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I. INTRODUCTION

The experimental and theoretical work carried out during this contract period continues the fundamental studies of the interaction mechanisms by which neutrons exchange energy with hydrogen atoms. Above about 1 ev these interactions may be regarded as collisions of neutrons with free hydrogen atoms, but at lower energies they must be regarded in terms of excitation of molecular, liquid or crystal, thermal motions determined by binding forces. The problem of neutron thermalization is therefore concerned with the distribution of energy levels and the probability of excitation by neutron collision. Particular emphasis has been given to studies of scattering in very low temperature materials, where binding effects are most apparent, and of scattering of very low energy neutrons. These two interests are in accord in that the best method of obtaining high intensity of the desired slow neutrons is from moderators at low temperature, and the studies of neutron energy exchange in cold materials lead to information required to design an optimum cold neutron source.

The elastic and inelastic scattering of low energy neutrons has been applied to a wide range of studies of solid-state phenomena. In all cases these studies have been carried out with neutrons derived from a thermal spectrum with equilibrium temperatures somewhere near room temperature of 295°K , corresponding to a $kT = 0.025$ ev. In a number of cases it has been desirable to use neutrons with longer wavelength and still lower energy and to study their interaction with materials in which the thermal vibration energy, kT , is itself correspondingly low. Indeed, many such experiments have been performed and are in progress using crystal filter, crystal spectrometer, and time-of-flight methods, but always with the inherent limitation and handicap of the low flux of neutrons available in the low-energy range of the thermal spectrum. It would be desirable to attain a

spectrum with more abundant very slow neutrons. A method, obvious in principle but quite difficult in practice, is to maintain a moderator at a very low temperature inside a reactor. Moderators at liquid nitrogen temperature have been used with cyclotrons⁽¹⁾ to enhance the low-energy neutron flux, and a liquid nitrogen temperature cryostat installed in the Brookhaven reactor⁽²⁾ for radiation-effects studies gave slightly increased low-energy flux. More recently, liquid hydrogen sources in the Harwell⁽³⁾ and Saclay⁽⁴⁾ reactors have yielded slow neutron fluxes 10 to 25 times higher than available from an equivalent warm source but still far short of that attainable if moderation were complete.

To carry out the experiments in the areas of interest described above, the experiments to be described were undertaken with neutrons from a pulsed electron linear accelerator. Energy distributions in various moderating materials at temperatures from 295°K down to 4°K have been studied by time-of-flight analysis. Various arrangements for deriving inelastic scattering data have been devised and used for additional experiments. Several cross-section measurements on a number of pertinent materials have also been completed as part of the study. The arrangements for the various types of experimental observations will be described in Section II.

At this point, it may be useful to point out the several advantages of the accelerator source in observations on cold moderating materials. Whereas the installation of devices in a reactor which can maintain a moderately large volume at low temperature is a difficult engineering task, the region immediately adjacent to the accelerator target is easily accessible. This accessibility facilitates the installation, easy removal, and modification of various cryostats with specimens of a variety of dimensions and shapes. Such flexibility is necessary in the present exploratory stage of this experimental program, one object of which is to determine an optimum configuration of the cold source. In a reactor the gamma-ray heating of the cryostat and its coolant consumes quantities of liquid coolant

many times the order of radiation losses in a normal cryostat. Because of the pulsed operation with an accelerator, the consumption is at least a factor of 100 less at the accelerator, eliminating the necessity for frequent replenishment of the coolants. The 25-Mev electron linear accelerator at General Atomic is utilized to give a copious supply of thermal neutrons for the measurements to be described. For the results presented in this paper, the basic instrumentation was that for pulsed time-of-flight measurements. For the various studies reported, a variety of flight paths was used varying the distances before and after the scattering sample according to the type of measurement being made.

Prior to this contract year, only monthly letter progress reports were required. Since this is the first annual report under this contract, which has been in progress for several years, the material to be presented will not in all cases be confined only to that obtained or developed during the last twelve months. In a number of cases, a better basis for understanding the results will be obtained by starting with measurements performed in earlier phases of the current program. In some cases, techniques now considered standard will be described in sufficient detail to give the reader appropriate background. An effort has been made to present a logical and unified development of the material, with no particular attention given to indicating earlier measurements in comparison with more recent data.

II. EXPERIMENTAL TECHNIQUES

A number of techniques and pieces of apparatus used for the performance of experiments will be described in this section. When it is preferable to describe a specific technique with reference to the specific measurements being made, such a description will be deferred to the appropriate section.

The discussion will start with a description of the facility. The accelerator used is a high-current, traveling-wave, S-band (2800 Mc), two-segment machine, which presently gives square wave pulses of about 20 Mev of energy and about 60 ma of current. For a duration of 5 μ sec per pulse and a repetition rate of 120/sec the power corresponds to about 1.5 kw average power. Until the end of 1960, the beam energy was usually in excess of 25 Mev and the beam current occasionally reached 100 ma. The lower values given above were more usual after January, 1961. In July, 1961, the linear accelerator was shut down for conversion from the two-stage S-band to a three-stage L-band operation. After the conversion, electron beam energies as high as 45 Mev are expected. The yield in neutrons should be increased by about a factor of 10.

Figure 1 shows the beam ports in the experimental area appropriate for present experimentation on differential neutron thermalization. Area A was a temporary position that is no longer used in these studies. Area B is the current location for both elastic and inelastic neutron-scattering measurements. Position C gave a rather long flight path (7 m) which was used for precision cross-section measurements, but it is no longer available since additional linear accelerator apparatus has been installed in this general area. By magnetic deflection, the electron pulse can be delivered to any of the desired and available experimental ports.

Fig. 1--Floor plan of experimental area for high-current electron linear accelerator; Positions A, B, and C show three different ports used for different differential neutron thermalization experiments; Position C gives a 7-meter flight path useful for higher-energy cross-section measurements

NEUTRON PRODUCTION

The electron beam emerges from a port through a thin metal foil. Deceleration of the electrons in traversing the first one or two centimeters of a 5-cm-thick heavy-nuclei target yields high-energy bremsstrahlung, primarily in the forward direction. The bremsstrahlung energy is high enough to be in the range of the giant resonance for (γ, n) reactions with heavy target nuclei. An approximate fission spectrum of neutrons is emitted isotropically, the yield being about 0.01 to 0.02 neutrons/electron, or about 10^{16} neutrons/sec, giving 5×10^{10} for the 5- μ sec pulse or an average of 5×10^{12} /sec with the 120-pulse/sec repetition rate. The neutron source is confined to a volume of a few cubic centimeters of the target.

For thermal-neutron research it is necessary that a moderator intercept and thermalize as many as possible of the primary fast neutrons. To meet this requirement alone, it would be desirable that the electron target be completely surrounded by moderator and reflector of the order of 20 to 50 cm thick. A second consideration, however, is the pulse length, since energy analysis of the emergent slow neutrons must be done by a time-of-flight method. It is necessary for attainment of satisfactory resolution that the pulse length be short compared to the flight time for a few meters of flight path. The stringency of this condition varies with the experiment but generally requires that the duration of the thermalized pulse be of the order of 10 to 50 μ sec, similar to that which would be transmitted by the rotor of a slow chopper. The slowing-down time to reach thermal-energy levels is of the order of 10 to 15 μ sec. If a large moderator were used, allowing further diffusion of neutrons, the die-away time might be of the order of 200 μ sec or more. This means that the moderator configuration must be carefully arranged. Figure 2 shows a typical neutron source arrangement. An outer layer of lead serves both as a fast-neutron reflector and a gamma-ray shield. A hydrogenous inner material--usually polyethylene--provides for the first few collisions to approach thermal equilibrium. The moderator viewed by the beam port is surrounded with cadmium so that

Fig. 2--Arrangement for source of cold-neutron spectrum; bremsstrahlung from 28-Mev electrons yields fast neutrons which are then moderated in cold CH₂; the large lead shield reduces significantly the gamma-radiation problems in the neutron detectors

the incident neutrons must be above the cadmium absorption cutoff energy, and the final few collisions to reach thermal energy must be in a relatively small region. This arrangement has the effect of allowing the desired neutrons which have reached thermal energy within a few microseconds to emerge, but of eliminating those thermal neutrons which would prolong the pulse by emerging after diffusions through a large moderator.

A special effort was made to optimize the source arrangement, which consists of an electron target, a large block of lead gamma-ray shield, and a low-temperature moderator placed appropriately near the electron target. Since the thermal shield for most cold moderators must occupy space which would preferably be used otherwise, a considerable effort was devoted to studying the various effects of geometry on neutron production. For a fairly extensive variety of experimental conditions, indium foils were exposed with and without cadmium to permit a quantitative analysis of the fast- and slow-neutron fluxes in the general area of the neutron source and moderator. The results of this study were rather helpful in pointing out that the exact location of the cold moderator was of secondary importance since the entire assembly was surrounded by a massive fast-neutron reflector. Within the well occupied by the low-temperature moderator the intensity of fast neutrons was reasonably constant, being more intense near the source, as would be expected. Over a distance from about 4 in. to about 12 in. from the electron target the intensity of fast neutrons (as well as the intensity of slow neutrons) varied by only about a factor of 2. In conclusion, one notes that the price paid for good thermal insulation is relatively small. As a result, the cold source has generally been constructed with walls of plastic Styrofoam rather than with the more compact arrangement having metal walls and using vacuum as the insulator.

DETECTORS AND ELECTRONIC CIRCUITRY

With regard to detectors, the linear accelerator has advantages relative to reactors but also has some special problems. Accelerator pulses

are more desirable than those from a chopper in that production of fast neutrons stops completely between accelerator pulses, which reduces the background, whereas the reactor-and-chopper system transmits the signal for only one percent of the time and the background is continuous. On the other hand, to take advantage of the accessibility of the accelerator neutron source, which depends on the absence of shielding walls, it is necessary to deal with gamma-ray background. Each neutron pulse is accompanied by a simultaneous pulse of penetrating high-energy bremsstrahlung, which may so overload the detecting instrumentation that its response is affected for hundreds of microseconds. In using boron-loaded scintillators, this overloading is a particular problem because of the activation during the pulse and subsequent decay of phosphorescence in the scintillator. A lead shield 2 in. thick located at the scintillator but not in the neutron beam has helped to reduce this response to gamma rays. It has also been found necessary to provide lead shielding for the BF_3 counters and their electronic preamplifiers, and in some cases to gate the detector so that it is inactivated during the electron pulse.

For a variety of experiments two types of slow-neutron detectors have been employed. One is a NE-401 5-in.-diam scintillator attached to a 5-in. Dumont photomultiplier tube (ABC7600). The other detector consists of a bank of BF_3 proportional counters (from 6 to 20, depending on the need) filled to about 1.5 atm of pressure. At the linear accelerator, the scintillator has considerable disadvantage because of its rather great sensitivity to γ -rays, as mentioned above. Its primary advantage is that of geometry. The 5-in.-diam tube fits easily into a 6 by 6 in. drift tube and simplifies the neutron shielding around the detector. As a consequence, the scintillator has been used extensively for cross-section measurements employing a 7-m flight path in Position C shown in Fig. 1. In this arrangement, a shielding wall about 8 ft thick separates the accelerator room from the experimental room and reduces the gamma-radiation problems. The preamplifier associated with the photomultiplier is a standard White cathode

follower, followed by a Hamner N354 preamplifier with clipping diodes to minimize the dead time in the main amplifier caused by the gamma radiation. Although BF_3 counters are less sensitive to gamma radiation than a scintillator, they nevertheless require considerable attention when employed in a region close to a linear accelerator. With the most recent arrangement of electron target employed in Position B in Fig. 1 there is no longer need to surround the BF_3 counters with lead. However, it is necessary to use diode clipping in the preamplifier to reduce the amplifier dead time. The recovery of the counter bank from gamma radiation limits the time measurements to flight times greater than 100 or 200 μsec , depending on exact circumstances. This is of little significance since it corresponds to only three or four 64- μsec channels out of 256 channels. Figure 3 shows the electronic system for a typical bank of BF_3 counters and for the simple monitor counter. An oversimplified wiring diagram (Fig. 4) for the preamplifier shows the diode clipping which significantly improved the operation with the gamma radiation.

TIME-OF-FLIGHT ANALYZER AND DATA HANDLING

The various electronic components in the time-of-flight system are shown in the block diagram (Fig. 3). In a typical operation the sequence of events is as follows. The linear accelerator runs on a regular repetition rate of either 30, 60, or 120 pulses/sec, depending on (1) the flight path being used, (2) the energy of neutrons of interest, and (3) the pulse overlap permissible. About 5 μsec prior to the acceleration of the electron beam, the linear accelerator provides a timing pulse, called a prepulse, which is used to trigger the time-of-flight analyzer. The fastest neutrons detected in the present apparatus (of the order 1.0 ev) are recorded soon after recovery from the gamma flash at times of the order of 100 μsec . A 256-channel time-of-flight analyzer (RCL) is used to provide 16-, 32-, 64-, or 128- μsec timing channels. A separate beam-monitor detector is used to give a signal proportional to the number of electrons hitting the electron

Fig. 3--Block diagram of the time-of-flight system showing the BF₃ counter detector bank and the single BF₃ counter beam monitor

Fig. 4--Over-simplified wiring diagram of the preamplifier for the BF_3 counters, which shows the location of the clipping diodes that significantly reduce the effect of gamma radiation

target. The location of this single BF_3 counter is such that it responds directly to the initial fast-neutron beam and is essentially independent of any changes in position for moderators or specimen samples. After blanking for the first 100 μsec with an off-gate unit to reduce the effects caused by gamma radiation, the monitor-counter signal feeds directly into a scaler with a capacity of 10^6 and equipped with adjustable preselectors to provide an arbitrary number of beam counts up to one million. After the monitor has accumulated the appropriate preselected number of counts, the time-of-flight analyzer is automatically shut off and is ready for recording the accumulated data.

The data-recording system for the time-of-flight analyzer includes a time analyzer, mentioned above, which can record up to one count per channel per pulse with a maximum of 256 channels. From there it is displayed on a scope and is recorded on printed tape or on an automatic plot or is punched on paper tape. The punched tape is, in turn, automatically converted to cards for an IBM-704 computer program. Recently, the IBM-704 has been replaced with an IBM-7090 with some consequent changes required in the programming. Soon an RPC-4000 computer will be available directly at the linear accelerator facility. It will presumably be programmed to take data directly from the 256-channel analyzer for immediate computations. Codes have been formulated for the IBM-704 for the computation of neutron-beam intensities, neutron energies, and neutron cross sections. Since the data-recording system accepts only one neutron per 16- μsec channel per pulse, the computer program must include a correction for coincidence loss in those channels where the count rate approaches one per pulse. Time for computation for intensity spectrum or a cross-section curve for 256 individual energies is of the order of 0.01 hr.

CRYOSTATIC EQUIPMENT

As compared with reactors, the necessary equipment for maintenance

of low temperatures is simple. For liquid nitrogen temperature (77°K) a vacuum-insulated cryostat is not essential. A view from above an open aluminum box insulated with a 2-in. outer layer of low-density plastic, Styrofoam, is shown in Fig. 2. The polyethylene moderator is completely immersed on three sides in liquid nitrogen, the fourth side being in contact with the aluminum walls and opposite a neutron window where multiple layers of aluminum foil substitute for the Styrofoam thermal insulation. Reduction of temperatures from 295° to 77°K by addition of the liquid nitrogen substantially reduces the abundant flux in the range $>0.05\text{ ev}$, but enhances the weak flux below 0.005 ev by at least a factor of 4. Such a cold source has therefore been in regular use.

For lower temperatures, the use of liquid hydrogen and liquid helium requires development of a suitable cryostat, such as that shown in Fig. 5. In order to minimize neutron attenuation, the walls of the lower portion are made of aluminum except for the inner cylinder of 0.4-mm stainless steel. Since a surrounding layer of liquid nitrogen is precluded for the same reasons, the radiation shield around the low-temperature area is maintained at low temperature by conduction from the annular nitrogen tank above. The inner cylinder is of uniform 6-in. diam to make possible substitutions of new moderating assemblies. Despite these departures from conventional cryostat design principles, the thermal conduction and radiation losses are acceptably low. As already discussed, the gamma-radiation heating is not serious because of the intermittent operation of the accelerator. In normal operation with the cryostat adjacent to the electron target, consumption of liquid hydrogen was found to be of the order of 2 liters during the 24-hr period of usual accelerator experiments.

The cryostat described above provides a great flexibility in arranging samples to be used in the experiments. In the process of conducting these many experiments, a number of different sample holders have been constructed or modified from previous holders. Figure 6 shows a photograph of a number of such holders, each of which will fit one at a time inside the

Fig. 5--Cryostat for maintaining moderators at 4°K or 20°K; the walls of aluminum and thin steel are relatively transparent to neutrons

Fig. 6--Six typical holders used for various low-temperature measurements of neutron spectra and neutron cross sections; each holder can be inserted in the cryostat shown in Fig. 5

cryostat. The large variety of holders was constructed to provide appropriate thicknesses for cross-section measurements, geometrical shapes for neutron-spectrum measurements, and means to alter the specimen temperatures from room temperature to 4°K .

Measurements have been made of both the total neutron cross section and neutron spectrum for liquid methane, solid methane, and CH_4 . Figure 7 shows the condenser, constructed from 1/8-in. copper used for liquefying the methane. When in use, the condenser is placed in 1 or 2 in. of liquid nitrogen, which conducts heat out of the lower metal skirt and thus the upper volume is cooled. When pure gas from a room-temperature tank of methane was communicated to the upper, closed, cooled region, it quickly condensed. In a period of 4 hr, approximately 8 liters of liquid methane were obtained.

For the purposes of the experiments conducted so far, very rudimentary temperature-measuring equipment was sufficient. Both small resistances and thermocouples were used. Small resistances (1/10 w, 47 ohm, IRC composition carbon) have proven useful as level indicators for liquid hydrogen or helium. Figure 8 shows a wiring diagram used to indicate liquid level. Table 1 gives typical values of the current in the resistor in various cold liquids, using nine 3-v-potential sources. Such resistances have proven ineffective in ordinary applications for accurately measuring absolute temperatures. When it has become necessary to cool

Table 1
CURRENT IN 47-OHM RESISTANCE FOR
VARIOUS COLD LIQUIDS

Liquid	Temp. ($^{\circ}\text{K}$)	Current (ma)	
		In	Out
N_2	77	46.7	56.7
H_2	20	30.6	38.0
He	4	23.5	32.7

Fig. 7--The copper condenser used for the liquefaction of methane; the perforated lower cooling skirt provides a heat leak for the rough control of the liquefying temperature

x - 47-OHM, 1/10-WATT RESISTORS

Fig. 8--Schematic of the method used for measuring liquid levels; the currents produced by a 3-volt potential gave indications of the liquid level

a large volume of material, such as 1 or 2 liters of H_2O , to the temperature of liquid hydrogen or helium, we have preferred to wait an appropriate length of time for thermal equilibrium, observing conditions of coolant boil-off and other phenomena to assure proper isothermal conditions. In a number of cases, however, such as cooling from a known temperature, that of liquid methane ($111.5^\circ K$), for example, to the temperature of liquid hydrogen ($20^\circ K$), a chromel-alumel thermocouple has been used. In spite of the fact that thermocouples become relatively insensitive to the temperature changes in the very low temperature region, the change of potential with temperature is still sufficient to permit reasonably accurate temperature measurements.

CROSS-SECTION AND SPECTRUM MEASUREMENT TECHNIQUES

The neutron cross section is determined from measurements of the beam transmission of a sample. The transmission T_i is related to the cross section σ_i as follows:

$$T_i = e^{-nx\sigma_i}, \quad (1)$$

where n is the number of nuclei per cubic centimeter having a cross section σ_i , and x is the thickness of the sample (in centimeters). The transmission is given experimentally as

$$T_i = \frac{(R_1 - R_2)_i}{(R_3 - R_4)_i}, \quad (2)$$

where $(R_1 - R_2)_i$ is the counting rate minus its background at the detector in time channel i with the sample in the beam, and $(R_3 - R_4)_i$ is the counting rate minus its background with the sample removed.

Experimental measurements of transmission were made at each of the three positions shown in Fig. 1, with flight paths of about 250 cm at Positions A and B and 700 cm at Position C. The technique at the accelerator is to allow a burst of thermal neutrons ($\sim 100\text{-}\mu\text{sec}$ duration at source)

to drift down the beam port to the detector. In traveling a flight path, d , the neutrons are time-analyzed so that neutrons arriving in the time-analyzer channel i (channel width τ) have a velocity v_i given as

$$v_i = \frac{d}{\tau_i} \quad (3)$$

The neutron energy is therefore related directly to the channel in which it is detected.

A good check on the reliability of the procedure is given by making measurements on a substance whose transmission is accurately known. For this purpose, an object was constructed with a known transmission (0.42) at all energies of interest. This consisted of a thick sheet of cadmium perforated with very many small holes. Measurements for this sample were always interspersed among those for other samples being studied to check on systematic errors. Such data are shown in Fig. 9. The computer evaluated Eqs. (1) and (2), using an arbitrary value of n_x in order to represent the results in terms of cross section. The value for the cross section in Fig. 9 is seen to be quite accurately independent of energy and corresponds to the transmission of 0.42 known for the sample. Results of this kind aid in placing reliance on measurements of unknown specimens.

In Eq. (2), the values of $(R_3 - R_4)_i$ represent the components of the spectrum emitted from the neutron source arriving in channel i . Measurements over an appropriate time duration give the time distribution of emitted neutrons. This time distribution can be converted to an energy distribution, noting that the differential energy distribution dN/dE is related to the measured time distribution dN/dt as follows:

$$\frac{dN}{dE} = \frac{dN}{dt} \times kt^3 \quad (4)$$

where k is a constant involving the neutron mass. The energy-spectra measurements to be reported are computed in this manner.

Fig. 9--A plot of transmission of a thick piece of cadmium perforated to have a known transmission of 0.42; except for systematic errors, the transmission, and hence "cross section" computed from this transmission, should everywhere be constant; the fluctuation for energies less than 0.001 ev represents statistical uncertainties in the data

III. TOTAL NEUTRON CROSS SECTIONS AT LOW ENERGY FOR SEVERAL HYDROGENOUS SUBSTANCES

One of the interests for the present experimental program has been in ascertaining the differences in neutron interactions in liquid para-hydrogen and liquid ortho-para-hydrogen and in comparing these results with those for other hydrogenous substances such as H_2O , CH_2 , and ZrH . Before discussing the cross-section results, it seems worthwhile to digress briefly to recall for the reader the pertinent physical facts about liquid hydrogen.

Farkas⁽⁵⁾ has given a wave mechanical description of ortho-hydrogen and para-hydrogen. It is shown that the para- system has even molecular rotation states ($J = 0, 2, 4, 6, \dots$) and that the ortho- system has odd states ($J = 1, 3, 5, \dots$). The lowest energy state available to para-hydrogen is $J = 0$, whereas the lowest for ortho-hydrogen is $J = 1$. Condensation of hydrogen gas gives 3:1 for the ratio of liquid ortho-hydrogen to liquid para-hydrogen. Transitions gradually occur from $J = 1$ to $J = 0$, with the ortho-hydrogen gradually converting to para-hydrogen. While it is possible to obtain liquid hydrogen in a practically pure para- state (>99%), it is impossible to obtain a concentration of ortho-hydrogen greater than 75%. In the further discussions, unless otherwise noted, para-hydrogen will imply full conversion (>99%) and ortho-para-hydrogen will imply a mixture.

CROSS SECTION OF PARA- AND ORTHO-HYDROGEN

One of the areas in which significant differences between the two states of hydrogen can be found is the total cross section for neutron scattering as a function of energy. An experimental program has been carried out to measure these two cross sections using the time-of-flight technique. • A variety of sample thicknesses was used. Ortho-para-hydrogen

measurements were made using sample holder 6 in Fig. 6; sample holders 1 and 3 were used for para-hydrogen. Figure 10 shows the experimental results for the total cross section of liquid para-hydrogen and liquid ortho-para-hydrogen at their normal boiling points. Also shown in the same graph are the results obtained by Sutton, et al.,⁽¹⁾ for para- and ortho-hydrogen gas. The latter measurements were made to check the theoretical calculations based on the Schwinger-Teller theory. To a rather good approximation it is seen that the liquid para-hydrogen cross section and, to a lesser extent, that for liquid ortho-hydrogen agree with the respective gas cross-section measurements.

It should be pointed out that the absolute values of the cross sections for ortho-para-hydrogen and para-hydrogen are somewhat different from those plotted in Fig. 10. For the para-hydrogen specimen, it was assumed that all molecules were in the para- state. It is possible that the amount of the specimen in the ortho- state was a few percent, though it is more likely that this fraction was of the order of 0.4%. Even such a small admixture of ortho-hydrogen with its large cross section would affect the measurement of the small para-hydrogen cross section for energies below 0.015 ev. No correction is made for this because of the lack of information on the components of the mixture. The cross section was computed for the ortho-para-hydrogen case on the assumption that all molecules were ortho-hydrogen. For energies below 0.015 ev, the ortho-hydrogen cross section is a great deal larger than shown, since at least one-fourth of the molecules had the very low cross section of para-hydrogen.

The cross section for liquid ortho-para-hydrogen behaves very much the same as most hydrogenous materials. The usual effects of chemical binding appear. Liquid para-hydrogen, in contrast to other materials studied, exhibits a cross section which decreases rapidly with decreasing energy to reach a minimum of about 3 barns per hydrogen atom at about 0.013 ev, then rises with further decreasing energy. The significance of this behavior is that decrease of the hydrogen cross section below the free-atom value of 20 barns necessarily implies a coherent scattering phenomenon.

Fig. 10--Cross section for para-hydrogen and a mixture of ortho-para-hydrogen; the cross section in the energy interval 0.010 to 0.020 ev was measured with considerably higher resolution and is shown in the insert

In general, the large scattering cross section of hydrogen is almost entirely incoherent. That is, there is no constructive or destructive interference between scattering from neighboring atoms because of the random orientation of the nuclear spin. In hydrogen molecules, however, spins of the two atoms are necessarily in the anti-parallel para-position or in the parallel ortho-position, such that there is coherence in at least the scattering of the two atoms within the molecule. The sharp rise from about 3 barns to considerably higher values appears to be due to the excitation from the $J = 0$ rotational level of the para-state to the first level, $J = 1$, of the ortho-state. Since the Doppler broadening of the level lowers the observed threshold for the cross-section rise below the energy of the level by some fraction of the thermal energy (0.0016 eV), the rotational level should be of the order 0.014 to 0.015 eV from the cross-section measurement. Measurements of specific heat in liquid hydrogen give a value of about 0.015 eV for this level, in good agreement with the present data.

Some features of the rapid rise in cross section for para-hydrogen are of interest. In the first place, the exact energy associated with this rapid rise in cross section is of consequence. The sharpness of the threshold is also of interest. In order to calibrate the energy scale for the cross-section curve, the beam was filtered through iron, which has sharp Bragg cutoff energies at 0.005 eV, 0.010 eV, and 0.015 eV, as shown in Fig. 11. With the information provided by this data, calibrations of the energy scale were made for the cross-section and spectra measurements.

The sharpness of the onset of the rise from the low value of cross section to higher values can give information on the rotations of the hydrogen molecule as hindered by intermolecular forces. In order to check on this feature experimentally, a small, rotating, mechanical chopper was inserted in the neutron beam close to the neutron source to improve significantly the resolution by reducing the duration of the neutron pulse to about 20 microseconds instead of 100 microseconds. In this case it became possible to examine meaningfully the cross section by making measurements

Fig. 11--Spectrum of neutrons transmitted through 1/2 in. of iron, showing the Bragg cutoff values at 0.005 ev, 0.010 ev, 0.015 ev, and 0.020 ev

with 16-microsecond channels in the time-of-flight analyzer, resulting in an energy resolution of about 0.0002 ev. Unfortunately, the chopper so reduced the data accumulation rate that rather poor statistics were available. In addition, the chopper also significantly limited the energy region over which cross-section measurements were made. Therefore, it is not possible with the presently available data to compute the cross-section curve in the whole region of interest. The chopper has shown, however, that the cross section starts to rise quite abruptly, within an energy interval of less than 1 mv at 0.013 ev. One concludes, therefore, that the molecular rotations are not strongly hindered.

CROSS SECTIONS OF LIQUID AND SOLID H₂O

For a number of considerations connected with the use of water and ice as a neutron moderator, it is useful to know the temperature dependence of neutron scattering caused by the water molecule, H₂O. For instance, in considering water cooled to very low temperatures as a useful neutron source, it is useful to know the energy dependence of the scattering cross section at low temperature. In addition, the shape of the cross-section curve as a function of neutron energy may reveal some of the properties of the crystalline structure for ice.

In view of the above considerations, cross-section determinations were made for water at +5°C, -5°C, -196°C, and -253°C, using an experimental arrangement such as that shown at Position A in Fig. 1. The source of neutrons was sample holder 4 in Fig. 6, with H₂O in the bottom portion and H₂ in the top. The total path length from source to the BF₃ detector was 253 centimeters. The distilled water specimen was mounted between two plates of aluminum, each 1/4 in. thick and 8 in. in diameter, of which the central 3-in. -diam portion was used. The water thickness was approximately 0.100 in. The desired temperature, measured with copper-constantan thermocouples, was maintained by an adjustment between a heater placed on the circumference of the water holder and a cooling

atmosphere maintained by a refrigerator box cooled variously with dry ice or liquid nitrogen. The sample for the lowest temperatures was held in the thinnest portion of holder 3 and cooled with liquid hydrogen. The neutron beam passed through thin aluminum windows placed on two sides of the refrigerator box. Considerable care was exercised in the manipulation of the scatterer to prevent the accumulation of frost on the beam windows and the sample itself.

A particularly difficult experimental problem is connected with the number of scattering molecules in the beam. The number, n , of molecules per cubic centimeter and the sample thickness, x , enter the transmission formula as $T = e^{-nx\sigma}$, where σ is the cross section. From the construction of the holder, which permitted easy expansion in the beam direction, it is expected that the expansion of the ice is mainly in this direction. Consequently, one can assume that nx is constant, independent of temperature, in our experiment. This circumstance makes it unnecessary to consider which of the seven types of ice⁽⁶⁾ is present and the appropriate density. Because the exact shape of the sample was not determined or controlled precisely during the freezing, there is some possibility of error in assuming for the different temperatures a constant value of nx . However, other measurements of the cross section using a crystal monochromator at the General Atomic TRIGA reactor indicate that the above assumption is valid for ice at -5°C .

In order to obtain both an independent confirmation of the constancy of nx for the ice sample and additional cross-section data to supplement those from the linear accelerator, measurements of the ice and water cross section were made at the General Atomic TRIGA reactor, using a high-resolution monochromator to select a number of well-defined neutron energies. The time-of-flight technique for measuring the cross section gives an accurate indication of the variation of cross section with energy but makes absolute cross-section determinations for a specific energy somewhat less reliable than those obtained using a crystal monochromator technique at a

reactor. For absolute calibrations or for relative calibrations of different samples at a single energy, a continuous reactor source is advantageous because of the greater precision in monitoring the beam. At a number of energies in the range 0.05 to 0.13 ev, the cross section was measured for $+5^{\circ}\text{C}$ and -5°C H_2O , and it agrees well with that determined from the linear accelerator time-of-flight measurements.

The experimental results for H_2O are shown in Fig. 12. It is to be noted that the data for liquid water agree exactly with the published data of Melkonian and of the Brookhaven slow-chopper group, as read from the cross-section compilation BNL-325.⁽⁷⁾ In addition, these results agree with the theoretical computations of Nelkin.⁽⁸⁾ Results for ice at -5°C , -196°C , and -253°C are also shown. The general behavior of the curves is what is expected. For energies near 0.1 ev, the various curves tend to merge, although the cross section of -196°C and -253°C ice is greater than that for ice at -5°C and for water. For neutron energies intermediate between about 0.006 ev and 0.1 ev, all curves for solid H_2O exhibit a substantial increase in cross section over that for liquid H_2O , the change being decidedly nonlinear with temperature difference. This general behavior is to be expected because of the more tightly bound hydrogen atoms in the ice molecule. For energies below about 0.006 ev, in which the inelastically scattered neutrons mainly gain energy, all of the ices show a scattering reduced below that for water. This is to be expected because the solid forms of water have higher Debye temperatures than liquid water. As a consequence, the solid forms have a smaller number of low-energy levels. At low temperatures, there is a smaller population of the energy levels from which neutrons can scatter inelastically to gain energy. The magnitude of this effect is seen to be dependent on the temperature of the ice, as expected. For ice at -196°C , the cross section for energies below 0.006 ev rises only slowly with decreasing energy. For ice at -253°C , the cross section for energies below 0.006 ev remains nearly constant. Ice at -5°C and water at $+5^{\circ}\text{C}$ have cross sections which rise much more rapidly with decreasing energy.

Fig. 12--Cross section of H_2O molecules at +5°C, -5°C, -196°C, and -253°C

CROSS SECTION OF LIQUID AND SOLID METHANE

Another substance, methane (CH_4) in liquid and solid forms, is of interest for the production of low-energy neutrons. The group at the EURATOM Laboratory at Mol have reported⁽⁹⁾ results which show that methane has good low-temperature properties. Several liters of pure liquid methane were prepared for use both for spectrum and total cross-section measurements. The methane spectra will be reported in a later section of this report. The cross-section results are reported here. Figure 13 shows the results for the total cross section of both liquid and solid methane. The temperature of the liquid was the normal boiling point; the temperature of the solid was that of liquid hydrogen (20°K). For comparison purposes, the cross section of CH_2 at 295°K is also shown in Fig. 13. The cross section is for the whole molecule, CH_4 . It is of interest to note that neither the liquid nor the solid show any abrupt breaks or rapid energy-dependent deviations of cross section as were noted for para-hydrogen. Like water, methane has a monotonically rising cross section, showing no abrupt changes attributable to low-energy vibrational or rotational states. It is to be noted that the cross section for solid methane for the lower energies is lower than that for the liquid state. This situation was noted for H_2O in its different phases and was discussed above. The explanation is the same for the case of methane. The cross sections were measured with two thicknesses of methane. These thicknesses were chosen to give a variety of conditions for transmission at both the high- and low-energy regions. Consistency between the various measurements was obtained. Appropriate checks were made using the perforated cadmium sample, as discussed in Section II, "Experimental Techniques."

CROSS SECTION OF LIQUID AND SOLID D_2O

Cross-section data have been obtained for heavy water for temperatures of $+9^\circ\text{C}$, -1°C , and -196°C . The specimen, 1.55-cm thick, was

Fig. 13--Cross section for CH_4 molecule at the boiling point, 111°K, and at 20°K; for comparison, the cross section for CH_2 molecule is also shown

mounted between the same aluminum plates used for the water specimen. In addition to the thermocouples attached to the outside of the aluminum holder, a thermocouple was placed in the center of the liquid to permit a temperature measurement on the D_2O itself. The same potential difficulties with the n_x determination described above for the water specimen were present for the D_2O specimen. As in the preceding case, the value of n_x was considered to be independent of the temperature. On this basis, the data were analyzed with the results exhibited in Fig. 14.

The cross section of D_2O at $+9^\circ C$ agrees well with the data in BNL-325.⁽⁷⁾ The three curves merge in the energy region above about 0.02 ev. This is the region for which solid H_2O showed an increase in cross section compared to liquid H_2O . Such an increase is not expected for deuterium because (1) the binding effect given by $[(A + 1)/A]^2$, A = atomic mass, is about half as great as for hydrogen, and (2) the oxygen cross section is a much larger fraction of the total cross section for D_2O as compared to H_2O . The curve for liquid D_2O exhibits diffraction effects, as expected, since the scattering from deuterium is mainly coherent. The results for solid D_2O at energies below 0.01 ev also exhibit diffraction effects. These are not increased by going from liquid to solid at about the same temperature. However, the magnitude of the scattering is considerably reduced for the solid, presumably indicating fewer scattering levels in the solid. With further cooling to $-196^\circ C$, the cross section in the region around 0.001 ev is greatly reduced. This is in contrast to the results for H_2O where the temperature variation of the cross section at 0.001 ev is proportionately a much smaller fraction of the liquid cross section. No theoretical treatment of the cross sections for the solid D_2O lattice is now available.

CROSS SECTION OF METAL HYDRIDES

The total scattering cross section increases with decreasing neutron energy from a value characteristic of the free atom to a value characteristic of rigid binding, with the ratio $\sigma_b/\sigma_f = (A + 1)^2/A^2$, where A is the atomic

Fig. 14--Cross section of D_2O molecule at $+9^{\circ}C$, $-1^{\circ}C$, and $-196^{\circ}C$

mass and the subscripts b and f refer to the bound and free states, respectively. In Fermi's original treatment⁽¹⁰⁾ of neutron slowing-down, the hydrogen was regarded as a harmonic oscillator bound to an infinitely heavy atom and having energy levels at multiples of $h\nu$. In that case, he showed the form of the expected variation of cross section with energy to be as illustrated in Fig. 15, with sharp minima at the threshold for excitation of each level. The details of this result could hardly be expected to apply to water, for which the assumed binding is not a reasonable approximation. In zirconium hydride, with the hydrogen confined at the center of a tetrahedron of zirconium atoms, the hydrogen behavior could be expected to be more like that of an isotropic harmonic oscillator. Cross sections have been measured for a sample of $\text{ZrH}_{1.5}$, using a high-resolution neutron crystal spectrometer. In Fig. 16 is shown the variation of cross section with energy, which exhibits the general features shown in Fig. 15 if the energy levels are taken to be at multiples of about 0.13 ev. The solid curve represents a reasonable fit to the experimental points. Inclusion of the Doppler broadening due to acoustical vibrations of the lattice brings the Fermi model into better agreement with the experimental cross sections.

A similar measurement of the total cross section for MgH_2 in Fig. 17 shows some irregularity in the energy variation of cross section, but no well-defined energy levels as in the case of $\text{ZrH}_{1.5}$. This difference is to be expected from the difference in the structures. Whereas the tetrahedral symmetry of $\text{ZrH}_{1.5}$ results in an isotropic potential well for the hydrogen, MgH_2 has a layer structure in which the hydrogen binding is nonisotropic. The cross section is therefore represented by a superposition of several curves of the type shown in Fig. 15, and the detailed structure is obscured. Also, the overlap of adjacent levels, arising from Doppler broadening by thermal motion of the lattice, should be greater for magnesium hydride than for zirconium hydride because of the lower mass of the magnesium atom.

The above results show that the total neutron cross section of

Fig. 15--Theoretical neutron cross section of bound proton, with frequency ν of isotropic harmonic oscillation

Fig. 16--The solid curve gives the total neutron cross section of hydrogen in zirconium hydride ($\text{ZrH}_{1.5}$) obtained from total cross section by subtracting the constant cross section of zirconium; the Fermi theoretical curve corrected for Doppler broadening has been fitted to the above data with $h\nu = 0.137$ eV and it agrees with the experimental curve except as shown by the dashed curve

Fig. 17--Total neutron cross section for magnesium hydride (MgH₂); the anomaly between 0.12 and 0.20 ev indicates partly resolved excitation levels for vibrations in MgH₂ lattice

hydrogen in zirconium hydride depends significantly on the energy of the incident neutron. The experimentally measured cross section shows the predicted variations which appear at multiples of the Einstein energy level, $h\nu$, which is about 0.130 ± 0.005 eV for ZrH. (Additional measurements using a cold neutron time-of-flight technique verified this energy level.⁽¹¹⁾) The cross section for lower neutron energies has been determined experimentally using the accelerator time-of-flight technique. For neutrons incident with lower energy ($< h\nu$), one expects, to first approximation, that the cross section of hydrogen bound as a harmonic oscillator should increase to the fully bound hydrogen cross section of 80.4 barns as the incident energy is decreased. The slow increase of the total cross section above the value for a fully bound hydrogen atom is due to inelastic scattering from the acoustical modes and is determined by the frequency distribution in the solid.

To compute the cross section of zirconium hydride, we have used the results of Marshall and Stuart⁽¹²⁾ with appropriate modifications for the case of a bound hydrogen atom. On the assumption of a Debye frequency distribution for the incoherent scattering, the cross section has been computed. For this computation there are three factors to be considered: (1) a form factor due to optical vibrations, (2) inelastic scattering from acoustical modes, and (3) diffraction effects in zirconium. The latter are small and can be approximately corrected by subtracting the observed elastic cross section for zirconium for neutron energies above the Debye cutoff. On these bases, we derived the following expression for the total cross section:

$$\sigma = \sigma_b f\left(\frac{E}{h\nu}\right) \left[1 + A_1(\chi, t) \left(\frac{m}{M}\right) + A_2(\chi, t) \left(\frac{m}{M}\right)^2 + A_3(\chi, t) \left(\frac{m}{M}\right)^3 + \dots \right]$$

where $\sigma_b = 81.4$ barns/H atom = bound cross section,

$$\chi = (E/\theta)^{1/2},$$

$$t = T/\theta,$$

θ = Debye temperature,

E = incident neutron energy,

T = temperature of scatterer,

m = neutron mass (1),

M = Zr mass (91),

$$f(E/h\nu) = \left(1 - e^{-4E/h\nu}\right) \frac{4E}{h\nu},$$

$h\nu$ = energy of Einstein level (0.13 ev for ZrH).

The parameters A_1 , A_2 , A_3 in this expression have been computed and tabulated by Marshall and Stuart. ⁽¹²⁾

During the experiment, the sample was kept at room temperature. The only remaining parameter subject to choice is the Debye temperature to be assumed for the zirconium hydride lattice. The cross sections have been computed for three different assumptions of reasonable Debye temperatures: 197°K (0.0170 ev), 296°K (0.0255 ev), 370°K (0.0319 ev). Both theoretical and experimental results are shown in Fig. 18 and are observed to be in relatively good agreement with the shape of the observed experimental curve.

A comparison of theoretical and experimental results shows that they are in quite good qualitative and quantitative agreement, both in regard to the shape and the absolute magnitude of the curve. The predicted cross section is found, however, to be relatively insensitive to the choice of Debye temperature for the lattice. This has the consequence that the agreement between experiment and theory may be regarded as real, rather than resulting from a fortuitous choice of Debye frequency. From the practical viewpoint of reactor theory, this is an advantage in that the cross sections may be regarded with confidence for computations. However, from the pure solid-state physics viewpoint, it is a disadvantage in that little additional information about the frequency distributions in the solid is to be expected from further measurements of this kind without extremely precise cross-section measurements.

Fig. 18--Cross section of H in ZrH at low energy

A number of additional cross sections were measured immediately preceding this contract period and have been reported.⁽¹³⁾ The temperature and energy dependence of the BeO cross section were reported there, and a theoretical computation of the thermal elastic scattering in BeO was discussed. BeO cross-section measurements were made because of the large interest in this material as a high-temperature reactor moderator.

IV. NEUTRON SPECTRA FROM HYDROGENOUS, LOW-TEMPERATURE MODERATORS

Neutron spectra from low-temperature moderators provide much information concerning low-energy neutron sources. This information, when combined with inelastic scattering data and total neutron cross sections, can be used to predict effective moderator sizes. Neither the energy distribution of vibrational states in solids and liquids nor the probability of phonon exchange with incident neutrons is sufficiently understood, particularly in the low-energy region, to permit computation of neutron spectra in cold moderators. This is the reason for the present interest in these measurements.

COLD-NEUTRON SOURCES

Consider first the general features of thermal equilibrium between neutrons and moderator. Assuming complete thermal equilibrium between neutrons and moderator, with no perturbation of the spectrum by absorption effects, the flux, N , at temperature T is given by

$$dN = E e^{-E/kT} dE \quad \text{or} \quad dN = (1/t^5) e^{-E/kT} dt \quad (5)$$

in terms of energy and time of flight, respectively; here proportionality constants are omitted. The theoretical energy distributions are shown in Fig. 19 for temperatures of 295° , 77° , 20.4° , and 4.2° K, corresponding to room temperature and the boiling points of liquid nitrogen, liquid hydrogen, and liquid helium, respectively. These show that for very slow neutrons it is possible, in principle, to increase the flux per unit energy range by three orders of magnitude. It should be noted that this does not imply an increase in total neutron flux; in fact, the assumption in Fig. 19 is that the total number of neutrons remains constant but concentrated in a

Fig. 19--Ideal Maxwell-Boltzmann energy distribution of neutron flux for equilibrium temperatures 295°, 77°, 20°, and 4.2° K

narrower energy range. In a 295°K thermal distribution, a fraction of 0.02 lies below the 0.005-ev beryllium-filter cutoff, so that the flux of beryllium-filtered neutrons might be increased by a factor of 50. There are, of course, practical limitations in attaining these ideal increases of flux. As the neutron decreases in energy, the number of vibrational states of the moderator to which it can lose energy usually diminishes rapidly, so that a larger number of collisions, and therefore longer path length in the moderator, is required to approach equilibrium. For this reason, and also because the neutron absorption by the moderator and any impurities it contains increases with $1/v$, a greater loss of neutrons by absorption occurs than at higher temperatures.

NEUTRON SPECTRA FROM HYDROGEN AND SOLID AND LIQUID H_2O

In Fig. 20, the velocity, or time-of-flight, distributions of neutrons from solid H_2O , from para-hydrogen, and from ortho-para-hydrogen (holder 3 in Fig. 6) all at 20°K , show large differences in behavior of the moderators. For thermal equilibrium, the maximum flux per unit time should be at the time corresponding to an energy of 2.5 kT, or 0.0045 ev for temperature 20°K . The total distribution should include an ideal Maxwellian spectrum, but be shifted toward higher energies by the superposition of the $1/E$ slowing-down spectrum. It is not surprising that the peaks of the distributions are in the range 0.008 to 0.012 ev, but it is significant that the shapes of Curve 1 for H_2O and Curves 3 and 4 for H_2 are quite different. The H_2O spectrum is depressed strongly in the region of about 0.06 to 0.1 ev and weakly around 0.021 ev, corresponding approximately to the strong hindered rotational level at 0.061 ev, known from previous measurements, ^(14, 15, 16) and a weaker level at 0.021 ev measured by Hughes, et al., ⁽¹⁶⁾ for the case of liquid H_2O . Although these energy levels introduce no anomalies in the spectra from liquid H_2O , it is reasonable that the levels, if they exist at all in ice, would be sharper and would result in scattering of neutrons out of the energy region immediately above

Fig. 20--Time-of-flight distributions of neutrons from para-hydrogen, ortho-para-hydrogen, and H₂O moderators at 20°K; the curves have been normalized to the same number of incident fast neutrons, and the spectra from the side and the interior of the para-hydrogen sample are identical within 10%

threshold. There is little evidence in Fig. 20 for similar anomalies corresponding to the 0.008, 0.005, and 0.0005 ev weak levels also observed by Hughes, et al.⁽¹⁶⁾

Curve 5 in Fig. 20, for para-hydrogen, exhibits a peak at 0.014 ev much sharper than expected from any form of statistical energy distribution, suggesting that the abrupt change in cross section for para-hydrogen at about 0.015 ev indicates a rotational energy state of the ortho-para-hydrogen system, possibly with other energy levels in the region 0.014 to 0.060 ev, for all of which the flux is depressed as compared with water. (An inelastic-scattering measurement reported in a later section confirms the presence of a strong scattering level at about 0.015 ev.) This strong energy level at low energy indicates a significant advantage of hydrogen over water for moderation to very low energies. The ortho-para-hydrogen results in Curves 3 and 4 show a slight broadening of the peak for pure para-hydrogen (Curve 5). Except for very long flight times where the ortho-para-hydrogen curve indicates slightly more cold neutrons, the two modifications of hydrogen are very similar. Although both spectra from liquid hydrogen are shifted toward lower energies as compared with water, the total scattered intensity is lower. This difference is caused possibly by its lower density and consequent decreased effectiveness in slowing down incident fast neutrons from the source. This factor should not be applicable to a source in a reactor isotropically irradiated with thermal neutrons.

An item which may be significant for design of cold-neutron facilities is seen in comparison of intensities from the side and interior. For solid H_2O and ortho-para-hydrogen, the neutron currents from the surface and from the interior of the moderator are quite different, whereas for para-hydrogen there is negligible difference. Interpretation of the basic significance will require further experiments, but there is one conclusion of importance in the design of cold-neutron sources in reactors. The relative effectiveness of various moderators may be quite dependent on the geometrical arrangement of the source. Solid H_2O is relatively much more effective

for Configuration 1 in Fig. 20, rather than Configuration 2, which approximates the geometry used by Butterworth, et al.,⁽³⁾ in evaluating the effectiveness of various media.

For a better comparison of intensities at low energies, some of the data of Fig. 20 are shown also in Fig. 21, but on a logarithmic scale and with the curves normalized to represent the same number of incident fast neutrons. This normalization is somewhat arbitrary but appears to approximate best the case of moderators inside a reactor thermal flux. It is seen that the intensity in range 0.005 to 0.015 ev, and hence the integrated flux, is very nearly equal for solid H₂O and hydrogen, but that para-hydrogen is better by a factor of 3 at very low energies.

For comparison of energy distributions from H₂O at various temperatures, Fig. 22 is plotted in the same form as the theoretical distributions in Fig. 19. Time-of-flight data on neutrons per time channel are converted to flux per unit energy range by multiplication by a factor proportional to t^3 , in accordance with Eq. (4). The energy distributions* in Fig. 22 are qualitatively similar to the theoretical Maxwellian distributions, but with significant quantitative differences. As expected, all curves converge at higher energies, where the spectrum is primarily a $1/E$ slowing-down spectrum rather than in thermal equilibrium. Although the peak of the 77°K (liquid nitrogen temperature) spectrum is only slightly higher than the energy 0.0065 ev, corresponding to equilibrium, the maximum for 20°K is around 0.004 ev, corresponding to equilibrium at 50°K, and peak intensity is correspondingly lower. This indicates that moderation is no longer effective enough for a close approach to equilibrium. The broken curve, as indicated, refers to neutrons emitted from the outer surface of the cylinder. Intensity at the peak and at lower energies is lower by a factor of 6 than from the interior, and the peak energy is about 0.0046 ev, corresponding to 57°K equilibrium.

The enhancement of a slow-neutron flux using a liquid para-hydrogen source rather than a conventional liquid moderator at room temperature

*All plotted energy distributions have been corrected for air absorption.

Fig. 21--Time-of-flight distributions of neutrons from para-hydrogen and H₂O moderators at 20°K; curves have been normalized to equal integrated flux from the moderator

Fig. 22--Energy distribution of the neutron spectrum from H₂O at 295°, 77°, and 20°K

is illustrated by Fig. 23. For energies beyond the Bragg diffraction cutoff of beryllium (i. e., <0.005 ev), intensity is increased by a factor of about 60 to 75. The small sharp breaks in the para-hydrogen curves represent the diffraction cutoff at 4 Å (0.005 ev) for the stainless steel inner walls of the cryostat. Intensity of scattering from liquid H_2O is almost the same from the surface of the 6-in. -diam cylinder as from the 2.5-in. -diam cylinder, and scattering from liquid hydrogen differs only by about a factor of 2 for the two cases, a circumstance fortunate for the design of cold facilities in the limited space of reactor beam holes.

Comparative spectra for para-hydrogen, ortho-para-hydrogen, and solid H_2O are again presented in Fig. 24, normalized such that the integrated flux is the same for the four experimental spectra. It is seen that para-hydrogen has a peak in the distribution at very nearly the energy 0.0032 ev, corresponding to the value of kT for $37^\circ K$, and, further, that over the range of energies 0.004 ev down to 0.0002 ev the spectrum follows very closely the form of an ideal Maxwellian distribution, despite the lack of complete thermal equilibrium between the neutrons and the scattering medium. The theoretical curve shown has been adjusted only in amplitude to fit the experimental peak. Above 0.004 ev the experimental points deviate from the theoretical curve somewhat more than can be accounted for by the $1/E$ slowing-down distribution, indicating that thermalization is not complete, however. The spectrum from ortho-para-hydrogen does not follow accurately a Maxwellian distribution near the peak, although its behavior for higher energy is similar to that for para-hydrogen. The conclusion is that thermalization is incomplete for both para-hydrogen and ortho-para-hydrogen. By contrast with the spectra from hydrogen, for the solid H_2O spectrum neither the energy peak intensity nor the shape corresponds to the expectation for thermal equilibrium at $37^\circ K$. Also, the spectra for solid H_2O at $4^\circ K$ and $20^\circ K$ are identical within the rather small statistical error. It must be concluded that the failure of the neutron population to attain as low energy as in the case of hydrogen is

Fig. 23--Energy spectra of neutrons scattered from H_2O at $295^\circ K$ and H_2 at $20^\circ K$; energy distribution is independent of moderator size and intensity is only slightly dependent

Fig. 24--Energy spectra of neutrons scattered from liquid para-hydrogen and liquid ortho-para-hydrogen at 20°K and solid H_2O at 20°K and 4°K, normalized to the same integrated flux from the moderators; theoretical Maxwellian distributions for 37° and 295°K are shown

mostly due to the absence of low-energy vibrational states in the solid H_2O to provide a mechanism for phonon emission by neutrons below a certain energy, and that at 20°K the process of scattering transitions to higher energy by phonon absorption is already sufficiently suppressed that it has no appreciable effect on the energy spectrum. For a solid H_2O cold-neutron source, there is, therefore, no gain in reduction of the temperature below 20°K or probably below 40°K .

Figure 25a shows the spectra from para-hydrogen, ortho-para-hydrogen, and H_2O , all at 20°K , normalized to the same incident fast neutron flux. Sample holder 4 in Fig. 6 was used with H_2O in the bottom section. Figure 25b shows the spectra from the surface of the moderator. Effort was made to obtain experimentally comparative spectra under conditions to give correct relative intensities. The neutron intensity from the interior of para-hydrogen and ortho-para-hydrogen is similar for all neutron energies greater than about 10^{-2} ev. This result is rather expected since the total cross section for each is about the same for energies greater than 0.025 ev. For energies below 0.015 ev, the ortho-para-hydrogen cross section is much larger than that for para-hydrogen. One expects that the neutron spectrum from ortho-para-hydrogen should show an increased number of slow neutrons for these lower energies. Experimentally, the ortho-para-hydrogen specimen gives a more copious supply of neutrons, reaching a maximum at about 2.5×10^{-3} ev, where the intensity is larger by about a factor of 2 than that from the spectrum for para-hydrogen. Both spectra decrease for still lower neutron energies. At the lowest energy observed (about 2×10^{-4} ev) the ortho-para-hydrogen spectrum slowly approaches the para-hydrogen spectrum and is about 1.5 times as intense at this energy.

The experimental data for para-hydrogen and ortho-para-hydrogen can be compared in another, illuminating, manner. The spectra from the outer surface and re-entrant hole in a specimen of para-hydrogen are very similar, being less than 10% more intense from the re-entrant hole. The

Fig. 25a -Energy spectra of neutrons scattered from liquid para-hydrogen and liquid ortho-para-hydrogen at 20°K and solid H_2O at 20°K, normalized to the same incident flux of fast neutrons; the spectra are from the interior of the specimen

Fig. 25b--Energy spectra of neutrons scattered from liquid para-hydrogen and liquid ortho-para-hydrogen at 20°K and solid H_2O at 20°K, normalized to the same flux of fast incident neutrons; the flux is from the surface of the moderator in each case (it is to be noted that the flux from the interior and surface of liquid para-hydrogen is approximately the same)

spectrum from a re-entrant tube in a specimen of ortho-para-hydrogen is about a factor of 2 greater than that for para-hydrogen. However, the spectrum from the surface of a 6-in. -diam specimen is about a factor of 2 less than that for para-hydrogen. The conclusion is that geometrical shapes have a decided effect on the intensity of neutron production for specimens of ortho-para-hydrogen, but a much smaller effect for para-hydrogen.

Since it was observed that the spectrum of neutrons moderated by liquid para-hydrogen had a peak at 0.0032 ev and that the flux of neutrons with lower energy was substantially increased over that from H_2O at $4^\circ K$, it seemed useful to investigate further improvements which might accrue from the use of liquid para-hydrogen at temperatures lower than its boiling point. A sample holder similar to holder 5 shown in Fig. 6 was used which permitted successive measurements on liquid hydrogen at 20° , 11° , and $4^\circ K$. The results are shown in Fig. 26 as Curves 1, 2, and 3, respectively. The curves for temperatures at 20° and $4^\circ K$ were obtained with identical-size specimens. The curve for liquid hydrogen at an intermediate temperature was obtained for a much larger amount of liquid para-hydrogen. Since the relative intensities are consequently meaningless, the curves have all been normalized to the same total number of neutrons in the spectra. From the curves, it is clear that the colder liquid hydrogen gives an increased flux for energies below 0.002 to 0.003 ev, of the order of a factor of 2. An important point to note from the above curves is that solid para-hydrogen is a poorer moderator than liquid para-hydrogen. This, of course, is to be expected from the fact that the Debye temperature is higher for the solid. Consequently, there are fewer lower-energy levels from which the neutrons can scatter in the moderation process. From the results shown here, one can conclude that liquid hydrogen is a better moderator for cold neutrons than solid hydrogen and enhancement of the cold neutron flux (<0.001 ev) is greatest for liquid hydrogen just above its melting point.

Fig. 26--Energy spectra of neutrons scattered from liquid para-hydrogen at 20° , 11° , and 4° K, normalized to the same integrated flux from the moderator

NEUTRON SPECTRA FROM LIQUID AND SOLID METHANE

Another substance frequently discussed as a good possibility for low-temperature neutron moderators is methane (CH_4), either liquid or solid. The group at the EURATOM Laboratory at Mol has reported good low-temperature properties for methane. In order to check these results and to compare them with the present results, spectra runs were made with pure liquid and solid methane, using holder 3 in Fig. 6. The spectrum measurements were made for liquid methane at its normal boiling point (111.5°K) and at 20°K . Figure 27 shows the resulting spectra for para-hydrogen and solid methane normalized to the same number of incident fast neutrons. It is to be observed that the spectrum from the re-entrant hole in solid methane at 20°K has its peak energy at lower energy than para-hydrogen. For energies near its peak, the intensity of neutrons is at least a factor of 2 greater than from para-hydrogen. However, it is important to note that the relative intensity from the surface of the methane volume is lower than that from para-hydrogen. Figure 28 exhibits the spectra from the surface and interior of liquid and solid methane. Cooling methane from 111° to 20°K is necessary before methane can compete with para-hydrogen as an effective cold-neutron source. The use of methane in preference to liquid hydrogen as a cold-neutron source requires the use of a re-entrant hole geometry and a refrigerant such as liquid hydrogen.

NEUTRON SPECTRA FROM POLYETHYLENE AND LIQUID HELIUM

Since polyethylene (CH_2) is frequently used as an effective neutron moderator at room temperature, it is reasonable to study the production of cold neutrons by thermalization in cold CH_2 . Figure 29 displays unnormalized experimental spectra from a slab 12 by 6 by 2 in.³ of CH_2 at $\sim 77^\circ\text{K}$ and from the interior of a 2-in.-diameter CH_2 cylinder at 4°K . Theoretical curves, also unnormalized, are drawn for 4° , 80° , and 300°K . One observes that the spectrum from the slab at 77°K

Fig. 27--Energy spectra from solid CH_4 at 20°K, liquid ortho-para-hydrogen, and liquid para-hydrogen, normalized to the same flux of incident fast neutrons; the spectrum from the surface of solid methane is also shown for comparison

Fig. 28--Energy spectra of neutrons scattered from CH₄, both liquid and solid, and from the interior and surface of the moderator; normalization was for the same flux of incident fast neutrons

Fig. 29--Energy spectra of neutrons scattered from polyethylene (CH₂) at 4°K and 77°K; also shown are theoretical spectra for 4°, 80°, and 300°K (it is to be noted carefully that the experimental and theoretical curves are all plotted on arbitrary scales)

is not in thermal equilibrium, but it does have a substantial peak intensity at about 0.010 ev. More spectacular, however, is the complete lack of thermal equilibrium for 4°K. The experimental spectrum has a peak at more than ten times the expected energy of about 0.0003 ev. The failure to produce thermal equilibrium in low-temperature solid CH₂ is due undoubtedly to the same lack of low-lying vibrational levels as in solid H₂O. It is more pronounced than in this case, however, probably due to the more tightly bound, complex molecule.

V. INELASTIC SCATTERING OF NEUTRONS

Inelastic scattering measurements, which provide direct evidence on the dynamic nature of neutron interactions, have been exploited in a number of fields as evidenced by the large variety of papers in a recent symposium.⁽¹⁷⁾ A number of measurements using the inelastic-scattering techniques reveal information directly bearing on moderators. Some very impressive techniques have been used to study features of one or two prominent neutron moderators. The Harwell group has studied the scattering in water and graphite in considerable detail. Pertinent information on a number of other useful scatterers has not as yet been measured in detail. No measurements on moderators at low temperatures, have, so far, been reported by other workers. A significant portion of the effort in this program is devoted to obtaining information on inelastic interactions in hydrogenous materials, including H_2 , CH_4 , H_2O , and metal hydrides.

EXPERIMENTAL ARRANGEMENT

Two rather different techniques were applied to the measurement of inelastic scattering. For one, modifications of the time-of-flight instrumentation were used; for the other, measurements were made using a monoenergetic beam from a reactor.

Linear Accelerator Measurements

The inelastic-scattering measurements performed at the linear accelerator were variations of the usual time-of-flight technique. As has been pointed out earlier (Section II), the initial pulse of neutrons is of the order of 50 to 100 μ sec. For the neutrons of interest a flight time of the order of a few thousand microseconds is arranged through the selection of a proper flight path. One has a choice as to the location of the scattering

sample. It can be placed near the source or near the detector, depending on the kind of measurement to be required. If the neutron beam is filtered through a cold beryllium-oxide block to give only neutrons with energy less than a few millivolts and if the scatterer is placed close to the source, then measurements of neutron-energy gain can be made as follows. Those slow neutrons which scatter elastically in the sample arrive at the detector after a long flight time. Neutrons which have gained energy in the scatterer will arrive at the detector at an earlier time. The separation of the times of arrival of those neutrons scattering elastically and with energy gain depends on the flight path between the detector and the scatterer. For these measurements, this distance is made as long as possible compared to the source scatterer distance. The sketch in Fig. 30 shows an arrangement which accomplishes this and which was used to measure the inelastic energy-gain scattering in polyethylene, in water, and in para-hydrogen.

The inelastic-scattering measurements require the largest possible beam intensity for satisfactory statistics, because only very thin scattering samples are used in order to reduce multiple scattering in the sample. The specimen thickness is chosen sufficiently thin to scatter only about 10% of the incident neutrons. Of the more or less isotropically scattered neutrons, only a small fraction fall in the solid angle subtended by the detector, usually less than one steradian. The beam of neutrons reaching the detector is usually less than 10^{-2} times the incident beam. In view of the fact that the General Atomic linear accelerator is being increased in beam current by perhaps as much as a factor of 10, only preliminary measurements were made during this contract period with the lower-power accelerator. In order to use the lower-power accelerator as efficiently as possible, several procedures were used to increase the flux of slow neutrons arriving at the scattering target. In Section II, the procedure has already been described by which the optimum cold-source location was chosen, using activation foils to study both fast- and slow-neutron distributions around the electron target. The largest possible beam size (4×12 in.²)

Fig. 30--Time-of-flight distribution of neutrons scattered with energy gain from H_2O and CH_2 , both at room temperature

was used in order to increase the available neutrons.

Reactor Measurements

The arrangement shown schematically in Fig. 31 was used to obtain a monoenergetic beam of neutrons from a reactor by use of a crystal monochromator. A rotating mechanical chopper (also used as described earlier in connection with the linear accelerator cross-section measurements) interrupted the beam periodically to give pulses of about 20- μ sec duration. After scattering from the thin specimen, the neutrons drift to the bank of BF_3 counters. A time-of-flight analyzer provides data which permits calculation of the energy spectrum of scattered neutrons. This technique was used for the study of inelastic scattering in a number of metal hydrides.

ENERGY-GAIN SCATTERING IN H_2O AND CH_2

The resultant time-of-flight spectrum for room-temperature water and polyethylene after subtraction of background is given in Fig. 30. In both cases the scattered spectrum clearly consists of an elastically scattered component represented by a peak beginning at about channel 100 and an inelastically scattered component represented by the counts in channels beginning at about number 40 and extending through the region of the elastic peak. At times shorter than channel 40, inelastic scattering may also be present, but it is difficult to measure in the presence of a relatively high background. It has been observed throughout the program that water and polyethylene differ significantly, and this fact is confirmed in more detail in the present measurements, which showed a more sharply defined peak of inelastic scattering as well as a higher elastic peak than for the case of water. In both cases it is seen that the elastic and inelastic spectrum are not completely resolved and separated as would be the case if the incident neutron beam were monoenergetic and if there were no low-energy states for exchange of small quanta of energy. The overlap of these two spectra, however, does not imply the existence of very low energy states, but

Fig. 31--Arrangement using a reactor neutron beam and crystal monochromator technique to provide a monoenergetic beam of neutrons; a rotating chopper provides a pulse of neutrons which is then scattered from the sample and drifts to the detector; inelastic-scattering gain measurements were performed with this apparatus

rather the fact that the incident spectrum includes all energies below 0.005 ev, allowing those of lower energy which take longer flight time from the source to the scatterer to appear in channels near the position of the Bragg cutoff.

ENERGY-GAIN SCATTERING IN PARA-HYDROGEN

Another measurement of particular interest was made to determine the elastic or inelastic nature of the scattering from liquid hydrogen. One experimental difficulty in such measurements is that the liquid hydrogen must, of course, be contained inside the neutron cryostat. Although it has been designed for minimum attenuation of neutron beams, it nevertheless includes multiple aluminum walls and a stainless steel inner wall 0.015 in. thick. Scattering for these surfaces is not negligible in comparison with the very thin scattering samples necessary to eliminate multiple hydrogen scattering. A second difficulty is that only one such cryostat is presently available, and when it is used to contain the scattering sample, it is no longer possible for use as a liquid-hydrogen source to get high intensity of low-energy neutrons. A neutron source of polyethylene (77°K) has therefore been substituted. A third, and much more serious, difficulty is in the nature of the liquid-hydrogen scattering sample. As indicated in Section III, the scattering cross section of liquid para-hydrogen is very low in the region of 0.005 ev, and therefore a sample of the order of several millimeters thickness must be used to get scattered intensities equivalent to the 0.25-mm thickness of liquid H_2O and CH_2 . The background, however, derives from scattering of random neutrons, mostly of higher energy in the sample, and for these higher-energy neutrons (0.012 ev and higher) the scattering is emphasized in comparison with the scattered intensity from the incident filtered beam. Furthermore, these higher-energy neutrons, having scattered in the sample, lose energy and approach the energy of the filtered beam. For these reasons, background has proved much more difficult to

eliminate in the case of para-hydrogen inelastic-scattering measurements.

Figure 32 displays the results of an inelastic-scattering measurement where the scattered spectrum from 20°K liquid para-hydrogen is compared with the spectrum from CH₂ at 295°K. These data appear to indicate at least some elastic scattering from the liquid hydrogen, although considerably weaker than that from CH₂. The data are not sufficient to draw firm conclusions about the inelastically scattered component from liquid para-hydrogen, but appear to indicate a substantially reduced inelastic-scattering component compared to that from room-temperature CH₂. This was to be expected since the low-temperature H₂ molecules were not expected to lose substantial additional energy to the incident neutrons. Further measurements with the higher-beam-current linear accelerator, either with a crystal monochromator before the scattering sample or a large-aperture mechanical chopper near the scattering sample must be used to provide accurate information on the detailed nature of energy-gain scattering of neutrons from liquid hydrogen.

TECHNIQUE FOR MEASURING ENERGY-LOSS SCATTERING

Inelastic-scattering measurements in which the incident neutron loses energy are also of importance in the study of a number of hydrogenous materials. For the case of low-energy incident neutrons (of the order 0.005 eV or less), the usual slow-chopper techniques employing crystal-filtered neutrons cannot be used because of the lack of monoenergetic incident neutrons. An adaptation of the linear accelerator time-of-flight technique can be used to obtain information on such energy-loss scattering. For these measurements, the flight path between the neutron source and scatterer is made long compared to the flight path from the scatterer to the detector. Position B in Fig. 1 shows a typical arrangement for the source scatterer and detector. In operation this arrangement works as follows. The neutrons arriving at the scatterer are energy-analyzed

Fig. 32--Time distribution of neutrons scattering with energy gain from polyethylene at 295° K and from liquid para-hydrogen

by virtue of the long time of flight. After scattering, the neutrons travel a very short distance to the detector, arriving at the detector within a short time after their scattering, quite independent of whether they have scattered elastically or inelastically. It is only necessary to choose the channel width for the time-of-flight analyzer as approximately equal to the average flight time for neutrons from the scatterer to detector. If one now places an energy-dependent filter (gadolinium or boron) between the scatterer and detector, it will be possible to measure for each time channel the average energy of the neutrons. Comparing for each channel the transmission obtained in this way with the transmission obtained by placing the filter between the source and scatterer, one can evaluate the average energy change for the neutrons in each time channel. Although this measurement does not measure the energy spectrum of the scattered neutrons for each incident monochromatic energy group, it does give a significant amount of information on the magnitude of energy changes. These values are of particular significance for small energy changes such as are involved with incident neutrons of a few millivolts.

The analysis of the data obtained with the above arrangement proceeds as follows. Let T_1 be the transmission of the energy-dependent filter when placed between the source and scatterer and T_2 be the transmission of the same filter when placed between the scatterer and detector. Then

$$T_{1(i)} = \exp [-nx\sigma(E_1)_i] , \quad (6)$$

where i is the time channel, n is the number of nuclei per cubic centimeter, and $\sigma(E_1)$ is the cross section for neutrons with energy E_1 . A similar formula holds for the filter placed in the second position, where E_2 is the average energy of the scattered neutrons. The ratio of the transmission before and after the scatterer is given as follows:

$$\gamma_i = \left(\frac{T_i}{T_2} \right)_2 = \frac{\exp [-nx\sigma(E_1)]}{\exp [-nx\sigma(E_2)]} = \exp \{-nx[\sigma(E_1) - \sigma(E_2)]_i\} . \quad (7)$$

A value of $\gamma \cong 1.0$ indicates elastic scattering with no energy loss, whereas values of $\gamma > 1$ correspond to energy loss in the scattering. Taking logarithms of both sides of Eq. (7) we obtain

$$\ln \gamma_i = -nx [\sigma(E_1) - \sigma(E_2)]_i. \quad (8)$$

In Eq. (8), γ is an experimentally determined ratio of the transmissions for each channel. The value nx is known from the concentration of absorber in the filter, and $\sigma(E_1)_i$ is known from the characteristics of the filter as a function of energy. It is then possible to determine from $\sigma(E_2)_i$ and Eq. (6), the value of E_2 , the average energy of the scattered neutrons. Then one can evaluate the logarithmic energy decrement, ξ , defined as

$$\xi = \ln \frac{E_1}{E_2}. \quad (9)$$

The reader should note that ξ is usually integrated over all angles of scattering and is usually used for neutrons of intermediate energy for which $\xi \leq 1$. The use of ξ for a single scattering angle and for those cases where the incident neutron energy is reduced to a very low value may not be particularly meaningful.

SCATTERING WITH ENERGY LOSS FOR PARA-HYDROGEN AND CH₂

The experimental data obtained from inelastic-scattering measurements for liquid para-hydrogen and CH₂ at 20°K are displayed in Fig. 33, where γ is plotted as a function of incident energy. It is observed that through the energy range from 0.008 to about 0.045 ev the value for γ is about 1.00 for polyethylene. On the other hand, γ for liquid para-hydrogen shows a significant rise between energies of 0.013 and 0.016 ev and continues at about 1.20 for energies up to 0.045 ev, the highest measured. Additional data not shown in this graph indicate that the value of γ for liquid para-hydrogen for considerably lower energies may be of the order

Fig. 33--Variation of gamma with incident neutron energy for liquid para-hydrogen and polyethylene at 20°K; gamma is the ratio of the transmission of an energy-dependent filter before and after scatter in the sample; large values of gamma signify significant energy loss in the scattering process and $\gamma = 1.0$ signifies no energy exchange in the scattering process

of 1.10, whereas that for polyethylene as well as for vanadium, an incoherent, heavy-nucleus scatterer, is about 1.0 for the same low-energy regions.

The data in Fig. 33 are to be interpreted in the following way. The value of γ approximately equal to 1.00 for polyethylene for energies between 0.008 and 0.030 ev indicates no energy gain or loss for neutron scattering on polyethylene, as one expects from hydrogen bound in solid polyethylene. The slight increase in γ for polyethylene between 0.030 ev and 0.045 ev indicates the beginning of energy loss for neutrons scattering in polyethylene. The curve of γ for liquid para-hydrogen giving $\gamma = 1.10$ for the lowest energies exhibited in Fig. 33 indicates that even for these low energies, para-hydrogen molecules scatter neutrons with energy loss for the incident neutron. The rapid rise between the energy 0.013 ev and 0.016 ev for liquid para-hydrogen indicates the excitation due to the transition of the $J = 1 \rightarrow J = 0$ for the ortho-para-hydrogen system. That γ continues to be fairly large, of the order of 1.2, for energies up to 0.045 ev indicates a substantial energy loss for neutrons with this incident energy. The conclusion is that para-hydrogen, for all energy regions included in this study, scatters with larger energy loss than polyethylene. This conclusion is consistent with the observed neutron spectrum from polyethylene, which has a demonstrated scarcity of low-energy neutrons (Fig. 29).

Unfortunately, it is not possible to convert the data from Fig. 33 to quantitative values of energy loss because sufficient data at very low energies is lacking for evaluation of Eq. (6). This is a consequence of the preliminary nature of the run. The average energy for scattered neutrons extends to lower energies than were investigated. Only for the highest incident energy, 0.044 ev, can the low energy of the scattered neutron be evaluated. This gives $E_2 = 0.010$ ev. All observed incident neutrons of lower energy give lower average scattered energies. The analysis indicates that the value of E_2 for $E_1 \sim 0.016$ to 0.018 ev is less than for all slower and faster incident neutrons in the range 0.009 to

0.044 ev. This energy loss, interpreted as the excitation of the $J=0 \rightarrow J=1$ transition, provides an efficient moderating mechanism for enhancing the low-energy spectra at the expense of neutrons with energy of ~ 0.017 ev.

SCATTERING WITH ENERGY LOSS FOR WATER, POLYETHYLENE, GRAPHITE, AND VANADIUM

Inelastic scattering measurements have been made for water, polyethylene, vanadium, and graphite using the linear-accelerator technique discussed above. Figure 34 shows a plot of γ for vanadium and graphite as a function of energy. Within the accuracy of the measurements, there is some evidence that graphite shows a small energy loss per collision. Additional measurements have been made comparing water and polyethylene. Figure 35 shows a comparison of these data, giving the logarithmic energy decrement at 90° as a function of energy for these two substances. The difficulties in analyzing the data experienced in the preceding section were absent here. It is readily evident that the logarithmic energy decrement is considerably greater for H_2O . Throughout the program there has been continued evidence that polyethylene is a substantially poorer moderator in the energy region from thermal (0.025 ev) to higher energies of the order of 0.1 ev.

The values of ξ must be determined for all angles to derive the quantity usually used in reactor calculations. With the higher-current linear accelerator soon to be available, it will be possible to obtain better measurements for a variety of angles. Of even more value for reactor computations is the variation of $\xi\sigma$ as a function of energy. An earlier effort in this program provided a comparison of $\xi\sigma$ for H_2O and ZrH , shown in Fig. 36. The prominent effect on energy loss of the vibrational levels in ZrH is readily seen. It is also clear that H_2O is a better neutron moderator for neutron energies less than 0.13 ev. Similar results for other hydrogenous substances should soon be available from this program.

Fig. 34--Variation of gamma with incident neutron energy for graphite and vanadium at room temperature; gamma is the ratio of the transmission in an energy-dependent filter before and after scattering; the scattering in vanadium should be essentially elastic with no energy loss, giving values of 1.0 for gamma; the small positive values of gamma in graphite for all energies investigated here represent a small energy loss in the scattering process

Fig. 35--Energy dependence of the logarithmic energy decrement for CH_2 and H_2O for 90° scattering (part of the data for H_2O was taken at Brookhaven National Laboratory in an earlier phase of the program)

Fig. 36--Logarithmic energy decrement $\xi\sigma$ for neutron inelastic scattering from H_2O and $\text{ZrH}_{1.5}$ obtained by averaging over all scattering angles (the experimental data were taken at Brookhaven National Laboratory in an earlier phase of the present program)

ENERGY LEVELS IN SOME METAL HYDRIDES

The general treatment of the energy gain and loss by neutrons in solids has been given in a number of places. ^{(18)*} The general formulation shows that there is a considerable dependence of the cross sections on the specimen temperature. For the case of a sharp Einstein level, $h\nu$, which is several times kT , such considerations show that the cross section for energy gain can be increased by heating the sample. In other terms, this is understood as increasing the population of the Einstein levels. However, the observed levels would thereby be broadened by Doppler effects. This latter effect is not particularly troublesome because only moderate resolution was used in the present experiment. Consequently, all specimens were heated, but by not more than 100°C because of the potential fire hazard.

Experimental measurements were made in an earlier part of the program using the reactor technique described above. For each of the specimens studied, the time-of-flight spectrum of the scattered beam was compared with the spectrum of the incident beam scattered by a thin vanadium slab. From this latter distribution, the energy of the incident neutron was determined. Table 2 shows the experimental data obtained in this

Table 2
RESULTS FOR INELASTIC SCATTERING OF NEUTRONS
BY SOME METAL HYDRIDES

Material	Energy of Scattered Neutron(ev)	Energy of Phonon (ev)	Type of Bond
YH_x	0.190	0.122 ± 0.007	Transition
ZrH_x	0.199	0.130 ± 0.007	Transition
LiH	0.153	0.085 ± 0.005	Ionic
UH_3	0.164	0.095 ± 0.005	Bridge
TiH_2	0.197	0.129 ± 0.007	Transition

*See references cited in Ref. 18.

manner. It is interesting to note that the value for zirconium hydride agrees well with that determined previously from measurements of the total cross section and the scattering of cold neutrons.⁽¹¹⁾

In addition to the substances reported in Table 2, some others were investigated. Calcium hydride also shows evidence of an energy level. However, this run was interrupted before it was possible to accumulate sufficient data to compute the energy level. For the case of magnesium hydride, however, there was no evidence for a sharp energy level. This result was to be expected in view of the fact that the total cross section as a function of energy (see Fig. 17) also shows no effects which would be caused by a sharp Einstein level. With the above resolution, no evidence was found for a predominant hydrogen level in KH_2PO_4 . A subsequent preliminary measurement made by scattering cold neutrons from KH_2PO_4 has shown evidence for small peaks at 0.0098, 0.0144, 0.02, 0.043, and 0.124 eV superimposed on a broad continuum of acoustical transitions. The apparatus described above could not be expected to resolve this spectrum.

The development of the theory of the metal hydrides has not progressed sufficiently far at present to permit one to predict the value of the Einstein level for these various metal hydrides or the relation between the hydrogen bond and the energy level. Table 2 shows that several types of bonding are involved for these five specimens.^{(19)*} For the case of lithium hydride, however, a simple treatment[†] which relates the acoustical and optical frequencies with the measured⁽²⁰⁾ characteristic temperature of LiH and LiD (851°K and 638°K, respectively) can be used to evaluate the optical energy level. In this manner, one computes a reasonable value for the acoustical characteristic temperature (250°K) and a value of 0.093 eV for the optical energy. The latter is in agreement with the experimental value of 0.085 ± 0.005 eV.

* A discussion of the chemical bonds is given in Ref. 19.

† Developed by I. Pelah, Weizmann Institute, Rehovot, Israel, when he was participating in this program.

VI. DISCUSSION

From the results presented above, we may derive some conclusions about the dynamic behavior of a variety of hydrogenous substances and particularly conclusions relevant to the selection of optimum cold-neutron sources for use both at a reactor and a linear accelerator.

The dynamics of the hydrogen in molecules of H_2O , CH_4 , CH_2 , H_2 , and ZrH affect directly the interaction with neutrons. For neutron energies above 1.0 ev, the interaction is primarily with free atomic hydrogen. For lower energies, the effect of chemical binding is to increase the cross section per hydrogen atom for most hydrogenous substances from 20 barns per atom at high energy to about 80 barns per atom in the fully bound state. The presence of energy levels associated with bound vibrational states or with rotation modifies the resulting cross section markedly in a number of cases. In the absence of strong rotational or vibrational levels, the interaction with neutrons is mainly by a collision with mass M . From this consideration, a hydrogen molecule with atomic mass 2 should provide much larger energy exchange per collision (up to 8/9 of the incident energy) than H_2O and CH_4 molecules for which the energy exchange is less than 1/4 the incident energy. The latter assumes that H_2O and CH_4 molecules are not further associated with other molecules as is almost certainly the case for some solids. In these cases, the collision loss would be further reduced below that for H_2 molecules. It is likely that this is the reason for the lack of moderation in cold CH_2 , evidenced by the lack of cold neutrons in the spectrum from CH_2 at 4°K , even though the smaller mass for CH_2 should improve the moderation compared to solid CH_4 .

The presence of vibrational or rotational states significantly modifies the energy loss in neutron scattering for para-hydrogen, solid CH_4 , and ZrH . Inelastic-scattering measurements have not been made as yet for

liquid and solid CH_4 to locate such levels, and the total cross-section curve gives no evidence for them. However, the observed energy spectrum, with maximum at about the same energy as for para-hydrogen and ortho-para-hydrogen, indicates their presence. This is particularly obvious when comparison of the spectra is made with those from solid H_2O and CH_2 , both at 4°K . Inelastic scattering measurements have confirmed the strong inelastic level in liquid para-hydrogen at about 0.014 ev. This is almost certainly the same level indicated by the rapid change in total cross section for para-hydrogen starting at 0.013 ev, and is in agreement with the value of the energy difference between the ortho- and para- ground states as determined from specific heat measurements. A number of similar measurements made with ZrH reveal strong inelastic-scattering levels at multiples of 0.130 ev. These levels are observed also in the total cross section and are of importance in the use of ZrH as a reactor moderator.

Some conclusions follow for selection of a cold-neutron source. It can be seen from Figs. 25a and 25b that if the objective is to enhance flux of beryllium-filtered neutrons ($E \lesssim 0.005$ ev), the advantage of H_2 over H_2O is relatively small. The slight decrease in intensity might be acceptable in consideration of other factors, such as the possibility of cooling with recirculated helium and without safety precautions necessary for hydrogen. In design of an H_2O cold-neutron source, it is more important than for H_2 that the configuration be such that the observed neutron beam is emitted from a re-entrant hole extending to the interior of the moderator rather than from the surface, but the required thickness of the solid H_2O , which would determine gamma-radiation heating, need not be large. It is, of course, not clear that the use of solid H_2O would not introduce problems, such as radiation-damage effects by molecular dissociation, which are not present in liquid H_2 . Even though recirculated helium coolant were used, there appears to be no advantage in reducing the temperature below 20° to 30°K for an H_2O source. There also seems to be little reason to substitute D_2O for H_2O , since the same limitation on the spectrum because of the lack

of energy-exchange mechanisms would be equally applicable to D_2O .

It appears that the use of solid methane at $20^\circ K$ can yield a flux of neutrons with energies below 10^{-3} ev equal to the flux from a similar volume of liquid ortho-para-hydrogen and greater than for para-hydrogen. Further measurements yet to be performed may indicate a still larger improvement in the low-energy flux ($<10^{-3}$ ev) for temperatures as low as $4^\circ K$. However, the use of a re-entrant hole to view the interior of the source is even more important than for cold H_2O or ortho-para-hydrogen. Failure to provide this arrangement can reduce the intensity by a factor of 5. Methane then becomes much poorer than para-hydrogen. Furthermore, the use of CH_4 at $20^\circ K$ requires hydrogen or helium as a coolant. As with the use of solid H_2O in a reactor, radiation-damage effects may be serious for solid CH_4 .

With particular reference to the electron linear accelerator, the available intensity of slow neutrons depends on a compact geometrical arrangement in which the slow-neutron-emitting surface is as close as possible to the primary point source of fast neutrons, and the cold moderator must therefore be efficient in both the early stages of slowing down fast and epithermal neutrons and in the final stages of approach to thermal equilibrium. For this combined function an H_2O source may ultimately prove superior to H_2 for production of cold neutrons in the intermediate energy range of 0.002 to 0.005 ev. A second factor, relevant to pulsed accelerators, in which the cold source is useful, is time resolution. Whereas the emission, or "die-away," time of each neutron pulse may be undesirably long for a room-temperature moderator, especially for slow neutrons, it is much shorter for a $20^\circ K$ moderator. Even 0.005-ev neutrons are in the epithermal range and are emitted in a few microseconds. It is for this reason that in Fig. 23 the iron Bragg diffraction cutoff appears as a one-channel-wide discontinuity in the H_2 spectra, although it is not visible in the warm H_2O spectra. In these preliminary experiments it has been possible to attain intensities sufficient that even at very low energies around 0.0001 ev,

corresponding to about 30 Å wavelength, enough counts for good statistical accuracy can be registered in a period of the order of 10 to 30 min for measurement of complete spectra, such as those illustrated in Fig. 24. Quantitatively, the presently available flux through a 1-in. -diam, 24-in. -long collimating tube gives counting rates amounting to 100 counts/sec/mv energy range at energies near the maximum at 0.0026 ev, and gives 14 counts/sec/mv for 0.0001 ev, 30 Å neutrons. It appears that the accelerator cold-neutron source will be useful in further studies of the dynamics of the interactions of slow neutrons with liquids and solids.

VII. SUMMARY

1. In zirconium hydride the hydrogen atoms act as independent oscillators, as in the Einstein crystal model, having discrete "optical" energy levels at multiples of 0.130 ev.⁽¹¹⁾
2. The total neutron cross section of zirconium hydride follows rather closely the original Fermi theory, based on the independent oscillator model, of neutron scattering in hydrogenous media (Figs. 15 and 16).
3. Neutron spectra indicate that a strong energy level of the solid H_2O near 0.06 ev and weaker level near 0.02 ev are the mechanisms for neutron-energy loss (Fig. 20).
4. The total scattering cross section for H_2O shows no evidence for energy levels at 0.06 ev and 0.02 ev nor for the weaker reported levels at 0.008, 0.005, and 0.0005 ev (Fig. 12).
5. A strong energy level at about 0.014 ev in liquid para-hydrogen is indicated both by a sharp break in the energy spectrum after multiple scattering and by energy loss on single scattering (Figs. 20 and 33).
6. The total cross section of liquid para-hydrogen exhibits a corresponding sharp minimum of about 3 barns at 0.013 ev, representing the threshold for excitation (Fig. 10). The low total cross section below 0.013 ev is proportional to $1/v$ and derives from coherence between the antiparallel protons in para-hydrogen.
7. The 0.014-ev energy level is interpreted as corresponding to the transition from the $J = 0$ ground level of para-hydrogen to the $J = 1$ ground rotational level of ortho-para-hydrogen, with spin reversal of the scattered neutron.

8. Consequently, no equivalent energy level is to be expected in liquid ortho-hydrogen, although it is still observed in the 3:1 ortho-para-hydrogen mixture, which represents the closest approach to pure ortho-hydrogen feasible to prepare. Total cross sections of ortho-para-hydrogen give no evidence of energy levels (Fig. 10).
9. Although 0.014 ev is the lowest rotational excitation of liquid hydrogen, evidence of inelastic scattering at lower energies indicates that energy exchange with scattered neutrons approximates the characteristic of a gas of free molecules of mass 2.
10. No evidence for low-energy levels in either CH_2 or CH_4 is found in the cross sections or in discontinuities in the spectra; however, the low energy of the spectrum from CH_4 indicates the presence of low-energy levels. Both the higher-energy spectrum and the absence of inelastic scattering indicate the absence of such levels in polyethylene, CH_2 .
11. Moderation in H_2O at 20°K does not produce an equilibrium spectrum, but one for which the intensity is maximum at about 0.004 ev and which drops off more rapidly than a Maxwellian spectrum at lower energies.
12. Further cooling of the solid H_2O moderator from 20°K to 4°K causes negligible change in the energy spectrum.
13. The energy spectrum of neutrons from a linear accelerator after moderation in liquid para-hydrogen at 20°K , as determined by time-of-flight analysis, approximates thermal equilibrium for temperature 37°K and energy $kT = 0.0032$ ev over the range 0.0001 to 0.004 ev, but includes an appreciable component of epithermal neutrons from 0.004 to 0.02 ev. The source was about 6 in. in diam and 6 in. high.
14. Liquid ortho-para-hydrogen mixture gives approximately the same spectral distribution as pure para-hydrogen, but with

higher intensity, as expected from the higher ortho-hydrogen cross section.

15. Cooling para-hydrogen below its boiling point (20°K) to 11°K results in more low-energy neutrons. Solid para-hydrogen at 4°K is a poorer moderator than liquid hydrogen at either 11°K or 20°K .
16. Moderation in CH_4 at 20°K produces an approximate thermal equilibrium for an apparent temperature 30°K and an energy $kT = 0.0025$ ev. As in the case of H_2O at low temperature, an appreciable number of epithermal neutrons are included from 0.004 to 0.020 ev.
17. Neutron flux over the range 0.0001 to 0.004 ev from liquid para-hydrogen and ortho-para-hydrogen exceeds by a factor of 50 to 100 the flux from an equivalent volume of H_2O at room temperature (Fig. 24).
18. The flux from H_2O and ortho-para-hydrogen at 20°K is strongly dependent on the geometrical arrangement of the moderator, being higher and also at lower energies from a re-entrant hole than from the outer surface of the moderator. This dependence is still greater in CH_4 , but is almost negligible for liquid para-hydrogen (Figs. 25a and 25b).
19. The relative intensities from the various moderators therefore depend on the configuration. Roughly, it is indicated that for a linear-accelerator source, the intensity at 0.00025 ev is about the same for an ortho-para-hydrogen mixture and for solid CH_4 at 20°K , but lower by a factor of 2.4 for solid H_2O ; however, the total flux of neutrons of energy less than 0.004 ev is nearly the same for water.
20. These relative intensities may be different for a cold-moderator reactor source surrounded by an isotropic source of room

temperature "thermal" neutrons, than for the cold-moderator accelerator source studied here, where irradiation is from an adjacent point source of fast neutrons.

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