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ON THE SLOWING DOWN AND CAPTURE OF NEUTRONS IN HYDROGENOUS SUBSTANCES

BY

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1. Introduction.

FERMI and his coworkers (1) have discovered that neutrons can be slowed down by surrounding their source with substances containing hydrogen such as water or paraffin wax. This discovery has been followed by a number of investigations (2) which have thrown much light on the phenomenon so that we have now a fairly complete picture of the processes involved.

Fast neutrons in a large container filled with water will collide with the protons of the water losing, on an average, half of their kinetic energy at each collision (3); they will also lose a (smaller) fraction of their energy in collisions with the oxygen nuclei (3). When the neutrons have come down to about 1 eV., energy transfer to the protons is hampered by the chemical bond forces (4); nevertheless, most of the neutrons get into thermal equilibrium (kT = 0.026 eV. at room temperature) and diffuse like atoms of a dissolved substance until finally, after another 100—200 collisions, they are captured by some of the nuclei present.

We have investigated the capture of neutrons in different liquids containing hydrogen. By varying the chemical composition of the liquid we have been able to separate the effects due to the different elementary constituents and to determine the capture cross-section of the nuclei of H, D, C, N, and O relative to the capture cross-section of Li and B which are known from absorption experiments with slow neutron beams (5). Some information on the interaction of fast neutrons with these nuclei and on the disintegration of D and Be by gamma-rays (6, 7) has also been obtained.

Furthermore, we have tried to obtain information of a qualitative character on the number of collisions required to bring a neutron through the "chemical bond region" and into thermal equilibrium. For this purpose we have studied the absorption, in different substances, of neutrons slowed down at different temperatures (8, 9, 10) and under various geometrical conditions.

2. Capture of neutrons in different liquids.

If a source of neutrons is placed in a large container filled with water (11), a spherically symmetrical density distribution of neutrons around the source will be established, the density $\varrho(r)$ decreasing with increasing distance r from the source. The exact shape of the density distribution depends on a number of parameters, such as the mean free path at all the neutron energies involved, the directional distribution of scattered neutrons, and so on. The total number $n = 4 \pi \int_{0}^{\infty} \varrho(r) r^2 dr$ of neutrons present at any instant, however, depends only on their mean life τ and the number q of neutrons produced per unit time:

$$n = q \cdot \tau = 4 \pi \int_{0}^{\infty} \varphi(r) r^{2} dr$$
(1)

Taking τ from diffusion experiments, AMALDI and FERMI(11) and recently AMALDI, TUVE and HAFSTAD(12) have used this method for determining the number q of neutrons emitted by different sources per unit time. For

exploring the density distribution they have made use of the activity induced in rhodium.

In our experiments we have made a somewhat different use of equation (1). By comparing n with different liquids but the same source (q constant), we have obtained relative values for τ in the liquids investigated. For exploring the density distribution we used a small boron-lined ionisation chamber which was immersed in the liquid. The number of counts J is then proportional to the neutron density ρ ; in the equation $J = s \cdot \rho$ the factor s (the sensitivity of the chamber) is independent of the velocity of the neutrons, provided that the absorption of boron follows the "1/v-law" (13, 14, 4) for all the neutron velocities involved (i.e. if its cross-section is inversely proportional to the velocity of the neutrons). As a matter of fact, we found that the number of counts is reduced by a factor of about 100 (in water) on surrounding the chamber with cadmium; this shows that about 99 per cent of the neutron density is made up of neutrons with energies below 1 eV. (13, 15), in which region we can safely adopt the 1/v-law for boron. The chamber sensitivity s need not be known since we are only interested in relative measurements. A rough estimate of s can, however, be obtained from the area of the boron layer and the known range of the alpha-particles emitted under the action of neutrons.

Assuming the capture cross-sections σ_c of all nuclei present in the liquid to be inversely proportional to the velocity of the neutrons, the chance per unit time of their getting captured does not depend on the velocity of the neutrons. (This corresponds to the assumption that resonance capture is of no importance (4).) On introducing the "capture probability per nucleus" $p_k = v \cdot \sigma_{ck}(v)$ (the index k referring to the different kinds of nuclei), the chance per unit time of being captured becomes $\chi = \sum_{k} p_k N_k$ (N = number of nuclei per cm.³). The mean life is then

$$\tau = \frac{1}{\chi} = \frac{1}{\sum_{k} p_k N_k} \tag{2}$$

and the total number of neutrons present at any instant is

$$n = q \cdot \tau = \frac{q}{\sum_{k} p_k N_k}.$$
(3)

By comparing the values obtained for n in two different liquids we find

$$\frac{n_1}{n_2} = \frac{\sum_k p_k N_{k2}}{\sum_k p_k N_{k1}}.$$
 (4)

If the two liquids are both composed of the same two constituents A and B, the relative proportion only being different, then the equation can evidently be used to determine $\frac{p_A}{p_B} = \frac{\sigma_{cA}}{\sigma_{cB}}$ = the ratio of the capture cross-sections of the two constituents.

In this way we have determined the ratio between the capture cross-sections of H and C, by comparing measurements on benzene and liquid paraffin. We have also made measurements on water and aqueous solutions of boric acid (16, 17), lithium hydroxide, deuterium oxide, and ammonium nitrate; this enabled us to find the capture cross-sections of oxygen, boron, lithium, deuterium and nitrogen, with respect to hydrogen. Since boron and lithium show hardly any scattering (18), their capture cross-sections may be identified with their total cross-sections obtained from

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absorption experiments on beams of thermal neutrons; in this way we obtained absolute values for the capture crosssections of the light elements we have investigated.

3. Experimental arrangement for the capture experiments.

As a source of neutrons we used a 100 mCurie of $RaSO_4$ mixed with about 1 gm. of thoroughly powdered beryllium contained in a platinum tube, 25 mm. long and 8 mm. wide,



Fig. 1. Small boron chamber. a, outer brass tube; b, soft glass tube providing insulation between a (earthed) and c; c, the high tension (100 Volt) electrode with the boron lining d; e, collecting electrode, connected to the amplifier by the thin wire k; f, grounded shield, insulated from c by the glass tube j, which is punched (h) to keep the insulating glass bead g in position.

with 0.3 mm. walls. The platinum tube was enclosed in a brass container which in turn was put in a soft glass tube (of 13 mm. outer diameter) to protect it from contact with the liquid.

The boron-lined ionisation chamber was made quite small so as to disturb the neutron distribution as little as possible, the construction of the chamber being shown by the scale drawing fig. 1. The chamber was connected to a Wynn-Williams linear amplifier and scale-of-eight counter system. The entire arrangement is shown in fig. 2.

Several times a day the sensitivity of the counting arrangement was checked with a "standard neutron density": the chamber was placed at the center of a tin can (20 cm. high, 20 cm. diameter) filled with water and the source was placed at a distance of 6.0 cm. from the chamber. The number of counts per minute under these conditions varied between 1000 and 800, during the course of



Fig. 2. Arrangement for capture experiments. a, container filled with 60 ltrs. liquid (galvanised iron, painted with paraffin wax inside when used for aqueous liquids); b, boron chamber (see fig. 1); c, neutron source; d, first amplifier stage; e, brass rod with scale; f, slider supporting source; g, rubber suspension; h, counterweight.

the investigation, but was in general constant to within a few per cent during one day.

4. Results of the capture experiments.

In fig. 3, 4, 5 the function $\rho(r) r^2$ is plotted for all the liquids investigated. The neutron densities ρ are measured in units of the "standard density" and all lengths are measured in cms.

In order to determine the areas below the curves, which represent the total number n of neutrons present at any instant in the liquid (see formula (1)), two alternative methods were used. In the "numerical method" the area of the measured part of the curve was calculated by the "trapezoidal rule"



Fig. 3. Neutron density times squared distance from source plotted with respect to distance. A, water (distilled); B, benzene; C, liquid paraffin.

using as ordinates the measured (not smoothed) values of $\rho(r) r^2$. To obtain the (small) area under the tail of the curve beyond r = 40 cm. an exponential form was assumed.

The "geometrical method" was based on the striking similarity in the shapes of the curves. This similarity is borne out by plotting the curves on a double logarithmic scale: it is then possible to bring the different curves into coincidence by shifting the origin; fig. 6 shows how good a fit can be obtained. This means that, if we return to the original curves (fig. 3, 4, 5), any of them can be made to

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coincide with the water curve A by changing the scale of the abscissae and ordinates by suitable factors F_x and F_y . By such a change of scale, any area will be changed by a factor $Q = F_x \cdot F_y$ which is, therefore, the area of the given curve relative to the area of the water curve.



Fig. 4. Neutron density times squared distance from source; A, water; B, $10^{0/0}$ heavy water; C, $25^{0/0}$ sugar solution; D, 7.6 $^{0/0}$ ammonium nitrate solution; E, $0.41^{0/0}$ lithium hydroxide solution.

In table 1 all the figures required for the later discussions are collected. It is seen that the areas determined numerically and geometrically agree quite well in a number of cases. Where there was a small difference, as in the case of the heavy water, the arithmetical mean has been adopted.

With the boric acid solutions, however, the deviations are large and obviously systematic. In fact we cannot expect the boric acid curves to be quite similar in shape to the water curve since the addition of the boron consider-

| | q | | |
|---|---|---|--|
| | - | | |
| - | - | | |
| | 5 | 0 | |

Results of neutron capture experiments. The composition of the liquid paraffin was found, by chemical analysis, to correspond to the formula $CH_{1.87}$.

| | | | | | | 10-22 | dmun × | er of a | atoms p | er cm ³ | |
|--|--------|---------|--|--------|------|-------|--------|---------|---------|--------------------|-------|
| Liquid | F_x | F_y | $\stackrel{\text{geom.}}{_{F_xF_y}} =$ | numer. | Н | С | 0 | D | Li | В | Z |
| H ₂ O | 1.00 | 1.000 | 1.000 | 1.000 | 6.70 | : | 3.35 | : | : | : | : |
| $H_2O + 10 ~^{0/0} D_2O \dots$ | 1.04 | 1.10 | 1.15 | 1.13 | 6.10 | : | 3.35 | 0.60 | : | : | : |
| $H_2O + 0.1^{-0/0} H_3BO_3 \dots$ | 0.96 | 0.79 | 0.77 | 0.785 | | : | _ | : | : | $0.0_{3}98$ | : |
| $H_2O + 0.5$ % $H_3BO_3 \dots$ | 0.91 | 0.435 | 0.396 | 0.415 | | : | | : | : | $0.0_{2}49$ | : |
| $H_2O + 1.0^{-0/0} H_3BO_3 \dots$ | 0.84 | 0.282 | 0.236 | 0.254 | 6 70 | : | 2 25 | : | : | $0.0_{2}98$ | : |
| $H_2O + 2 ^{0/0} H_3BO_3 \dots$ | 0.82 | 0.173 | 0.142 | 0.154 | 01.0 | : | 0.00 | : | : | 0.0196 | : |
| $H_2O + 4 ~^{0/0} H_3BO_3 \dots$ | 0.80 | 0.094 | 0.075 | 0.083 | | : | | : | : | 0.0392 | : |
| $H_2O + 0.41^{0/0} Li OH \dots$ | 0.95 | 0.79 | 0.75 | (0.76) | | : | | : | 0.0104 | : | : |
| $H_2O + 7.6 ^{\circ/0} NH_4 NO_3$ | 0.98 | 0.945 | 0.93 | I | 6.63 | : | 3.37 | : | : | : | 0.118 |
| $H_2O + 25 ~^{0/0} C_{12}H_{22}O_{11} \dots$ | 0.98 | 1.02 | 1.00 | 1.01 | 6.63 | 0.58 | 3.31 | : | : | : | : |
| liquid paraffin | 06.0 | 1.02 | 0.92 | 0.92 | 7.15 | 3.82 | : | : | : | : | : |
| Benzene corrected | 1.34 | 1.22 | 1.63 | I | 4.05 | 4.05 | : | : | : | : | : |
| (» uncorrected) | (1.32) | [(1.17) | (1.55) | (1.55) | : | : | : | : | : | : | : |

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ably shortens the diffusion path of the slow neutrons, thereby shifting the maximum towards the origin, and at the same time has practically no influence on the slowing down of the fast neutrons which is the determining factor for the



Fig. 5. Neutron density times squared distance from source; Boric acid solutions: $A_{x} 0^{0}/_{0}$; $B, 0.1^{0}/_{0}$; $C, 0.5^{0}/_{0}$; $D, 1^{0}/_{0}$; $E, 2^{0}/_{0}$; $F, 4^{0}/_{0}$.

slope of the curve at large distances from the source. Applying the "geometrical method" to the boric acid curves means extrapolating with a slope which is too steep and thereby obtaining values for the area which are too small. In the case of the boric acid solutions we have therefore based the further calculations on the numerically calculated values only; the part of the curves between 20 and 40

cm. which had not been measured was extrapolated assuming the same logarithmic slope as in the water curve.

The area of the LiOH-curve was determined geometrically and then corrected assuming the same difference between geometrical and numerical method as in the case of the 0.1 per cent H_3BO_3 solution; in this way the figure in brackets (0.76) was obtained.

5. Corrections.

The error introduced by the limited size of the container (in theory it should be infinite) was found, by remeasuring the water curve with a much larger container (about 80 cm. in all directions), to be of the order of one per cent or less. Hence it would appear that the error is equally small, and therefore negligible, in all the other liquids, with the exception of benzene. In benzene the hy-

drogen density is much smaller than in the other liquids investigated; the mean free path for neutrons is larger and consequently the whole curve is more stretched (F_r) is considerably larger than unity). A considerable increase in nmight therefore be anticipated for measurements carried out in a much larger volume of benzene.

In order to avoid



Fig. 6. Same as fig. 3 (with corresponding letters) but on double logarithmic plot. In plotting B and C the frame has been shifted in such a way that the point (10, 10) lies for the three sets of points at the positions indicated by the arrows A, B, C. It is possible in this way to bring the curves into coincidence.

this costly and dangerous experiment (a fire is bad enough with 60 ltrs. of benzene) we tried to determine the necessary correction in an indirect way, based on a similarity argument. We have remeasured the water curve using a container which was similar in shape to the old one but smaller by a factor of 1.32 (the value of F_x for benzene) in all directions. We believe then that we commit the same error as we committed in the benzene measurement¹. The

¹ This would be exactly correct if the slowing-down process in benzene were exactly the same as in water with the sole difference that the

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value for n obtained with the small water container was found 5 per cent smaller than with the large container, and consequently we have increased the value of Q for benzene by 5 per cent, from 1.55 to 1.63.

The error introduced by the limited size of the ionisation chamber (in theory it should be a point) was estimated in the following way. The chamber was immersed in water and a thin-walled brass tube of twice the chamber's diameter was placed around it. If the space between the chamber and the tube was filled with air (instead of water), the number of counts was increased by 4 per cent, with the source at a distance of 10 cm. Since the disturbance introduced by a cylindrical "air bubble" should be roughly proportional to the square of its diameter, it was concluded that the disturbance introduced by the ionisation chamber was of the order of one per cent only; no correction has been applied for this. The disturbance caused by the absorption in the chamber walls and the boron layer was estimated to be negligible.

6. Capture cross-sections.

From the figures given in table 1, relative capture crosssections have been calculated using equation (4). First we compared benzene and liquid paraffin; this gives the equation

$$\frac{n_1}{n_2} = \frac{1.63}{0.92} = 1.77 = \frac{(3.82 \, p_C + 7.15 \, p_H)}{(4.05 \, p_C + 4.05 \, p_H)}$$

and therefrom

$$rac{p_{C}}{p_{H}} = rac{\sigma_{cC}}{\sigma_{cH}} = 0.005 \pm 0.02.$$

mean free path for every neutron velocity is longer by a factor 1.32. This is certainly not exactly true, but the good fit of fig. 6 indicates that it is at least roughly true.

This means that the absorption cross-section of carbon is negligible with respect to that of hydrogen, in fact probably smaller than one fiftieth. Qualitatively, the vanishing capture cross-section of carbon is evident from the fact that the ratio of the neutron numbers (1.77) is practically identical with the inverse ratio of the hydrogen densities $\left(\frac{7.15}{4.05} = 1.76_5\right)$ in the two liquids.

Assuming $\sigma_{cC} = 0$ we can proceed to determine $\frac{\sigma_{cO}}{\sigma_{cH}}$ by comparing water and benzene. From the equation

$$\frac{n_1}{n_2} = \frac{1.63}{1.00} = 1.63 = \frac{(6.70 \, p_H + 3.35 \, p_O)}{4.05 \, p_H}$$

we get

$$rac{p_{O}}{p_{H}} = rac{\sigma_{cO}}{\sigma_{cH}} = - \ 0.03 \ \pm \ 0.02$$

which means that the capture cross-section of oxygen, like that of carbon, is negligible, at any rate probably smaller than $0.02 \cdot \sigma_{cH}$. Again, the ratio of the neutron numbers (1.63) is practically identical with the inverse ratio of the hydrogen densities $\left(\frac{6.70}{4.05} = 1.65\right)$.

The sugar experiment does not give additional information because the amount of carbon contained in the sugar solution is too small. The result is, however, in agreement with the foregoing for the neutron number n as well as the hydrogen density are not considerably changed by dissolving the sugar; only the shape of the curve is slightly modified ($F_x = 0.98$) because the additional carbon shortens the mean free path of the neutrons (especially of the fast ones, which are decisive for the shape).

On dissolving ammonium nitrate in water, however, the

neutron number is considerably reduced (Q = 0.93), while the slight decrease (one per cent) in hydrogen density would cause in itself a corresponding increase in neutron number. From the equation

$$\frac{n_1}{n_2} = 0.93 = \frac{6.70 \, p_H}{(6.63 \, p_H + 0.118 \, p_N)}$$

we get

$$\frac{p_N}{p_H} = \frac{\sigma_{cN}}{\sigma_{cH}} = 5 \pm 1.$$

The heavy water experiment will be discussed later in connection with the experiments with photo-neutrons.

The boric acid and lithium hydroxide experiments are of special interest because they enable one to place all the measured cross-sections on an absolute scale. On comparing the five boric acid solutions (0.1, 0.5, 1, 2, 4 per cent) with water, the following values for $\frac{p_B}{p_{H_{2}0}} = \frac{p_B}{2 p_H}$ are obtained: 935, 970, 1010, 945, 940. The agreement is satisfactory and as a weighted mean the value of 970 \pm 50 was accepted. On comparing the lithium hydroxide solution with water we get $\frac{p_{H_20}}{p} = 102 \pm 10$.

It is known that thermal neutrons are very little scattered in boron and lithium (18) and we may therefore identify their capture cross-sections with their total crosssections for thermal neutrons as it is determined by absorption experiments on beams of thermal neutrons. Assuming a value of $(500 \pm 30) \cdot 10^{-24}$ cm.² for σ_B (see § 10) we find $\sigma_{cH_20} = (0.51 \pm 0.04) \cdot 10^{-24}$ cm.² and consequently $\sigma_{cH} = (0.26 \pm 0.02) \cdot 10^{-24}$ cm.². Taking, furthermore, $\sigma_{Li} =$ $= (63 \pm 4) \cdot 10^{-24}$ cm.² (see § 10) we get $\sigma_{cH} = (0.30 \pm 0.04) \cdot$

 $\cdot 10^{-24}$ cm.². As the most probable value we suggest the weighted mean $\sigma_{cH} = (0.27 \pm 0.02) \cdot 10^{-24}$ cm.².

From this the absolute capture cross-sections of C, N, and O can be calculated; the results are collected in table 2 (including D, see § 7).

Our value of σ_{cH} is somewhat smaller than the value given by AMALDI and FERMI (0.31 \cdot 10⁻²⁴ cm.²), which fur-

Table 2.

Capture cross-sections of light elements for neutrons of $\sqrt{2 kT/m} = 2.2 \cdot 10^5$ cm./sec. (see § 10).

| Element | Н | D | С | N | 0 |
|--|-----------------|--------|--------|---------------|--------|
| $\sigma_c \cdot 10^{24} \text{ cm}^{-2}$ | 0.27 ± 0.02 | < 0.03 | < 0.01 | 1.3 ± 0.3 | < 0.01 |

thermore corresponds (4) to a velocity of $2.5 \cdot 10^5$ cm.sec.⁻¹ and should therefore be increased by a factor of 1.13 to be compared with our value. Our figure for σ_{cN} can be compared with the cross-section of nitrogen for disintegration by thermal neutrons which has been determined by BURCHAM and GOLDHABER (19) by counting the protons emitted in the process; the agreement between their value ($\infty 1 \cdot 10^{-24}$ cm.²) and our figure indicates that most of the slow neutrons captured by a nitrogen nucleus give rise to the emission of a proton rather than of a gamma-quantum, just as one would expect it to be on general theoretical reasons, for such a light nucleus.

7. Experiments with photo-neutrons.

In order to discuss the heavy water experiment described in the last section we have to take account of the

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neutrons which are created throughout the liquid under the action of the radium gamma-rays (6). The possibility of creation of neutrons when fast neutrons from the source collide with deuterons has also to be kept in mind. In order to get information on both these effects we have made some measurements where the Ra-Be source was replaced by a neutron-free gamma-ray source; furthermore by surrounding this source with beryllium, low energy neutrons were obtained (7) which are certainly not able to



Fig. 7. Neutron-free gamma-ray source. a, glass tube; b, chrom-nickel tube; c, radon; d, mercury.

disintegrate deuterons by their impact.

The neutron-free gamma-ray source (see fig. 7) consisted of a small tube of a

chrom-nickel alloy sealed into glass. The tube was filled with about 300 mCurie radon; a mercury seal prevented the radon from coming in contact with the glass (which would have caused neutron emission); the glass was sealed off on top of the mercury, for safety. The source was found to emit one thousandth of the neutrons emitted by a source in which the same amount of radon was mixed with beryllium.

The source was placed at the center of either of two identical thin-walled brass containers (cylindrical with 1.8 cm. height and 1.9 cm. diameter); one of them was filled with 2.7 gr. metallic beryllium, the other with 3.0 gr. carbon which has scattering properties for neutrons and gammarays similar to those of beryllium but emits no photoneutrons. Measurements were carried out in the same way as described above, with ordinary and "heavy" water; the

decay of the radon was taken into account and the results were always expressed per 100 mCurie.

The results are shown in fig. 8. Because of the smaller energy of the neutrons, curves A and B have a much steeper slope and the maximum lies at smaller distances from the origin than in the curves shown in fig. 3, 4 and 5. Curve D, however, is especially flat; this is due to the fact that the neutrons are produced throughout the liquid and that it is not their absorption but that of the gamma-rays which is responsible for the slope. The half-value thickness of water for gamma-rays of 2.2 MeV is¹ 15 cm., which is in good agreement with the final slope of curve D (see curve G). In curve B at small distances from the source the neutrons come mostly from the beryllium while for large distances the curve flattens out, most of the neutrons being then due to photo-disintegration of the deuterons.

The difference of A and C (curve E) corresponds to the photo-neutrons from the beryllium only and curve E is therefore very similar in shape to the corresponding curve F = B - D. The areas of curve F and E have been determined numerically and their ratio is found to be 1.11. From this ratio we obtain from $1.11 = \frac{6.70 \ p_H}{(6.10 \ p_H + 0.60 \ p_D)}$ the value $\frac{p_D}{p_H} = \frac{\sigma_{cD}}{\sigma_{cH}} = -0.1 \pm 0.1$.

In the experiments with the Ra + Be source a slightly higher value for the ratio of the areas of the heavy and ordinary water curves was found (1.13; see table 1). If we, however, deduct from the heavy water curve the photo-

¹ Calculated from figures given by L. MEITNER and H. H. HUPFELD, ZS. f. Physik, **67**, 147 (1931). It should be noted that γ -rays which have been scattered once have almost certainly not enough energy left to disintegrate deuterons.

neutrons produced in the deuterium, that is, the area of $D-1.11 \cdot C$ (fig. 8), then the ratio becomes 1.11 in agreement with the ratio found when using the low energy beryllium photo-neutrons. We conclude from this that the number of neutrons created by collisions between fast neutrons and deuterons is too small to be detected in this way. This is not surprising since only about one out of ten fast neutrons will happen to hit a deuteron before having lost part of its energy and because only a small fraction of the primary neutrons have sufficient energy to disintegrate the deuteron (~ 3.2 MeV).

It is assumed in the calculation of the capture crosssections that all the neutrons emitted by the source reach low energies where the 1/v-law can be applied while only a negligible fraction vanishes before getting slow. Although for example nitrogen is known to offer a considerable crosssection for disintegration by fast neutrons (22), still the number of neutrons which get lost, in this way, in our NH₄NO₃-solution can be estimated to be very small, just as the number of neutrons created in collisions with deuterons in our heavy water was found experimentally to be negligible. Furthermore, our result that the total number *n* of neutrons found in water, benzene and paraffin oil is inversely proportional to the hydrogen densities in the three liquids excludes any considerable loss of neutrons on account of the presence of oxygen and carbon.

8. Yields.

The relative neutron yields of the sources investigated by us can be deduced fairly accurately from the experiments. From the ratio of the areas of curve E (fig. 8) and curve A (fig. 4) which is 0.039 we see that the $\text{Rn}_{\gamma} + \text{Be}$ source emits 3.9 per cent of the neutrons emitted by the Ra + Be source. The total area of curve $D - 1.11 \cdot C$ (fig. 8) (indicating the production of photo-neutrons in our heavy

water) can be estimated to be 0.020 of that of curve B (fig. 4). Furthermore, we have found that our Ra + Be source emitted about 65 times as many neutrons as 1 mCurie radon mixed with beryllium. The total number of neutrons emitted by such a source has been measured (21, 11) and values ranging from 7.000 to 27.000 per second have been obtained. Assuming a value of 20.000 we can calculate the cross-sections of deuterium and beryllium for photo-disintegration bv the gamma-rays of Ra



Fig. 8. Neutron density times squared distance from source. A and B: gammaray source + beryllium; C and D: without beryllium; A and C in water; B and D in 10 % heavy water. E = A - C; F = B - D. G indicates the calculated absorption of the hardest gamma-ray line of radium C (2.2 MeV.) in water.

B+C. The intensities of the two hardest lines (2.2 and 1.8 MeV.) have been found (22) to correspond to the emission of 0.074 and 0.26 quanta per disintegration, respectively. With deuterium the hardest line only can be effective and the cross-section is found equal to $7 \cdot 10^{-28}$ cm.² (see however, the note at the end of the paper). For beryllium we have to introduce two cross-sections $\sigma_{1.8}$ and $\sigma_{2.2}$ corresponding to the two lines of 1.8 and 2.2 MeV.; from the

observed total number of neutrons we then get the equation $\sigma_{1,8} + 0.3 \cdot \sigma_{2,2} = 2 \cdot 10^{-27} \text{ cm.}^2$. Should the number of





Fig. 9. Arrangement for neutron beam experiments. *a*, boron trifluoride chamber (see fig. 10); *b*, cadmium shield; *c*, absorber; *d*, boron shield (about 1 gr./cm.²); *e*, lead block (the chamber is fairly sensitive for gamma-rays); *f*, paraffin wax; *g*, sources; *h*, copper beaker soldered to copper bar *j*; *k*, wooden sup-

port; l, liquid air.

neutrons emitted by 1 mCurie of Rn + Be turn out to be larger than 20.000 per second then the cross-sections given have to be increased in the same proportion.

9. Neutron beam experiments.

In addition to the experiments on the capture of neutrons described above we have carried out some experiments on the absorption (including scattering) of slow neutron beams in a number of substances. On the one hand we wanted to redetermine the cross-sections of boron and lithium, the values published so far disagreeing considerably among themselves; on the other hand we tried to get additional information on the influence of the temperature of the paraffin on the absorbability in different elements of the neutrons stopped by cadmium ("C-neutrons") (11).

A first set of measurement was carried out using, as neutron

source, a paraffin howitzer the inner part of which could be cooled down to the temperature of liquid air (fig. 9). The copper beaker covered, on the inside, with a one cm. layer of paraffin wax was soldered on a copper bar of

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4 cm. diameter, the lower end of which was immersed in liquid air. The temperature of the beaker was within a few degrees that of the liquid air, varying slightly with the liquid air level; about 2 ltrs. of liquid air per hour were required.

As detector for the slow neutrons an ionisation chamber filled with boron trifluoride was used. The construction of the chamber may be taken from fig. 10. The chamber was



Fig. 10. Boron trifluoride chamber. a, steel cylinder; b, steel collecting electrode; c, glass tube (may be unnecessary); d, ebonite rings; e, grounded ring; f, copper capillary (through which the gas is introduced), squeezed and sealed with solder. The chamber was made gas-tight by apiezon wax W, which also covers the inside surface of the ebonite and protects it from contact with the BF₈.

at first filled to about 10 atm. pressure; excessive potentials were, however, required to collect the ions in a sufficiently short time and the pressure was therefore lowered to about $1^{1/2}$ atm., about 800 Volt still being required for satisfactory operation. The chamber has proved very stable and reliable during nearly a year's work and the apiezon wax W used for insulation does not seem to deteriorate in contact with the boron trifluoride.

A cadmium screen (0.4 gm./cm.²) was used as usually to discriminate between fast and slow neutrons (C-neutrons). The cadmium was always placed below (instead of above) the absorbing sample; this is important in the case of paraffin where the slowing down of neutrons in the sample may considerably lower the apparent absorption if the Cd is

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placed on top of the paraffin. All the substances were investigated as pure elements, except for Cl where CCl_4 was used.

10. Neutron beam experiments: Results.

The results are collected in table 3. The absorber thickness was always chosen such as to reduce the beam intensity to about one half; from the absorber thickness and the intensity reduction the values of μ were calculated.

Table 3.

Mass absorption coefficients (of the compounds used) in $\text{cm.}^2/\text{g}$. and total cross-sections (of the elements) for beams of thermal neutrons. The subscripts give the temperature of the paraffin used in slowing down the neutrons.

| Absorber | μ_{290} | μ_{80} | $ \frac{\mu_{80}}{\mu_{290}} $ | $\sigma_{290} \cdot 10^{24} \mathrm{cm}^{-2}$ |
|-------------|-------------------|--|--------------------------------|---|
| H (paraf- | | | | |
| fin wax) | 4.3 ± 0.2 | 4.7 ± 0.1 | 1.09 ± 0.05 | 48 ± 3 |
| Li | 5.5 ± 0.2 | 8.3 ± 0.3 | 1.51 ± 0.10 | 63 ± 4 |
| B | 27.7 ± 0.9 | 42.3 ± 1.2 | 1.53 ± 0.07 | 500 ± 30 |
| Al | 0.035 ± 0.002 | 0.039 ± 0.002 | 1.10 ± 0.10 | 1.6 ± 0.1 |
| $Cl(CCl_4)$ | 0.82 ± 0.02 | 1.32 ± 0.04 | 1.61 ± 0.06 | 51 ± 2 |
| Fe | 0.115 ± 0.007 | 0.12 ± 0.007 | 1.05 ± 0.10 | 12 ± 0.7 |
| As | 0.070 ± 0.006 | 0.094 ± 0.004 | 1.34 ± 0.13 | 8.8 ± 0.8 |
| Ag | 0.34 ± 0.015 | 0.52 ± 0.02 | 1.53 ± 0.09 | 60 ± 4 |
| Cd | 16.6 ± 0.7 | 16.6 ± 0.5 | 1.00 ± 0.05 | 3100 ± 150 |
| Au | 0.29 ± 0.015 | $0.43 \hspace{0.2cm} \pm \hspace{0.2cm} 0.03 \hspace{0.2cm}$ | 1.48 ± 0.15 | 90 ± 5 |

It must, however, be kept in mind that the C-neutrons are inhomogeneous and that their velocity distribution is changed when they pass through an absorber the crosssection of which depends on their velocity. If the neutrons originally have a MAXWELLIAN distribution and if the absorber follows the "1/v-law" (i. e. if its cross-section is inversely proportional to the velocity of the neutrons), then the cross-section obtained when using a very thin absorber corresponds to a neutron velocity of $\sqrt{\pi kT/2 m} = 1.95 \cdot 10^5$ cm.sec.⁻¹; with thicker absorbers, however, a hardening occurs such that the value which is found using a "half-value" absorber corresponds (23) to a velocity which is about 1.13 times larger and is, in fact, very nearly = $\sqrt{2 kT/m} = 2.2 \cdot 10^5$ cm.sec.⁻¹.

It is seen that some of the substances (Li, B, Cl, Ag, and Au) show a "temperature effect" (8, 9, 10) (increase of absorption coefficient when the paraffin wax from which the neutrons come is cooled to liquid air temperature) of about 1.5 while some other substances show practically no temperature effect (paraffin wax, Al, Fe); with As an intermediate value is obtained.

In fact, no temperature effect should be expected if the observed cross-section is all due to scattering; the strong scattering of slow neutrons by aluminium and iron is well known (24, 18). In paraffin wax, however, although absorption is negligible compared with scattering, a small temperature effect should be expected on account of the chemical bond action (4); calculations carried out by Mr. N. ARLEY (to appear shortly in Kgl. Danske Vid. Selsk. Math. Phys. Medd.) give a factor of about 1.3. Such an effect has also been observed by FINK (9).

In the case of lithium and boron, there are strong theoretical reasons (4) to expect their cross-sections to be inversely proportional to the neutron velocity, over a considerable velocity range; also in the case of silver and gold one would expect the 1/v-law to be valid for neutrons of thermal energies. It is then very satisfactory that all these elements show the same amount of temperature effect. That chlor-

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ine shows it as well is in agreement with the fact (25) that also in chlorine most of the cross-section is due to capture. In the case of arsenic, the value of 1.34 seems to indicate that both capture and scattering contribute to the observed cross-section; indeed, unpublished experiments of one of us (O. R. F.) on the activation of As by C-neutrons



Fig. 11. Absorption coefficient of silver plotted with respect to $T^{-\frac{1}{2}}$ (T = absolute temperature of the paraffin). Dots, "flat" paraffin block; crosses, thick-walled howitzer; open circle, boric acid difference experiment. (The straight line would correspond to the " $T^{-\frac{1}{2}}$ -law").

indicated a capture cross-section of about one half the total cross-section.

However, the amount of the temperature effect observed (1.5) is not equal to $\left(\frac{T_1}{T_2}\right)^{-\frac{1}{2}} = \left(\frac{80}{290}\right)^{-\frac{1}{2}} = 1.9$, the inverse ratio of the mean velocities of neutrons in temperature equilibrium (28). Since this discrepancy might be due to incomplete temperature equilibrium of the neutrons cooled in a thin layer of paraffin only (see fig. 9), we have made some

experiments (27) with a flat block of paraffin of 5—6 cm. thickness which could be kept at four different temperatures: 80° (liquid air), 195° (solid carbon dioxide), 290° (room temperature), and 415° (the paraffin being molten, in this case). The absorption coefficient of silver μ_{Ag} for C-neutrons was measured for each temperature. If the neutrons were in thermal equilibrium one would expect μ_{Ag} to be proportional to $T^{-\frac{1}{2}}$. From fig. 11 this relation is seen to hold fairly well for the three highest temperatures while the lowest point strongly deviates from the " $T^{-\frac{1}{2}}$ -law", the ratio being again about 1.5 only, instead of 1.9.

This discrepancy has been observed before. FOMIN and his colleagues (10) found a temperature effect of 2.2 when comparing the absorption of silver for neutrons of 20° (liquid hydrogen) and 290° while $\left(\frac{T_1}{T_2}\right)^{-\frac{1}{2}} = 3.8$ in this case. Furthermore for the absorption of boron the factor was only 1.65 ± 0.2 . In our experiments, no significant difference between silver and boron was found, comparing neutrons at liquid air temperature (80°) and 290°. FOMIN et al. do not explicitly give the temperature effect for the step from 80° to 290°, but from their absorption figure (on silver) a value of about 1.3 seems to follow. FINK (9) using an arrangement quite similar to ours, obtains for Li, B, and Ag values of 1.20, 1.32, and 1.20, respectively, which are all lower than our figures; we do not know the reason for this difference.

The discrepancy between the observed temperature effects and the value of $\left(\frac{T_1}{T_2}\right)^{-\frac{1}{2}}$ has sometimes been taken as an argument against the validity of the 1/v-law. We believe, however, that the explanation lies in the fact that the neutrons defined by their strong absorption in cadmium

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(C-neutrons) have not thermal energies throughout but include a "tail" of neutrons with energies up to the cadmium absorption limit (about 1 volt.) which is more intense than would follow from MAXWELL's distribution law.

In order to calculate the number of "tail neutrons" relative to the thermal ones a detailed knowledge is required of the slowing down mechanism which is not available at present. A very rough estimate, however, indicating the order of magnitude may be obtained as follows. We know that neutrons with energies below 0.1 eV. lose their energy at a much lower rate than fast ones because the protons in the paraffin, on account of their chemical binding, cannot be regarded as free when offered energy amounts less than one quantum of oscillation energy; they should rather be regarded as rigidly connected to the carbon atom to which they belong. The carbon atoms having much lower oscillation frequencies may be regarded as free, in first approximation. One may then get a rough picture of the slowing down mechanism by assuming (2) that the neutrons collide with particles of mass 14, (considering the paraffin as a structure of CH₂-molecules).

In collisions with such particles, neutrons will lose one eighth of their energy at every impact, at an average, and about ten collisions will be required to bring a neutron from 0.1 Volt down to 0.03 Volt (mean energy at room temperature). On the other hand a neutron, on leaving the paraffin, has, on the average, suffered about 100 collisions (see the next section). There is therefore, roughly speaking, a 10 per cent chance of a neutron emerging from the paraffin before having reached thermal equilibrium, and consequently 10 per cent of the neutrons which get into the boron chamber will be "tail neutrons". Taking their greater velocity into account

(which is of importance not only as regards the penetrating of the absorber but also the sensitivity of the boron chamber) we get the result that the mean absorption coefficient of C-neutrons of room temperature is about 5 per cent less than for neutrons in perfect thermal equilibrium.

At liquid air temperature, however, a much larger difference should result. Firstly, about twice as many collisions should be required (about 20) for the neutrons to get into thermal equilibrium, starting from 0.1 Volt; secondly, the number of collisions which they survive should be considerably smaller (perhaps 60) on account of the increased capture cross-section of the protons for slower neutrons.

It has been suggested (28) that the capture cross-section of the protons at low temperature may be decreased on account of their zero-point oscillations. However, a detailed discussion shows that quite generally the mean life of slow neutrons in any substance which has no resonances in the energy range concerned should be entirely independent of temperature, depending only on the density of absorbing nuclei (in the way discussed in the first chapter); and there are certainly no resonances near the thermal range, in the case of paraffin.

Furthermore, the slowing down of neutrons below room temperature is additionally hampered by the fact that the carbon atoms in the paraffin can be no more regarded as free to take up such small energy amounts (one eighth of 0.03 volt or less). This effect should be expected to cause rapidly increasing deviations from the $T^{-\frac{1}{2}}$ -law as temperatures goes down, in agreement with our experiments.

If this explanation is true, one would expect the absorbability of the neutrons to be higher when the arrang-

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ement is such that the mean life of the neutrons in the cold paraffin is increased. We have therefore measured μ_{Ag} , using a thick-walled ($\infty 4$ cm.) paraffin howitzer which could be cooled as a whole; the neutron sources were placed outside the Dewar flask, below the howitzer, with more paraffin wax underneath. The values obtained (see the crosses fig. 11) are in fact larger than with the flat block, especially at the low temperature.

We have also tried to measure the absorption of thermal neutrons at room temperature without using cadmium to distinguish them from the faster ones. We have used two equal howitzers made of water and a 0.5 per cent boric acid solution, respectively; the liquids were contained in thin-walled metal forms. The boric acid should have very little influence on the intensity of the fast and "tail" neutrons emitted from the howitzer since those neutrons stay only very short time in the liquid; the difference in intensity (which was about 50 per cent) should be almost entirely due to thermal neutrons. The absorption of these "difference" neutrons in silver was determined by measuring the intensity difference between the two howitzers with and without a silver absorber in front of the boron chamber. The absorption coefficient obtained (see the open circle in fig. 11) was practically equal to the value obtained for C-neutrons using a howitzer; this value may therefore be regarded as the value for true thermal neutrons, with an error less than 5 per cent. This is of some importance because all the neutron energy determinations based on the 1/v-absorption of boron (13, 14, 28, 23, 29, 15) depend on the assumption of thermal equilibrium of C-neutrons at room temperature, an assumption which gains strong support by our measurements.

11. Mean life-time of neutrons in water.

From the measurements reported in § 6 and § 9 we can calculate the mean life-time τ_0 of neutrons in water. The life-time in a boric acid solution is given by

$$r = \frac{1}{\left(\frac{1}{r_0} + p_B N_B\right)} \tag{5}$$

(this follows from equation (2)). In the graphical representation given in fig. 12 which is, of course, essentially identical with the calculation given in § 6 the intersection of the straight line indicates the boron density N'_B which by itself would permit the neutrons to live just as long as they do in pure water; the life-time is then found $\tau_0 = 1/p_B N'_B$. The value for p_B is taken from § 10: $p_B = \sigma_B \cdot v = 500 \cdot 10^{-24} \text{ cm.}^2 \cdot 2.2 \cdot 10^5 \text{ cm.sec.}^{-1} = 1.1 \cdot 10^{-16} \text{ cm.}^3 \text{ sec.}^{-1}$. From this (and a corresponding calculation making use of the LiOH experiment, see § 6) we get $\tau_0 = (2.7 \pm 0.2) \cdot 10^{-4} \text{ sec.}$, which is somewhat larger than the value $1.7.10^{-4} \text{ sec.}$ given by AMALDI and FERMI.

The average number of collisions suffered by a neutron is then equal to $N = \frac{\overline{v} \tau_0}{\lambda}$, where $\overline{v} = \sqrt{\frac{8 kT}{\pi m}} = 2.4_8 \cdot 10^5 \text{ cm.sec.}^{-1}$. Taking the mean free path $\lambda = 0.31 \pm 0.02 \text{ cm.}$ according to our value of the hydrogen scattering cross-section (table 3) we find $N = 205 \pm 20$, which is again somewhat larger than the value (N = 140) obtained by AMALDI and FERMI.

We have also made similar measurements where a boron chamber was placed outside a can (28 cm. high, 19 cm. diameter) filled with water or boric acid solution and with

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the source placed in the center. The results are also shown in fig. 12 (open circles). The life is seen to be shorter just as one should expect since the life of the neutrons is limited not only by capture but also by escaping through the surface. A more detailed discussion of similar experi-



Fig. 12. Abscissa, Amount of boron added to water; ordinate, Reciprocal of the intensity reduction caused by adding the boron. Filled circles, Intensity integrated over large volume (cf. table 1); Open circles, Intensity observed outside water can; Cross, Water howitzer (§ 10).

ments, taking into account the diffusion of the neutrons, has been given by WESTCOTT (30).

In a howitzer the surface should but little shorten the life of the neutrons as only those which pass the bottom almost normally will escape for ever. However from the intensity reduction caused by the boron in the waterhowitzer experiment described at the end of § 10 a rather

short life-time would follow (see the cross in fig. 12). This probably comes from the fact that the source was placed very near the bottom of the bore in this experiment, so that there was a strong neutron density gradient; such a gradient would reduce the life in a similar way as a free surface, or in other words, the neutrons observed near the source are preferably those which have suffered only few collisions.

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SUMMARY

1. The density distribution of slow neutrons around a source was studied, the source being placed in large volumes of different liquids. From the relative total numbers of neutrons their relative life in the liquids could be obtained. Assuming the life to be limited only by the capture of the neutrons while slow, the capture cross-sections of a number of light elements were obtained, relative to the capture cross-sections of boron and lithium, which are known absolutely from neutron beam absorption experiments. The results are collected in table 2.

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2. The yield of photo-neutrons from beryllium and deuterium irradiated with radium gamma-rays has been compared with the yield of neutrons from beryllium mixed with radon. Assuming the latter yield as 20 000 neutrons per second per mCurie, the cross-section of deuterium for dissociation by the gamma quantum of 2.2 MeV. is found to be $7 \cdot 10^{-28}$ cm². In the case of beryllium the two hardest lines of radium with 1.8 and 2.2 MeV. are energetic enough to cause dissociation; in this case we find $\sigma_{1.8} + 0.3 \cdot \sigma_{2.2} = 2 \cdot 10^{-27}$ cm.² where $\sigma_{1.8}$ and $\sigma_{2.2}$ correspond to the 1.8 and to the 2.2 MeV. line respectively.

3. The total cross-sections for C-neutrons of a number of elements have been measured by the neutron beam method, the neutron source being a paraffin howitzer the inner part of which could be cooled to liquid air temperature. The cross-section was found to be larger by a factor of about 1.5 for "cold" neutrons, in some of the elements, including those (Li, B, Ag, Au) which are known to show strong absorption and little scattering. In the elements where scattering is known to be strong (H, Fe, Al) and in Cd little or no influence of temperature was found. Arsenic gave an intermediate value.

4. The absorption coefficient of silver has been measured, the neutrons being slowed down in paraffin at different temperatures between 80° and 415° , and under different geometrical conditions. The results (which are shown graphically in fig. 11) support the view that the C-neutrons though containing mostly neutrons with thermal energies include a "tail" of faster neutrons extending to the cadmium absorption limit of about 1 volt. This tail does not change the absorption coefficient of thermal neutrons by more than a few per cent, at room temperature, but becomes very important at lower temperatures.

5. From our measurements the mean life-time of thermal neutrons in water is found to be $(2.7 \pm 0.2) \cdot 10^{-4}$ sec. and the mean number of collisions is found equal to 205 ± 20 .

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NOTE ADDED IN PROOF

In a recent paper of CHADWICK, FEATHER and BRETSCHER (Proc. Roy. Soc. A 163, 366, 1937) the binding energy of the deuteron has been redetermined and found to be 2.25 ± 0.05 MeV (instead of the formerly accepted value of 2.14 MeV). Hence it appears that the neutrons obtained (see p. 19—21) on irradiating heavy water with the gamma-rays of RaB + C cannot be due to the gamma-line of 2.198 MeV but must be produced by some harder gamma-rays. Some evidence for the existence of such gamma-rays has been given in papers of SKOBELZYN (ZS. f. Physik 43, 354, 1927), ELLIS (Proc. Roy. Soc. A 143, 350, 1934), and GRAY (Proc. Roy. Soc. A 159, 263, 1937). Too little, however, is known about their energies and intensities to give a basis for a modified calculation of the cross-sections of the deuteron and of the beryllium nucleus.

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