ATOMIC MOTIONS IN WATER BY SCATTERING OF COLD NEUTRONS

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The scattering of very slow, or "cold," neutrons is a useful technique for study of the motions of atoms in solids and liquids. In this method, as developed[1] at Brookhaven, a beam of approximately 0.005-ev neutrons, produced by beryllium filtration, is incident on a thin sample. Measurement of the changes in energy at scattering gives directly the desired information concerning the atomic motions in the sample. As the energy changes are usually large compared with the incident energy, they can be measured accurately. In the case of an incoherent scatterer, such as hydrogen, a further advantage of the cold neutron technique obtains; essentially all motions of the atoms can be measured because all momentum gains are possible. The cold-neutron method was applied to water in the present experiments because of the intrinsic interest of the atomic motions and because the inelastic scattering of neutrons in water has an important bearing on its function as a reactor moderator.

The water samples, which were extremely thin, ranging down to 0.2 mm, were formed by an aluminum sample holder containing milled portions of the correct thickness separated by supporting ribs. The neutrons scattered at 90° were passed through a slow chopper and the energies measured by time of flight to detectors five meters distant, as already described.[1] Careful background runs were made with empty sample holders and the spectrum shape of the incident neutrons was determined by scattering from a thin sheet of vanadium, which is essentially an elastic incoherent scatterer. The spectrum of scattered neutrons is shown on a time-of-flight scale in Fig. 1, corrected for instrumental effects. The results were somewhat unexpected in that they show a number of definite energy levels, rather than a continuum of energy changes that would be expected from a classical liquid. Because of the quantum nature of water thus revealed it appears that it is not a suitable material for simple application of the theory of scattering of neutrons by liquids developed by Vineyard,[2] on the basis of the more general theory of Van Hove.[3]

Further investigations are being made on these various features, particularly as a function of sample temperature, and only the most signifi-

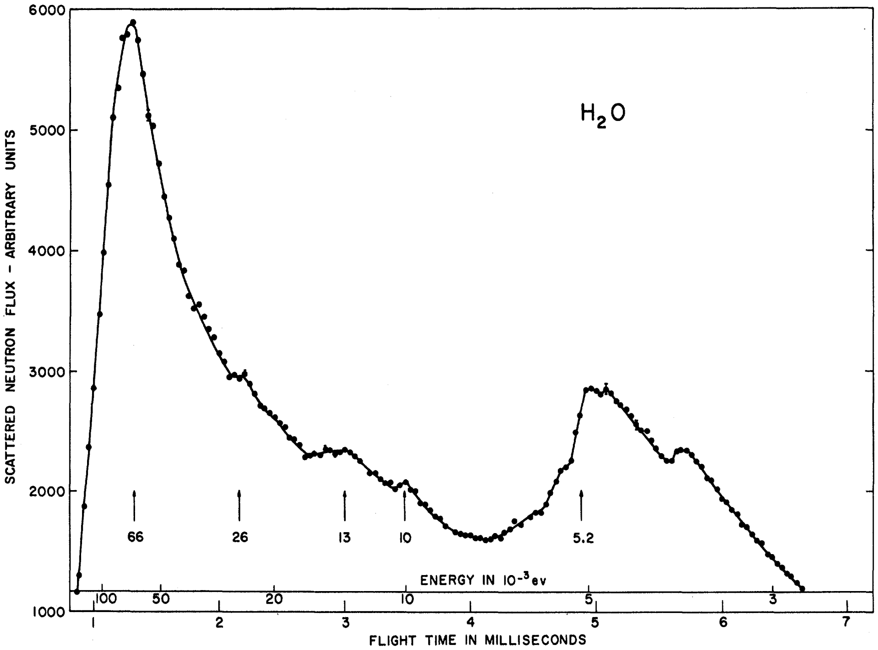

FIG. 1. Neutron flux observed at 90° from incident beam direction. Data are corrected for counter efficiency and chopper transmission. The arrow at 5.2 $\times 10^{-3}$ gives the position of the Bragg cutoff for the Be-filtered incident spectrum. The other arrows give the measured neutron energies corresponding to hindered motions of the liquid. The quantum energy of these states is the difference between the indicated energy and 5.2×10^{-3} ev, the incident energy.

cant will be considered here. The prominent energy gain of magnitude 0.061 ev has been found much earlier by infrared measurements[4] and interpreted as a hindered rotation, that is, a rotatory oscillation of one water molecule in the field of its neighbors. Infrared measurements are not possible at energies much lower than that of the hindered rotations, whereas the present technique extends about a hundredfold lower, to the region of microwave absorption. At energies below that of the hindered rotation there are a number of peaks, which are probably to be associated with various types of translatory motions of water molecules in the potential of their neighbors.[4]

A most surprising effect observed is the extremely small energy changes shown by the two peaks near the energy of the incident neutrons (Figs. 1 and 2), representing the transfer of energy from and to the liquid of the same amount, 0.7×10^{-3} ev. This energy is much less than that of the other energy changes found in water and there is no known transition in liquid water that could account for it. The observed energy actually corresponds closely to a line recently observed[5] in the microwave spectrum of water vapor, although it would be very surprising for this energy transition to appear in the liquid.

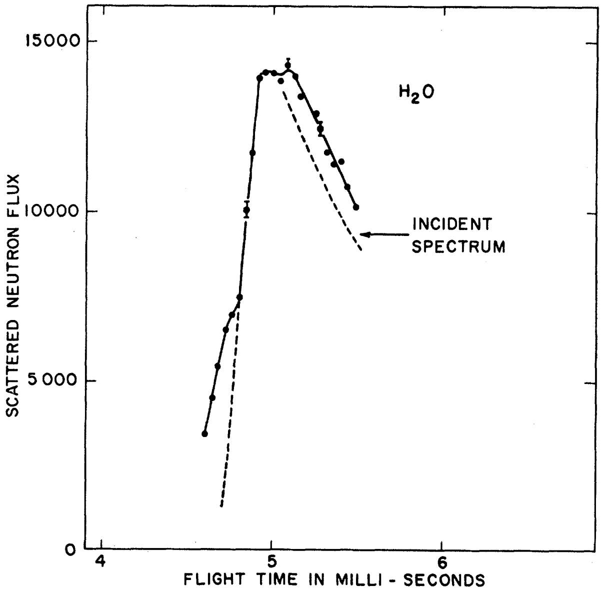

FIG. 2. The details of the scattered spectrum near the "elastic peak." The dashed curve shows the position of the incident spectrum as would be measured with a thin vanadium scatterer. The water data are seen to coincide with the incident spectrum in the region of the sharp rise, indicating little diffusive scattering. The two small peaks at energies lower and higher than the sharp edge correspond to neutron energy loss and gain of 0.7×10^{-3} ev.

The low incident energy in the present experiment should be particularly suitable for observing the small energy changes arising from diffusive motions of the atoms. Vineyard has considered the slight energy broadening of the incident spectrum and in one simple model the energy spread, $\Delta\epsilon$, is given directly by the diffusion constant

$$\Delta\epsilon \cong 2\hbar D k_0^2, \qquad (1)$$

where $\hbar k_0$ represents the momentum transferred to the scattered neutron. The surprising finding of the present work is, however, that no spreading of the incident beam is observed (Fig. 2), although for the experimental conditions one would expect from Eq. (1) a spreading of 0.9×10^{-3} ev, which would be easily observed. A careful analysis of the present results shows that a diffusion spreading of 0.3×10^{-3} ev could have been detected, and hence the observed spread is certainly less than one-third of the calculated value. In similar measurements with water at 45°C, the upper limit is even lower, about one-sixth.

The present results indicate that the so-called "jump time" for diffusion, instead of being equal to the reciprocal of the Debye frequency, which is about 10^{-12} second in water, is at least six times longer. This finding indicates that the water molecule remains in its initial position for a number of vibrations before jumping to a new position. The magnitude of the jump would then have to extend over several molecules in order to obtain agreement with the known diffusion constant.

In order to clarify further the results just discussed, additional measurements are being made at higher temperatures and for vapor and ice as well as for liquid D_2O. As far as the application to moderation of neutrons in reactors is concerned, the small energy changes here presented are of little significance relative to the extremely prominent interchange of energy with the hindered rotation. The results also show that the motions in water are not represented well by a gas consisting of points of mass 18, an approximation often used in reactor calculations.

Brockhouse[6] has made similar measurements on water, with both low- and high-energy incident neutrons. His results with high-energy incident neutrons, as expected, do not reveal the details shown by the present measurements.

Both his results and ours with cold neutrons reveal the hindered rotation but there are marked differences concerning the other features. His measurements do not show the sharp peaks that we have observed, particularly those corresponding to the small energy changes. Instead of the latter, he observes a general broadening of the elastic peak, which is somewhat smaller than that expected from diffusive motions. This result is in sharp contrast to ours, which shows no evidence of broadening related to diffusive motions.

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[1] Eisenhauer, Pelah, Hughes, and Palevsky, Phys. Rev. 109, 1046 (1958).

[2] G. H. Vineyard, Phys. Rev. 110, 999 (1958).

[3] L. Van Hove, Phys. Rev. 95, 249 (1954).

[4] M. Magat, Ann. Physik 6, 108 (1936).

[5] C. H. Townes (private communication).

[6] B. N. Brockhouse, Phys. Rev. Letters 2, 287 (1959).

SCATTERING OF 50- to 140-Mev PHOTONS BY PROTONS AND DEUTERONS[*]

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The Compton scattering of photons by protons has been studied in several experiments[1-4] below π-meson photothreshold, and in two experiments[5,6] above. We report here further work below meson threshold using both protons and deuterons as targets.

The techniques of identifying and measuring the energy of the bremsstrahlung x-rays from the M.I.T. synchrotron after they are scattered by thin targets are essentially as reported in Pugh et al.[1] The many changes which have been made in the synchrotron itself, giving it greater stability and intensity, have been described in recent M.I.T. progress reports.[7] The many changes in construction and performance of the photon telescope, energy-measuring liquid scintillator, and especially electronics, are being reported elsewhere.[8]

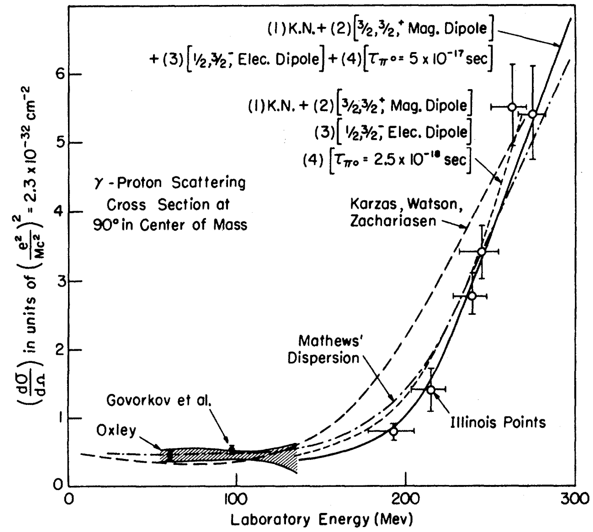

FIG. 1. Differential cross sections for scattering of 50-280 Mev photons by hydrogen at 90° c.m.

In Fig. 1 experimental and theoretical cross sections for scattering of photons by hydrogen are plotted as a function of lab energy at a c.m. angle of 90°. The cross sections are given per unit solid angle in the c.m. system for direct comparison with the Illinois points[5] and those of other groups.[2-4] Our results are given as a shaded area within which the cross section must lie in order to give, after appropriate folding, a fit to a smooth curve through our observed data. A discussion of this technique, which of necessity assumes a fairly smooth cross section, is given by Pugh et al.[1] The statistical standard deviation is represented by the height of the shaded area, but statistical errors are thereby exaggerated somewhat, since the data were taken in channels of approximately 10 Mev, but the cross section at most energies is averaged by the counter response over an energy band of about 20 Mev. On the other hand, a possible 10% error in absolute normalization is not displayed. We see in Fig. 1 that these new data, and the results of the other experiments at this angle, are all in good agreement below meson threshold and fit in smoothly with the Illinois data.