

## The Production and Properties of Low Temperature Neutrons

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Evidence has been found that (a) hydrogenous substances so cold that nearly all the molecules are in the ground state can cool only those neutrons whose kinetic energies are great enough to excite the molecules to their first state, e.g., the temperature of neutrons cooled by a crystal of such molecules at the absolute zero will approximate the temperature at which the specific heat begins to rise above the normal curve of the Debye type for crystalline specific heat; (b) the probability of energy loss to a liquid or crystal as a whole seems to be small; (c) for hydrocarbons, the limiting temperature for cooling neutrons appears to decrease with increase in length of the hydrocarbon chain. A neutron thermometer, using the absorption by boron, has been tested and calibrated approximately.

### INTRODUCTION

THE evidence reported by many investigators,<sup>1-7</sup> that neutrons emerging from thick layers of hydrogenous matter have approximately thermal energies, suggested the importance of measuring neutron temperatures.

It is necessary that the qualified way in which the term "temperature" of neutrons is used in this discussion be made clear. The processes of absorption of the slower neutrons and of production by slowing of the fast neutrons both act to distort the velocity distribution from the Maxwellian. Velocity selector experiments (reference 6) and data for lower temperatures presented below both reveal this. The term "neutron temperature" is used only to specify the ap-

proximate average neutron energy. Some of the results below were reported briefly several months ago.<sup>8, 9</sup>

### PRELIMINARY EXPERIMENTS

Among the first experiments tried was an attempt to use the gamma-radiation produced by the capture of slow neutrons by protons in paraffin as a means of studying the rate of capture and therefore the average velocity of the neutrons. The assumption that the probability of capture is inversely proportional to the velocity of the neutron (the "1/v" law of capture), should then allow a study of the variation of neutron velocity to be made. The arrangement and apparatus are shown in Fig. 1, and the data in Table I.

At a given temperature, counts were taken, first with the Dewars empty, and second with paraffin plugs (3.5 inches in diameter and 16 inches long, coated with sufficient H<sub>3</sub>BO<sub>3</sub> to prevent escape of slow neutrons) in place inside each of the Dewars. The control experiment with CCl<sub>4</sub> gives an upper limit to the effect of scattering of gamma-radiation from the source and leads to a prediction of approximately 25 counts per minute as the effect to be expected at 25°K, if the capture production of gamma-radiation were to vary as the reciprocal of the square root of the absolute temperature. The data show quite clearly that the effect is much smaller. This apparent temperature independ-

TABLE I. *Data on temperature variation of gamma-radiation from capture of slow neutrons in paraffin.*

DIFFERENCE IN COUNT RATE, WITH AND WITHOUT PARAFFIN PLUGS. (Counts/Minute.)	TEMPERATURE
9.2 ± 1.5	300°K
8.2 ± 2	90°K
8.0 ± 2	25°K
WITH AND WITHOUT CCl <sub>4</sub> INSTEAD OF PARAFFIN	
2.4 ± 1	300°K

<sup>1</sup> Dunning, Pegram, Mitchell and Fink, Phys. Rev. **47**, 888 (1935).

<sup>2</sup> Moon and Tillman, Nature **135**, 904 (1935).

<sup>3</sup> Dunning, Pegram, Fink, Mitchell and Segrè, Phys. Rev. **48**, 704 (1935).

<sup>4</sup> Westcott and Niewodniczanski, Proc. Camb. Phil. Soc. **31**, 617 (1935).

<sup>5</sup> Moon and Tillman, Proc. Roy. Soc. **A153**, 476 (1936).

<sup>6</sup> Fink, Phys. Rev. **50**, 738 (1936).

<sup>7</sup> Preiswerk and von Halban, J. de phys. et rad. **7**, 29 (1937).

<sup>8</sup> Libby and Long, Phys. Rev. **50**, 575 (1936).

<sup>9</sup> Libby and Long, Phys. Rev. **50**, 576 (1936).

ence of the capture can be understood as evidence that even at 300°K a large fraction of the thermal neutrons produced are captured in so large a block of paraffin, eliminating the possibility of any large increase on cooling. This agrees with the considerations of Fermi<sup>10</sup> on the diffusion of thermal neutrons in paraffin. Calcula-

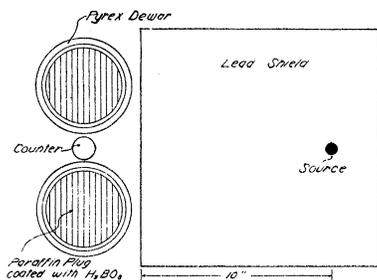


FIG. 1. Paraffin neutron-proton gamma-radiation assembly.

lation for the arrangement used, assuming the "1/v" law of capture, gave 46 percent as the expected increase of gamma-radiation intensity on cooling from 300°K to 25°K, in agreement with the measured effects within the rather large limits of error. Consequently, it was concluded that any measurement of temperature effects must be accomplished by observing effects produced in a small region of the larger system used to cool the neutrons, e.g., activation of a target imbedded in the system or preferably exposed to the neutrons emitted from the surface of such a system. Some of the later experiments further illustrate this point.

#### TEMPERATURE CURVES FOR DIFFERENT HYDROGENOUS MOLECULES

The apparatus finally selected for comparison studies of several different hydrogenous molecules is shown in Fig. 2. A metal Dewar of 5.25 inches inside diameter, with inner wall of brass and outer of monel metal, each about one millimeter thick, was surrounded with a paraffin cylinder of 10 cm wall thickness. The neutron source, either radon-beryllium (60–210 millicuries) or radium-beryllium (200 millicuries) was placed in the paraffin jacket as shown. This jacket, surrounding the whole Dewar, produced slow neutrons which diffused into the Dewar space, activating the silver target. Inside

the metal Dewar a soft glass Dewar was mounted to the Dewar cap to serve as protection for the silver target cylinders which were dropped into place inside the surrounding can of cold hydrocarbon. The walls of the soft glass Dewar absorbed about seven percent of room temperature neutrons. The copper can used to hold the cooling hydrocarbon in position had 0.3 mm walls, thick ends (approximately 1 mm), and a set of radial vanes inside to facilitate the establishment of temperature equilibrium. Thirteen hundred cubic centimeters was the total volume. When in position, at least ninety percent of the neutrons activating the target passed through the can. In order to estimate this fraction, the can was filled with 3.7 molal  $\text{CdCl}_2$  solution, which diminished the activity by eighty percent. Half of the residual twenty percent was not absorbed by cadmium.

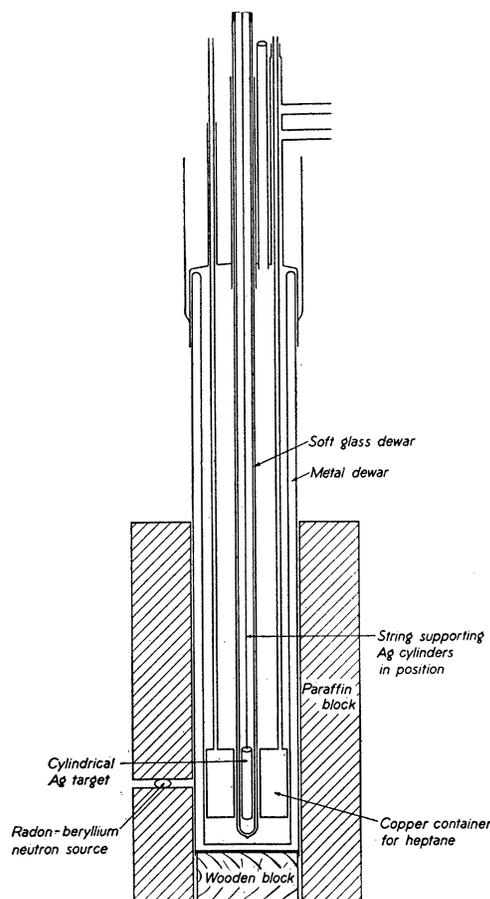


FIG. 2. Large Dewar assembly.

<sup>10</sup> Fermi, *Ricerca Scient.* 7, 13 (1936).

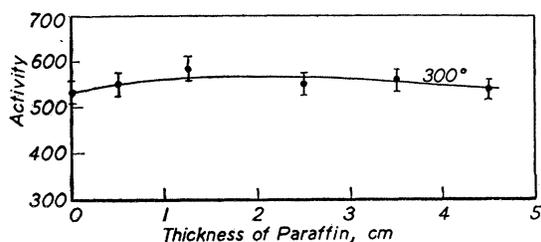


FIG. 3. Effect of introduction of paraffin cylinders of different thicknesses on activity of target.

The procedure used was to activate the silver cylinder (11 cm long, 1 cm in diameter, and approximately 0.1 mm thick) for 7 minutes, with the hydrogenous substance at a temperature constant within one or two degrees during the activation period. The Dewar contents were generally cooled with either liquid air or liquid hydrogen, and allowed to come to thermal equilibrium. The cooling liquid was then removed and silver activations were taken as the contents slowly warmed. Since the temperature drift in the important low temperature range was considerably greater than at higher temperatures, an accurate point at 20°K was obtained by making activation runs while the liquid hydrogen was still surrounding the can (as shown by the thermocouples), and then extrapolating the activity data to the time the last hydrogen left. Several runs from 20° to 300° gave enough data to average over each interval of a few degrees. The target activities were measured with a solid-wall tube counter for the interval of nine minutes between the end of the second and the beginning of the twelfth minute after the end of the activation period. This assured that the effects measured were mainly due to the 2.3 minute silver activity. For purposes of comparison a theoretical curve which was calculated on the assumption that all neutrons captured were absorbed by "1/v" law nuclei is shown with each set of data. The expression for the curve is

$$\text{Activity} \sim e^{-\gamma T^{-1}}(1 - e^{-0.77 T^{-1}}), \quad (1)$$

where  $T$  is the absolute temperature and 0.77 was determined by the dimensions of the target and the absorption cross section of silver for neutrons exciting the 2.3-minute period. This treatment obviously cannot be expected to be

rigorous because it assumes that the neutrons pass through the target only once. However, at the lower temperatures where the absorption cross sections are larger, it might be expected to be more rigorous. The smeared velocity distribution resulting from the capture and creation processes introduces an additional reason for this relation's being only approximate. The constant  $\gamma$ , presumably measuring the thickness of hydrogenous material traversed on

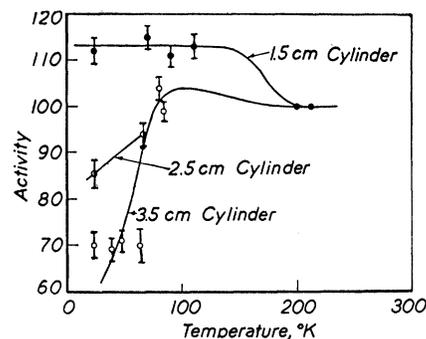


FIG. 4. Temperature effects with different thicknesses of cold paraffin.

the average by the neutrons after they have been cooled, is fitted to the data in each case.

#### DATA FOR PARAFFIN

Paraffin was studied by replacing the copper can shown in Fig. 2 with paraffin cylinders of the same height and various thicknesses. At room temperature, introduction of the paraffin cylinders inside the Dewar altered the activity of the target very little, as shown in Fig. 3. The data for the cold paraffin cylinders are shown in Fig. 4. These effects are similar to those reported by Moon and Tillman, but somewhat smaller, presumably because the neutrons were able to pass through the target several times. As shown later, the effect of such multiple passage would tend to decrease the temperature effects.

The general character of the curves in Fig. 3 suggests that the initial rise on cooling is due to the effect of slowing the neutrons on the capture probabilities for both the silver target (2.3 minute period) and the paraffin protons, the two capture effects being so balanced as to lead to an initial rise in target capture which is more

than compensated at lower temperatures by capture by the protons and soft glass Dewar. The paraffin temperatures were measured by two copper-constantan thermocouples imbedded at the top and bottom of the cylinders. The temperatures differed by less than two degrees during the runs and their mean was used.

#### DATA FOR *n* HEPTANE

Normal heptane was selected for careful study because it has a straight chain molecule of considerable length and was rather easily obtained. The sample was 99 percent pure. Fig. 5 shows the data. They obviously closely resemble those for paraffin.

Data for a 1.5 cm layer of heptane in a can similar to the 4.5 cm can described are also shown. Again, they seem to show the same characteristics as those found with paraffin of approximately the same thickness, an initial rise

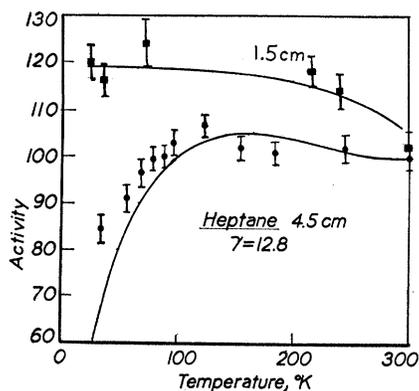


FIG. 5. Heptane data for 4.5 cm and 1.5 cm thicknesses.

and then temperature independence at lower temperatures. This is expected for small  $\gamma$  values from Eq. (1) and checks our supposition that the  $\gamma$  values increase with the average thickness of hydrogenous material through which the cold neutrons diffuse.

#### DATA FOR ETHANE

Ethane was introduced into the apparatus by immersing the 4.5 cm thick copper can in liquid air and condensing gas from a commercial cylinder. Measurements of the boiling point gave an estimate of less than three percent of impurities. Fig. 6 shows the data.

Comparison of these data with those shown in Fig. 5 for heptane reveals a definite difference below 80°K. For ethane, there seems to exist a temperature below which no further cooling changes the activity. Since for all the hydrogenous substances used, except liquid hydrogen, the numbers of protons per cubic centimeter were the same within four percent, it seems evident that at low temperatures the ethane ceases to cool the neutrons as well as does heptane. The curve was drawn for  $\gamma=13.5$ , approximately the same as for heptane. The data of Witt and Kemp<sup>11</sup> on the heat capacity of ethane show that the heat capacity first rises above the Debye curve for heat capacity due to crystalline vibrations at about 50°K, indicating the appearance of some new type of excitation, probably molecular, at about this temperature. This temperature agrees fairly well with the temperature at which ethane appeared to stop cooling and seems to point to a connection between ability to cool neutrons and this new type of excitation.

#### DATA FOR METHANE

Methane condensed from natural gas, pure to at least 95 percent, was condensed in the can just as in the case of ethane. Since it boils at 112°K no data for higher temperatures were obtained. The data for the lower range are shown in Fig. 6. The point for 300°K was written in on the basis of the observation mentioned above, that the placing of 300° paraffin, heptane, or water around the target cylinder caused no appreciable change in the activity produced.

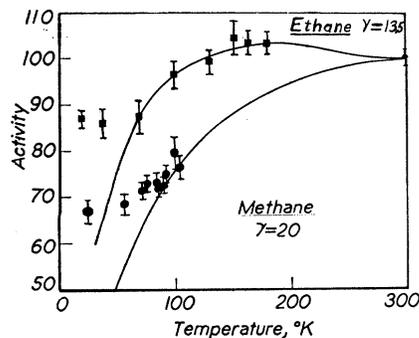


FIG. 6. Ethane and methane data.

<sup>11</sup> Witt and Kemp, J. Am. Chem. Soc. 59, 273 (1936).

Assuming that 300° methane would not differ appreciably from these substances, the value of the activity at 100°K becomes remarkably low. Apparently the methane is cooling more rapidly than either heptane or paraffin, so that the neutrons have a thicker average layer of hydrocarbon to traverse before reaching the target. It is also clear that at some temperature around 80°K the ability to cool neutrons further is being lost rapidly. However, since the number of carbon nuclei per unit volume is considerably smaller in this case an effect similar to that found with pure liquid hydrogen reported elsewhere in this paper might be expected. The hydrogen experiment seems to show that the absence of any heavy nuclei to scatter the neutrons and keep them in the cooling medium as long as they would have remained otherwise made it difficult for the hydrogen to cool them. Such an effect in the methane would have given too low an activity at 300°K and our assumption that the methane would be quite equivalent to heptane at 300°K would then be wrong. Against this view, experiments reported later, in which liquid hydrogen itself was used in the Dewar instead of the hydrocarbon, and compared directly with an hydrocarbon solution of the same proton density at 300°K, showed that the activity due to the liquid hydrogen (which was unable to cool to temperatures lower than about 120°K, and was therefore not greatly different from 300°K liquid hydrogen) was nearly the same as that for the hydrocarbon solution. The surrounding paraffin jacket seems to help keep the neutrons in the cooling medium in both cases. Consequently, the authors believe the best explanation is that methane has more ways

of absorbing neutron energy, possibly because its molecular rotation is so easily excited.

The solid curve on the graph is for  $\gamma$  equal to 20, agreeing with the previous interpretation of  $\gamma$  as a measure of the average thickness of hydrocarbon traversed after the average neutron was cooled.

#### DATA FOR WATER

The principal difficulty in the work with water was the expansion on freezing, which stretched the can and changed the geometry to a slight extent. However, the distortion, occurring principally in the radial direction perpendicular to the surface of the target, could hardly have caused a serious change in the shape of the temperature curve. The data are shown in Fig. 7.

On the lines of the previous interpretations the temperature independence of the activities with water in the can probably means that it has ceased to cool at some temperature not lower than 100°K where the large temperature effects are expected to appear first (with this apparatus). In addition, it is clear that water does not cool as rapidly as does methane, for there is no appreciable drop between 300°K and 100°K. It appears that the two molecules differ markedly in the rate at which they cool thermal neutrons.

These conclusions are not surprising in view of the well-known character of the ice crystal. Investigations of the temperature variation of the dielectric constant (for infinite frequency) with temperature shows that a constant value is reached on cooling to 110°K. In addition, the heat capacity of water first rises above the Debye curve at about 185°K.<sup>12</sup> One would expect, therefore, that any neutron cooling process depending on excitation of states of individual molecules rather than of the ice crystal as a whole (Debye curve) would cease to function at some neutron temperature between 110°K and 185°K, agreeing with the results obtained.

In more detail, the lethargic slowing by water as compared to ammonia can be understood on the basis of the crystal structure of ice. Bernal and Fowler<sup>13</sup> have shown that a structure in-

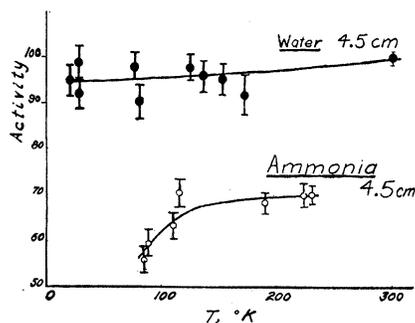


FIG. 7. Data on water and ammonia.

<sup>12</sup> Giaque and Stout, *J. Am. Chem. Soc.* **58**, 1144 (1936).

<sup>13</sup> Bernal and Fowler, *J. Chem. Phys.* **1**, 515 (1933).

TABLE II. Data on first liquid  $H_2$  experiment.

CONDITIONS	ACTIVITY
Empty Dewar (300°K)	400±15
Liquid $H_2$ (20°K)	500±15
Water (300°K)	540±15

volving random orientation of hydrogen bonds<sup>14</sup> accounts very well for the known properties of water. The verifications<sup>12, 15</sup> of Pauling's<sup>16</sup> prediction on the basis of this model, of a discrepancy between the entropy of ice deduced from specific heat data and the third law of thermodynamics and the entropy from spectroscopic data, has established the model even more firmly. Therefore any mechanism of transfer of energy from a neutron to a water molecule which requires excitation of rotational states would be expected to be quite inhibited in the ice crystal. On the other hand, the methane crystal is known to allow molecular rotation, agreeing with our observation that methane apparently cooled more rapidly than ice. The indication afforded by these facts, that the excitation of rotational states in the cooling molecule is an important mechanism for neutron cooling, is further born out by work described below on the relative effects with *ortho* and *para* liquid hydrogen.

#### DATA ON AMMONIA

Ammonia from a commercial cylinder (99 percent pure) was condensed in the can immersed in a bath of ether cooled with solid carbon dioxide. The corrosive action of the liquid and solid ammonia on the copper made the manipulation somewhat difficult, so data were not obtained at liquid hydrogen temperatures. The data down to 80°K are shown in Fig. 7.

One might suppose at first sight that the difference between the empty can activity and the asymptotic value of 80 percent for the 300° ammonia is due to rapid cooling of the type shown by methane. However, it has been shown by several observers that nitrogen has a rather large cross section for capture ( $\sim 8 \times 10^{-24}$  cm<sup>2</sup>).

<sup>14</sup> Latimer and Rodebush, J. Am. Chem. Soc. **42**, 1529 (1920).

<sup>15</sup> Long and Kemp, J. Am. Chem. Soc. **58**, 829 (1936).

<sup>16</sup> Pauling, J. Am. Chem. Soc. **57**, 2680 (1935).

The simplest explanation seems to be that the nitrogen absorption in the ammonia caused this decrease in activity. The character of the data indicates that it is still cooling at liquid-air temperatures, though it will probably soon stop because the specific heat of ammonia drops to the Debye limiting curve at about this temperature.

#### DATA ON DIATOMIC HYDROGEN

Since the investigation of liquid and solid hydrogen over any appreciable range of temperatures was quite impossible, it was necessary to use special experiments employing some of the results reported above to study this very important molecule. The first experiment tried was performed in the small assembly shown in Fig. 8. The Dewar was filled with liquid hydrogen to a level two or three inches above the top of the silver target. The activities so obtained (2.3-min. activity, after 7 minutes activation) were compared with activities from the same arrangement when the liquid hydrogen was replaced with water at room temperature. Table II contains the data.

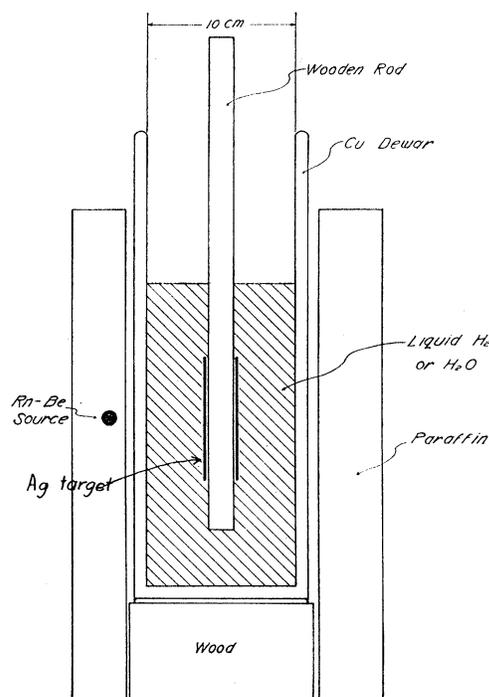


FIG. 8. First experiment on cooling by liquid hydrogen. Small copper Dewar assembly.

TABLE III. Data for liquid H<sub>2</sub> runs in large assembly.

CONDITIONS	ACTIVITY
Empty Dewar (300°K)	540±15
Liquid H <sub>2</sub> (20°K)	730±15
Solid H <sub>2</sub> (13°K)	720±15
"Equivalent" (300°K) Liquid (21.2% CH <sub>3</sub> OH in acetic acid)	700±15
Paraffin Cylinder (1.5 cm thick) +Liquid H <sub>2</sub> (20°K)	560±15
Carbon Cylinder (0.5 cm thick)+Liquid H <sub>2</sub> (20°K)	700±15

Since the relative proton densities of liquid hydrogen and water are 1 and 1.4, from these data it seemed probable that the liquid hydrogen and water were having about the same effect, allowing for the higher proton concentration of the water. To further elucidate this point, the large assembly shown in Fig. 2 was used with the copper sample can removed. The level of liquid hydrogen was adjusted by placing two thermocouples approximately two inches apart on the soft glass Dewar, at a level about three inches above the top of the silver target. The experiments described above were then repeated with this assembly. In addition, runs were made with solid hydrogen (obtained by pumping till the vapor pressure fell to 4 cm) and with a solution of methyl alcohol in acetic acid which had the same number of protons per cubic centimeter as the liquid hydrogen. The composition of this solution is 21.2 percent alcohol by volume. The data are shown in Table III.

Table III also includes results from an experiment in which a paraffin cylinder 1.5 cm thick and of the same height as the target, was placed around the soft glass Dewar so as to cover the target as completely as possible, and the rest of the volume filled with liquid hydrogen.

It is apparent from the data that liquid and solid hydrogen both have nearly the same effect as a room temperature hydrocarbon liquid of the same proton density. The interposition of the paraffin cylinder caused a large decrease incommensurate with its dimensions, in view of the experiments with paraffin and pure hydrocarbons described above. One is led to conclude that the process probably consists of a cooling by liquid hydrogen to a certain temperature, e.g. 180°K, well above 20°K and a further cooling by the paraffin. This further cooling would lead to high

absorption by the paraffin protons and by the soft glass Dewar, causing the observed drop in activity. An alternative explanation, of course, is that the carbon in the paraffin cylinder is absorbing or scattering to such an extent that the activity is reduced. This point was settled, however, by replacing the paraffin cylinder with a thinner one of carbon containing the same number of carbon atoms. The result, shown in Table III, is conclusively in favor of the explanation given.

The correlations already obtained between the ability to cool neutrons and the specific heats involving excitation of the individual molecules, rather than crystals as a whole, made the inability of the hydrogen molecule to cool to low temperatures quite understandable. It also suggested that in this case a more direct check on the mechanism of the energy transfer might be made by studying the relative effects of *ortho* and *para* liquid hydrogen in the Dewar.

This was done by filling the Dewar with non-equilibrium liquid hydrogen (75 percent *ortho*) and converting it to *para* by dropping active charcoal into the Dewar and allowing it to stand for approximately an hour. Samples of the gas were taken in both cases for subsequent analysis by the standard heat conductivity method. The

TABLE IV. Data for runs with equilibrium (75% *ortho*) and *para* liquid H<sub>2</sub> in large assembly.

75% <i>ortho</i>	ACTIVITIES	100% <i>para</i>
2998		2858
2861		3140
2854		2975
2766		3115
2610		3150
2620		2990
2980		3188
2837		2900
—		3160
2817±35		3018
		3049±27
		% Increase on conversion to <i>para</i> state=8.3±1.5

TABLE V. Data on production of slow neutrons by water and liquid hydrogen.

LIQUID	ACTIVITY
Water (300°K)	14±1
Liquid H <sub>2</sub> (20°K)	1±1

compositions were found to be 75 percent *ortho* in the first case and 99 percent *para* in the second, within one or two percent. The data for the runs on the activities in the two cases are given in Table IV.

Since the first excited state of the *para* molecule loses its population at a much lower temperature than does the *ortho* molecule, these results are in complete agreement with the hypothesis advanced. Fig. 9 shows the populations of the ground and first excited rotational state for the two molecules at various temperatures. The curves were calculated from the data of Giaque.<sup>17</sup>

From these data a lower limit of approximately 100°K for *para* hydrogen and approximately 200°K for the nonequilibrium mixture (75 percent *ortho*) might be expected. The agreement with the results seems to indicate, in this case at least, that the cooling mechanism is mainly the excitation of rotation in the cooling molecule.

Another experiment was done to determine the ability of liquid hydrogen (75 percent *ortho*) to slow the very fast neutrons from the source. The large metal Dewar was used without the surrounding paraffin cylinder, and the source was placed inside a Pyrex Dewar of the same dimensions as the soft glass Dewar described previously. Targets of dysprosium oxide mounted on varnished linen with rubber cement (0.20 g of Dy<sub>2</sub>O<sub>3</sub>/cm<sup>2</sup>) were placed in the cooling liquid. The Dewar was filled to a level about 20 inches from the bottom. A cross section of the assembly is shown in Fig. 10 and the data in Table V.

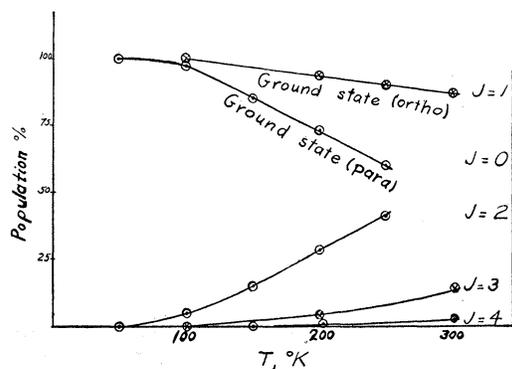


FIG. 9. Level populations for ortho- and parahydrogen.

<sup>17</sup> Giaque, J. Am. Chem. Soc. 52, 4816 (1930).

These results agree with those of Fomin, Houtermans, Leipunsky, and Schubnikow,<sup>18</sup> who also found that water is much more efficient in

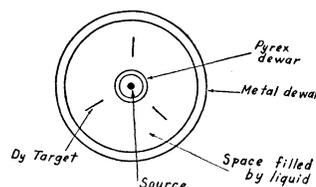


FIG. 10. Arrangement for determining relative effects of water and liquid hydrogen in slowing fast neutrons.

producing slow neutrons. A plausible explanation seems to be that the smaller mass of the H<sub>2</sub> molecule and the absence of the additional scattering due to O prevents the neutrons from staying in the cooling liquid as long as in the case of water, and consequently allows them less chance to attain temperature equilibrium. The experiment is interesting in that it indicates, as did all the previous experiments with heavier hydrogenous molecules, that cooling by recoil of the whole molecule is of small importance in the liquid or solid state. It seems probable that this conclusion should necessarily apply to all heavier molecules in the liquid or solid states.

#### CONCLUSIONS FROM COOLING EXPERIMENTS WITH DIFFERENT HYDROGENOUS MOLECULES

Figure 11 shows the collected data for the molecules most thoroughly studied, other than H<sub>2</sub>. These curves in conjunction with the detailed considerations given above seem to justify the following conclusions:

1. An hydrogenous molecule in its ground state can cool only those neutrons whose kinetic energies are at least equal to the energy necessary to excite the molecule to its first state, e.g., the temperature of neutrons emerging from a crystal of such molecules at 0°K will approximate the temperature at which the specific heat curve begins to rise above the Debye curve for the crystalline specific heat.
2. The probability of energy loss to a crystal or liquid as a whole seems to be small.
3. For hydrocarbons, the limiting temperature for cooling appears to decrease with increase in length of the hydrocarbon chain.

<sup>18</sup> Fomin, Houtermans, Leipunsky, and Schubnikow, Physik Zeits. Sowjetunion 9, 696 (1936).

4. The speed of cooling by hydrogenous solids, i.e., the probability of energy loss by a high temperature neutron, seems to be highest for those molecules having the largest number of rotational degrees of freedom in the solid state.

#### MEASUREMENT OF NEUTRON TEMPERATURE

If the absorption of slow neutrons by boron nuclei really obeys the "1/v" law, it should be possible to use this absorption as a rather sensitive neutron "thermometer." A first attempt was made to study the absorption in  $H_3BO_3$  sheets (absorbed on filter paper) of the neutrons passing through the walls of the large metal Dewar shown in Fig. 3, when it was filled with paraffin and all surrounding hydrogenous material removed to a distance of at least two feet. The absorption curve was run at both  $300^\circ K$  and  $90^\circ K$ , using rhodium as a detector. No change in absorption coefficient greater than about five percent was found. Presumably this was due to absorption of the slower neutrons in the Dewar walls at both temperatures, reducing the effect of temperature change. The large component of neutrons not absorbable in cadmium exciting the rhodium also made the measurements more uncertain. For this reason the target was changed in the later work to dysprosium.

About 98 percent of the neutrons exciting dysprosium are absorbed by cadmium. At first it seemed that this choice would lead to difficulties because it had been commonly supposed that the very high absorption cross section for

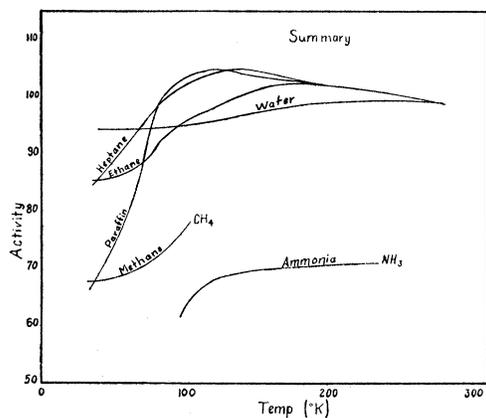


FIG. 11. Summary of data on hydrogenous molecules.

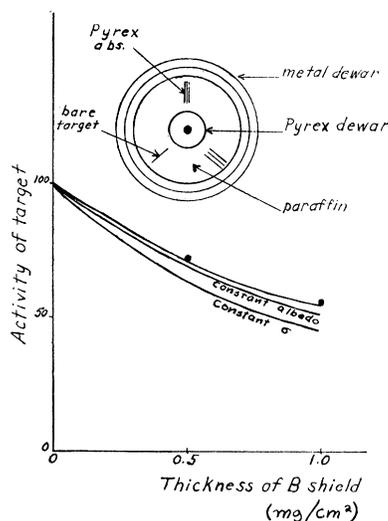


FIG. 12. Arrangement and  $90^\circ K$  curve for multiple passage boron absorption.

this element must be due to the existence, for one of the isotopes, of a resonance level in or near the thermal region, which would necessarily distort the absorption from the "1/v" law. However, test runs made in collaboration with Dr. P. W. Schutz, with dysprosium and silver targets placed between paraffin blocks, first at room temperature, and then at liquid-air temperature, gave the same relative change (approximately 10 percent increase) for the activity of the dysprosium as for the 2.3-minute activity of silver. Data described later on a comparison between B and Dy further check the "1/v" character of the dysprosium absorption. The activities of the targets were measured with a Lauritsen quartz fiber electroscop.

#### "MULTIPLE PASSAGE" BORON ABSORPTION

Figure 12 shows a cross section of the apparatus used in the first experiment with the dysprosium targets. The long period (2.3 hours) of the dysprosium activity allowed the target to be placed inside the Dewar and so eliminated Dewar wall absorption. Three targets were used. All were squares with 3 cm sides and having a total of 0.20 g of dysprosium, as  $Dy_2O_3$ , mounted on varnished linen with a thin layer of rubber cement. Calibration showed the three targets to be identical within a small error. The targets were placed symmetrically around the source (in the central small Pyrex Dewar) in slits cut in

the paraffin cylinder just wide enough for the targets and absorbers. In this way each run gave two points on the absorption curve at each temperature.

Since the thickness and absorption cross sections for the  $H_3BO_3$  absorbers and  $Dy_2O_3$  absorbers as well as the reflection coefficient (albedo) for the neutrons succeeding in passing through both absorbers and the target were all known for 300°K neutrons it seemed possible to calculate the absorption curve. This proved to be the case. Furthermore, by assuming the "1/v" law throughout, also that the albedo varies as indicated by Fermi's considerations, and that the neutrons were at thermal equilibrium inside the block, the curves could be calculated for 90°K and 20°K. If the absorption coefficient of the dysprosium target is expressed as  $bT^{-1/2}$  and for the boron absorbers as  $taT^{-1/2}$ , where  $b$  and  $a$  are constants and  $t$  is the thickness of the boron absorbers, the activity of the target,  $A$ , is given at 300°K by

$$A = K(1 - [\exp -b(300)^{-1/2}] \times \frac{[\exp -ta(300)^{-1/2}]}{1 - \zeta[\exp - (b+2ta)(300)^{-1/2}]}, \quad (2)$$

in which  $K$  is a constant and  $\zeta$  is the albedo coefficient (0.82 for 300°K neutrons). At any other temperature  $\zeta$  should be calculated, on the above assumptions, from the expression

$$\frac{2}{1 - \zeta} = (N)^{1/2}, \quad (3)$$

where  $N$  is the average number of collisions of the neutron with protons necessary for capture by the protons. The assumption of the "1/v" law for proton capture requires  $N$  to vary as the reciprocal of the square root of the temperature, so that

$$\zeta = 1 - 0.75T^{-1/2}. \quad (4)$$

Substitution of expression 4 in Eq. (2) gives the final equation used for calculating the expected curves. Figs. 12 and 13 show the curves and data for 90°K and 20°K, respectively. Curves are also shown for the effect of assuming the dysprosium cross section,  $\sigma$ , to be independent of temperature in one case, and the albedo to be constant in the other case. The experimental

error of the points is about equal to their width so that the agreement seems to be satisfactory in both cases. For purposes of comparison, one point of the 300°K curve is shown on the 20°K plot to indicate how small the temperature effect is in this arrangement where the neutrons are allowed to make as many passages as possible through the target and absorbers. For this reason the arrangement was discarded as a means of measuring neutron temperatures and the following single passage arrangement was substituted.

### SINGLE PASSAGE BORON ABSORPTION

The arrangement for this series of experiments is shown in Fig. 14. The three dysprosium targets were placed symmetrically around the source at the surface of the 4.5 cm paraffin cylinder. Each had a sheet of cadmium behind it to prevent multiple passage. The whole assembly was moved as far as possible (approximately two feet) from the nearest hydrogenous material. Thin Pyrex plates, made by etching thicker plates with hydrofluoric acid, were used as absorbers. Use of several thicknesses showed the absorption to be exponential at each temperature. The averages from twenty runs are shown in Fig. 14, with the probable errors indicated by the vertical lines through each point. The two low points refer to measurements described later, made with a  $BF_3$  neutron counter.

Apparently the data do not fit the straight line expected if the "1/v" law holds for B and Dy and the neutrons are at the temperature of the block. It seemed probable that the assumption of the "1/v" law for dysprosium was the weakest of the three assumptions involved, so

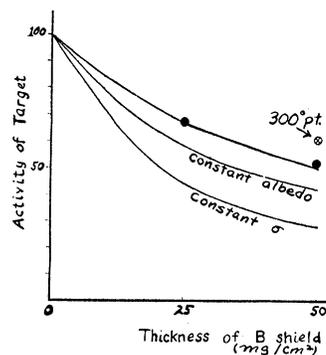


FIG. 13. Multiple passage boron absorption curve for 20°K.

determinations of the absorption coefficients were made with a neutron counter filled with boron trifluoride, instead of the dysprosium.

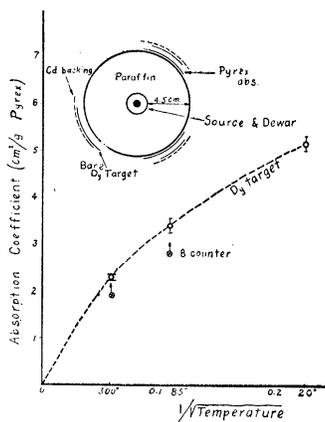


FIG. 14. "Single passage" boron absorption assembly. Data obtained.

The counter is an ordinary tube counter with an iron wire, 0.22 mm in diameter, filled to 12.5 cm pressure with  $\text{BF}_3$  gas. It is quite possible by use of a linear amplifier of standard type to adjust the counter to respond to slow neutrons quite efficiently (probably 5 percent of the 300°K neutrons traversing the counter register), without giving counts of more than two or three per minute due to the gamma-radiation from 200 mg of  $\text{Ra}+\text{Be}$  at a distance of six inches or less even when the counter is unshielded with lead. The voltage adjustment for the counter is not very critical, a variation of twenty volts in the counting voltage of 1600 being quite unimportant. It is necessary to remove all hydrogenous material, such as paraffin, from contact with the gas to prevent its decomposition and destruction of the selective characteristics of the instrument.

The arrangement used for the boron counter work consisted of the paraffin plug and Pyrex Dewar for the source as shown in Fig. 14. The paraffin was cooled, for the 90°K run, by immersion in liquid air, and was then removed to a position in mid-air at considerable distance (approximately two feet) from hydrogenous material. The boron counter, 17 cm long and 2.3 cm in diameter, was placed 8 inches distant with its axis parallel to that of the paraffin cylinder. A thick Pyrex plate was interposed at intervals between the paraffin and the counter

to absorb the neutrons coming directly from the paraffin, and to allow the slow neutrons being scattered from the floor to be counted. The cold paraffin runs were only two minutes long and a test run with a thermocouple imbedded in the block showed that the temperature rise during this interval was certainly less than 5°.

The absorption curves were obtained for both the paraffin and "floor" neutrons by slipping Pyrex cylinders (etched to appropriate thickness with hydrofluoric acid), around the counter. Fig. 15 is a logarithmic plot of the data obtained. The data obviously check the dysprosium data within the limits of error, for the ratio of the absorption coefficient for 90°K to that for 300°K was  $1.5 \pm 0.1$  in the case of the dysprosium targets and is  $1.44 \pm 0.05$  in this case, indicating again that dysprosium acts like a "1/v" absorber. However, this test, using the B and Dy as targets rather than absorbers, is not as reliable as a test using them as absorbers would be. The small amount of dysprosium available (approximately 1 gram) prevented the latter experiment. The two coefficients are plotted in Fig. 14. They are lower in absolute value than those for the Dy work by about the fraction one would expect from the difference in the two cases in the average angles between the paths of the incident neutrons and the normals to the Pyrex absorbers. The average angle in the case of the counter work was smaller.

The most plausible view now remaining seems to be that the neutrons emerging from such a

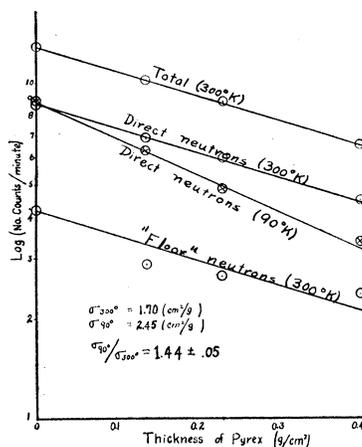


FIG. 15. Data for boron counter determination of boron absorption coefficient.

block have not attained thermal equilibrium. The experiments on "multiple passage" seemed to show that in this case thermal equilibrium was nearly attained, though the low sensitivity of the measured effects to change in temperature may have masked any small failure to cool. It seems reasonable, however, that in the case of multiple passage the repeated reflections may offer more opportunity for the neutrons to cool and even select those that have cooled. Further evidence for this conclusion was obtained by consideration of the results of Amaldi and Segrè,<sup>19</sup> on the apparent equality of the albedo coefficient for 90°K to that for 300°K.

TEMPERATURE VARIATION OF ALBEDO (REFLECTION) COEFFICIENT

The results of Amaldi and Segrè were checked in the following way. A dysprosium target was activated when exposed on the top of a cube of paraffin (approximately 8 cm on a side) which was so placed beside the radium-beryllium source that a second identical cube placed on top of the target would occupy a position symmetrical to that of the first cube with respect to the source. The arrangements are shown in Fig. 16.

The ratio of the activity with two blocks to that with one block was obtained first with the blocks at 300°K and later with them immersed in liquid air at 90°K. The ratio was  $8.1 \pm 0.2$  in both cases, a somewhat lower value than reported by Amaldi and Fermi,<sup>20</sup> probably because the effect of neutrons of intermediate energy above thermal produced in one block which are cooled on addition of the second block is neglected. This was justified because such an effect should be expected to be nearly independent of temperature and should also be small in absolute magnitude. This ratio,  $R$ , is easily shown<sup>10</sup> to satisfy the relation

$$R = 2/(1 - \zeta), \tag{5}$$

if the absorption cross section of the target for neutrons present inside a block of paraffin and that for those emitted from the surface can be assumed to be the same. This, however, is the point discussed above. If  $\sigma_i$  represents the cross section inside the block and  $\sigma_e$  that for the neutrons coming from the surface, Eq. (5)

becomes

$$R = (\sigma_i/\sigma_e)(2/(1 - \zeta)), \tag{5'}$$

where  $\zeta$  is the albedo coefficient, as usual.

Assuming that  $\sigma_i$  and  $\zeta$  vary with the temperature in the way expected from the "1/v" law, and that the neutrons inside the block are at the temperature of the paraffin block, the ratio of  $R$  for 90°K to  $R$  for 300°K can be calculated from the experimental value of 1.47 for the ratio of  $\sigma_e$  at 90° to  $\sigma_e$  for 300°. The result is

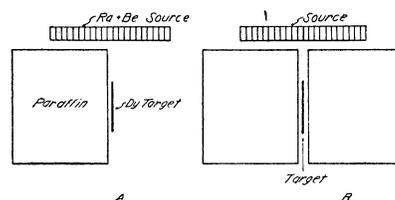


FIG. 16. Arrangements for measuring temperature variation of the "albedo" coefficient.

unity within the error of measurement, agreeing with the experimental results.

In addition to such a failure to cool due to proximity to the surface, there probably is an effect in the paraffin analogous to that found in lighter molecules which limits the cooling because of absence of likely mechanisms at the lower energies. The authors are much indebted to Professor J. R. Oppenheimer and Mr. W. E. Lamb of the physics department for helpful discussions and suggestions about this and other phases of the problem.

TEMPERATURE OF "ICE NEUTRONS"

Having a temperature scale empirically calibrated, the paraffin cylinder in Fig. 14 was replaced by one of ice of the same dimensions and the absorption curve obtained when the ice was at 20°K. The value for the absorption coefficient was  $3.4 \pm 0.4$  cm<sup>2</sup>/g of Pyrex, corresponding to a neutron temperature of about 100°K, in agreement with the previous conclusions.

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<sup>19</sup> Amaldi and Segrè, Phys. Rev. 50, 571 (1936).

<sup>20</sup> Amaldi and Fermi, Phys. Rev. 50, 899 (1936).