APPENDIX D. TEMPERATURE DEPENDENCE OF LINE WIDTHS

The pressure normalized impact parameter is expressed as: $\Delta\nu/P=(nv/P\sqrt{2})b_{\text{eff}}^2$. In the classical kinetic gas theory model b is independent of temperature. Since $n/P\propto T^{-1}$, $v\propto T^{\frac{1}{2}}$, it follows that $\Delta\nu/P\propto T^{-\frac{1}{2}}$. From the more sophisticated line-breadth theories $b\propto T^{-1/2(n-1)}$, therefore $\Delta\nu/P\propto T^{-(n+1)/2(n-1)}$, for $n\rightarrow\infty$, $\Delta\nu/P\propto T^{-\frac{1}{2}}$. The temperature dependence of polarizability-exchange line breadths is found to be $\Delta\nu/P\propto T^{-0.63}$. This corresponds to an interaction varying as $1/r^8$ or $1/r^9$, which is not unexpected since $H_{\text{pol}}\propto 1/r^6$, $H_{\text{exch}}\propto 1/r^9$. The agreement between this theoretical value and several recent measurements[35,36] which yielded results near $T^{-0.9}$ is not too good.

[35] R. Beringer and J. G. Castle, Jr., Phys. Rev. **81**, 82 (1951).
[36] R. M. Hill and W. Gordy, Phys. Rev. **91**, 222 (1953).

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Diffusion Parameters of Thermal Neutrons in Water

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A method is described for the determination of neutron diffusion parameters by measurements of the decay of thermal neutrons in a finite moderator with a pulsed-neutron source and a time analyzer. In the theory for the method one takes into account the "diffusion cooling" effect, which decreases the temperature of thermal neutrons below the temperature of the moderator. Measurements are reported on a cylindrical geometry, involving between 1 and 4 liters of distilled water. Analysis of the result yields a value of 0.333 $\pm$0.003 barns for the neutron-proton capture cross section, in good agreement with other measurements, and $(36\,340\pm750)[1-(0.20\pm0.04)\kappa_0^2]$ cm^2/sec for the diffusion constant of thermal neutrons in water. The term involving the "buckling" κ_0^2 of the geometry is due to the diffusion cooling effect. The value of the diffusion constant for $\kappa_0^2=0$ leads to a value for the diffusion length in excellent agreement with the result of other measurements. The diffusion cooling effect is larger by a factor of 3.5 than the theoretical value, calculated for the model of a monatomic gas.

ONE of us[1] had previously suggested a detailed study, using a pulsed neutron source, of the decay rate of the neutron flux from a moderator as a means of determining the mean life for absorption, the diffusion constant, and the related diffusion length for neutrons in the moderator. The present article reports the result of such measurements on distilled water in a cylindrical geometry.

THEORY

When the diffusion theory approximation is used the decay of the thermal neutron flux from a finite homogeneous moderator can be described as the exponential decay of a number of modes, each corresponding to a different neutron distribution in the moderator. Each of these modes vanishes on the extrapolated boundary of the moderator, which lies at the extrapolation distance $0.71l_t$ outside the true boundary. The transport mean free path of the neutrons is l_t.

For the decay constant of the νth mode we obtain, using the diffusion theory,

$$\lambda_\nu=\lambda_a+D\kappa_\nu^2, \tag{1}$$

where λ_a is the absorption probability per unit time (the inverse of the mean life for absorption θ). In a moderator containing n_i nuclei of type "i" per cm^3 with the absorption cross section σ_{ai}, the absorption probability λ_a is $\Sigma n_i\sigma_{ai}v$ and since, as is mostly the case, σ_{ai} varies as $1/v$, it follows that λ_a is independent of the velocity of the neutrons.

In Eq. (1) κ_ν^2 is the geometric buckling coefficient for the geometry in the νth mode. In the cylindrical geometry used in the present investigation we have for the buckling coefficient of the lowest, fundamental, mode:

$$\kappa_0^2=(2.405/R')^2+(\pi/H')^2, \tag{2}$$

where R' and H' are the extrapolated radius and height of the cylinder.

D in Eq. (1) is the diffusion constant of the neutrons in the moderator, which is proportional to the transport mean free path and the velocity of the neutrons. D will thus depend on the "temperature" T_n of the neutrons and is related to the diffusion constant D_0 at a standard temperature T_0 by the formula:

$$D=D_0[1+\beta(T_n-T_0)-\gamma(T_n-T_0)^2]. \tag{3}$$

It is usually assumed that thermal neutrons have a Maxwellian velocity distribution of the moderator temperature T_0, irrespective of the size of the moderating geometry. In reference 1 it was shown, however, that diffusion cooling will decrease the temperature T_n of thermal neutrons below the temperature T_0 of the moderator. The temperature difference increases in

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[1] G. von Dardel, Trans. Roy. Inst. Technol. **75**, 1 (1954).

the first approximation proportionally to the geometric buckling of the geometry. In the second approximation we obtain, by an obvious extension of the formulae in reference 1 for the special model of a monatomic moderating gas of nuclei of mass M:

$$T_0-T_n=T_0\frac{2l_tl_b}{9\pi^{\frac{1}{2}}C}\kappa_0^2 \times\left[1-\left(1-\frac{1}{2(1+m/M)}\right)\frac{2l_tl_b}{9\pi^{\frac{1}{2}}C}\kappa_0^2\right], \quad (4)$$

where l_b is the neutron mean free path if the nuclei were rigidly bound, and C is a constant which depends on M/m and is listed in reference 1. Because of the variations in neutron temperature the diffusion constant will also depend on the buckling of the geometry:

$$D=D_0\left\{1-E\kappa_0^2+E^2\kappa_0^4 \times\left[\frac{1}{\beta T_0}\left(1-\frac{1}{2(1+m/M)}\right)-\frac{\gamma}{\beta^2}\right]\right\}, \quad (5)$$

where

$$E=\beta T_0(2l_tl_b/9\pi^{\frac{1}{2}}C). \quad (6)$$

APPARATUS

A schematic view of the apparatus is shown in Fig. 1. Neutrons are generated in a heavy-ice target bombarded by the pulsed deuteron beam[2] of a 150-kv accelerator.[3] Fast neutrons from the target penetrate a cadmium shield into a cylindrical aluminum vessel containing the moderator, where the neutrons are slowed down to thermal energies. The flux of thermal neutrons which emerges from the moderator is detected with $B^{10}F_3$ counters of 150-cc sensitive volume and the time distribution of the pulses from the detectors is investigated by means of a multichannel time analyzer.[4] A shield of cadmium sheet next to the container and the detectors, tanks filled with boric acid solution below and around the container, and boron and paraffin bricks on top of the measuring space decrease the background of stray neutrons so effectively that the neutron flux can in most cases be followed over a factor of 10^5.

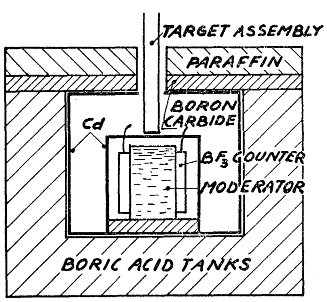

FIG. 1. A schematic view of the experimental arrangement.

Measurements were made with two cylindrical containers for the water moderator, of 18 and 14.5 cm diameter respectively. The water level was varied between 3.5 and 15 cm above the bottom of the container. Before and after a measurement the amount of water was determined by weighing and the temperature was measured. For each water level a run of 2–4 hours was made, which was sufficient to register about 10^6 pulses in the usable portion of the decay curve. The length of the neutron burst and the channel width were for each measurement chosen so that the decay was followed over at least a factor of 10^3 before the advent of the next neutron burst.

SUPPRESSION OF HARMONIC MODES

The presence of harmonic modes in the neutron distribution will make the decay curve depart from the pure exponential decay of the fundamental mode, and will make the measurements more difficult to interpret. In geometries of the small sizes studied in the present investigation, the amplitude of the harmonic modes will be considerably reduced during the slowing-down process. Their influence on the experimental results can be reduced further by a suitable choice of the experimental conditions. The target of the accelerator is situated on the axis of symmetry of the arrangement. Hence, for symmetry reasons, no harmonic modes having radial nodal planes through the axis will be excited.

By a suitable arrangement of the detector it is possible to eliminate the influence of one further set of harmonic modes, and this possibility should be used to suppress the harmonic mode with the smallest decay constant. If the water level in the container is high, this will be the mode with one nodal plane perpendicular to the axis. This mode is suppressed if the BF_3 chambers are placed along the cylindrical surface of the container symmetrically with respect to the nodal plane. For lower water levels the dominant harmonic modes have cylindrical, concentric nodal surfaces. These modes are eliminated by putting the detector below the bottom of the container, behind an opening in a cadmium mask, cut according to the function $rJ_0(2.405r/R')$.

With these precautions the experimentally determined decay curves which had first been corrected for counting rate losses (up to 3 percent in the first channels) are exponential except for the first time intervals. During about 40 μsec after the end of the neutron burst, the neutron spectrum and thus the diffusion constant changes with time, as the neutrons approach thermal equilibrium. This part of the decay curve was therefore always discarded.

In the remaining curve a small residual exponential component with a fast decay was usually found superimposed on a main exponential decay. The period of

[2] von Dardel, Hellstrand, and Taylor, Appl. Sci. Research. **B3**, 35 (1952).
[3] E. Blomsjö and G. von Dardel, Appl. Sci. Research **B4**, 49 (1954).
[4] G. von Dardel, Appl. Sci. Research **B3**, 209 (1953).

this component agreed with the calculated period for the lowest harmonic mode not eliminated by the above precautions.

TREATMENT OF DATA

The harmonic component and in a few cases a small background, were subtracted from the decay curve. The decay constant λ_0 of the corrected curve which is due entirely to the decay of the fundamental mode was calculated by the method of Peierls.[5]

The experimentally determined decay constants, corrected to a temperature of 22°C, are shown as a function of the buckling in Fig. 2. The curvature of the experimental points is apparent, showing that the diffusion constant in Eq. (1) decreases with increasing buckling, as expected theoretically, because of the diffusion cooling effect.

The experimental data have been fitted to a 3-term power series expression in κ_0^2,

$$\lambda_0 = \lambda_a' + D_0'\kappa_0^2(1 - E'\kappa_0^2), \tag{7}$$

by the method of least squares. The best values for the parameters are

$$\lambda_a' = 4907 \pm 6 \text{ sec}^{-1},$$
$$D_0' = 36\,160 \pm 50 \text{ cm}^2/\text{sec},$$
$$E' = 0.175 \pm 0.002 \text{ cm}^2.$$

In Fig. 2 are also shown the deviations of the experimental points from the best parabolic curve (7).

ERRORS AND CORRECTIONS

The errors of the experimental points in Fig. 2 include the statistical errors in λ_0, as calculated by Peierls'[5] method, and the uncertainty in the water level due to the evaporation of water during the experiment. The errors given in the values for the parameters λ_a', D_0', and E' are calculated on the basis of these errors alone. The spread of the experimental points in Fig. 2 is about twice as large as would be expected on the basis of the calculated random errors, indicating the presence of other, unknown, sources of random errors. We take these into account by increasing the uncertainty limits of the parameters λ_a', D_0' and E' by a factor of two.

We further have to take into account various systematic errors. The electronic apparatus was frequently checked during the measurements and was found very reliable. The calibration and stability of the oscillator in the time analyzer is better than 10^{-4}. We have confidence that no significant systematic errors are introduced by malfunctioning of the apparatus.

The measured decay constants would be too small if some of the neutrons which emerge from the moderator are reflected back into the geometry. For thermal neutrons the effect of neutrons reflected by the moderating material outside the cadmium screen will be very small, since these neutrons have to pass twice through the cadmium in order to affect the measurements. We verified this point by measuring the decay rate with and without paraffin blocks on the outside of the cadmium shield and found the same result within the error of the measurement. Some neutrons will be reflected by the aluminum wall of the container and by the cadmium shield but this will have a negligible effect on the decay constant.

For the extrapolation length we used the value 0.32 cm as calculated from the transport mean free path. The uncertainty in this value is probably 10 percent, which will give rise to uncertainties of ± 35 sec^{-1} in λ_a', ± 650 cm^2/sec in D_0', and ± 0.018 cm^2 in E'.

Additional errors are due to the theoretical assumptions which have to be made in interpreting the data. In the first place we have used ordinary diffusion theory. This is permissible since even for the lowest water level the dimensions of the geometry are much larger than the mean free path. We further checked this point by making calculations for the most unfavorable case by the spherical harmonic method,[6] taking into account the first four spherical harmonics, and found negligible deviations from the results of simple diffusion theory with the ordinary choice of extrapolated boundaries.

Before the experimentally determined parameters λ_a', D_0', and E' can be identified with the corresponding quantities of Eqs. (1) and (5), we have to consider the effect of the higher terms in the expression (5) for the diffusion constant D. From published curves[7] of the scattering cross section we estimate for the first and second temperature coefficients of the diffusion constant the values $\beta = 2.4 \times 10^{-3}$ per °C and $\gamma = 0.2\beta^2$. Assuming the mass of the scattering centers to be large compared to the neutron mass we find from Eq. (5)

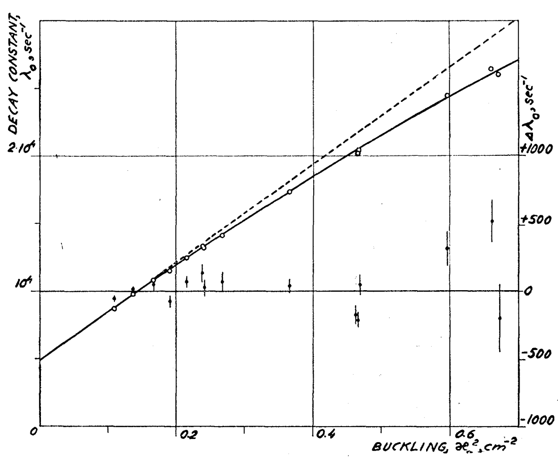

FIG. 2. Measured decay constant λ_0 and deviations $\Delta\lambda_0$ from the parabola $\lambda_0 = 4907 + 36\,160\kappa_0^2(1 - 0.175\kappa_0^2)$ sec^{-1} as functions of the buckling κ_0^2. Vertical lines on the $\Delta\lambda_0$ points indicate the calculated random errors.

[5] R. Peierls, Proc. Roy. Soc. (London) **A149**, 467 (1935).

[6] B. Carlson, U. S. Atomic Energy Commission declassified report MDDC-236 (LA-571) (unpublished).

[7] *Neutron Cross Sections*, U. S. Atomic Energy Commission Report AECU-2040 (Office of Technical Services, Department of Commerce, Washington, D. C., 1952).

for the second-order term in D the value $0.5E^2\kappa_0^4$. Since this is a rough estimate only we assign correspondingly generous uncertainty limits $\pm E^2\kappa_0^4$. The presence of a cubic term,

$$(0.5\pm 1.0)D_0'E'^2\kappa_0^6,$$

in the expression (1) will influence the determination of the coefficients of the first three terms from the experimental data. If we approximate the cubic term by a parabola, using the method of least squares in the same way as for the experimental data, we find the following corrections to the parameters λ_a', D_0' and E':

$$\Delta\lambda_a' = -15\pm 30 \text{ sec}^{-1},$$
$$\Delta D_0' = 180\pm 350 \text{ cm}^2/\text{sec},$$
$$\Delta E' = 0.02\pm 0.03 \text{ cm}^2.$$

With these corrections and combining the various sources of errors we obtain as our final result

$$\lambda_a = 4892\pm 50 \text{ sec}^{-1},$$
$$D_0 = 36\,340\pm 750 \text{ cm}^2/\text{sec},$$
$$E = 0.20\pm 0.04 \text{ cm}^2.$$

THE ABSORPTION PROBABILITY

From the value for the absorption probability per unit time λ_a, we derive the mean life for absorption $\theta = 204.4\pm 2.0$ μsec and the neutron-proton absorption cross section 0.333 ± 0.003 barns at 2200 m/sec in excellent agreement with the values 0.329 ± 0.004 obtained by Hamermesh, Ringo, and Wexler,[8] and 0.332 ± 0.007 barns obtained by Harris *et al.*[9] A previous measurement in our laboratory, by von Dardel and Waltner,[10] who also used a pulsed-neutron method, yielded the somewhat lower value 0.321 ± 0.005 barns. In this investigation the decay of the neutron flux was measured in a large water tank, so that the leakage of neutrons was negligible. The effects of diffusion were eliminated by integrating over the whole volume of the tank. The major uncertainty in this measurement is due to the effect of the detectors on the neutron distribution. An additional uncertainty lies in the presence of a background component which revealed itself in deviations of the decay curve from an exponential at low counting rates. We assumed this background to be constant with time and corrected the decay curve accordingly. If instead the background decreased with time, as may well be the case, the correction for it should be larger. This would increase the cross-section value. We propose to repeat the experiment at a later occasion and will then try to minimize the background.

The two pulsed-neutron methods have the advantage over most other methods in that the absorption cross section is determined directly without an intermediate determination of, for example, the boron absorption cross section. The method described in the present article has the advantage over our previous method that the target and the detectors are positioned outside the moderator and thus have no influence on the measurement. Very efficient detectors can therefore be used so that the counting rate is high, and any background will be negligible. The larger geometry used in the other method is also more difficult to screen efficiently.

On the other hand, we must in the present method apply diffusion theory in order to correct the measured decay for the leakage out of the tank. Furthermore, the interpretation is complicated by the diffusion cooling effect. If a firmer theoretical basis for the interpretation could be established, the uncertainty of the result of the present method would be decreased appreciably. Extending the measurements to smaller values of the buckling κ_0^2 (i.e., to larger geometries), would also increase the accuracy in the determination of λ_a, since the influence of the uncertainty in extrapolation length and in the correction for the cubic term in κ_0^2 would then be decreased. On the other hand, the harmonic modes will increase when the buckling is decreased and an optimum will soon be reached.

Scott, Thomson and Wright[11] have recently performed measurements very similar to ours using a pulsed fission neutron source. Their value for the neutron-proton absorption cross section, 0.323 ± 0.008 barn, is somewhat lower than ours, and the measurements extend over a range of buckling below the lowest value investigated by us. As discussed above, the presence of harmonic modes may make the experimental results uncertain in this region.

THE DIFFUSION CONSTANT D_0

The diffusion constant D_0 has not previously been determined directly. However, from D_0 and θ we can calculate the diffusion length $L = (D_0\cdot\theta)^{\frac{1}{2}}$, which has been studied extensively in the past. We find for L the value 2.725 ± 0.03 cm at 22°C. The most reliable result of a direct measurement of the diffusion length is probably the value 2.763 ± 0.015 cm found by DeJuren and Rosenwasser.[12] The effect of the buckling on the neutron temperature was, however, not taken into account in their measurements. While in our measurement the buckling is positive and equal to the geometric buckling, in their measurements it will be negative and equal to the material buckling L^{-2}. Thus in their case the temperature of thermal neutrons is higher than the moderator temperature. Using Eq. (5) and the value of E, we find that their diffusion constant is larger by 2.6 percent and their diffusion length by 0.035 cm than for the case where the buckling is zero. Correcting for this effect leads to excellent agreement with our result.

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[8] Hamermesh, Ringo, and Wexler, Phys. Rev. **90**, 603 (1953).
[9] Harris, Muehlhause, Rose, Schroeder, Thomas, and Wexler, Phys. Rev. **91**, 125 (1953).
[10] G. von Dardel and A. W. Waltner, Phys. Rev. **91**, 1284 (1953).
[11] Scott, Thomson and Wright, Phys. Rev. **95**, 582 (1954).
[12] J. A. DeJuren and H. Rosenwasser, J. Research Natl. Bur. Standards **51**, 203 (1953).

We have also investigated the temperature dependence of the decay constant over the interval 10–35°C. The absorption probability λ_a is temperature-independent apart from a small effect due to the density change. From the results we can therefore calculate the temperature coefficient of the diffusion constant D. We find a value of $+(2.4\pm0.4)\times10^{-3}$ per °C in agreement with the value calculated from the scattering cross section curves. This value leads to a temperature coefficient of the diffusion length of +0.003 cm per °C, a factor of two smaller than the value quoted by Fermi.[13]

DIFFUSION COOLING

From the experimentally determined parameter E we can, knowing the temperature coefficient β of the diffusion constant, calculate the temperature decrease of the neutrons with increasing buckling. By Eqs. (4) and (6) we obtain in the first approximation:

$$T_n = T_0 - 83\kappa_0^2, \quad (\kappa_0^2 \text{ in cm}^{-2},\ T_n \text{ and } T_0 \text{ in °K}), \qquad (8)$$

which indicates a by no means negligible temperature difference between neutrons and moderator for the region of κ_0^2 investigated.

Using an independent method, we have demonstrated this decrease in neutron temperature by measurements of the transmission through a pyrex absorber.[1] The experimentally determined neutron temperatures are shown in Fig. 3 as a function of the buckling and are in good agreement with the theoretical curve predicted by Eq. (8). In interpreting the transmission measurements in terms of neutron temperature, the tables for perpendicular incidence given in reference 1 were used, and the neutron temperature for zero buckling was made to agree with the moderator temperature by normalizing the transmission data. It was shown that other assumptions concerning the angular distribution of the neutrons do not influence the result materially.

Finally, we can compare the experimentally determined value for the diffusion-cooling parameter E with the theoretical value given by (6). This expression is valid for the simple model of a monatomic gas consisting of nuclei of mass M. If we regard the water molecules as rigid scattering centers and neglect the bonds between them, we have $M = 18m$. For this case

[13] E. Fermi, Los Alamos Report LADC-255, 1946 (MDDC-320) (unpublished).

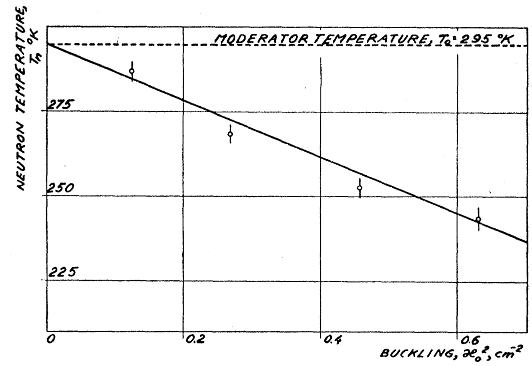

FIG. 3. Neutron temperature as a function of buckling calculated by Eq. (8) (solid line) and from transmission measurements.

we have $C = 0.1383$ according to reference 1. With $l_b = 0.2$ cm and $l_t = 0.45$ cm, we obtain the theoretical value 0.059 cm² for E which is considerably lower than the experimental value 0.20 ± 0.04 cm². This disagreement would indicate that the model of the monatomic gas is not very accurate for water of room temperature. As the temperature of the neutrons falls to the neighborhood of the Debye temperature, about 300°K for water,[14] we would expect the bonds between the molecules to become effective. This will tend to decrease the energy transfer per collision, and consequently the diffusion cooling will increase, as we observe experimentally.

On the other hand, the theoretical results for the monatomic gas model were in fairly satisfactory agreement with the measurements described in reference 1, on the approach to equilibrium of the neutron temperature, a process which is closely related to the diffusion-cooling effect. More theoretical and experimental work is obviously needed for the detailed understanding of the phenomena, and will probably also allow the limits of error on the results of the present investigation to be narrowed.

ACKNOWLEDGMENTS

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[14] N. Bjerrum, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 1 (1951).