

# KERNFORSCHUNGSZENTRUM

## KARLSRUHE

April 1967

KFK 600

Institut für Angewandte Kernphysik

Experimental Investigations of Angular Dependent Thermal Neutron Leakage Spectra

J.M. Kallfelz



GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.

KARLSRUHE

## Sonderdruck aus "Nukleonik", 9. Band, 6. Hejt, 1967, S. 273—281

Springer-Verlag, Berlin · Heidelberg · New York

## Experimental Investigations of Angular Dependent Thermal Neutron Leakage Spectra\*

#### J. M. KALLFELZ\*\*

Institut für Angewandte Kernphysik, Kernforschungszentrum Karlsruhe

#### Received October 17, 1966

Abstract. Using the pulsed source-chopper technique, asymptotic angular dependent neutron leakage spectra have been measured. The spectra for perpendicular leakage from quasi-infinite samples of  $H_2O$ , benzene and Dowtherm A are practically the same. These spectra and diffusion-cooled spectra from thin slabs of  $H_2O$  and benzene are well described by results of the program ADALS, using realistic scattering kernels. The measured angular dependence of leakage spectra from quasi-infinite samples of  $H_2O$  and plexiglass, which correspond to those of the Milne problem, is relatively weak. The average neutron energy for perpendicular leakage from  $H_2O$  is about 19% higher than that of a Maxwellian at moderator temperature,  $\overline{E}_M$ , while for emission parallel to the surface, the average energy is still about 10% higher than  $\overline{E}_M$ . This weak angular dependence is in agreement with the results of the program IMALS, using the Goldman-Nelkin scattering kernel.

#### 1. Introduction

The theoretical situation for the Milne problem for thermal neutrons has been described in a previous article [1]. It concerns the stationary distribution of neutrons in a semi-infinite, nonabsorbing medium in the half-space x > 0; the other half-space is vacuum. This paper will describe experimental investigations of the "leakage spectra",  $\varphi(x=0, E, \mu)$ ,  $\mu < 0$ , (where the notation of [1] has been used), and their agreement with theoretical results.

The first experimental investigations of Milne problem leakage spectra for thermal neutrons were made by BECKURTS [2] in H<sub>2</sub>O. The spectra investigated were asymptotic, that is they were measured when the thermalization process was complete. The results of [2] for neutrons leaking perpendicularly to the surface agreed well with calculations of KIEF-HABER [3]; this spectrum was about 18% "hotter"<sup>1</sup> than the spectrum in the middle of a large sample, which was a Maxwell distribution<sup>2</sup>. (This heating of the spectrum is due primarily to the energy dependence of the transport mean free path and boundary effects.) However, before the work reported here, no known measurements had been made of the angular dependence of asymptotic spectra. Angular leakage spectra reported by others [4] had a slowing-down component in the spectrum.

The theoretical solutions for this problem available when these measurements were started gave only very approximate, and rather varying results for this dependence (see [I]). Most solutions predicted a comparatively strong angular dependence, with the average energy of the spectrum decreasing with decreasing  $|\mu|$ , and a Maxwellian or near-Maxwellian for grazing emission ( $\mu = 0$ ). However, this result depended on the method of solution or simple scattering kernel used; no argument could be given for the necessity of a Maxwellian at grazing emission. Thus, there was considerable interest in an experimental determination of this dependence, which has been investigated in plexiglass and H<sub>2</sub>O.

For thin moderator slabs another phenomen, the diffusion cooling effect, also first measured directly in  $H_2O$  in [2], influences the form of the leakage spectra. This effect is caused by the fact that the leakage rate for high energy neutrons is greater than for those with low energies, so that the equilibrium spectrum in the slab middle is "cooler"<sup>2</sup> than a Maxwell distribution. This effect becomes particularly noticable when the leakage makes a significant contribution to the neutron losses. The spectrum in the slab middle can be considered as the "source" for the leakage spectrum, which, although still hotter than the spectrum in the side middle, is nevertheless cooler than that of the semi-infinite medium.

Milne problem and diffusion cooled perpendicular leakage spectra have been measured from  $H_2O$ , benzene, and Dowtherm A, three hydrogenous moderators with quite different frequency distributions

<sup>\*</sup> Based on work contained in a dissertation at the Technische Hochschule Karlsruhe. \*\* Present address — School of Nuclear Engineering,

<sup>\*\*</sup> Present address — School of Nuclear Engineering, Georgia Institute of Technology, Atlanta, Georgia, U.S.A. <sup>1</sup> The expressions "hotter" and "cooler" mean here that

<sup>&</sup>lt;sup>1</sup> The expressions "hotter" and "cooler" mean here that the neutrons of the spectrum have an average energy higher or lower, respectively, than the spectrum with which it is compared. <sup>2</sup> When a Maxwell distribution is referred to in this work,

 $<sup>\</sup>overline{2}$  When a Maxwell distribution is referred to in this work, one with the temperature of the moderator is meant, unless otherwise stated.

[5, 6], from which the scattering kernel is derived, to determine if the leakage spectra are very sensitive to the detailed form of the frequency distribution.

## 2. General Experimental Methods

2.1. Experimental Equivalence to Milne Problem. The experimental situation which is governed by the same form of the transport equation as that of the Milne problem can be obtained with a pulsed neutron source for moderators with 1/v-absorption, as the following considerations show. A sufficiently long time after the injection of a pulse of fast neutrons into a finite moderator, the energetic and spatial higher modes have decayed so that their contribution is negligible, and essentially only the time-asymptotic distribution is present. For the case of a one-dimensional geometry, the time-dependent neutron flux can then be written in the following form<sup>3</sup>:

$$\Phi(x, E, \mu; t) = \varphi(x, E, \mu) e^{-\alpha t}$$
(1)

where  $\alpha$  is the asymptotic decay constant, and the other symbols are as in [1].  $\varphi(x, E, \mu)$  is a solution of the transport equation in the following form:

$$\begin{bmatrix} \mu \frac{\partial}{\partial x} + \Sigma_i(E) - \alpha/v \end{bmatrix} \varphi(x, E, \mu) \\ = \int_0^\infty \int_{-1}^1 \Sigma(E', \mu' \to E, \mu) \varphi(x, E', \mu') dE' d\mu'.$$
(2)

 $\Sigma_t(E)$  is the total cross section and  $\Sigma(E', \mu' \to E, \mu)$  is the "scattering kernel". For the case of a 1/v absorber the absorption components of  $\Sigma_t(E)$  and  $\alpha/v$ cancel each other out, so that  $\Sigma_t(E) - \alpha/v = \Sigma_s(E) - \alpha_t/v$ , where  $\alpha_t$  is the part of  $\alpha$  caused by leakage. For large moderator samples,  $\alpha_t/v$  is negligible compared to  $\Sigma_s(E)^4$ , and Eq. (2) becomes that of the Milne problem, Eq. (1) of [1]. The diffusion cooling is also negligible for a large sample, so that the spectrum in the middle is a Maxwellian. The leakage spectra for this case are equivalent to those of the Milne problem.

2.2. Basic Methods and Apparatus. The time-offlight assembly used was that of BECKURTS and REICHARDT [2, 7]. The cadmium "shutter" described in [2] has been replaced by a cadmium chopper [8]. Basically, the method is as follows:

A 400 keV Cockcroft-Walton accelerator [9] delivers a short burst of 14 MeV neutrons through the (D, T) reaction. Some of these neutrons enter the moderator sample, where they diffuse and are thermalized. After a sufficient waiting time the chopper opens for a few microseconds, and lets a short pulse of neutrons leaking from the moderator enter the threemeter evacuated flight path. The BF<sub>3</sub> detector bank at the end of the flight path is connected to a TMC-256 time analyser. After several corrections, the leakage spectrum can be obtained from the time-of-flight spectrum of the analyser.

The appropriate waiting time,  $t_w$ , between the neutron pulse and chopper opening must consider the

flight time for the distance d between the extraction point and the chopper. This flight time causes a distortion of the spectrum, which can however be eliminated by a correction discussed below (par. 4.2).

A further factor which must be considered in the determination of the appropriate value for  $t_w$  is the time necessary for complete thermalization of the neutrons,  $t_{\rm ct}$ . For H<sub>2</sub>O, MÖLLER and SJÖSTRAND [10] have determined this time to be 25—30 µsec. The time allowed in these measurements for thermalization,  $\Theta_{\rm th}$ , is defined for an associated minimum velocity:  $v_{\rm min} = d/(t_w - \Theta_{\rm th})$ . For neutrons with  $v > v_{\rm min}$ , a longer time for thermalization was obviously allowed.  $v_{\rm min}$  was 1200 m/sec or less for all these measurements. For the H<sub>2</sub>O measurements,  $\Theta_{\rm th}$  was always at least 50 µsec.

For the other moderators considered here, no measurements of  $t_{\rm ct}$  are known. However, a rough estimate of its values can be obtained using the known value of  $t_{\rm ct}$  for H<sub>2</sub>O, and measured values of the second energy exchange moment  $M_2$  (see [11]). For the measurements in benzene and Dowtherm A, a value of  $\Theta_{\rm th} \geq 50$  µsec was used, while for plexiglass  $\Theta_{\rm th}$  was always at least 65 µsec. For all measurements spectra were measured at two different delay times, to insure that the thermalization was complete, i. e., that the spectrum form no longer changed with time.

Lifetime measurements were made to determine the time behaviour of the higher spatial modes.  $t_{v}$ was long enough so that the influence of these higher modes on the leakage spectra was negligible [11].

The moderator temperature was measured with a copper-constantan thermocouple. The average moderator temperature for a particular measurement,  $\overline{T}_0$ , was between 24° C and 25° C for all measurements except several of the perpendicular leakage spectra of paragraph 5, which were measured at about 27° C, and corrected to 24° C. The temperature during a particular measurement was constant to within  $\pm 1^{\circ}$ C about  $\overline{T}_0$  for the angular dependent spectra of paragraph 6, and to within  $\pm 1.5^{\circ}$  C for the other measurements.

#### 3. Special Methods for Particular Measurements

3.1. Perpendicular Leakage Spectra. For the measurements of the perpendicular leakage spectra from  $H_2O$ , benzene, and Dowtherm A, cadmium-covered aluminium containers approximately  $30 \times 30$  cm, with thicknesses varying from 15 cm to 3 cm were used. The neutrons were extracted from the center of one of the  $30 \times 30$  cm sides. The distance d between the extraction point and chopper was 2 cm, the minimum value possible for this assembly.

3.2. Angular Spectra from Plexiglass. As previously mentioned, there was particular interest in the measurement of the spectrum at the grazing angle ( $\mu = 0$ ). The measurement of this spectrum for the ideal case is obviously impossible, since the effective area of an extraction window is proportional to  $\mu$ . For liquid moderators the effective container thickness (in the extraction direction) approaches  $\infty$  for  $\mu \rightarrow 0$ . The ideal case can be more closely approached for a solid moderator without container sides, which complicate the situation. For this reason measurements at almost-grazing emission ( $\bar{\mu} = -0.03$ ) were carried out

<sup>&</sup>lt;sup>3</sup> Presuming that a discrete energy primary mode exists. For the moderator samples considered in this work, this is the case.

<sup>&</sup>lt;sup>4</sup> For instance, for a  $12 \times 22 \times 22$  cm sample of H<sub>2</sub>O,  $\alpha_f \approx 3500 \text{ sec}^{-1}$ . For neutrons with the most probable energy of a Maxwell distribution,  $v = 2.2 \cdot 10^5$  cm/sec, and  $\Sigma_s(E) \approx 3.8$  cm<sup>-1</sup>.

on plexiglass, a hydrogenous moderator whose total cross section is similar to that of  $H_2O$  [12].

Fig. 1 shows the assembly used for this experiment. The moderator sample consisted of two  $10 \times 15 \times 15$  cm plexiglass blocks, 10 mm apart. Two blocks were used to increase the neutron intensity; for neutrons whose energy was above the cadmium cut-off, the probe was effectively one  $20 \times 15 \times 15$  cm block, so that the leakage during the slowing down was smaller. The cadmium between the two blocks prevented an exchange of thermal neutrons through the extraction windows, which would have caused a deviation from the Milne problem situation.

The extraction window was designed for a minimum distortion of the Milne problem geometry. For the first 3 cm from the front and rear sides of the window, (in the extraction direction), the cadmium had a thickness of only 0.1 mm. Therefore, by milling the plexiglass only 0.1 mm, the surface of the cadmium could be brought in the same plane as the surface of the plexiglass in the extraction window. Thus, neutrons in angles down to  $\mu = 0$  could leave the block; the extracted neutrons were collimated so that the maximum value for  $|\mu|$  was 0.06.

For the measurement at perpendicular leakage  $(\mu = -1.0)$  the spectrum was extracted from a  $15 \times 15$  cm side of a  $10 \times 15 \times 15$  cm block. Furthermore, the spectrum in the middle of this block was measured using a 1.3 cm diameter extraction canal, which was perpendicular to a  $15 \times 15$  cm side.

3.3. Angular Spectra from  $H_2O$ . For the angular measurements in H<sub>2</sub>O down to  $\mu = -0.2$ , the assembly shown in Fig. 2 was used. The fixed pivot points were so located that the moderator sample turned on an axis which ran through the middle of the extraction area on the H<sub>2</sub>O surface. Two sample sizes,  $15 \times 30 \times 30$  cm, and  $12 \times 22 \times 22$  cm were used; both samples approximated well an infinite medium. The smaller sample had the advantage that for large values of the angle  $\delta$  (shown in Fig. 2) the moderator could be brought nearer to the chopper, and that the higher spatial modes delayed more rapidly. Using two different geometries with different distances from the chopper served also as a check to insure that the influence of the moderator size was negligible, and that the effect of the flight time to the chopper was properly corrected for. For the smaller sample, all measurements up to  $\delta = 78.5^{\circ}$  ( $\mu = -0.2$ ) were made with a chopper-extraction point distance d of 13.3 cm.

Since the neutron generator could not be moved, the entire assembly on which the flight path, chopper, and moderator were mounted was moved in the vertical and horizontal direction after each measurement, so that the target orientation with relation to the  $H_2O$ sample was the same for all angles.

To determine the relative intensity of the leakage spectra at each angle, the normal monitor system was not sufficient, since slight changes in the position of moderator, target, and shielding, relative to the location of the three independent normal monitors, for different angles would have a slight influence on the monitor counting rate. For this reason, a BF<sub>3</sub> counter was fixed to the back of the moderator sample opposite the extraction surface, as shown in Fig. 2. The intensity of lifetime measurements with this counter

18\*

at each angle was a measure of the neutrons which were actually injected into the moderator.

Grazing emission  $(\mu = 0)$  is a special case, which can not be measured in the ideal situation; the problems involved were already discussed in conjunction with the measurements in plexiglass. For the measurements in H<sub>2</sub>O a 2.4 cm wide extraction canal was used which ran parallel to and slightly under the water surface, as shown in Fig. 3. Thus at the extraction point



Fig. 1. Assembly for the measurement of the leakage spectrum from plexi-glass at almost-grazing emission ( $\ddot{\mu}=-0.03$ )



Fig. 2. Assembly for the measurement of angular-dependent leakage spectra from  $\rm H_2O$ 



Fig. 3. Assembly for the measurement of the leakage spectrum from  $\rm H_2O$  at grazing emission

there was a slight step in the H<sub>2</sub>O surface, which formed a finite emission area for the neutrons at  $\mu = 0$ . The penetration depth of the canal below the surface should be kept at a minimum, for several reasons. The step in the H<sub>2</sub>O surface is a perturbation of the ideal situation; furthermore, for the nonperturbed case the change in the spectrum form due to the surface dies off quite rapidly with increasing distance from the surface [12, 13]. The minimum step depth was determined by the smallest intensity that could still give an acceptable measuring time and spectrum-to-background ratio. Spectra were measured for extraction steps of 4 and 2 mm. Because of the low intensity, the measurement for 2 mm had a very long measuring time (about 160 hours). In order to determine the background as exactly as possible, the spectrum and background measurements were altered about every two hours.

Except for the last 8 mm from the extraction point, the extraction canal was lined with cadmium, to insure that only neutrons from the extraction point could pass through the chopper. The wall thickness of the canal was milled to  $\frac{1}{2}$  mm for the last three centimeters from the extraction end, to keep the canal perturbation as small as possible.

#### 4. Evaluation of Measurements

In order to obtain the energy-dependent neutron spectrum at the extraction point,  $\varphi(0, E, \mu)$ , from the counting rate of the time analyzer, Z(t), several corrections must be applied; an IBM 7074 program



Fig. 4. Terminology for the correction of the spectra for the length of the extraction window



Fig. 5. Quotient of two leakage spectra measurements from  $H_2O$ , with different lengths 2b of the extraction windows. 2b = 4.0 cm and 2.7 cm for  $\varphi_1$  and  $\varphi_2$ , respectively

was written for this purpose. These corrections are for subtraction of background, scattering and absorption in the flight path, sensitivity of the detector bank, conversion from the time scale to the energy scale, energy resolution, and the flight time to the chopper. These corrections have been discussed by BECKURTS [2, 14] and the author [11], and only the latter two will be mentioned briefly here.

4.1. Energy Resolution. Because of the finite width of the chopper opening and the time analyser channel  $(\varDelta t' = 32 \ \mu sec)$ , a correction for the energy resolution is necessary. The relation between the corrected value  $Z_0(t)$  and the measured value Z(t) is given through the following expression ([15], p. 323):

$$Z_0(t) = Z(t) \left[ 1 - \frac{Z''(t)}{Z(t)} \frac{\Delta^2}{2} \right]. \tag{4}$$

 $\Delta^2$  is a constant which depends on the resolution function of the chopper and the channel width of the time analyser. For our case the resolution function is well approximated by an isosceles triangle with a base width of  $2 \cdot \Delta t \ (\Delta t = 45 \text{ or } 60 \ \mu\text{sec})$  and  $\Delta^2 = (1/6) \cdot (\Delta t)^2 + (1/3) \cdot (\Delta t')^2$ . The corrections which were calculated using the expression in Eq. (4) were negligible for E < 0.14 eV, and were less than 18% for E < 0.20 eV. 4.2. Correction for the Time of Flight to Chopper. As mentioned in [2], for an asymptotically decaying flux the relation between the energy distribution of neutrons at the chopper  $\varphi_d(E)$ , and at the extraction point,  $\varphi_0(E)$ , is given by:

$$\varphi_d(E) = \varphi_0(E) e^{\alpha d/v} \tag{5}$$

were d is the distance from the moderator to the chopper, and  $\alpha$  is the known asymptotic decay constant [see Eq. (1)].  $\varphi_0(E)$  signifies  $\varphi(x, E, \mu)$  from Eq. (1) for x = 0 and some particular value of  $\mu$ .

In assemblies for the measurement of perpendicular leakage spectra, the distance d has the same value  $(d_0)$  for all neutrons, and the correction is given by the factor  $\exp(-\alpha d_0/v)$ . For  $\delta \neq 0^\circ$ , however, the extraction area has a finite projection on the extraction axis (see Fig. 4), so that the distance from different positions on the extraction area is different  $(d_0 + y \sin \delta)$ . Then, analog to Eq. (5), the energy distribution at the chopper is

$$I_d(E) = \operatorname{const} \int_{-b}^{b} \varphi_0(E, y) e^{\alpha \langle d_0 + y \sin \delta \rangle / v} \cos \delta \, dy.$$
 (6)

y is the coordinate parallel to the moderator surface and perpendicular to the rotation axis (see Fig. 4). Presuming that the y dependence of  $\varphi_0(E, y)$  is given by a cosine distribution  $[\varphi_0(E, y) = \varphi_0(E) \cos B_y y]$ then from Eq. (6) follows:

where

$$\varphi_0(E) = I_d(E) e^{-\alpha d_0/v} H(E) / \text{const}$$
(7)

$$H(E) = \left[\int_{-b}^{b} e^{\frac{\alpha}{v} y \sin \delta} \cos B_{y} y \cos \delta \, dy\right]^{-1},$$

and  $B_y^2$  is the buckling in the y direction. This expression is the same as for the case of perpendicular leakage, Eq. (5), except for the factor H(E). The somewhat involved expression for H(E) was calculated in the evaluation computer program. To keep this correction to a minimum, the windows in the cadmium were made as short as possible, consistent with an acceptable measuring time. For  $\delta = 0^{\circ}$ ,  $b = b_0 = 4$  mm. For the other angles, b was determined by  $b = b_0/\cos \delta$ , so that the maximum value of b, for  $\delta = 78.5^{\circ}$ , was 2.0 cm. For all measurements,  $H(E) \cdot 2b_0$  was in the interval between 1.00 and 1.01, and had therefore a very small influence on the form of the spectrum.

The assumption of a cosine distribution for  $\varphi_0(E, y)$ , which led to Eq. (7), is not a priori valid, since gradients in the *y*-direction could influence the form of the leakage. Calculations showed, however, that their influence was negligible for the small values of *b* used in these experiments.

In order to check the accuracy of this correction, for the extreme case of the  $\delta = 78.5^{\circ}$  measurement two different window lengths (2b = 2.67 and 4.0 cm)were used<sup>5</sup>. In Fig. 5 the quotient of the two resulting spectra is shown; it can be seen that they are identical, within experimental accuracy. Therefore the influence of the window length on the corrected spectrum is negligible.

<sup>&</sup>lt;sup>5</sup> The final results given are for the smaller window.

9. Bd., Heft 6, 1967

## 5. Results for Perpendicular Leakage Spectra

5.1. Quasi-infinite Medium Spectra. Fig. 6 shows the perpendicular leakage spectra ( $\mu = -1.0$ ) from quasi-infinite ( $15 \times 30 \times 30$  cm) samples of H<sub>2</sub>O, benzene, and Dowtherm A. The wings in this figure and all other spectra results contain contributions due to the estimated errors in the energy resolution correction and normalization of the background curve, but the major contribution is the statistical error, represented by the standard deviation. The spectrum for H<sub>2</sub>O, Fig. 6—I, is about 18% hotter than a Max-

well distribution (dotted line) and agrees well with the experimental results of BECKURTS [2] for this case. The results for Dowtherm A and benzene, shown in Fig. 6-II and 6-III, are practically the same as those of  $H_2O$ , as can be seen in Fig. 6-IV and 6-V, where the various results are compared. Thus the perpendicular leakage spectra are apparently relatively insensitive to the detailed differences in the frequency distribution of these moderators, caused by varying chemical binding. This result agrees with those of BIGHAM [12], who made integral measurements of the temperature changes of spectra near an absorber boundary for four hydrogenous moderators, and noted no significant differences because of chemical binding effects.

The solid curves of Fig. 6—I—III are theoretical results of the program ADALS of KLADNIK [16]. For  $H_2O$  the Goldman-Nelkin kernel<sup>6</sup> [17] was used in the calculations, while for the organic moderators the kernels of GLÄSER [5] derived from experimental scattering laws were used. For these and all other theoretical results shown in this paper, the kernels include linearly anisotropic scattering. As can be seen, the agreement with the measurements is satisfactory in all cases.

5.2. Diffusion Cooled Spectra. Fig. 7 shows results for the diffusion-cooled spectra. The results for the 3 cm and 2.1 cm slabs of  $H_2O$ are from REICHARDT [18]. The effect of diffusion cooling is evident, as can be seen in Fig. 7—V, where the leakage spectrum from

the 3.1 cm benzene slab is compared with that of the quasi-infinite medium. The maximum of the spectrum for the thin slab is at about 0.025 eV, whereas the maximum for the quasi-infinite medium is at about 0.03 eV.

Just as for the case of the thick slabs, there is no fundamental difference between the spectra for  $H_2O$ and benzene. For the same dimensions, however, the cooling for benzene is greater than for  $H_2O$ . Fig. 7—VI shows that the spectra for the 2.1 cm  $H_2O$  slab and the 3.1 cm benzene slab are approximately the same, that is both spectra have about the same cooling in relation to the spectra for a quasi-infinite moderator. This is primarily due to the lower hydrogen density  $(N_{\rm H})$  for benzene, and can be roughly explained as follows. Since, as mentioned above, the spectrum in the middle of the slab can be thought of as a source for the neutron leakage, a cooling of this source

 $^{6}$  A free gas correction for oxygen was added to this kernel for all calculations.

results in a cooling of the leakage spectra. Using the assumption that the spectrum in the middle can be represented by a Maxwell distribution with a temperature T, the following expression can be derived ([15], p. 215):

$$\frac{T-T_0}{T_0} = \frac{-\overline{D}B^2 \left(1+2\frac{d\ln D}{d\ln T}\right)}{N_H M_2}$$

where  $T_0$  is the moderator temperature,  $B^2$  is the sample buckling, and  $\overline{D}$  is the diffusion coefficient.



Fig. 6. Perpendicular leakage spectra from a large sample  $(15 \times 30 \times 30 \text{ cm})$  at room temperature  $(24^{\circ} \text{ C})$ . The vertical scale is arbitrary. (-----) Theoretical calculations with ADALS [16]. I:  $\text{H}_{20}$  (...); Maxwell distribution at the moderator temperature (---); II: Dowtherm A; III: benzene; IV:  $\text{H}_{20}$  (...) compared with Dowtherm A (+++); V:  $\text{H}_{20}$  (...) compared with benzene (+++)

Furthermore, the following expression can be developed for C, the diffusion cooling coefficient:

$$C = \frac{\overline{v} \, \overline{D}^2}{2 \, N_H M_2} \left( 1 + 2 \, \frac{d \ln \overline{D}}{d \ln T} \right)^2.$$

Table 1 shows experimental values for  $M_2$  for  $H_2O$ and benzene. Although the experimental values do not agree with each other particularly well, based on the average values it appears that  $M_2$  for the two moderators can be assumed to be roughly the same. For this assumption, the ratio  $(T - T_0)/T_0$  for the two

Table 1. Values for the second energy exchange moment,  $M_2$ 

(in barn)				
	H <sub>2</sub> O	Ref.	Benzene	Ref.
From pulsed neutron measurements	31	[ <i>21</i> ]	$28\pm 6$	[20]
From scattering law	46.5	[5]	52	[5]
Average Value	39		40	



Fig. 7. Perpendicular leakage spectra from thin slabs at 24° C. The vertical scale is arbitrary. In I to V the solid curves are theoretical calculations with ADAIS [16]. I:  $H_2O$ , 4.2 cm slab; II:  $H_2O$ , 3 cm slab, from REICHARDT [18]; III:  $H_2O$ , 2.1 cm slab, from REICHARDT [18]; IV: benzene, 4.2 cm slab; V: benzene, 3.1 cm slab (...); empirical curve fitted to the points in Fig. 6-V (---); VI:  $H_2O$ , 2.1 cm slab (...) compared with benzene, 3.1 cm slab (+++); Maxwell distribution at moderator temperature (\_\_\_\_\_)



slabs varies relative to each other as their values  $B^2 \cdot \sqrt{C/N_H}$ . For benzene,  $\sqrt{C/N_H}$  is about  $2.5 \pm 0.4$  times as large as that for H<sub>2</sub>O [19, 20]. On the other hand, the value for  $B^2$  for the H<sub>2</sub>O slab is about 2.1 times as large as for the benzene slab. Hence one would expect roughly the same cooling for both slabs.

The spectra in Fig. 7—VI can be fitted fairly well by a Maxwell distribution with the moderator temperature, shown by the solid line. The spectrum in the middle of the slab would however be much cooler than a Maxwell distribution, so that the heating of the leakage spectrum with relation to the "source" spectrum in the slab middle is present, as expected.

The solid curves in Fig. 7—I—V are also ADALS results, using the Goldman-Nelkin kernel for  $H_2O$  and the Gläser kernel for benzene. As can be seen, ADALS describes the diffusion cooling effect quite well; for both benzene and  $H_2O$  [the calculated results agree well with experiment. Thus the accuracy of the ADALS calculations for the case of perpendicular leakage is good for both quasi-infinite and thin slabs.

## 6. Results for Angular Dependent Milne Problem Spectra

6.1. Plexiglass. The spectrum from the middle of the plexiglass block was measured to determine how much it varied from the Maxwell distribution of the ideal quasi-infinite case. As can be seen in Fig. 8—I, this spectrum is well fitted by a Maxwell distribution with  $T = 23^{\circ}$  C, which is about 1.3% lower than the temperature of the moderator. The diffusion cooling is therefore very slight, and the leakage spectra are essentially those of the ideal Milne problem case.

Fig. 8—II shows the perpendicular leakage spectrum from the surface, compared with a curve representing the same spectrum for  $H_2O$  and benzene. Just as for the previously discussed organic moderators, the spectrum for plexiglass is practically the same as that for  $H_2O$ .

The spectrum for almost-grazing emission ( $\bar{\mu} = -0.03$ ) is shown in Fig. 8—III. Although the points of this spectrum scatter somewhat, due to the low intensity, it is evident that this spectrum is much hotter than a Maxwellian, given by the dotted line. One can also see from Fig. 8—II that the spectrum for near-grazing emission is cooler than that for perpendicular emission.

6.2.  $H_2O$ . The results of the measurements in  $H_2O$  for  $\mu$  values from -1.0 to -0.2 with the smaller sample  $(12 \times 22 \times 22 \text{ cm})$  are shown in Fig. 9. The results with the larger sample were the same, within the experimental accuracy. The solid curves are results from IMALS [1] using the Goldman-Nelkin kernel<sup>6</sup>. The values of the average neutron energy,

Fig. 8. Spectra from plexiglass,  $10 \times 15 \times 15$  cm sample, at room temperature. The vertical scale is arbitrary. I: Spectrum from the sample middle  $(\dots)$   $T_0 = 27^{\circ}$  C; Maxwell distribution at  $T = 23^{\circ}$  C (——); II: Perpendicular leakage spectrum  $(\dots)$   $T_0 = 24^{\circ}$  C; Empirical curve fitted to the points in Fig. 6—V (——); solid curve from III (——); III: Spectrum at almost-grazing emission ( $\bar{\mu} = -0.03$ )  $(\dots)$   $T_0 = 24^{\circ}$  C; Empirical curve fitted to the points in Fig. 11—I (——); Maxwell distribution at  $T = 24^{\circ}$  C

 $\overline{E}(\mu)^{7}$ , for these curves are given in Table 4 of [1]. The agreement between the theory and experiment in Fig. 9 is excellent. The experimental values for  $\overline{E}(\mu)$ agree with those of the theoretical curves within the experimental accuracy  $(\pm 0.02 \ \overline{E}_{M})$  in all cases<sup>8</sup>. ( $\overline{E}_{M}$ is the average value of a Maxwell density distribution.)



Fig. 9. Angular dependent leakage spectra from a  $12 \times 22 \times 22 \operatorname{cm} H_2O$ sample at 24° C. The vertical scale is arbitrary. (-----) Theoretical calculations with IMALS [1] for the Goldman-Nelkin kernel. I:  $\mu = -1.0(\cdots)$ ; Empirical curve fitted to the points in Fig. 11-1 (---); II:  $\mu = -0.7$ ; III:  $\mu = -0.5$ ; IV:  $\mu = -0.3$ ; V:  $\mu = -0.2$ 

The change in the experimental spectra down to  $\mu = -0.2$  is quite small.  $\overline{E}(\mu)$  is  $1.19 \pm 0.02 \overline{E}_M$  and  $1.13 \pm 0.02 \overline{E}_M$  for  $\mu = -1.0$  and -0.2, respectively. This change is so small that it is hard to detect in Fig. 9. That the spectra become cooler with decreasing  $|\mu|$  can best be seen from the quotients of different results. The ratio of the results for the two extreme values,  $\mu = -1.0$  and -0.2, are shown in Fig. 10—I. The two IMALS spectra from which the theoretical curve was obtained have a difference in their average energy,  $\Delta \overline{E}$ , of 0.053  $\overline{E}_M$ . In Fig. 10—II and 10—III

the results for the two extreme  $\mu$  values are compared with that of an intermediate value,  $\mu = -0.5$ . Both theoretical curves have a  $\Delta \overline{E}$  of about  $0.025 \overline{E}_M$ . Although the points scatter somewhat, the experimental results agree with the curves satisfactorily, within the experimental accuracy (about  $\pm 0.01 \overline{E}_M$ in  $\Delta \overline{E}$ ). As can be seen from Fig. 10—IV, the difference between the spectra for  $\mu = -0.3$  and -0.2 lies within the accuracy of the experiment.



Fig. 10. Quotients of the leakage spectra from Fig. 9 at various  $\mu$ -values. The vertical scale is arbitrary. (\_\_\_\_\_) Theoretical calculations with IMALS [1] for the Goldman-Nelkin kernel. I:  $\mu_i = -0.2$ ;  $\mu_k = -1.0$ ; III:  $\mu_i = -0.2$ ;  $\mu_k = -0.2$ ;

Fig. 11 shows the results for the special case of  $\mu = 0$ , for extraction steps of 4 and 2 mm; these results are the same, within the experimental accuracy. Due to the low intensity, the measurements are rather inaccurate for  $E \gtrsim 0.1$  eV. Because of the previously mentioned problems associated with the depth of the extraction step, these results must be interpreted with care. However, since the difference between the two spectra is minimal, it was felt justified to extrapolate, and assume that the 2 mm measurement gives essentially the form of the spectrum for the ideal case, i. e., no extraction step.

The comparison in Fig. 11—I shows that the  $\mu = 0$  spectrum is somewhat cooler than for  $\mu = -1.0$ . The grazing angle spectrum is however considerably hotter than a Maxwellian, as is shown in Fig. 11—II. The points fit the theoretical IMALS result reasonably well. The experimental value for  $\overline{E}(0)$  is  $1.10 \pm 0.03 \, \overline{E}_M$ . The grazing angle spectrum for H<sub>2</sub>O is the same as the near-grazing spectrum for plexiglass, within the experimental accuracy (see Fig. 8—III).

 $<sup>{}^{7}\</sup>overline{E}(\mu)$  is averaged over the neutron density. Since the form of the leakage spectra is considerably different from that of a Maxwellian in a large energy range, the average energy values relative to that of a Maxwellian are different if the averaging is done over the flux. For instance, the average energy of the theoretical curve in Fig. 9—I is 17.9% higher than that of a Maxwellian for averaging over the neutron density, and 13.3% for averaging over the flux.

<sup>&</sup>lt;sup>8</sup> To determine the values of  $\overline{E}(\mu)$ , the experimental results were extrapolated at low and high energies, using the theoretical curves.





Fig. 11. Leakage spectra from  $H_2O$  at grazing emission ( $\mu = 0$ );  $T_0 = 24^\circ$  C. The vertical scale is arbitrary. (----) Theoretical calculation with IMALS [1] for the Goldman-Nelkin kernel. I: Extraction step in H<sub>2</sub>O surface of 4 mm (...); Empirical curve fitted to the points for  $\mu = -1.0$ (Fig. 9-I) (----); II: Extraction step in H<sub>2</sub>O surface of 2 mm (...); Maxwell distribution at moderator temperature (---)



Fig. 12. Relative intensity of the energy-integrated leakage,  $\varphi(\mu)$ .  $\triangle$  Measurements from a  $12 \times 22 \times 22$  cm H<sub>2</sub>O sample; (\_\_\_\_\_) Theoretical results from IMALS [1] for the Goldman-Nelkin kernel

Table 2. Relative intensity  $\star$  of the energy-integrated flux,  $\varphi(\mu) = const \int_{0}^{\infty} \varphi(E,\mu) dE$  and the monoenergetic solution,  $\varphi(\mu)_{ME}$ 

	$\operatorname{const} \cdot \int\limits_{0}^{\infty} \varphi(E,\mu) dE$		$\varphi(\mu)_{ME}$	
	Experiment	Theory IMALS [1]	PLACZEK [22]	
-1.0	0.974 + .025	1.000	1.000	
-0.7	$0.816 \pm .025$	0.816	0.816	
-0.5	0.715 + .02	0.689	0.692	
- 0.3	0.572 + .017	0.561	0.565	
-0.2	0.524 + .017	0.495	0.499	
0.0		0.341	0.340	

\* The experimental values were normalized to those of the theory at  $\mu = -0.7$ .

The relative intensity of the spectra between  $\mu = -1.0$  and -0.2 was measured, in order to determine the relative intensity of the energy-integrated flux,  $\varphi(\mu) = \text{const} \int_{0}^{\infty} \varphi(E,\mu) dE$ .  $\varphi(\mu)$  decreases almost linearly with  $|\mu|$ , and agrees fairly well with the IMALS results for the Goldman-Nelkin kernel, given in Table 2 and Fig. 12. The values for the solution of PLACZEK [22] for the monoenergetic case with isotropic scattering, also given in Table 2, agree with the IMALS values within 1% for all cases.

To summarize the results for the angular variation, the dependence of the spectra on  $\mu$  was rather weak,  $\overline{E}(\mu)$  varying between about 1.19  $\overline{E}_M$  and 1.10  $\overline{E}_M$ between perpendicular and grazing emission. The grazing angle spectrum was considerably hotter than the Maxwellian or near-Maxwellian predicted by many of the earlier theories, which used simple scattering kernels.

Thus the results of IMALS for realistic scattering kernels, which for  $H_2O$  predicted just the behaviour observed experimentally, are substantiated. This weak angular dependence is understandable physically; as explained in [1], the strength of the dependence varies with the thermalization power of the moderator.

Acknowledgements. The author would like to thank Prof. K. H. BECKURTS for suggesting these investigations, and for many valuable discussions. I am grateful to Prof. K. WIRTZ for the opportunity to use the experimental facilities of his institute. I am particularly indebted to Dr. W. REICHARDT for his advice and help in all phases of the investigations. Dr. W. EYRICH gave much assistance during the experiments.

I would like to thank all at Karlsruhe who helped to make my stay in Germany memorable and rewarding.

References. [1] KALLFELZ, J. M., and W. REICHARDT: An improved variational solution to the energy-dependent Milne problem for realistic scattering kernels. Nukleonik 9, 148 (1967). — [2] BECKURTS, K. H.: Z. Naturforsch. 16a, 611 (1961). — [3] KIEFHABER, E.: Nukleonik 6, 262 (1964). — [4] SINCLAIR, R. N., and P. J. WILLIAMS: In: Pulsed Neutron Research, p. 581. Vienna: I. A. E. A. 1965. — [5] GLÄSER, W., and K. H. BECKURTS: Nuclear Sci. and Eng. 20, 235 (1964). — [6] HAYWOOD, B. C., and I. M. THORSON: BNL 719, vol. 1, p. 26. Brookhaven National Laboratory 1962. — [7] BE-CKURTS, K. H., and W. REICHARDT: In: Neutron time-offlight Methods (J. SPAEPEN, ed.), p. 239. Brussels: Euratom 1961. — [8] REICHARDT, W.: Untersuchungen zur Neutronenthermalisierung in Zirkonhydrid und Eis. Diss. Techn. Hochschule Karlsruhe 1966. — [9] EYRICH, W.: Nukleonik 4, 167 (1962). — [10] MÖLLER, E., u. N. G. SJÖSTRAND: Arkiv Fysik 27, 501 (1965). — [11] KALLFELZ, J. M.: Das Milne-Problem für Thermische Neutronen in Wasserstoffhaltigen Moderatoren. Diss. Techn. Hochschule Karlsruhe 1966. — [12] BIG- HAM, C. B.: Nuclear Sci. and Eng. 21, 106 (1965). — [13] RENZ, K.: Untersuchung des energieabhängigen MILNE-Problems für thermische Neutronen in Wasser. Diss. Techn. Hochschule Aachen 1966. — [14] BECKURTS, K. H.: Untersuchung des Diffusionskühlungseffektes durch direkte Beobachtung von Neutronenspektren. Habil.-Schr. Techn. Hochschule Karlsruhe 1960. — [15] BECKURTS, K. H., u. K. WIRTZ: Neutron Physics. Berlin-Göttingen-Heidelberg: Springer 1964. — [16] KLADNIK, R.: Nuclear Sci. and Eng. 23, 291 (1965). — [17] FEDERIGHI, F. D., and D. T. GOLDMAN: KER-NEL and PAM-Programs for Use in the Calculation of the

Thermal Scattering Matrix for Chemically Bound Systems. KAPL-2225, Knolls Atomic Power Laboratory 1962. — [18] REICHARDT, W.: Private communication. — [19] ARAI, E., and M. KÜCHLE: Nukleonik 7, 416 (1964). — [20] KÜCHLE, M., and G. KUSSMAUL: Nukleonik 6, 329 (1964). — [21] KÜCHLE, M.: Nukleonik 2, 131 (1960). — [22] PLACZEK, G.: Phys. Rev. 72, 556 (1947). Dr. J. M. KALLEELZ

Dr. J. M. KALLFELZ School of Nuclear Engineering Georgia Institute of Technology Atlanta, Georgia, U.S.A

Druck der Universitätsdruckerei H. Stürtz AG., Würzburg