining the final scattered-neutron energy with a resonant detector (using neutron resonance in gold at 4.91 eV). The results of the scattering measurements in water confirm the model in which a chemically bound nucleus that scatters electron-volt neutrons behaves like a quasifree nucleus.

1. INTRODUCTION

MOST experimental investigations devoted to the interaction of neutrons with nuclei contained in molecules pertain to the region of low neutron energies, smaller than 1 eV. This is due primarily to the relative simplicity of obtaining and detecting strong beams of such neutrons. Yet the energy region above 1 eV is no less interesting for the investigation of molecular media and makes it possible to analyze various subtle effects, the interpretation of which is practically impossible using slow-neutron spectra.

On the basis of a number of theoretical investigations summarized in the review[1], the general notions concerning the scattering of neutrons of electron-volt energy by nuclei contained in molecules can be formulated as follows. At sufficiently large energy of the incident neutron, and consequently at sufficiently large (as a rule) transfer of energy ϵ (on the order of several eV) from the neutron to the scattering nucleus, the effective energy transfer time $\sim\hbar/\epsilon$ turns out to be much shorter than the period of the molecular oscillations, i.e., $\hbar/\epsilon \ll 1/\omega$, where ω is the characteristic frequency of the molecular oscillations of the scattering nucleus. In other words, during the time of energy transfer the position of the nucleus (and consequently the quantum state of the molecule) does not have time to change significantly. Therefore in the case when $\hbar/\epsilon \ll 1/\omega$ the chemically bound nucleus behaves like a quasifree nucleus, and the molecular bond becomes manifest only via the momentum scatter possessed by the nucleus as a result of the presence of the chemical bonds.

It is obvious that under the foregoing conditions the calculations of the second-differential neutron scattering cross section $d^2\sigma/d\epsilon do$ can be much simpler than in the case of slow neutrons, since they reduce only to a correct allowance for the momentum distributions of the scattering "quasifree" nuclei.

In their turn, such momentum distributions, which generally speaking can take into account all kinds of subtle peculiarities of the molecular structure (for example, different anharmonicity effects, effects of coupling between the rotation and vibration of the molecule, and also effects of interaction between the molecules in condensed media) can be interpreted relatively easily within the framework of the indicated picture on the basis of the measured neutron spectra. We note that these considerations served as the basis for an idea advanced by Gol'danskiĭ back in 1956[2] of investigating the properties of molecules by means of the spectra of the scattered "fast" neutrons.

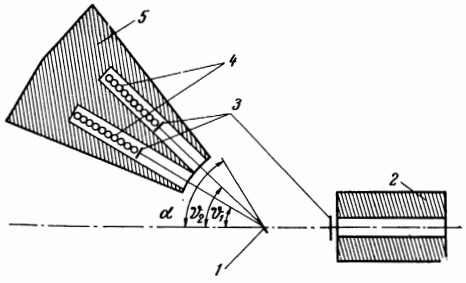

FIG. 1. Diagram of experimental setup: 1—scatterer, 2—collimator, 3—filters of gold foil, 4—neutron detectors, 5—detector shield.

The experiments reported below were aimed at developing a procedure for an investigation of the scattering spectra of electron-volt neutrons, proposed by Shapiro[3] and based on a determination of the final energy of the neutron with the aid of a resonant detector, and also a verification of the aforementioned theoretical premises, which are summarized in the review[1]. The object of the investigation was water, in view of the relative ease with which experiments can be performed with it and also because all the H_2^2O-molecule parameters needed for the calculation can be regarded as known.

2. EXPERIMENTAL PROCEDURE

The gist of the resonant-detector method can be made clear by describing the experimental setup, which is shown in Fig. 1.[1)] A beam of neutrons from the IBR pulsed reactor[4] traversed a distance of 250 m through collimator 2 and was incident on sample 1, which was a flat aluminum container of area 7×20 cm, installed at an angle α to the direction of the beam and filled with distilled water. The neutrons scattered by the sample at angles ϑ_1 and ϑ_2 are registered by two detectors 4, each of which is made up of ten proportional counters of type SNMO-5, filled with $B^{10}F_3$. To reduce the background of extraneous neutrons, the deters were

[1)] Machekhina, Ogzheval'skiĭ, and Shapiro[3] used a similar setup and performed measurements on graphite and LiF.

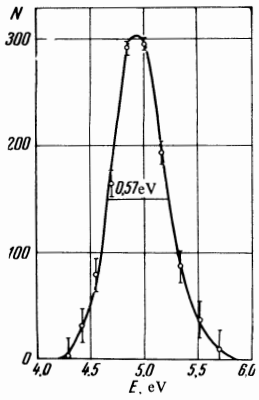

FIG. 2. Spectrum of neutron absorption in the filter, obtained with the filter placed in the primary neutron beam.

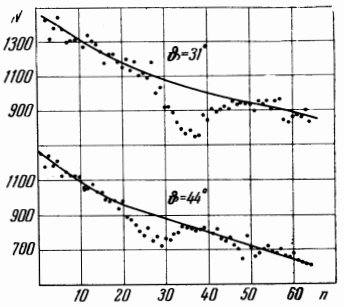

FIG. 3. Time spectra of scattered neutrons, obtained with a sample 1 mm thick. N—number of registered counts, n—number of analyzer channel.

placed in a shield 5 made of paraffin with B_4C and of cadmium. The pulses of both detectors were sorted with a time analyzer with respect to the time of flight of the neutrons from the reactor to the detectors. Inasmuch as the distance from the sample to the detectors (80 cm) was much shorter than the total transit base, the obtained time distributions of the registered neutrons characterize the spectrum of the energies possessed by these neutrons prior to scattering by the sample.

If we take a filter with a narrow neutron absorption band (we used a gold foil 0.04 mm thick, which has a strong neutron resonance at 4.91 eV) and place it in the neutron beam ahead of the sample, then a deep trough will appear in the spectra of the registered neutrons from both detectors, at an energy equal to the energy of this band. Figure 2 shows the absorption spectrum of the gold foil, converted in terms of the energy scale. The dip in the corresponding transmission spectrum practially reached zero, and its width, 0.57 eV, was determined by the properties of the filter and by the energy resolution of the entire setup.

On the other hand, if such filters are placed directly ahead of the detectors, then the positions of the dips in the spectra will correspond to the energies of those neutrons which fall into the absorption band of the filter after being scattered from the sample at a specified angle, and their form is now determined also by the properties of the sample, i.e., by the magnitude of the energy transfer between the neutrons and the nuclei.

Thus, by performing the measurements with filters ahead of the detectors, it is possible to obtain information on the scattering of neutrons by different substances. It is important here that the width of the filter absorption line be considerably smaller than the average energy transfer during the scattering act.

3. DATA REDUCTION AND RESULTS

We obtained eight time spectra at scattering angles $\vartheta_1 = 31^\circ$ and $\vartheta_2 = 44^\circ$ for three samples of water 1, 2, and 4 mm thick. Figure 3 shows by way of an example the experimental distributions for the 1 mm sample. Each of the two spectra shows one broad dip of depth on the order of 20% corresponding to the scattering of the neutrons by the hydrogen atoms contained in the water molecules. The dips due to the scattering by oxygen and by the aluminum of the container were not observed, within the limits of errors. All the remaining curves are quite similar to those shown in the figure, and the positions of the maxima on all the curves are in satisfactory agreement with the quantities obtained from the formula for the scattering of a neutron by a proton at rest:

$$E_0 = E' / \cos^2 \vartheta, \qquad (1)$$

where the final energy of the scattered neutron is $E' = 4.91$ eV as follows from the theory[1]).

Further reduction of the data consisted of the following. A smooth curve was drawn freehand through the points on both sides of the dip, as indicated in Fig. 3, and the ordinates of the points in the region of the dip were subtracted from the corresponding ordinates of the curve. The spectra obtained in this manner for the absorption of neutrons by the gold filter, normalized beforehand for the non-uniform spectrum of the neutrons registered by the detector (the same smooth curve), were plotted on an energy scale (see example on Fig. 4). Smooth curves were again drawn through

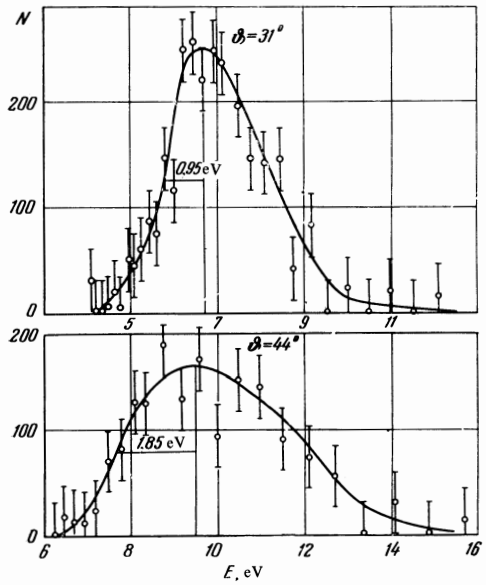

FIG. 4. Energy spectra of the neutrons absorbed by the filter after being scattered by the sample. The spectra were obtained by reducing the apparatus spectra shown in Fig. 3.

the obtained points; they are marked with the half-width ΔE_{exp} on the left slope at half the height of the peak.

The values of ΔE_{exp} were corrected for the apparatus line width ΔE_{app} and the width ΔE_ϑ connected with the finite angular resolution of the setup. The corrected curve was calculated from the formula

$$\Delta E_{corr} = \sqrt{\Delta E^2_{exp} - \Delta E^2_{app} - \Delta E^2_\vartheta}.$$

The numerical values of ΔE_{app} were obtained from the peak width 0.57 eV of Fig. 2, corresponding to a neutron energy 4.9 eV, which was recalculated for the energies 6.7 and 9.5 eV by using the known function of the energy resolution of the setup, and ΔE_ϑ was calculated by differentiating (1) with respect to ϑ and using the geometry of the sample and of the detectors.

All the numerical results are summarized in the table. As seen from the table, all the values of ΔE_{corr} for a given angle ϑ are in reasonable agreement with one another, and the magnitudes of the corrections, i.e.,

Sample thickness, nm	$\vartheta°$	$\alpha°$	$\Delta E_{exp}/\partial\varepsilon$	$\Delta E_{app}/\partial\varepsilon$	$\Delta E_\vartheta/\partial\varepsilon$	$\Delta E_{corr}/\partial\varepsilon$	Measurement time, hr
1	31	60	1.9	0.58	0.28	1.78	32.0
2	31	60	2.2	0.58	0.28	2.10	39.5
4	31	60	1.8	0.58	0.28	1.68	20.6
4	31	135	2.2	0.58	0.41	2.08	15.5
1	44	60	3.7	0.60	0.49	3.62	32.0
2	44	60	3.4	0.60	0.49	3.31	39.5
4	44	60	3.8	0.60	0.49	3.73	34.8
4	44	135	3.2	0.60	0.97	3.00	15.5

the differences $\Delta E_{exp} - \Delta E_{corr}$ do not exceed the scatter between the individual values of ΔE_{corr}.

One might expect the width of the peak to increase with increasing thickness of the sample, owing to the contribution made by multiple scattering of the neutrons in the sample. However, neither ΔE_{exp} nor the total widths of the peaks with allowance for both slopes, which are not presented here, reveal a noticeable correlation with the sample thickness. Under these conditions we are justified in assuming that the influence of the multiple scattering does not exceed the limits of the errors, in averaging the obtained results, and in representing the error as the mean deviation from the mean:

$$\Delta E = \overline{\Delta E_{corr}} = \begin{cases} (1.9 \pm 0.2)\ \text{eV for } \vartheta = 31°, \\ (3.4 \pm 0.3)\ \text{eV for } \vartheta = 44° \end{cases}$$

4. THEORETICAL FORMULAS AND THEIR COMPARISON WITH THE EXPERIMENTAL RESULTS

The intramolecular oscillations for water at room temperature (which was used in the experiments) can be regarded as fully harmonic. In this case the cross section for the scattering of a neutron by a hydrogen atom in a solid-angle element do with loss of energy ϵ is given by a formula (see [1], p. 73) corresponding to the model of the quasifree scattering and valid with relative accuracy on the order of $\sqrt{\omega/\epsilon}$ [2]:

[2] We shall henceforth use a system of units in which Planck's constant $\hbar$ and the Boltzmann constant are assumed equal to unity.

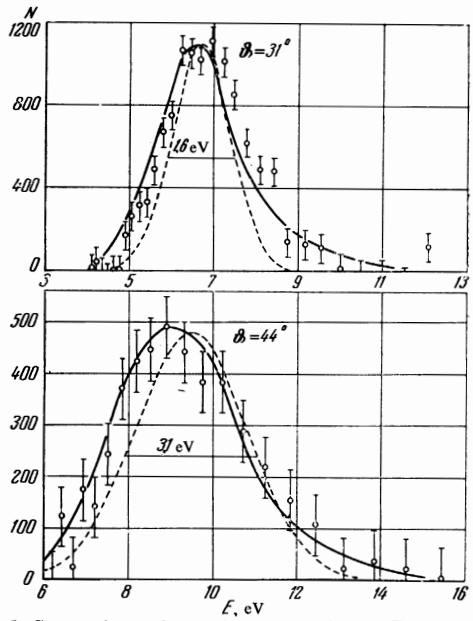

FIG. 5. Comparison of experiment and theory. Experimental points—sum of all the measurements at a given scattering angle. The curves show theoretical calculations: solid curves—formula (2), dashed curves—formula (3).

$$\frac{d^2\sigma}{d\varepsilon\, do} = 4|F_n|^2 \frac{k'}{k} \left\langle \frac{1}{\sqrt{2\pi R\omega(\mathbf{n})}} \exp\left[-\frac{(\varepsilon - R)^2}{2R\omega(\mathbf{n})}\right] \right\rangle, \qquad (2)$$

where F_n is the amplitude for neutron scattering by a free proton, k and k' are the neutron momenta before and after scattering, $R = (k' - k)^2/2m$ is the recoil energy of the hydrogen atom, m is the mass of the proton, and $\omega(\mathbf{n})$ is the effective energy of the scattering atom, including both effects of molecular vibration and effects of translational and rotational motions:

$$\omega(\mathbf{n}) = \sum_i (\mathbf{e}^i\mathbf{n})^2 \omega_i \operatorname{cth}\frac{\omega_i}{2T} + 2T\mathbf{n}\hat{m}^{-1}\mathbf{n}.$$

Here $\mathbf{e}^i$—amplitude vectors for the scattering atom, expressed in units of $1/m$ and connected with the force constants of the molecule, T--temperature, and $\hat{m}$—mass tensor for the scattering nucleus, whose components are expressed in units of m. The vector $\mathbf{n}$, which assumes arbitrary directions in space, characterizes the orientation of the molecule, and the symbol $\langle\ \rangle$ denotes averaging over the molecule orientations, i.e., integration with respect to $d\mathbf{n}/4\pi$. We note that in formula (2) the translational and rotational motions of the molecule are assumed free.

The results of a comparison of theory with experiment is shown in Fig. 5, where the experimental points constitute the sum of all the spectra pertaining to a given scattering angle. The solid curves in the figure were obtained on the basis of formula (2) by numerical integration, and the dashed curves correspond to the simplified theoretical formula

$$\frac{d^2\sigma}{d\varepsilon\, do} = 4|F_n|^2 \frac{1}{\sqrt{\pi}\Delta} \exp\left[-\frac{(E - E_0)^2}{\Delta^2}\right], \qquad (3)$$

where

$$\Delta^2 = \frac{2\sin^2\vartheta}{\cos^4\vartheta} E'\bar{\omega}, \qquad (4)$$

obtained from (2) by replacing the rigorous averaging

by introducing the mean value

$$\bar{\omega} = \langle \omega(n) \rangle \cong (0.17 + 1{,}1\,T) \text{ eV} \cong 0.2 \text{ eV (for } T = 300^\circ \text{ K)}$$

and using the assumption that $E - E_0 \lesssim \Delta$. The constants required for the calculation were obtained for the H_2O molecule from [5].

If we obtain the parameter Δ of the Gaussian distribution (3) from the experimental values of the width ΔE, then we can use (4) to determine the experimental values of the average frequency ω. The corresponding calculations yield the result

$$\bar{\omega} = (0.26 \pm 0.06) \text{ eV for } \vartheta = 31^\circ,$$
$$\bar{\omega} = (0.24 \pm 0.04) \text{ eV for } \vartheta = 44^\circ.$$

Thus, comparing theory with experiment, we can conclude that the position of the maxima of the observed distributions, and also their widths, are in satisfactory agreement with the theoretical ones. The ratio of the heights of the maxima of the distributions, corresponding to two different scattering angles 31 and 44° and to the same number of incident neutrons, equal to 2.0, also agrees approximately with the theoretical value of this ratio, equal to $\Delta(44^\circ)/\Delta(31^\circ) = 1.9$. The theory accounts also for the experimentally observed asymmetry in the energy distributions of the scattered neutrons. A certain quantitative disagreement, noticeable at a scattering angle 31°, is possibly due to some unaccounted for experimental errors, and also possibly to a manifestation of the effects of the liquid state, unaccounted for in the theory, and the errors connected with the use of the approximation of the quasifree particle, which can be of the order of 10%. On the whole, however, it seems possible to conclude that the experiment confirms all the qualitative conclusions of the theory [1] and is also in satisfactory quantitative agreement with it (taking into account the appreciable experimental errors). It also seems to us that further improvement of the described experimental procedure (an increase in the measurement accuracy, development of a procedure for the measurement of the absolute value of the scattering cross section) will make it possible to use the procedure for a quantitative determination of certain molecular constants (for example, the force constants), and also for an investigation of different effects of anharmonicity of the intramolecular vibrations).

We note also that the confirmation of the model of the "quasifree" scattering by bound nuclei makes it possible, in our opinion, to regard as correct also the general picture of the inelastic processes which occur in the molecule under the influence of electron-volt neutrons [6]. According to this picture, such inelastic processes break up into two independent stages: during the first stage the neutron transfers to the bound nucleus a certain energy, which subsequently (second stage) is lost to different inelastic processes occurring in the molecule. Inasmuch as these subsequent processes (e.g., dissociation of the molecule) do not affect at all the spectra of the scattered neutrons, it must be assumed that no information concerning these inelastic processes (e.g., concerning the binding energy of the molecule) can be obtained on the basis of these spectra.

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