Neutron thermalization in water using spherical geometry

Mohamed Shawky Fahim Nassar

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NEUTRON THERMALIZATION IN WATER USING SPHERICAL GEOMETRY

by

Mohamed Shawky Fahim Nassar

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Major Subject: Nuclear Engineering

Approved:

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In Charge of Major Work

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Head of Major Department

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Dean of Graduate College

Iowa State University Of Science and Technology Ames, Iowa 1967
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I. INTRODUCTION

The subject of neutron thermalization concerns the manner in which the "neutron gas" comes into thermal equilibrium (if it ever does!) with the medium which contains it. Its analysis shows a nice interplay between theories of neutron transport and theories of the solid and liquid state, and is also reminiscent of the classical kinetic theory of gases. More precisely, it resembles the "foreign-gas" problem, where a small number of gas molecules is introduced into a large collection of molecules already in equilibrium at some temperature.

The thermalization problem might be compared with a particularly simple, linearized version of the kinetic theory of gases, were it not for the feature of chemical binding. In all but the simplest models, the atoms of the moderator interact with one another and the complicated motions which result produce a complex scattering pattern in the laboratory system.

Recently, the subject has assumed various and interesting fields of neutron physics. One of these, which received extensive theoretical and experimental considerations, is the field dealing with the integral part of the thermalization problem and which is the main interest of this thesis. In the integral experiments the concentration is on the temporal
change of the energy-integrated flux rather than on the details of neutron spectra.

The most eminent technique for these experiments is the pulsing technique introduced by Von Dardel (54). The theory behind the method is summarized in a consideration of a burst of fast neutrons injected in the moderator. When a sufficiently long time has elapsed after the initiation of the pulse the flux decays as \( \exp(-\lambda t) \) where \( \lambda \) is the decay constant. A semi-log. plot of the flux, obtained by a "1/V" detector, versus time should give a straight line with a slope equal to \(-\lambda\).

von Dardel and Sjöstrand (55) found that \( \lambda \) could be expressed in terms of power series of the square of the geometric buckling, \( B_g^2 \):

\[
\lambda = \alpha + D_o B_g^2 + CB_g^4 + \ldots
\]

where \( \alpha \) is the decay constant which would be observed in an infinite medium, \( D_o \) is the diffusion coefficient and \( C \) is the diffusion cooling constant that describes the cooling phenomenon due to the preferential leakage of the high-energy neutrons from a finite medium. Nelkin (38) has obtained these coefficients by a variational approach. He used the "neutron temperature" as a variational parameter. Singwi (48), and Purohit (43), applying the diffusion approximation, have determined \( D_o \) and \( C \) with the help of the Laguerre expansion.
The experimental values of $a$ and $D_0$ were obtained with good accuracy and the values measured by different experimenters are in agreement with each others and with the simplest model of diffusion theory. On the other hand, the data reported for $C$ showed large discrepancies.

All pulse data available today have been obtained from multi-dimensional systems whose methods of analysis are, undoubtedly, complicated. To help in resolving the inconsistencies of the experimental values of the diffusion cooling constant, further work with spherical geometries is needed. This need has been emphasized by Gelbard et al. (24) because this shape would be amendable to theoretical interpretation and might yield more information on the thermalization parameters.

The purpose of this thesis is to study the feasibility of pulse experiments in spherical geometry. The study is based on both theoretical and experimental investigations. The former is treated in Chapter II while the latter is dealt with in Chapter V.

The theoretical model of the present work is the $P_1-L_1$ approximation developed in section II-A. In the next section the eigenvalue problem is defined and the behavior of the decay constants, as functions of the space eigenvalues, is studied. This study is felt to be of interest for two reasons. First, Travelli and Calame (52) pointed out that the plot of
the fundamental time-eigenvalue, as obtained by a 4-group $P_1$ approximation, versus the space eigenvalue gave a curve with a positive curvature at the origin in disagreement with the experimental results. The study is aimed to clarify the nature of this contradiction. Second, some understanding of the eigenvalue spectrum is needed to test the feasibility of the commonly used expansion of the decay constant in power series of the geometric buckling. The results inferred from this study are used in verifying the treatment in section II-C where the various diffusion parameters are derived.

In section II-D the space-energy dependence of the eigenfunctions is examined together with the concept of a unique buckling. Equations for the computation of the diffusion-cooled neutron spectra for the asymptotic and the transient distributions are developed. These equations are made use of, in section II-E, to calculate the spectrum-weighted average energy and effective buckling for a sphere.

The space-dependence of the extrapolation distances for a spherical geometry are compared with those of an "equivalent slab" in section II-F. The Marshak's boundary condition is used for the system-vacuum interface.

In the experimental part two different investigations of the die-away experiments are described. Section V-A deals with the determination of the diffusion parameters. The extrapolation distances for five spherical geometries are determined in section V-B.
II. GENERAL THEORY

A. The $P_1-L_1$ Equation in Spherical Geometry

1. The $P_1$ equation

The source-free integro-differential form of the Boltzmann transport equation describing the time, space, energy and angular dependence of neutron flux in a sphere of homogeneous materials is

$$
\frac{1}{V} \frac{\partial \phi(r,E,\mu,t)}{\partial t} + \mu \frac{\partial \phi(r,E,\mu,t)}{\partial r} + \frac{(1 - \mu^2)}{r} \frac{\partial \phi(r,E,\mu,t)}{\partial \mu} + \Sigma_T \phi(r,E,\mu,t) = 2\pi \int_{-1}^{1} \int_{E'} \Sigma_s(\mu, E' \rightarrow E) \phi(r,E',\mu',t) d\mu' dE' \tag{2.1}
$$

where

$\phi(r,E,\mu,t) =$ The angular flux

$\Sigma_s(\mu, E' \rightarrow E) =$ The scattering kernel

$= \Sigma_s(\mu', E' \rightarrow E) =$ Direction cosine

$\Sigma_T =$ Total macroscopic cross section

The solution of 2.1 should be made to satisfy the usual boundary conditions of transport theory, namely:

a. The neutron flux must be finite and non-negative in all regions of the medium since the medium contains no sources.

b. At the outer boundary, which faces a vacuum, there must be no neutrons returning to the medium from
the vacuum.

Since \( \phi(r,E,\mu,t) \) is time dependent, an initial condition must also be stated to solve the problem completely. However, this study is concerned only with the relative behavior of the angular flux as a function of the system size and some reference time; hence the complete solution of the problem is not required.

The angular dimension of 2.1 may be removed in the usual manner by expanding the angular flux and the scattering kernel in Legendre polynomials

\[
\phi(r,E,\mu,t) = \sum_{m} \frac{(2m + 1)}{4\pi} \phi_m(r,E,t) P_m(\mu) \quad \text{(2.2.a)}
\]

\[
\Sigma_s(\mu, E' \rightarrow E) = \sum_{m} \frac{(2m + 1)}{4\pi} \Sigma_m(E' \rightarrow E) P_m(\mu) \quad \text{(2.2.b)}
\]

This yields, after the insertion of the angular expansion, making use of the addition theorem of Legendre polynomials, multiplying through by \( P_n(\mu) \) and integrating over \(-1 \leq \mu \leq 1:\)

\[
(n + 1)[\frac{3}{r} + \frac{(n + 2)}{r}] \phi_{n+1}(r,E,t) + n[\frac{\partial}{\partial r} - \frac{(n-1)}{r}] \phi_n(r,E,t) + (2n + 1)[(\Sigma_T + \frac{1}{V} \frac{\partial}{\partial t}) \phi_n(r,E,t) - S_n \phi_n(r,E',t)] = 0 \quad \text{(2.3)}
\]

where \( n = 0, 1, 2, \ldots N \) for an \( N \)th order angular approximation,
\[ S_n \phi_n(r,E',t) = \int_{E'=-\infty}^{\infty} \sum_{n=0}^{\infty} (E'^E) \phi_n(r,E',t) dE' \quad (2.4) \]

and

\[ \phi_0(r,E,t) = 0 \]

By direct substitution, it can be shown that the solution of 2.3 is of the form

\[ \phi_n(r,E,t) = F_n(B,E,\lambda) j_n(Br)e^{-\lambda t} + G_n(B,E,\lambda) n_n(Br)e^{-\lambda t} \quad (2.5) \]

where \( j_n(Br) \) and \( n_n(Br) \) are the \( n^{th} \) order spherical Bessel functions of first and second type respectively. The parameter \( B \) is a space eigenvalue and \( \lambda \) is a time eigenvalue.

The asymptotic limits for spherical Bessel functions with small arguments are:

\[ j_n(Br) \xrightarrow{r \to 0} \frac{(Br)^n}{1 \cdot 3 \cdot 5 \cdots (2n + 1)} \]

\[ n_n(Br) \xrightarrow{r \to 0} \frac{1 \cdot 3 \cdot 5 \cdots (2n + 1)}{(Br)^n + 1} \]

Since \( n_n(Br) \) contains the argument in the denominator of its limit, it is seen that it would violate the first boundary condition, while \( j_n \) does not. Therefore the arbitrary constant \( G_n(Br) \) must be set identically equal to zero and \( \phi_n(r,E,t) \) becomes:

\[ \phi_n(r,E,t) = F_n(B,E,\lambda) j_n(Br)e^{-\lambda t} \quad (2.6) \]
The substitution of $\phi_n(r,E,t)$ as given by 2.6 into 2.3 gives a set of $N + 1$ homogeneous coupled equations relating the $F_n$'s for an $N^{th}$ order approximation:

$$(n+1)BF_{n+1}(B,E,\lambda) - nBF_{n-1}(B,E,\lambda) + (2n+1)[(\Sigma_T - \frac{\lambda}{V})F_n(B,E,\lambda) - S_nF_n(B,E',\lambda)] = 0$$

In the $P_1$ approximation, this set takes the form

$$BF(B,E,\lambda) + [(\Sigma_T - \frac{\lambda}{V})F_0(B,E,\lambda) - S_0F_0(B,E',\lambda)] = 0 \quad (2.8.a)$$

$$-BF_0(B,E,\lambda) + 3[(\Sigma_T - \frac{\lambda}{V})F_1(B,E,\lambda) - S_1F_1(B,E',\lambda)] = 0 \quad (2.8.b)$$

The $P_1$ component of the scattering kernel can be approximated by

$$\Sigma_{s1}(E'\rightarrow E) \approx \bar{\mu}(E)\Sigma_{so}(E'\rightarrow E)\delta(E'\rightarrow E)$$

where $\delta(E'\rightarrow E)$ is the Dirac delta function and $\bar{\mu}$ is the average of the cosine of the scattering angle.

With the above approximation the integral $S_1F_1$ becomes

$$S_1F_1(B,E',\lambda) \approx \Sigma_{so}(E)\bar{\mu}F_1(B,E,\lambda) \quad (2.9)$$

The substitution of this expression into 2.8 gives

$$F_1(B,E,\lambda) = BF_0(B,E,\lambda)/3(\Sigma_{TR} - \frac{\lambda}{V}) \quad (2.10.a)$$
\[ \left[ \frac{B^2}{3} (\Sigma_{TR} \cdot \frac{\lambda}{V}) + \Sigma_T - \frac{\lambda}{V} \right] F_0 (B, E, \lambda) = S_0 F_0 (B, E', \lambda) \]  
(2.10.b)

where

\[ \Sigma_{TR} (E) = \Sigma_a (E) + \Sigma_{so} (E) [1 - \tilde{\mu} (E)] \]  
(2.11)

The set 2.10 is closely related to that obtained by Nelkin (39) and Vertes (53) for an infinite slab using the Fourier transform. However, the above formalism has an advantage over Nelkin's; namely, the Fourier transform implies an infinite medium. By finding the set of eigenvalues of 2.10:

\[ B_0, B_1, \ldots, B_k \]

one can write down the total solution of 2.8 which will satisfy the boundary conditions of a finite sample.

2. Limiting value of the decay constant

According to Corngold and others (11-14) the discrete eigenvalues of the decay constant, \( \lambda \), are limited by

\[ \lambda_{\text{lim.}} = (V \Sigma_{s, \text{in}})_{\text{min.}} \]  
(2.12)

where \( \Sigma_{s, \text{in}} \) is the macroscopic inelastic scattering cross section for neutrons of speed \( V \). The minimum theoretical value of \( V \Sigma_{s, \text{in}} \) occurs for \( V \to 0 \), and for water it is close to 300,000 sec.\(^{-1}\). As Corngold and Michael (14) pointed out, the experimental values of the fundamental decay constant
exceed this limit in many cases. This means that if the experimental points are correct, they stand in direct contradiction to rather direct consequences of the Boltzmann equation. It is perhaps more reasonable to look at the problem from a different point of view.

In 2.10.b, $F_0(B,E,\lambda)$ is recognized as the energy component of the scalar flux at a given $B$ and $\lambda$. For this component to be real and finite the following should hold.

$$\lambda<[Vb^2/3(\Sigma_{TR} - \Sigma_{in}) + V(\Sigma_s + \Sigma_a)]_{E_o} = \lambda_{lim}. \quad (2.13)$$

where $E_o$ corresponds to the energy at which the quantities in brackets are at a minimum. Equation 2.13 explains why the experimental decay constant exceeds Corngold's limit in some cases. In case of water, however, both the absorption and the transport cross section behave like $1/V$ for small $V$ and $2.13$ would reduce to Corngold's limit as $V^0$ provided that $(V\Sigma_s)_{E_o}$ is recognized as $(V\Sigma_{s,\text{in.}}+\Sigma_{in.})_{E_o}$.

From the foregoing one concludes that $\lambda_{lim.}$ is a separating point between two types of spectra:

a. A discrete set of eigenvalues and a corresponding discrete spectrum of eigenfunctions in the range

$$\lambda<\lambda_{lim.}.$$  

b. A continuum of eigenvalues in the range

$$\lambda>\lambda_{lim.}.$$
The corresponding eigenfunctions are given by

\[ F_0(B,E,\lambda) = P \left[ \int_0^{\infty} \sum_{E' \rightarrow E} F_0(B, E', \lambda) dE' \right] \]

\[ + g(\lambda) \delta \left( \frac{\lambda}{V} - \Sigma_T + \frac{B^2}{3(\Sigma_{TR} - \frac{\lambda}{V})} \right) \]  

(2.14)

where \( P \) implies the principal value of the integral when the denominator appears as the integrand. The delta functions give the contribution of singularities other than the poles.

By the principle of superposition, the solution of the \( P_1 \) equation becomes

\[ \phi_n(r, E, t) = j_n(Br) \left[ \sum_{i=0}^{M} F_{ni}(B, E_i) e^{-\lambda_i t} + \int_{\lambda_{lim}}^{\infty} F_n(B, E, \lambda) e^{-\lambda t} d\lambda \right] \]

\( ; n = 1, 2 \)  

(2.15)

where \( M \) stands for the number of the discrete eigenvalues and the integral gives the contribution of the continuum.

Only for a proton gas case does there exist a satisfactory discussion of the character of the time eigenvalues associated with the energy modes, as given by Corngold et al. (14).

According to these authors the number of discrete eigenvalues between zero and \( \lambda_{lim} \) is infinite for an infinite medium. Shapiro (47) has obtained extensive numerical results
for the existence and convergence of discrete eigenvalues for the monatomic gas model. Numerical results for the eigenvalues of the bound proton model have been obtained by Ohanian and Daitch (41) using the diffusion approximation. The quoted paper indicated that the first eigenvalue always exists for a finite medium. There are also strong indications that there is always at least a second eigenvalue even though it may lie close to the limiting decay constant. Recently the thermal neutron space-time eigenvalue spectrum of the multi-group P_N approximations were investigated numerically by Travelli et al. (52), for a modified form of Radkowsky kernel. Both discrete and continuous eigenvalues were found.

3. Expansion of energy eigenfunctions in orthogonal polynomials

Expansion of $F_n(B,\varepsilon)$ in a complete set of orthogonal polynomials allows the solution of the eigenvalue problem. The choice of these polynomials is arbitrary. The Laguerre polynomials have been used widely, as they are the exact eigenfunctions of the Wilkin's heavy gas scattering operator. Let

$$F_n(B,\varepsilon) = \sum_{k=0}^{\infty} M(\varepsilon) F_n^k(\varepsilon)L^{(1)}_k(\varepsilon), \quad (2.16)$$

$$\Sigma_a(\varepsilon) = \Sigma_{ao} \sqrt{\varepsilon} (1/V \text{ absorber}) \quad (2.17)$$
\[ \Sigma_{TR}(E) = \Sigma_{TRO} \sqrt{E} \left( \frac{1}{V} \text{ transport cross section} \right) \quad (2.18) \]

and

\[ V = V_0 \sqrt{E} \quad (2.19) \]

where \( E \) is a dimensionless energy variable expressed in units of \( KT \),

\[ M(E) = \text{Maxwellian neutron distribution} \]
\[ = E e^{-E} \quad (2.20) \]

\[ L_k^{(1)}(E) = \text{Associated Laguerre polynomials of first order} \]
\[ = (k+1) \sum_{p=0}^{k} \frac{(-1)^p k!}{p! (1 + p)! (k - 1)!} E^p \quad (2.21) \]

\[ \Sigma_{a0} = \text{Absorption cross section at room temperature} \]
\[ \Sigma_{TRO} = \text{Transport cross section at room temperature} \]
\[ V_0 = \text{Neutron speed at room temperature} \]
\[ = 2.2 \times 10^5 \text{cm/sec.} \]

By rewriting the scattering cross section in the integral form

\[ \Sigma_{so}(E) = \int_0^\infty \Sigma_{so}(E' \rightarrow E) dE' \quad (2.22) \]

and substituting 2.16 through 2.22 into 2.10, one gets

\[ \Sigma_{k=0}^\infty \left[ (\frac{-V_0}{\sqrt{E}} B^2/3(V_0 \Sigma_{TRO} - \lambda) + (\lambda/V_0 - \Sigma_{a0}) M(E)L_k^{(1)}(E) \right] \]
According to the detailed balance theorem of statistical mechanics,

\[ M(E') \Sigma_{so}(E' \rightarrow E) = \Sigma_{so}(E') M(E') H(E' \rightarrow E) \]

\[ = \Sigma_{so}(E) M(E) H(E \rightarrow E') \quad (2.24) \]

where \( H(E \rightarrow E') dE \) is the probability that a neutron suffering a scattering collision at \( E' \) shall have an energy \( E \) in \( dE \).

By expanding

\[ L^{(1)}(E') = L^{(1)}(E) + \sum_{P=1}^{\infty} \frac{d^{P} L^{(1)}(E)}{dE^{P}} (E' - E)^{P} \]

and using the above results, the scattering terms can be expressed as a sum of energy transfer moments:

\[ \int_{0}^{\infty} [L^{(1)}(E') \Sigma_{so}(E' \rightarrow E) - L^{(1)}(E) M(E) \Sigma_{so}(E \rightarrow E')] dE' \]

\[ = \sum_{P=1}^{\infty} \frac{1}{P!} \frac{d^{P} L^{(1)}(E)}{dE^{P}} M(E) \Sigma_{so}(E) A_{P}(E) \quad (2.25.a) \]
\[
A_p(E) = \int_0^\infty (E' - E) P_{H(E' \rightarrow E)} dE' \quad (2.25b)
\]

\[= p^{th} \text{ energy transfer moment} \]

By substituting 2.25 into 2.23, multiplying through by \(L^{(1)}_i(E)\) and integrating over \(E\), one gets

\[
\sum_{k=0}^{\infty} \left[ t_{ik} B^{2/3} + (K/V_0) w_{ik} + F_{ik} \right] F_0^k(B) = 0 \quad (2.26.a)
\]

\[
F_1^i(B) = \frac{B^{2/3}}{3} \sum_{k=0}^{\infty} t_{ik} F_0^k(B) \quad (2.26.b)
\]

where

\[
t_{ik} = V_0 \int_0^\infty \left[ \sqrt{E} M(E)L^{(1)}_i(E)L^{(1)}_k(E)/(V_o \Sigma_{tro} - K) \right] dE
\]

\[= V_0 v_{ik}/(V_o \Sigma_{tro} - K), \quad (2.27)\]

\[
v_{ik} = \int_0^\infty \sqrt{E} M(E)L^{(1)}_i(E)L^{(1)}_k(E) dE
\]

\[
\Sigma_{tro} = \Sigma_{tro} - \Sigma_{ao}
\]

\[= \text{Transport cross section for zero absorption} \]

\[= \Sigma_{so}(1 - \mu) \quad (2.28)\]

\[
w_{ik} = \int_0^\infty (1/\sqrt{E}) M(E)L^{(1)}_i(E)L^{(1)}_k(E) dE \quad (2.29)
\]

\[
K = \lambda - V_o \Sigma_{ao} \quad (2.30)
\]
\[ F_{ik} = \sum_{P} \left[ \int_{0}^{\infty} \frac{d^{PL}(l)(E)}{dE} M(E)L_{k}(l)(E)\Sigma_{s0}A_{P}(E) \right] \] \hspace{1cm} (2.31)

Some values for \( w_{ik} \) and \( v_{ik} \) are listed in Table 2.1 and 2.2 respectively.

Equation 2.26 is the \( P_{1}-L_{k} \) equation in spherical geometry. For the \( L_{1} \) approximation it reduces to

\[ (t_{00}B^{2}/3 - Kw_{00}/V_{0})F_{0}^{0}(B) + (t_{01}B^{2}/3 - Kw_{01}/V_{0})F_{0}^{1}(B) = 0 \] \hspace{1cm} (2.32.a)

\[ (t_{10}B^{2}/3 - Kw_{10}/V_{0})F_{0}^{0} + (t_{11}B^{2}/3 - Kw_{11}/V_{0}) + |F_{11}|)F_{0}^{1}(B) = 0 \] \hspace{1cm} (2.32.b)

\[ F_{1}^{0}(B) = \frac{B}{3}(t_{00}F_{0}^{0}(B) + t_{01}F_{0}^{1}(B)) \] \hspace{1cm} (2.33.a)

\[ F_{1}^{1}(B) = \frac{B}{3}(t_{01}F_{0}^{0}(B) + t_{11}F_{0}^{1}(B)) \] \hspace{1cm} (2.33.b)

where

\[ F_{11} = -M_{2}/4 \] \hspace{1cm} (2.34.a)

\[ M_{2} = \int_{0}^{\infty} \int_{0}^{\infty} (E' - E)^{2} \tau_{s0}(E)M(E)H(E\rightarrow E')dE' dE \] \hspace{1cm} (2.34.b)

= Second energy transfer moment.

Equation 2.32 is a set of homogeneous equations. The roots of the characteristic equation are the possible values of \( B^{2} \).
Table 2.1. Values of $w_{ik}$

<table>
<thead>
<tr>
<th>$w_{00}$</th>
<th>$w_{01}$</th>
<th>$w_{02}$</th>
<th>$w_{03}$</th>
<th>$w_{11}$</th>
<th>$w_{12}$</th>
<th>$w_{13}$</th>
<th>$w_{22}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.8862</td>
<td>0.3134</td>
<td>0.1919</td>
<td>0.1384</td>
<td>0.7754</td>
<td>0.3392</td>
<td>0.2252</td>
<td>0.7061</td>
</tr>
</tbody>
</table>

*From Purohit (43).

Table 2.2. Values of $v_{ik}$

<table>
<thead>
<tr>
<th>$v_{00}$</th>
<th>$v_{01}$</th>
<th>$v_{11}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.330</td>
<td>-0.471</td>
<td>1.828</td>
</tr>
</tbody>
</table>

For other values of $v_{ik}$ use is made of the relations

$$v_{ik} = v_{00}[(w_{ik}/w_{00}) - T_{ik}], \quad T_{ik} = [(w_{ik}/w_{00}) - (v_{ik}/v_{00})]$$

Table 2.3. Values of $T_{ik}$

<table>
<thead>
<tr>
<th>$T_{00}$</th>
<th>$T_{01}$</th>
<th>$T_{02}$</th>
<th>$T_{03}$</th>
<th>$T_{11}$</th>
<th>$T_{12}$</th>
<th>$T_{13}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.7071</td>
<td>0.2887</td>
<td>0.1882</td>
<td>-0.5505</td>
<td>0.8674</td>
<td>0.3540</td>
</tr>
</tbody>
</table>

*From Perez (42).*
4. Matrix elements and associated integrals

The success of the $P_1-L_k$ equation in interpreting the experimental data depends on how accurately the matrix elements $F_{ik}$ of the scattering operator are determined. For $P_1-L_1$, only the element $F_{11}$ is required.

The elements can be obtained from the related energy transfer moments $M_{kp}$ or their associated integrals.

Nelkin (38) was the first to introduce $M_{20}$ or $(M_2)$ in estimating the thermalization parameters, by the use of variational principles. Using the detailed balance theorem, Purohit (44) gave recurrence formulas.

The values of the energy transfer moments depend on the scattering model used. For water, four models are tried in this thesis. These are:

a. Hydrogen gas: The motions of the protons in water are like a free gas (58) with no binding between protons and oxygen.

b. Mass-18 gas: The water molecules are considered as rigid structures and replaced by a gas of point particles with mass 18.

c. Brown-St. John (6): The water molecules are treated as rigid structures free to rotate. The rotator is then replaced by a free point particle with an "effective rotational mass" of 1.88. The model utilizes a trial cross section that contains adjustable
parameters so that the computed scattering cross section can be fitted to the experimental cross section.

d. The Nelkin water (40): The dynamics of protons in water are described by three harmonic oscillators, two for the vibrational levels at 0.205 ev and 0.48 ev and one for the hindered rotation at 0.06 ev. The motions of the molecules are described by a mass-18 gas.

The second energy transfer moment, \( M_2 \), for the above mentioned models is listed in Table 2.4. In all these cases a free gas kernel with mass 16 was considered to approximate the scattering from oxygen.

Throughout the remainder of this work, whenever there is a choice among the above models, the Nelkin water model will be the one selected. This is justified on the basis that this kernel predicts infinite medium spectra in good agreement with spectra measured by Beyste (5) over a wide range of poison types and concentrations. The kernel also gives diffusion coefficients and cooling coefficients in good agreement with values measured by Star and Koppel (49). Detailed comparisons of the Nelkin kernel with measurements by Eglestaff et al. (21) have been made by Goldman and Federighi (25). They found reasonable agreement between the theoretical predictions of the model and the experimental results at
Table 2.4. Second energy moment of the isotropic scattering kernel

\[ M_2 = \int_0^\infty \int_0^\infty M(E) \Sigma_s(E') H(E' - E) (E - E')^2 dE dE' \]

<table>
<thead>
<tr>
<th>Kernel</th>
<th>( M_2 ) (cm(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>H gas</td>
<td>3.85</td>
</tr>
<tr>
<td>Mass 18</td>
<td>0.67</td>
</tr>
<tr>
<td>Brown &amp; St.John</td>
<td>5.23</td>
</tr>
<tr>
<td>Nelkin</td>
<td>3.34</td>
</tr>
</tbody>
</table>

\(^a\)From Honeck (27).

all but small values of energy and moment transfer. Fortunately this region contributes little to the total cross section or to the energy transfer moments. Finally, the Nelkin water gives a total cross section over the entire thermal energy range.

B. Time Eigenvalues

A study of the eigenvalue spectra in the multigroup \( P_N \) approximations (15, 52) indicated the possibility of a traveling wave phenomenon at large bucklings. This work is extended here to the \( P_1-L_1 \) approximation for two reasons. First, Travelli and Calame (52) pointed out that the fundamental eigenvalue curve in the \((\lambda, B^2)\) plane, in a 4-group \( P_1 \) approxi-
mation, contradicts the experimental results in having a positive curvature at the origin. This study will explain the nature of this contradiction. Second, the split of energy into few groups does not reflect the usual diffusion cooling phenomena (15). The diffusion cooling is obtained in the continuous energy representation by expanding the lowest eigenvalue in power series of $B^2$. The radius of convergence of this expansion is evaluated by a knowledge of the eigenvalue spectra.

The eigenvalue problem relevant to this study is

$$\begin{bmatrix}
(t_{00}B^2/3 - K_{w00}/V_o) & (t_{01}B^2/3 - K_{w01}/V_o) \\
(t_{10}B^2/3 - K_{w01}/V_o) & (T_{11}B^2/3 - K_{w11}/V_o + |F_{11}|)
\end{bmatrix} \begin{bmatrix}
F_0^0(B) \\
F_0^1(B)
\end{bmatrix} = 0$$

(2.35)

Since equation 2.35 consists of two linear homogeneous equations, the eigenvalues are fixed by the condition that the determinant of the coefficients must vanish, i.e., by the equation

$$Q(K, B^2) = (t_{00}B^2/3 - K_{w00}/V_o)(t_{11}B^2/3 - K_{w11}/V_o + |F_{11}|) - (t_{10}B^2/3 - K_{w01}/V_o)^2 = 0$$

(2.36)

where $K$ is related to $\lambda$ by 2.30.

The function $Q(K, B^2)$ is a polynomial in $B^2$ of degree 2 and for a fixed $B^2$ it is a polynomial of degree 4 in $K$. In
general, for a \( P_N - L_M \) approximation there is a polynomial of degree \( \frac{1}{2} M(N + 1) \) in \( B^2 \) and of degree \( M(N + 1) \) in \( K \).

The expansion of (2.36) gives

\[
Q(K, B^2) = 3K^4(w_{00}w_{11} - w_{01}^2) \\
-3K^3V_0[2\Sigma_{\text{tr}}(w_{00}w_{11} - w_{01}^2) + w_{00} |F_{11}|] \\
+ K^2V_0^2[B^2(v_{11}w_{00} - 2v_{01}w_{01} + v_{00}w_{11}) \\
+ 3\Sigma_{\text{tr}}^2(w_{00}w_{11} - w_{01}^2) + 6\Sigma_{\text{tr}}w_{00}|F_{11}|] \\
- KV_0^3[\Sigma_{\text{tr}}B^2(v_{11}w_{00} - 2v_{01}w_{01} + v_{00}w_{11}) \\
+ |F_{11}|(3\Sigma_{\text{tr}}w_{00} + B^2V_0)] \\
+ B^2V_0^4[\frac{B^2}{3}(v_{00}v_{11} - v_{01}^2) + \Sigma_{\text{tr}}V_0 |F_{11}|] = 0
\]

(2.37)

First estimates of the roots of \( Q(K, B^2) \) were obtained by plotting this function versus \( K \) at specified values of \( B^2 \). Some of these plots are indicated in Figure 2.1. These roots were then taken as first trial values in Newton Raphson method (33). Iterations were carried out on the IBM-360 computer until the desired accuracy was obtained. Values of the various cross sections used in the numerical calculation are given in the Appendix.
Figure 2.1. Roots of the polynomial, $C(K, B^2)$, for various values of $B^2$ and $\mu_2 = 3.34 \text{ cm}^{-1}$
A study was made using the four scattering models listed in Table 2.4. The results are given in Table 2.5, 2.6, 2.7 and 2.8. The corresponding curves are shown in Figure 2.2, 2.3, 2.4 and 2.5 respectively. For the sake of comparison results obtained by the diffusion theory approximation are also listed in the respective tables. These tables show a number of features of interest:

a. The diffusion approximation gives only two eigenvalues compared to 4 in the $P_1$-$L_1$ approximation. This is because the latter approximation has the form of the telegrapher's equation (57) and differs from the diffusion equation by an additional term containing the second order time derivative.

b. The diffusion theory curves indicate that the decay constants increase indefinitely in direct proportion to $B^2$ and are always real. This behavior stems from the linear relationship between $B^2$ and $K$. For the $P_1$-$L_1$ approximation, on the other hand, there exist two limiting values of $B^2$, $B^2_{1,\text{max.}}$ and $B^2_{2,\text{max.}}$, such that:

i. For $B^2 < B^2_{1,\text{max.}}$, all eigenvalues, $K$, are real.

ii. For $B^2 > B^2_{2,\text{max.}}$, all eigenvalues are complex.

iii. For $B^2_{1,\text{max.}} < B^2 < B^2_{2,\text{max.}}$, two real eigenvalues exist.
Table 2.5. The tabular entries give the decay constants in (microseconds)^{-1} for M_2 = 0.67 cm^{-1}, the bucklings and theories indicated

| B^2     | K_0     | K_1    | K_2    | K_3    | Diff. Theory
<table>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
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<td>0.0 cm^{-2}</td>
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<td>0.05342</td>
<td>0.6216</td>
<td>0.6286</td>
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<td>0.05812</td>
<td>0.62080</td>
<td>0.62712</td>
<td>0.000949</td>
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<tr>
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<td>0.001898</td>
<td>0.06067</td>
<td>0.62000</td>
<td>0.62500</td>
<td>0.001878</td>
</tr>
<tr>
<td>0.10</td>
<td>---</td>
<td>---</td>
<td>0.6163</td>
<td>0.62150</td>
<td>---</td>
</tr>
<tr>
<td>0.20</td>
<td>0.007218</td>
<td>0.07698</td>
<td>0.6022</td>
<td>0.61714</td>
<td>0.007089</td>
</tr>
<tr>
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<td>0.01530</td>
<td>0.08883</td>
<td>0.5992</td>
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<td>0.010295</td>
</tr>
<tr>
<td>0.40</td>
<td>0.013920</td>
<td>0.09923</td>
<td>0.5762</td>
<td>0.6152</td>
<td>---</td>
</tr>
<tr>
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<td>0.11508</td>
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<td>0.18256</td>
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<td>0.027249</td>
</tr>
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<td>0.20410</td>
<td>0.4698</td>
<td>0.6046</td>
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</tr>
<tr>
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<td>0.5728</td>
<td>0.054918</td>
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<tr>
<td>3.0</td>
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<td>0.5420</td>
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</tr>
<tr>
<td>4.0</td>
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<td>0.5057</td>
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<td>-0.05300</td>
<td>0.8663</td>
<td>0.691</td>
<td>-2.70457</td>
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</tbody>
</table>

**The root has a complex value.**
Table 2.6. The tabular entries give the decay constants in (microseconds)$^{-1}$ for $M_2 = 3.34 \text{cm}^{-1}$, the bucklings and theories indicated.

<table>
<thead>
<tr>
<th>$B^2$</th>
<th>$P_{1-L_1}$</th>
<th>Diff. Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$K_0$</td>
<td>$K_1$ $K_2$ $K_3$</td>
</tr>
<tr>
<td>0.0 cm$^{-2}$</td>
<td>0.000000</td>
<td>0.27642 0.62150 0.63000</td>
</tr>
<tr>
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<td>0.00967</td>
<td>0.28052 --- ---</td>
</tr>
<tr>
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<td>0.001933</td>
<td>0.28474 0.61570 0.62500</td>
</tr>
<tr>
<td>0.100</td>
<td>---</td>
<td>--- 0.60670 0.62330</td>
</tr>
<tr>
<td>0.150</td>
<td>0.005780</td>
<td>0.30299 --- ---</td>
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<tr>
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<td>0.31314 0.58650 0.62050</td>
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</tr>
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<td>0.700</td>
<td>0.026609</td>
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<td>0.900</td>
<td>0.034087</td>
<td>--- --- ---</td>
</tr>
<tr>
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<td>---</td>
<td>--- --- 0.59875</td>
</tr>
<tr>
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<td>0.075289</td>
<td>--- --- ---</td>
</tr>
<tr>
<td>3.000</td>
<td>0.114870</td>
<td>--- --- 0.53450</td>
</tr>
<tr>
<td>4.000</td>
<td>---</td>
<td>--- --- 0.49340</td>
</tr>
<tr>
<td>5.000</td>
<td>0.219162</td>
<td>--- --- 0.43770</td>
</tr>
<tr>
<td>6.000</td>
<td>---</td>
<td>--- --- 0.33000</td>
</tr>
<tr>
<td>7.000</td>
<td>---</td>
<td>--- --- ---</td>
</tr>
<tr>
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<td>0.21180 0.69810 0.63840</td>
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<td>0.106300</td>
<td>0.06780 0.88250 0.68370</td>
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**The root has a complex value.
Table 2.7. The tabular entries give the time eigenvalues, \( K_i \), in \((\text{microseconds})^{-1}\) for \( M_2 = 3.85 \text{ cm}^{-1} \), the bucklings and theories indicated.

<table>
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<th>( P_1 - L_1 )</th>
<th>( K_0 )</th>
<th>( K_1 )</th>
<th>( K_2 )</th>
<th>( K_3 )</th>
<th>Diff. Theory</th>
</tr>
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<tbody>
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<td>***</td>
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<td>.34595</td>
</tr>
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**The root has a complex value.**
Table 2.8. The tabular entries give the time eigenvalues, $K$, in (microseconds)$^{-1}$ for $M_2 = 5.23$ cm$^{-1}$, the bucklings and the theories indicated.

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<th>$K_0$</th>
<th>$K_1$</th>
<th>$K_2$</th>
<th>$K_3$</th>
<th>Diff. Theory</th>
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<td>-**</td>
<td>.59987</td>
<td>.285304</td>
<td>1.01601</td>
</tr>
<tr>
<td>- .500</td>
<td>- .019300</td>
<td>---</td>
<td>.72680</td>
<td>.63870</td>
<td>- .019810</td>
<td>.38879</td>
</tr>
<tr>
<td>-1.500</td>
<td>- .058200</td>
<td>.23415</td>
<td>.84550</td>
<td>.66310</td>
<td>- .063741</td>
<td>.30466</td>
</tr>
<tr>
<td>-2.000</td>
<td>- .078000</td>
<td>.19690</td>
<td>.89050</td>
<td>.67440</td>
<td>- .088352</td>
<td>.26525</td>
</tr>
<tr>
<td>-2.500</td>
<td>- .097600</td>
<td>.16500</td>
<td>---</td>
<td>.68530</td>
<td>- .115049</td>
<td>.22792</td>
</tr>
<tr>
<td>-3.000</td>
<td>- .122400</td>
<td>.13770</td>
<td>---</td>
<td>.69620</td>
<td>- .144030</td>
<td>.19288</td>
</tr>
</tbody>
</table>

**The root has a complex value.
Figure 2.2. Eigenvalue curves for $M_2 = 0.67 \text{ cm}^{-1}$ and the theories indicated.
\[ K = \lambda - V_0 \Sigma_{ao-1} \] (microseconds)

\[ K_0, K_1, K_2, K_3 \]

\[ K_{lim} \]

--- F_1-L_1 approximation

--- Diffusion approximation
Figure 2.3. Eigenvalue curves for $M_2 = 3.34 \text{ cm}^{-1}$ and the theories indicated.
\[ K = \lambda - V_0 \sum_{a_0} (\text{microseconds})^{-1} \]
Figure 2.4. Eigenvalue curves for $M_2 = 3.85 \text{ cm}^{-1}$ and the theories indicated.
\[ K = \lambda - V_0 \Sigma_{ao}^{-1} \] (microseconds) \[ \approx \]

\[ K_{lim.} \]

---

\[ P_1 - L_1 \text{ approximation} \]  
\[ \text{Diffusion approximation} \]
Figure 2.5. Eigenvalue curves for $M_2 = 5.23 \text{ cm}^{-1}$ and the theories indicated
The diagram illustrates a relationship between variables $K_0$, $K_1$, and $K_2$, with $K = \lambda - V_0 \Sigma_{\alpha\beta}$ (in microseconds) as a function of $B^2$ (in $\text{cm}^{-2}$). The section shows the $P_1-L_1$ approximation and the diffusion approximation for different values of $B^2_{1,\text{max}}$ and $B^2_{2,\text{max}}$. The diagram is not labeled with specific values or units for these variables, but it provides a visual representation of how these approximations change with $B^2$.
These limiting values are listed in Table 2.9 as functions of $M_2$. It is clear from this table that, within the range of $M_2$'s investigated, the limiting values are monotonic decreasing functions of $M_2$. In addition, the table shows that $B_{2,\text{max}}^2$ is well above the experimental range of bucklings. Thus the usual expansion of the lowest eigenvalue in power series of $B^2$ is valid for the experimental ranges of $B^2$ reported in literature. On the other hand, $B_{1,\text{max}}^2$ is within the range of pulsing experiments. For Nelkin's scattering kernel, for example, it has a value of 0.45 cm$^{-2}$ and the expansion of the second lowest eigenvalue in power series of $B^2$ is doubtful.

c. The eigenvalue curve, which passes through $B^2 = k = 0$, has a negative curvature at this point for all the models in question. The amplitude of the curvature increases by decreasing $M_2$. For the Nelkin water, the curvature is in close agreement with the experimental results. Recently, Travelli and Calame (52) made a numerical investigation on thermal neutron space time eigenvalue spectrum of the multigroup $P_N$ approximations for a modified form of Radkowsky kernel. Their results, in general, agree with the present findings. However, in a 4-group $P_1$ calculation, although the curve corresponding to the fundamental
Table 2.9. The limiting values of buckling and the corresponding decay constants for various water scattering kernels. Values of $\lambda_{\text{lim}}$, obtained by other authors are also indicated.

<table>
<thead>
<tr>
<th>Kernel and references</th>
<th>$M_2$ (cm$^{-1}$)</th>
<th>$B_{1,\text{max}}^2$ (cm$^{-2}$)</th>
<th>$B_{2,\text{max}}^2$ (cm$^{-2}$)</th>
<th>$\lambda_{\text{lim}}$ ($10^{-4}$sec$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass-18</td>
<td>0.67</td>
<td>1.37 ± .03</td>
<td>6.67 ± .05</td>
<td>31.700</td>
</tr>
<tr>
<td>Nelkin</td>
<td>3.34</td>
<td>0.45 ± .02</td>
<td>5.87 ± .05</td>
<td>33.300</td>
</tr>
<tr>
<td>Mass-1</td>
<td>3.85</td>
<td>0.37 ± .02</td>
<td>5.80 ± .07</td>
<td>33.500</td>
</tr>
<tr>
<td>Brown &amp; St. John</td>
<td>5.23</td>
<td>0.15 ± .02</td>
<td>5.70 ± .07</td>
<td>33.800</td>
</tr>
<tr>
<td>Mass-1 (41)</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>33.650</td>
</tr>
<tr>
<td>GIN (41)$^a$</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>31.512</td>
</tr>
<tr>
<td>Gorngold and Michael (12)</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>30.000</td>
</tr>
<tr>
<td>Doppler-Corrected (45)</td>
<td>3.17</td>
<td>---</td>
<td>---</td>
<td>42.000</td>
</tr>
</tbody>
</table>

$^a$Goldman improved Nelkin kernel.

The eigenvalue correctly passes through the origin, its curvature is positive at that point, in disagreement with the experimental results. This leads to the conclusion that the continuous energy representation as given by two Laguerre polynomials predicts the results of pulsing experiments more accurately than a 4-group approximation.
With respect to the nature of the various eigenvalues in the $P_1L_1$ approximation, one has the following argument:

For the sake of simplicity let us identify them first by the sequence $K_0 < K_1 < K_2 < K_3$.

The $(K_0,K_3)$ pair is identified separately from the $(K_1,K_2)$ pair by virtue of the one-speed model. For this case equation 2.10.b reduces to the form

$$\frac{B^2}{3(\Sigma_{TR} - \frac{1}{V})} - \frac{1}{V} + \Sigma_a = 0$$

or

$$\left(\frac{1}{V}\right)^2 - \frac{1}{V}(\Sigma_{TR} + \Sigma_a) + \Sigma_a \Sigma_{TR} + \frac{B^2}{3} = 0 \quad (2.10.b)$$

For each value of $B^2$, this equation gives two roots,

$$K = \lambda - \sqrt{\Sigma_a} = \frac{V}{2}(\Sigma_{TR} - \Sigma_a)[1 \pm \sqrt{1 - \frac{4(3\Sigma_a \Sigma_{TR} + B^2)}{3(\Sigma_{TR} + \Sigma_a)^2}}], \quad (2.10.b')$$

which are identical to $K_0$ and $K_3$ except for some modifications due to the change in the energy spectrum. Equation 2.10.b' implies that $K$ is real only if

$$B^2 \leq \frac{3}{4}(\Sigma_{TR} - \Sigma_a)^2 = \frac{3 \Sigma_a^2}{4 \Sigma_{TR}}$$

This means that the basic physical phenomenon of traveling waves is not affected by the energy dependence as long as
the time behavior of the current is taken into consideration.

In case of no absorption and no leakage ($B^2 = 0$) the two eigenvalues of \(2.10.b\) become

\[ K_0 = \lambda_0 = 0, \quad K_1 = \lambda_1 = V\Sigma_{tr}. \]

The eigenvalue zero corresponds to a mode which persists indefinitely; the corresponding eigenfunction has an arbitrary amplitude to match the initial flux but has no current component. The eigenvalue \(V\Sigma_{tr}\) has an eigenfunction which has an arbitrary current amplitude to match the initial current. This second mode then is simply an angular transient which is due to the mismatch of the initial angular distribution of the persisting mode. The neutrons in the second mode do not leak out of the system nor are they absorbed since in this particular example $B^2 = \Sigma_a = 0$. In this case also the net current is zero according to equation \(2.10.a\).

The time taken for the angular transient to rearrange itself on the persisting distribution is of the order of \((V\Sigma_{tr})^{-1}\).

As the leakage and absorption terms increase from zero, the net number of neutrons in the current transient becomes non-zero. This net number is just the number of neutrons that will actually leak from and be absorbed in the second mode. For large enough leakage or absorption the net number of neutrons in the transient can be considerable and the
influence of leakage is to make the properties of this transient more like that of the persisting mode.

On the other hand, the \((K_0, K_1)\) pair can be identified from the \((K_2, K_3)\) pair by the diffusion-theory approximation which gives a pair of eigenvalues close to the \((K_0, K_1)\) pair at small values of \(B^2\). Since in this case the time dependence of the neutron current is neglected and since in one-group diffusion approximation there is only one eigenvalue which is closely related to \(K_0\) at small \(B^2\), then \(K_1\) is simply an eigenvalue of the energy transient. This transient is a distortion of the fundamental mode spectrum. It is essentially an eigenvalue of the scattering kernel. In conclusion the four eigenvalues are interpreted as follows:

\[
\begin{align*}
K_0 & = \text{The fundamental eigenvalue.} \\
K_1 & = \text{The eigenvalue of the energy transient.} \\
K_2 & = \text{The eigenvalue of the angular transient which is a distortion to the second energy mode (with an eigenvalue } K_1). \\
K_3 & = \text{The eigenvalue of the angular transient that distorts the fundamental mode.}
\end{align*}
\]

The value of \(K\) at \(B_{2,\text{max}}^2\) is the limiting value, \(K_{\text{lim}}\), for the fundamental eigenvalue \(K_0\). The corresponding value of \(\lambda_{\text{lim}}\) is obtained by adding the constant term \(V_0 \Sigma_{ao}\). Values of \(\lambda_{\text{lim}}\) obtained by this method and as reported by
different authors are listed in Table 2.9. This table indicates that the limiting value is a function of the chemical binding of the water molecules and that it increases slowly with \( M_2 \).

Ohanian and Daitch (41) calculated \( \lambda_{\text{lim}} \) for the Mass-1 and the GIN (for the Goldman improved Nelkin) scattering kernels. Their calculations were based upon a numerical method employing a discrete representation of the energy variable. Their reported value for the Mass-1 kernel is in excellent agreement with the present value as shown in Table 2.9. One can also observe the good agreement between the value for the Mass-18 and that for the GIN kernel. The two values differ by less than 1%.

While all values reported in Table 2.9 agree with each other within 10%, the value reported by Purohit and Sjöstrand (45) is out of this range. From the foregoing this value for the Doppler-corrected kernel should lie in the range \( 33.0 \times 10^4 - 33.3 \times 10^4 \) sec.\(^{-1} \)

C. Diffusion Parameters

In section B the discussion was mainly on the behavior of the time eigenvalues in the real \((K,B^2)\) plane. In the light of the observations in that section, analytical expressions for various diffusion parameters of water will be developed here.
The first step in this direction is to write 2.37 in the form:

\[ \kappa^2 - kV_0 \left[ \frac{B^2(t_{11} w_{00} - 2t_{01} w_{01} + t_{00} w_{11}) + 3w_{00} |F_{11}|}{3(w_{00} w_{11} - w_{01}^2)} \right] 
+ \frac{B^4(t_{00} t_{11} - t_{01}^2) + 3B^2 t_{00} |F_{11}|}{9(w_{00} w_{11} - w_{01}^2)} \]  

(2.38)

where \( t_{ik} \) is given by 2.27.

Equation 2.38 can be regarded as a transcendental quadratic equation with two roots given by

\[ K_0 = \lambda_0 - V_0 \sigma_{ao} \]
\[ = \frac{1}{2} [b_0 - (b_0^2 - 4c_0)^{1/2}] \]  

(2.39)

\[ K_1 = \lambda_1 - V_0 \sigma_{ao} \]
\[ = \frac{1}{2} [b_1 + (b_1^2 - 4c_1)^{1/2}] \]  

(2.40)

where

\[ b_n = V_o \left[ \frac{B^2(t(n) w_{00} - 2t(n) w_{01} + t(n) w_{11}) + 3w_{00} |F_{11}|}{3(w_{00} w_{11} - w_{01}^2)} \right], \]  

(2.41)

\[ c_n = V_o \left[ \frac{B^4(t(n) t_{00} - (t(n))^2) + 3B^2 t_{00} |F_{11}|}{9(w_{00} w_{11} - w_{01}^2)} \right], \]  

(2.42)

and
\[ t^{(n)}_{ik} = V_0 v_{ik} / (V_0 \Sigma^r - K_n) ; \quad n = 0, 1. \quad (2.43) \]

From these equations valuable information can be obtained.

1. **Diffusion cooling coefficient**

The expansion of 2.39 in power series of \( B^2 \) is guaranteed only if \( B^2 < B^2_{\text{max}} \). If, in addition, \( B^2 \) is small the expansion assumes the simple form

\[ K_0 = \lambda_0 - V_0 \Sigma_{ao} \]

\[ = D_T(B^2)B^2 - C_T(B^2)B^4 + 0B^6 \quad (2.44) \]

where

\[ D_T(B^2) = \frac{2}{3\sqrt{\pi}} \frac{V_0 t^{(0)}_{00}}{} \quad (2.45) \]

\[ C_T(B^2) = \frac{D_T^2(B)}{V_0 M_2} \quad (2.46) \]

\[ g = \frac{\sqrt{\pi}}{4} \left[ 1 + \frac{8}{\sqrt{2}} \frac{V_{01}}{V_{00}} \left( \sqrt{2} \frac{V_{01}}{V_{00}} - 1 \right) \right] \quad (2.47) \]

The numerical factor, \( g \), is a function of the energy dependence of the transport mean free path and the number of the Laguerre polynomials used in the series expansion of the flux. Values of the factor \( g \) are given in Table 2.10 for the several approximations used.
Table 2.10. Comparison of several expressions for the numerical factor $g$

<table>
<thead>
<tr>
<th>Values of $g$ under several approximations</th>
<th>Nelkin (38)</th>
<th>Purohit (43)</th>
<th>Perez et al. (42)</th>
<th>$P_1-L_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy dependent transport mean free path</td>
<td>1.772</td>
<td>---</td>
<td>1.772</td>
<td>1.776</td>
</tr>
<tr>
<td>Constant transport mean free path</td>
<td>0.443</td>
<td>0.443</td>
<td>0.443</td>
<td>0.443</td>
</tr>
</tbody>
</table>

The coefficient, $C_T$, of $B^4$ is the transport analog of the diffusion cooling coefficient $C_D$, obtained from the diffusion theory. $C_T$, however, differs from $C_D$ in its dependence on $B^2$. To suppress the $B^2$-dependence, one should write

$$D_T(B^2)B^2 = D_0B_T^2$$  \hspace{1cm} (2.48)

$$C_T(B^2)B^4 = C_B^4$$  \hspace{1cm} (2.49)

where

$$D_0 = \text{The diffusion coefficient}$$

$$= V_0 v'_{00} / 3 w_{00} \Sigma_{tro}$$  \hspace{1cm} (2.50)

$$C = \text{The diffusion cooling coefficient}$$

$$= gD_0^2 / V_0 M_2$$  \hspace{1cm} (2.51)

and $B_T^2$ is given, to the order of $B^6$, by
This expression shows that $B_t^2$ is a function of both the geometric and the scattering properties of the moderator. The ratio $B_t^2/B^2$ is plotted versus $B^2$, for $M_2 = 3.34$ cm$^{-1}$, in Figure 2.6. The curve shows that, for the values of $B^2$ considered, $B_t^2$ differs from $B^2$ by no more than 4%. Hence one can consider $B_t^2$ as a transport buckling with a non-diffusive correction for $B^2$. Furthermore, the values of $B_t^2/B^2$ reported in Table 2.11 show that the ratio is not sensitive to $M_2$ for small values of $B^2$.

The substitution of 2.48 and 2.49 into 2.44 gives

$$K_0 = \lambda_0 - V_o \Sigma_{ao}$$

$$= D_0 B_t^2 - CB_T^4 + OB_T^6$$

Both $D_0$ and $C$ are independent of $B^2$.

Expression 2.51 for $C$ was first derived by Nelkin (38) using the Rayleigh-Ritz variational principle, based upon the neutron temperature concept. Using the same concept, Mani (35) modified it to take into account the variation of the transport mean free path with energy. Singwi (48) developed a general theory of the diffusion cooling based upon the expansion of the asymptotic energy distribution by a sum of the associated Laguerre polynomials of order one. Häfele and Dresner (26) have also given a similar theory for the calcu-
Figure 2.6. A plot of the ratio $B_T^2/B^2$ versus $B^2$ for $\nu_2 = 3.34 \text{ cm}^{-1}$
Table 2.11. The ratios of $B_2^2/B^2$ for various values of $B^2$ and two values of $M_2$

<table>
<thead>
<tr>
<th>$B^2 (\text{cm}^{-2})$</th>
<th>$B_2^2/B^2$ for $M_2 = 3.34 \text{ cm}^{-1}$</th>
<th>$B_2^2/B^2$ for $M_2 = 3.85 \text{ cm}^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>1.0000</td>
<td>1.0000</td>
</tr>
<tr>
<td>0.025</td>
<td>1.0016</td>
<td>1.0016</td>
</tr>
<tr>
<td>0.050</td>
<td>1.0031</td>
<td>1.0031</td>
</tr>
<tr>
<td>0.150</td>
<td>1.0093</td>
<td>1.0093</td>
</tr>
<tr>
<td>0.200</td>
<td>1.0124</td>
<td>1.0124</td>
</tr>
<tr>
<td>0.300</td>
<td>1.0184</td>
<td>1.0184</td>
</tr>
<tr>
<td>0.500</td>
<td>1.0303</td>
<td>1.0305</td>
</tr>
<tr>
<td>0.700</td>
<td>1.0427</td>
<td>1.0431</td>
</tr>
</tbody>
</table>

Evaporation of the diffusion cooling coefficient in a monoatomic heavy gas, using the same expansion. Kazarnovsky et al. (30) also used the Laguerre polynomials in their study of neutron thermalization problems. All of these studies were undertaken for the finite medium under the diffusion approximation. Nelkin (39) studied the decay of a thermalized neutron pulse in an infinite plane geometry using the transport theory in the Fourier space. The Fourier variable, $B$, in Nelkin's formalism has been left ill-defined. The appli-
cation of this method to a moderator with finite size and energy dependent transport mean free path is not unique (59).

Expression 2.50 and 2.51 were derived using neither the neutron temperature concept nor the Fourier transform technique. By the application of the proper boundary conditions, it will be shown later that the variable $B$ in this thesis has a definite physical meaning.

Table 2.12 shows a comparison between values of $C$ and $D_0$, obtained by the suppression of the $B^2$-dependence and those reported by other authors. It is clear that there is an agreement between the present values for the Nelkin's water and the $P_3$ values obtained by Gelbard and Davis for the Radkowsky kernel.

As a further check on the validity of the above expansion, $\lambda_0$ was calculated from 2.53 and 2.39 with an iterative technique. The results are listed in table 2.13 for $M_2 = 3.34 \text{ cm}^{-1}$. The table indicates that, within the limits of accuracy set for the computer, both values agree quite satisfactorily.

2. Thermalization time constant

The thermalization time constant is defined as the time constant with which the neutrons attain an asymptotic energy distribution by colliding with the atoms of a moderator. In case of an infinite and non-absorbing medium equation 2.39
Table 2.12. Values of the diffusion cooling coefficient, $C$, and diffusion coefficient, $D_0$, for various methods

<table>
<thead>
<tr>
<th>Method and references</th>
<th>Kernel</th>
<th>$C$ cm$^{-1}$.sec.$^{-1}$</th>
<th>$D_0$ cm$^2$.sec.$^{-1}$</th>
<th>Range of bucklings cm$^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{1-L_1}$</td>
<td>H gas</td>
<td>3139</td>
<td>38692</td>
<td>---</td>
</tr>
<tr>
<td>&quot;</td>
<td>Mass-18</td>
<td>18038</td>
<td>38692</td>
<td>---</td>
</tr>
<tr>
<td>&quot;</td>
<td>Nelkin</td>
<td>3618</td>
<td>38692</td>
<td>---</td>
</tr>
<tr>
<td>&quot;</td>
<td>Brown &amp; St. John</td>
<td>2311</td>
<td>38692</td>
<td>---</td>
</tr>
<tr>
<td>$P_3$ (24)</td>
<td>Radkowsky</td>
<td>3614</td>
<td>38380</td>
<td>---</td>
</tr>
<tr>
<td>Calame (7)</td>
<td>Nelkin</td>
<td>2931</td>
<td>36810</td>
<td>---</td>
</tr>
<tr>
<td>Scott et al. (46)</td>
<td>---</td>
<td>---</td>
<td>38500±800</td>
<td>.006-.018</td>
</tr>
<tr>
<td>Antonov et al. (1)</td>
<td>---</td>
<td>4000±1000</td>
<td>35000±1000</td>
<td>.09 -.93</td>
</tr>
<tr>
<td>Lopez and Beyster (34)</td>
<td>---</td>
<td>4852±800</td>
<td>36700±370</td>
<td>.0 -1.00</td>
</tr>
</tbody>
</table>

Table 2.13. Values of the fundamental decay constant, $\lambda_0$, as obtained from the correct form 2.39 and the expansion form 2.53, for $M_2 = 3.34$ cm$^{-1}$.

<table>
<thead>
<tr>
<th>$B^2$ (cm$^{-2}$)</th>
<th>$\lambda_0$ (sec.$^{-1}$) Correct value</th>
<th>Expansion value</th>
<th>($\lambda_0$ corr - $\lambda_0$ expans.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>4876</td>
<td>4876</td>
<td>0.00</td>
</tr>
<tr>
<td>0.025</td>
<td>5843±5</td>
<td>5843</td>
<td>0.00</td>
</tr>
<tr>
<td>0.050</td>
<td>6809±5</td>
<td>6809</td>
<td>0.00</td>
</tr>
<tr>
<td>0.150</td>
<td>10656±5</td>
<td>10651</td>
<td>5.00±5</td>
</tr>
<tr>
<td>0.200</td>
<td>12572±5</td>
<td>12563</td>
<td>9.00±5</td>
</tr>
<tr>
<td>0.300</td>
<td>16388±5</td>
<td>16359</td>
<td>29.00±5</td>
</tr>
<tr>
<td>0.500</td>
<td>23965±5</td>
<td>23850</td>
<td>115.00±5</td>
</tr>
<tr>
<td>0.700</td>
<td>31485±5</td>
<td>31170</td>
<td>315.00±5</td>
</tr>
</tbody>
</table>

$^a$Nelkin scattering model.
and 2.40 reduce respectively to

\[ K_0 = \lambda_0 = 0, \]  
\[ K_1 = \lambda_1 = -V_0 w_{00} F_{11} \left( w_{00} w_{11} - w_{01}^2 \right) \]

\[ = 2V_0 M_2 / 3\sqrt{\pi} \]  
\[ (2.55) \]

In this case there is an asymptotic energy distribution. The decay of this distribution is governed by \( \lambda_0 \) equals to zero. If the atoms of the moderator have a Maxwellian velocity distribution, then the asymptotic distribution is also the Maxwellian distribution which is established with a thermalization time constant equals to the reciprocal of \( \lambda_1 \). In the case of a finite medium, the zeros eigenvalues associated with the higher spatial modes also play an important role in the establishment of the final asymptotic energy distribution. If the amplitudes of higher modes are very small compared with the fundamental spatial mode, then the first eigenvalue, \( \lambda_1 \), would give the thermalization time constant in the finite medium. The discussion, here, is limited to the time constant with which the Maxwellian distribution is established.

The thermalization time constant is given by the reciprocal of \( \lambda_1 \)

\[ t_{th} = \frac{3\sqrt{\pi}}{2V_0 M_2} f_t \]  
\[ (2.56) \]
a result which is identical with that derived from the diffusion theory (43). \( f_t \) is a correction factor due to higher order polynomials than \( L_1^{1/1} \). Its value is 1.15 (45).

Various models have been considered in calculating \( t_{th} \) according to equation 2.56. The results are presented in Table 2.14. Two cases were treated. In case I the transport mean free path is energy dependent; while in case II it is taken as a constant. As could be seen from this table, \( t_{th} \) for the constant \( \lambda_{th} \) is about 4 times as large as that for the energy dependent case for all the models used. Thus a comparison between these results and the experimental values of \( t_{th} \) would indicate the energy behavior of the transport mean free path. Fortunately, Möller and Sjöstrand (37) have measured \( t_{th} \) in light water by obtaining information on the change of the neutron spectrum with time from the reaction rate with spectrum indicators dissolved in the system. They reported a value of \( 4.1 \pm 0.4 \) usec. This agrees with the value of \( 4.16 \) usec obtained for \( M_2 = 3.34 \text{ cm}^{-1} \) in the energy dependent case. One concludes, therefore, that the transport cross section for light water behaves more or less like \( 1/V \).

DeJurene (17) took into account the spectral changes caused by diffusion-cooling and reported a value of \( 2.77 \pm 0.65 \) usec. Although this result disagrees with the experimental value reported by Möller, yet it is in accord with the theoretical value for \( M_2 = 5.23 \text{ cm}^{-1} \) (Brown and St. John
Table 2.14. Thermalization time constant for light water in the $P_1 - L_1$ approximation for various scattering models

<table>
<thead>
<tr>
<th>Kernel</th>
<th>$M_2$ (cm$^{-1}$)</th>
<th>$t_{th}$ (microseconds)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Case I ($\lambda_{tr} \alpha \sqrt{E}$)</td>
</tr>
<tr>
<td>Nelkin</td>
<td>3.34</td>
<td>4.16</td>
</tr>
<tr>
<td>Mass-1</td>
<td>3.85</td>
<td>3.61</td>
</tr>
<tr>
<td>Mass-18</td>
<td>0.67</td>
<td>13.85</td>
</tr>
<tr>
<td>Brown &amp; St.John</td>
<td>5.23</td>
<td>2.66</td>
</tr>
</tbody>
</table>

scattering model).

Wood (60) represented water with the effective width kernel of Egelstaff (22). He calculated $t_{th}$ for water as a function of the reciprocal of the effective width parameter, $d$, of the scattering law. His results are listed in Table 2.15 which shows that the value of $t_{th}$ at $d = 0.27$ is in agreement with the $P_1 - L_1$ value for the Nelkin's model.

The thermalization time constant can also be expressed in terms of the cooling coefficient by eliminating $M_2$ between 2.51 and 2.55. The result is

$$t_{th} = \frac{6C}{D^2_0} \left[ 1 + \frac{8}{\sqrt{2}} \frac{V_0}{V_00} \left( \frac{V_0}{V_00} - 1 \right) \right]^{-1} f_t, \quad (\lambda_{tr} \alpha \sqrt{E})$$

$$= \frac{6C}{D^2_0} f_t \quad (\lambda_{tr} = \text{const.}), \quad (2.57)$$
Table 2.15. Thermalization time constant for light water by other authors

<table>
<thead>
<tr>
<th>Reference</th>
<th>Kernel</th>
<th>( t_{th} ) (microseconds)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Möller &amp; Sjöstrand (37)</td>
<td>---</td>
<td>4.10 ± .4 (water)</td>
</tr>
<tr>
<td>DeJurene (17)</td>
<td>---</td>
<td>2.77 ± .65</td>
</tr>
<tr>
<td>Wood (60)</td>
<td>Effective width</td>
<td>4.50 (( d^a = .21 ))</td>
</tr>
<tr>
<td></td>
<td>&quot;</td>
<td>4.13 (( d = .27 ))</td>
</tr>
<tr>
<td></td>
<td>&quot;</td>
<td>3.71 (( d = .50 ))</td>
</tr>
<tr>
<td></td>
<td>Mass-1</td>
<td>3.60</td>
</tr>
<tr>
<td></td>
<td>Mass-1.88</td>
<td>4.89</td>
</tr>
<tr>
<td></td>
<td>Mass-2</td>
<td>5.05</td>
</tr>
</tbody>
</table>

\( d^a \) is the reciprocal of the effective width parameter of Egelstaff Kernel (22).

in agreement with the expression obtained by Purohit (43).

D. Space and Energy Dependent Eigenfunctions

The existence of a unique buckling depends on the validity of the first fundamental theorem of reactor theory \textit{vis.} space and energy are separable:

\[
\phi(\bar{\xi},E) = X(\bar{\xi})Y(E) \tag{2.58}
\]

This is true in a homogeneous infinite medium. It requires, however, some justification in finite systems. What is done is to seek an "asymptotic" region inside the medium far from the boundaries in which the first fundamental theorem
is valid. Experiments (29) have been performed to test this assumption. Those by Inönu at Oak Ridge are particularly interesting. Inönu measured the thermal and epithermal fluxes and showed that only if data within 3-3.5 inches of the boundary of a large critical aqueous U-235 solution were included was the extrapolation distance independent of energy i.e. equation 2.58 applies. This example is an extreme example; in non-multiplying medium the effect will be less.

On physical grounds the exact solution can be written in the form

\[ \phi(\xi, E) = \phi_{as}(\xi, E) + \phi_{trans}(\xi, E) \]

\[ = X(\xi)Y(E) + \phi_{trans}(\xi, E) \quad (2.59) \]

The asymptotic part establishes a unique extrapolation distance, \( d(B^2) \), for a given buckling and energy. Figure 2.7 shows how this extrapolation distance is related to the asymptotic flux. The distance \( S \) is the width of the zone in which the term \( \phi_{trans}(\xi, E) \) is important.

In this section analytical expressions for \( \phi_{as} \) and \( \phi_{trans} \) are established. In a latter section an expression for \( d(B^2) \) will be found.

As has been mentioned before, the polynomial \( C(K, B^2) \) given by equation 2.37 is of degree 2 in \( B^2 \). For \( K = K_0 \) (the lowest eigenvalue), the equation for \( B^2 \) becomes
Figure 2.7. The relation between the extrapolation distance, \( d(B^2) \), and the asymptotic flux distribution. The width, \( S \), of the transient zone is indicated.
\( (t_{00} t_{11} - t_{10} t_{01}) B^4 + \left( \frac{3t_{00} t_{11} - 3t_{01} t_{10}}{4} \right) W_{00} - 2t_{01} w_{01} \)

\( + t_{00} w_{00} ] B^2 + \frac{9K_0^2}{V_o^2} (w_{00} w_{11} - w_{01}^2) - \frac{9K_0 w_{00} M_2}{4V_o} = 0 \) (2.60)

where \( t_{ik}^{(0)} \) is given by 2.43.

For \( K_0 < K_{\text{lim}} \), all the coefficients in 2.60 are real. In addition, in case of light water, the term \( (t_{00} t_{11} - t_{01} t_{10}) \) is a positive quantity. The last coefficient is negative only if

\[ K_0 < \frac{V_o w_{00} M_2}{4(w_{00} w_{11} - w_{01}^2)} \] (2.61)

Thus it is easily established, by requiring that the coefficient of \( B^2 \) is positive, that for

\[ K_0 < \frac{V_o M_2}{4(v_{00} w_{11} + v_{11} w_{00} - 2v_{01} w_{01})} \] (2.62)

\[ \approx 1.88 \times 10^4 M_2 \]

there exist two values of \( B^2 \)

\[ B^2 = B_0^2, \quad B^2 = -U^2 \] (2.63)

where both \( B_0^2 \) and \( U^2 \) are positive quantities. The interpretation of \( B_0^2 \) as the geometric buckling, \( B_0^2 \), of the system depends on the application of the boundary condition at the vacuum-
matter interface. The commonly used outer face boundary conditions are the Marshak and the zero extrapolated flux boundary condition. The first boundary condition will be used in this thesis. Taking $B_0^2$ as $B_g^2$, one can write the scalar flux in the form

$$\phi_0(r,E) = F_0(B,E)j_0(Br)$$

$$F_0(B_g,E)j_0(B_g r) + F_0(U,E)j_0(iUr) \quad (2.64)$$

where

$$F_0(B_g,E) = A_{B_g}Ee^{-E[F_0^0(B_g) + \frac{1}{\sqrt{2}}(2 - E)F_0^1(B_g)]} \quad (2.65)$$

$$F_0(U,E) = A_UEe^{-E[F_0^0(U) + \frac{1}{\sqrt{2}}(2 - E)F_0^1(U)]} \quad (2.66)$$

Recalling equation 2.59, one can associate the first term of 2.64 with $X(r,Y(E)$ and the second term with $\phi(r,E)_\text{trans}$.

1. **The relaxation length**

   The physical meaning of $U^{-1}$ is understood by writing 2.64 in the form

   $$\phi_0(r,E) = A_{B_g}Ee^{-E}\left\{F_0^0(B_g) + \frac{1}{\sqrt{2}}(2 - E)F_0^1(B_g)\right\}j_0(B_g r)$$

   $$+ \frac{A_U}{A_{B_g}}\left[F_0^0(U) + \frac{1}{\sqrt{2}}(2 - E)F_0^1(U)\right]j_0(iUr) \quad (2.67)$$

The ratio $A_U/A_{B_g}$ is determined by applying the Marshak's boundary condition at the vacuum interface. It is found that
\[
\frac{A_U}{A_B} j_0(iUr) \approx \text{const} \times e^{-U(R-r)}
\]  (2.68)

Thus for distances \( > U^{-1} \) this term decays rapidly and \( U^{-1} \) is interpreted as a relaxation length which is a measure of the transient zone near the boundary. \( U^{-1} \) is given by

\[
U = \sqrt{u^2},
\]

\[
u^2 = 1.4027 \times 10^{-11} K_0 \left( V_0 \Sigma_{\text{tro}} - K_0 \right) \left( \frac{7.315 \times 10^4 M_2}{K_0} - 2.9465 \right)
\]

\[
x \left[ 1 + \left( \frac{1.3349 \times 10^6 (M_2/4K_0 - 3.0208 \times 10^{-6})}{(2.926 \times 10^5 M_2/4K_0 - 2.9465)^2} \right)^{1/2} \right]
\]  (2.69)

For the Nelkin's water, calculations indicate that \( U^{-1} = 0.483 \text{ cm} \) in the limit as \( B_g^2 \to 0 \). The results suggest that deviation from the asymptotic solution begins to become large at points of the order of 0.5 cm from the boundary. Table 2.16 shows the variation of \( U^{-1} \) with buckling. The observed increase of the relaxation length with \( B_g^2 \) is expected on the basis that the energy spectrum deviates more from the Maxwellian distribution for small systems. \( U^{-1} \) is very sensitive to the value of \( M_2 \) and the effect of chemical binding on the space-energy separability can be studies through this parameter. From Table 2.16 it can be seen that the smaller the value of \( M_2 \) the less accurate is the space-energy
Table 2.16. The variation of the relaxation length for water with buckling and $M_2^a$

<table>
<thead>
<tr>
<th>$B_g^2$ (cm$^2$)</th>
<th>$M_2 = 0.67 cm^{-1}$</th>
<th>$M_2 = 3.34 cm^{-1}$</th>
<th>$M_2 = 3.85 cm^{-1}$</th>
<th>$M_2 = 5.23 cm^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>1.07000</td>
<td>0.48300</td>
<td>0.44300</td>
<td>0.38200</td>
</tr>
<tr>
<td>0.025</td>
<td>1.10244</td>
<td>0.48557</td>
<td>0.45187</td>
<td>0.38732</td>
</tr>
<tr>
<td>0.050</td>
<td>1.12898</td>
<td>0.48818</td>
<td>0.45403</td>
<td>0.38876</td>
</tr>
<tr>
<td>0.150</td>
<td>1.24405</td>
<td>0.49889</td>
<td>0.46284</td>
<td>0.39462</td>
</tr>
<tr>
<td>0.200</td>
<td>1.30713</td>
<td>0.50439</td>
<td>0.46736</td>
<td>0.39762</td>
</tr>
<tr>
<td>0.300</td>
<td>1.44415</td>
<td>0.51571</td>
<td>0.47663</td>
<td>0.40374</td>
</tr>
<tr>
<td>0.500</td>
<td>1.73918</td>
<td>0.53967</td>
<td>0.49617</td>
<td>0.41656</td>
</tr>
<tr>
<td>0.700</td>
<td>-**</td>
<td>0.56552</td>
<td>0.51714</td>
<td>0.43017</td>
</tr>
<tr>
<td>0.900</td>
<td>-**</td>
<td>0.59339</td>
<td>0.53964</td>
<td>0.44466</td>
</tr>
</tbody>
</table>

$^a$The values of $M_2$ correspond to the scattering kernels listed in Table 2.4.

**A complex value.

separability, i.e., the smaller is the asymptotic region. For $M_2 = .67$ cm$^{-1}$ there is no unique buckling above a value of 0.5 cm$^{-2}$ where the inequality 2.62 is invalidated.

2. The asymptotic diffusion-cooled neutron spectrum

The spectrum of the asymptotic distribution is given by

$$F_0 (B_g, E) = A_{B_g} E e^{-E} \left[ F_0^0 (B_g) + \frac{1}{\sqrt{2}} (2 - E) F_0^1 (B_g) \right] \quad (2.70)$$
where $F_0^0(B_g)$ is an arbitrary constant that can be taken as unity since it originates in the set of two homogeneous equations given by 2.32. The quantity $F_0^1(B_g)$ has the form

$$F_0^1(B_g) = \frac{B^2 t_0(0)/3 - w_{01} K_0/V_0}{B^2 t_{11}(0)/3 - w_{11} K_0/V_0 + M_2/4}$$  \hspace{1cm} (2.71)

The latter expression is obtained from 2.32 and is considered as a measure of the deviation from the Maxwellian distribution. Figure 2.8 shows a plot of $F_0^1(B_g)$ versus $B^2_g$. It is clear from this Figure that $F_0^1(B_g)$ is zero only for an infinite medium where the energy distribution follows the Maxwellian distribution.

Examples of the diffusion-cooled spectra for the Nelkin's scattering model are shown in Figure 2.9. The corresponding data are listed in Table 2.17. In Figure 2.9, the shifting of the peak of the curve toward lower energy values with increasing $B^2_g$ is evident, and the change of the shape of the curve is indicated. The Maxwellian distribution is seen to represent the limiting value of $F_0^1(B_g E)$ as $B^2_g \rightarrow 0$. The magnitude of the cooling effect is indicated by the shifting of the peak value of the curve from one unit of KT for $B^2_g = 0$ to .87 units of KT for $B^2_g = .9$ cm$^{-2}$.

Recently, Clendenen (9) made similar calculations for light water treated with the Nelkin model. He used a new iterative method applied to a $P_{11}$ approximation although
Figure 2.8. Buckling dependence of $F_0(B_g)$ for various values of $M_2$. 

- $M_2 = 0.67$
- $M_2 = 3.34$
- $M_2 = 3.85$
- $M_2 = 5.23$
Figure 2.9. Diffusion-cooled neutron spectra for Nelkin model of water moderator at room temperature. Variation with buckling $B_g^2 (\text{cm}^{-2})$ is shown.
Table 2.17. Normalized values of diffusion-cooled neutron spectra for Nelkin model of water moderator at room temperature. Variation with buckling $B_g^2$ is indicated.

<table>
<thead>
<tr>
<th>Energy, $E$ (in units of $KT$)</th>
<th>$F_0(B_g,E)$ for $B_g^2 = 0.0 \text{cm}^{-2}$</th>
<th>$B_g^2 = 0.5 \text{cm}^{-2}$</th>
<th>$B_g^2 = 0.9 \text{cm}^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>0.0000</td>
<td>0.0000</td>
<td>0.0000</td>
</tr>
<tr>
<td>0.025</td>
<td>0.0245</td>
<td>0.0266</td>
<td>0.0280</td>
</tr>
<tr>
<td>0.500</td>
<td>0.3033</td>
<td>0.3235</td>
<td>0.3385</td>
</tr>
<tr>
<td>0.750</td>
<td>0.3543</td>
<td>0.3740</td>
<td>0.3885</td>
</tr>
<tr>
<td>1.000</td>
<td>0.3679</td>
<td>0.3842</td>
<td>0.3963</td>
</tr>
<tr>
<td>1.500</td>
<td>0.3347</td>
<td>0.3421</td>
<td>0.3476</td>
</tr>
<tr>
<td>2.000</td>
<td>0.2707</td>
<td>0.2707</td>
<td>0.2707</td>
</tr>
<tr>
<td>0.500</td>
<td>0.2052</td>
<td>0.2006</td>
<td>0.1973</td>
</tr>
<tr>
<td>3.000</td>
<td>0.1493</td>
<td>0.1427</td>
<td>0.1378</td>
</tr>
<tr>
<td>3.500</td>
<td>0.1057</td>
<td>0.0987</td>
<td>0.0930</td>
</tr>
<tr>
<td>4.000</td>
<td>0.0732</td>
<td>0.0667</td>
<td>0.0619</td>
</tr>
<tr>
<td>4.500</td>
<td>0.0500</td>
<td>0.0445</td>
<td>0.0403</td>
</tr>
<tr>
<td>5.000</td>
<td>0.0337</td>
<td>0.0292</td>
<td>0.0259</td>
</tr>
</tbody>
</table>

His results generally agree with the present findings, the method does not directly give the limiting value of $F_0(B_g,E)$ as $B_g^2 \to 0$. Besides, this method is only suited to high-order approximations to the transport equation.
3. **The energy spectrum of the transient component**

This spectrum is given by

\[ F_0(U,E) = A_0e^{-E}[1 + \frac{1}{\sqrt{2}}(2 - E)F_0^1(U)] \]  \hspace{1cm} (2.72)

where

\[ F_0^1(U) = \frac{U_0^2t_0/3 + w_0k_0/V_o}{U_0^2t_0/3 + w_1k_0/V_o} \]  \hspace{1cm} (2.73)

\( F_0^1(U) \) is plotted, for various scattering kernels, in Figure 2.10. It starts from a non-zero value for an infinite medium and increases steadily with decreasing the size of the system. The implication of these results is that the energy spectrum of the transient distribution is always a non-Maxwellian distribution and that its magnitude becomes significant for small systems. Examples of this spectrum, for the Nelkin's water, are shown in Figure 2.11. The corresponding data are listed in Table 2.18. Each of these curves is shown to have a minimum and a maximum the location of which depends on the value of \( B^2_g \), i.e., on the cooling effect.

Having discussed the energy spectra of the asymptotic and the transient components of the scalar flux, it is of practical importance to discuss this distribution as a function of the position \( r \). The flux \( \phi_0(r,E) \) as given by 2.63 is plotted in Figure 2.12 versus the ratio \( r/R \) at \( B^2_g = 0.5 \text{ cm}^{-2} \) and two different values of energy. For the sake of
Figure 2.10. Buckling dependence of $F_0^1(U)$ for various values of $M_2$. 
Figure 2.11. Transient energy spectra for Nelkin's water at room temperature. Variation with buckling \( B_g^2 (\text{cm}^{-2}) \) is shown.
Figure 2.12. Flux distribution as a function of space points at $B_q^2 = 0.5 \text{ cm}^{-2}$.
Table 2.18. Transient energy spectra at room temperature for the Neijkin's water. Variation with buckling $B_g^2(\text{cm}^{-2})$ is indicated.

| Energy, $E$, (KTUnits) | $F_0(U,E)$ (Arbitrary Units) $B_g^2 = 0.05 \text{ cm}^{-2}$ | $B_g^2 = 0.5 \text{ cm}^{-2}$ |
|------------------------|---------------------------------------------------------------|
| 0.000                  | 0.0000                                                        |
| 0.025                  | 0.1235                                                        |
| 0.500                  | 1.2342                                                        |
| 0.750                  | 1.2605                                                        |
| 1.000                  | 1.1207                                                        |
| 1.500                  | 0.6772                                                        |
| 2.000                  | 0.2707                                                        |
| 2.500                  | -0.0048                                                       |
| 3.000                  | -0.1562                                                       |
| 3.500                  | -0.2187                                                       |
| 4.000                  | -0.2264                                                       |
| 4.500                  | -0.2058                                                       |
| 5.000                  | -0.1732                                                       |

comparison, the asymptotic flux distribution is also plotted in the same Figure from which one can observe the following:

1. For points far from the physical boundary by amounts of the order of $U^{-1}$, the total flux, independent of energy, has the same value as the asymptotic distribution.

2. Close to the boundary the two fluxes differ from each other. The difference depends on the energy.
At low energies (close to KT), the asymptotic flux has a higher value; while at high energies (in the order of 5KT) the asymptotic flux has a lesser magnitude than the total flux.

Thus the effect of the transient flux near the boundary is a function of energy. At high energy the effect is subtractive and the predicted extrapolation length, according to the "total" flux distribution, is greater than that for the asymptotic distribution. On the other hand, at low energies the effect is additive giving rise to an extrapolation distance lower than the asymptotic value. In practice, however, the flux distribution is measured by a boron trifluoride detector over a range of energy. The net transient effect in this range is found to be additive. This point will be made more clear in the next section in the discussion of the effective average energy.

In support of the above observations, are the results of Walker (56) who illustrated the effect of the flux distortion on the extrapolated endpoint of a 4-in cubic container filled with water. His results indicated that the extrapolation distance increases steadily by including more points closer to the boundaries.

E. Spectrum-Averaged Parameters

Having obtained \( \phi_0(r, E) \), one is in a position to calculate various parameters averaged over the space-dependent
neutron spectrum. Two of these of interest in the thermalization problem are

1. Effective average energy.
2. Effective buckling.

1. **Effective average energy**

   The effective average energy is defined (59) by

   \[
   \bar{E}_{\text{eff}}(r, B_g^2) = \frac{\int_C E \phi_0(r, E) dE}{\int_0^\infty \phi_0(r, E) dE}
   \]  

   (2.74)

   This leads to

   \[
   \bar{E}_{\text{eff}}(r, B_g^2) = 2 \left[ 1 - \frac{1}{\sqrt{2}} \frac{\frac{A_U}{A_B} F_0(B_g) j_0(B_g r) + \frac{A_U}{A_B} F_0(U) j_0(i U r)}{j_0(B_g r) + \frac{A_U}{A_B} j_0(i U r)} \right]
   \]  

   (2.75)

   where

   \[
   \frac{A_U}{A_B} = -\frac{F_0(B_g) j_0(B_g R) + 2(t_{01}) + t_0 F_0(B_g) j_1(B_g R)}{F_0(U) j_0(i U R) - \frac{2}{3} (t_{01} + t_{11} F_0(U)) u j_1(i U R)}
   \]  

   (2.76)

   and

   \[
   j_1(x) = \text{First order spherical Bessel function}
   \]

   \[
   = \frac{\sin x}{x^2} - \frac{\cos x}{x}
   \]

   \[
   \bar{E}_{\text{eff}}(r, B_g^2)
   \]  

   is a measure of the way in which the neutron
energy spectrum changes with position and how accurately space and energy can be made separable. Two special cases are of interest: \( \bar{E}_{\text{eff}}(O, B_g^2) \) and \( \bar{E}_{\text{eff}}(R, B_g^2) \). These are respectively given by

\[
\bar{E}_{\text{eff}}(O, B_g^2) = 2\left[1 - \frac{1}{\sqrt{2}} \frac{F_0^1(B_g) + (U A_U / B_g A_B) F_0^1(U)}{1 + (U A_U / B_g A_B)}\right] \tag{2.77}
\]

\[
\bar{E}_{\text{eff}}(R, B_g^2) = 2\left[1 - \frac{1}{\sqrt{2}} \frac{F_0^1(B_g) + Y F_0^1(U)}{1 + Y}\right] \tag{2.78}
\]

where

\[
Y = - \frac{\frac{F_0^1(B_g) + 2 \left[ t^{(0)}_{01} + t^{(0)}_{11} F_0^1(B_g) \right] (B_g \cot B_g R - 1/R)}{F_0^1(U) + 2 \left[ t^{(0)}_{01} + t^{(0)}_{11} F_0^1(U) \right] (U \coth U R - 1/R)}}
\]

For an infinite medium

\[
\bar{E}_{\text{eff}}(R \to \infty, B_g^2 \to 0) = 2KT \tag{2.79}
\]

the same as that one obtained by assuming space and energy to be rigorously separable. This average is for the Maxwellian distribution. For a finite medium, however, there is a preferential leakage that depends strongly on the behavior of the transport mean free path with energy (59). For light water, \( \lambda_{tr} \) is proportional to the square root of energy and the preferential leak is in favor of the high
energy neutrons, a matter that results in a diffusion cooling phenomenon. For most of the crystalline moderators, like beryllium, the reverse is true (59) and one has a diffusion heating.

The effective average energy for light water is plotted as a function of position in Figure 2.13. The plot (for \( M_2 = 3.34 \text{ cm}^{-1} \)) shows the constancy of the average energy with position up to a distance from the boundary in the order of \( U^{-1} \). Close to the outer boundary, there is a marked increase in \( \bar{E}_{\text{eff}} \). A physical explanation of these results can be given in the light of certain experiments. According to Zinn (61) the temperature of neutrons emitted from a paraffin surface was 390°K whereas their temperature inside the medium was 300°K.

In conclusion, for a precise calculation of the neutron spectrum in a finite system it is necessary to take into account the changes in neutron density and spectrum in the vicinity of the boundary of the medium, which are caused by escape, and also the transfer of neutrons from a group with one energy to a group with another as a sequence of the energy exchange in the medium.

2. Effective buckling

The asymptotic region can be displayed quite easily by the expression (24)
Figure 2.13. Variation of the effective average energy with position for $M_2 = 3.34 \text{ cm}^{-1}$. Water is taken as $1/V$ scatterer.
\[ B_{\text{eff}}^2 = -\frac{\int_{0}^{\infty} \phi_0(r,E) dE}{\int_{0}^{\infty} \phi_0(r,E) dE} \]

\[ = \frac{B_g^2 \sin B_g r - \frac{U}{B_g} \sinh Ur}{\sin B_g r + \frac{B_g}{U} \sinh Ur} \]

(2.80)

disregarding the second term of 2.80, i.e., taking into account the asymptotic flux, then \( B_g^2 = B_{\text{eff}}^2 \). Thus the space variation of \( B_{\text{eff}}^2 \) and its departure from \( B_g^2 \) indicate the departure from the asymptotic region. \( B_{\text{eff}}^2 \) and \( -\Delta \phi_0(r,E)/\phi_0(r,E) \) for a sphere of water are plotted in Figure 2.14. Here \( B_g^2 = 0.5 \text{ cm}^{-2} \). The effective buckling behaves in a fashion similar to the effective average energy defined above. However, the relative variation of \( B_{\text{eff}}^2 \) is much greater than that of the average energy. The Figure also indicates that \( B_{\text{eff}}^2 \) corresponds to \( -\Delta \phi_0(r,E)/\phi_0(r,E) \) at \( E = 2KT \).

F. The Buckling Dependence of the Extrapolation Distance

To obtain the extrapolation distance in case of the existence of an asymptotic region the boundary conditions must be applied. For the vacuum-matter interface the Marshak's boundary condition can be used. In the \( P_1 \) approximation it has the form:
Figure 2.14. Variation of the effective buckling and 
$-\Delta \phi_0(r,E)/\phi_0(r,E)$ with position for $M_2 = 3.34 \text{ cm}^{-1}$. Water is taken as $1/V$ scatterer.
\( \phi_0(R,E) - 2\phi_1(R,E) = 0 \)  \hspace{1cm} (2.81)

and in the \( L_1 \) approximation:

\( \phi_0^0(R) - 2\phi_1^0(R) = 0 \)  \hspace{1cm} (2.82.a)

\( \phi_0^1(R) - 2\phi_1^1(R) = 0 \)  \hspace{1cm} (2.82.b)

From equation 2.6, 2.16, 2.33 and 2.63

\[ \phi_0^0 = A_B j_0(B_R) + A_U j_0(U) \]  \hspace{1cm} (2.83)

\[ \phi_0^1 = A_B F_0^1(B) j_0(B_R) + A_U F_0^1(U) j_0(U) \]  \hspace{1cm} (2.84)

\[ \phi_1^0(R) = \frac{A_B B_R}{3} [t_0^0 + t_0^1 F_0(B_R) j_1(B_R)] \]

\[ + \frac{A_U U}{3} [t_0^0 + t_0^1 F_0(U) j_1(U)] \]  \hspace{1cm} (2.85)

\[ \phi_1^1(R) = \frac{A_B B_R}{3} [t_1^0 + t_1^1 F_0(B_R) j_1(B_R)] \]

\[ + \frac{A_U U}{3} [t_1^0 + t_1^1 F_0(U) j_1(U)] \]  \hspace{1cm} (2.86)

By the substitution of these equations into 2.82, the following characteristic equation can be obtained.
The conventional definition of the geometric buckling for a sphere with a radius $R$ is

$$B_g^2 = \left( \frac{\pi}{R + d} \right)^2$$

This definition can only be retained if $d = d(B_g^2)$. The substitution of

$$j_0(B_g R)/n_0(B_g R) = \tan B_g d$$

into 2.87 yields

$$d(B_g) = \frac{1}{B_g} \left[ \frac{1}{\tan^{-1} \frac{2B_g^2(t(0) + t_{01} J_0(B_g))}{3}} \times \frac{1 - \frac{t_{01} F_0(B_g)}{t(0) + t_{01} F_0(B_g)} \times \frac{a}{h}}{1 - \frac{2}{3R(t(0) + t_{01} F_0(B_g)) - (F_0(B_g) - \frac{2}{3R(t_{01} + t_{11} F_0(B_g))) \frac{a}{h}}}} \right]$$

(2.89)
where

\[ g = R - \frac{2}{3} t^{(0)}_{00} + t^{(0)}_{01} F^1_0(U)[1 - UR \coth UR], \quad (2.90) \]

\[ h = RF^1_0(U) - \frac{2}{3} t^{(0)}_{01} + t^{(0)}_{11} F^1_0(U)[1 - UR \coth UR] \quad (2.91) \]

The physical meaning of the factor \((t^{(0)}_{00} - t^{(0)}_{01} F^1_0(B_g))\) is made clear by observing that

\[ \lambda_{tr}(E) = \frac{1}{\Sigma_{TR}(E) - \frac{\lambda_0}{V_0}} \]

and

\[ \frac{\int_0^\infty F_0(B_g; E) \lambda_{tr}(E) dE}{\int_0^\infty F_0(B_g; E) dE} = t^{(0)}_{00} + t^{(0)}_{01} F^1_0(B_g) = \langle \lambda_{tr}(E) \rangle_{F_0} \quad (2.92) \]

Thus this factor is the effective transport mean free path averaged over the asymptotic distribution. It gives the buckling dependence of the extrapolation distance resulting from the diffusion cooling.

Equation 2.89 is a transcendental equation since it explicitly contains \(d(B_g)\) in the right hand side. The extrapolation distance for a given buckling was obtained through an iterative technique carried out on the IBM-360 computer. The results, for various scattering kernels, are listed in Table 2.19 and plotted in Figure 2.15. Gelbard and Davis
Figure 2.15. The buckling dependence of the extrapolation distance in spherical geometry. The variation with $M_2$ is indicated.
Table 2.19. The buckling dependence of the extrapolation distance in spherical geometry for different values of $M_2$

<table>
<thead>
<tr>
<th>$B_g^2$ (cm$^{-2}$)</th>
<th>Extrapolation distance, $d(B_g^2)$ cm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$M_2 = 0.67$ cm$^{-1}$</td>
</tr>
<tr>
<td>0.000</td>
<td>0.3432</td>
</tr>
<tr>
<td>0.025</td>
<td>0.3430</td>
</tr>
<tr>
<td>0.050</td>
<td>0.3416</td>
</tr>
<tr>
<td>0.150</td>
<td>0.3326</td>
</tr>
<tr>
<td>0.200</td>
<td>0.3276</td>
</tr>
<tr>
<td>0.300</td>
<td>0.3182</td>
</tr>
<tr>
<td>0.500</td>
<td>--**</td>
</tr>
<tr>
<td>0.700</td>
<td>--**</td>
</tr>
<tr>
<td>0.900</td>
<td>--**</td>
</tr>
</tbody>
</table>

**The theory does not hold at this value of $B_g^2$.**

(24) have also calculated the extrapolation distance for the $P_3$ and the diffusion approximation. The dotted curves show their results. For the $P_3$ calculations, the authors employed the Radkowsky kernel and the Marshak's boundary condition. The diffusion theory curve was obtained by the delta method that consists of defining a linear extrapolation distance

$$ -\frac{1}{\delta} = \frac{d}{dr}\left(\frac{\sin B_gr}{r}\right) \bigg|_{r = R} /\left(\frac{\sin B_gr}{r}\right) \bigg|_{r = R} $$

and an augmentation length ($d(B_g)$ in this section). By
plotting $\theta$ versus $B_g^2$ and starting with a trial value equals to $d(0)$, $d(B_g)$ can be obtained by iterating between 2.93 and

$$B_g^2 = \left[ \frac{\pi}{R + d(B_g)} \right]^2$$

From Figure 2.15 it is clear that, while the $P_1-L_1$ curves have the general shape of the delta-curve, they markedly disagree with the $P_3$ curve. It can be concluded, therefore, that the $P_1-L_1$ approximation in spherical geometry with the Marshak's boundary condition over-estimates the buckling dependence of the extrapolation distance.

Gelbard (23) remarked that in a $P_N$ approximation of any order the eigenvalue of a spherical reactor is exactly equal to the eigenvalue of an "equivalent" slab reactor. An "equivalent" slab reactor is a slab reactor of half thickness $R$, having the same composition as the sphere. The flux in the "equivalent" reactor is constrained to be antisymmetric about its midplane. Thus the main mode of a bare sphere having a diameter $2R$ is equal to the second mode of a bare slab, of the same composition, with thickness $2R$. In another paper by Gelbard and Davis (24) it has been pointed out that the extrapolation distances for a sphere and the corresponding equivalent slab are exactly the same in the $P_3$ approximation.

To test the validity of Gelbard's observations in case of the $P_1-L_1$ approximation, the equivalent slab calculations
were carried out using (53):

\[ d(B_g) = \frac{1}{B_g} \left[ \tan^{-1} \left( \frac{2B_g(0)(0) + t_{01}F_{0}^{1}(B_g)}{3} \right) \right] \]

\[ 1 - \left( \frac{t_{01}F_{0}^{1}(B_g)}{t_{00} + t_{01}F_{0}^{1}(B_g)} \right) \frac{2}{h} \]

(2.94)

with \( g \) and \( h \) assuming the new forms:

\[ g = 1 + \frac{2}{3} \left[ t_{00} + t_{01}F_{0}^{1}(U) \right] \tanh UR \]

(2.95)

\[ h = F_{0}^{1}(U) + \frac{2}{3} \left[ t_{01} + t_{01}F_{0}^{1}(U) \right] \tanh UR \]

(2.96)

It should be noted that \( R \) in 2.94 is the half thickness of the "equivalent" slab and \( B_g^{2} \) is its second lowest eigenvalue. Furthermore this equation can be obtained from 2.89 by deleting all terms explicitly containing \( R \) and replacing \( \coth(UR) \) by \( \tanh(UR) \). Thus the extrapolation distance of the equivalent slab is a limiting case of the corresponding bare sphere.

The extrapolation distances for the equivalent slab are listed in Table 2.20 and plotted in Figure 2.16 from which one can observe the marked improvement in the behavior of the extrapolation distance as a function of buckling. In this case the curve for the Nelkin's water agrees qualitatively with the \( P_3 \) curve. The difference in the magnitude is partly because of the error involving the \( P_1 \) approximation and
Figure 2.16. The buckling dependence of the extrapolation distance in "equivalent" slab geometries. The variation with $M_2$ is indicated.
Table 2.20. The buckling dependence of the extrapolation distance in "equivalent" slab geometry for different values of $M_2$

<table>
<thead>
<tr>
<th>$\frac{B_g^2}{(\text{cm}^{-2})}$</th>
<th>Extrapolation distance, $d(B_g^2)\text{ cm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$M_2 = 0.67 \text{ cm}^{-1}$</td>
</tr>
<tr>
<td>0.000</td>
<td>0.3405</td>
</tr>
<tr>
<td>0.025</td>
<td>0.3366</td>
</tr>
<tr>
<td>0.050</td>
<td>0.3327</td>
</tr>
<tr>
<td>0.150</td>
<td>0.3180</td>
</tr>
<tr>
<td>0.200</td>
<td>0.3113</td>
</tr>
<tr>
<td>0.300</td>
<td>0.2994</td>
</tr>
<tr>
<td>0.500</td>
<td>-**</td>
</tr>
<tr>
<td>0.700</td>
<td>-**</td>
</tr>
<tr>
<td>0.900</td>
<td>-**</td>
</tr>
</tbody>
</table>

**The theory does not hold at this value of $\frac{B_g^2}{(\text{cm}^{-2})}$.

partly because of the error in the $L_1$ approximation; besides, the two kernels are different.

From the foregoing, it can be concluded that in order for the $P_1$-$L_1$ approximation in spherical geometry to be useful in the analysis of pulse experiments the extrapolation distance of an equivalent slab should be used. This is because the Marshak's boundary condition is more suited to a slab than to a spherical geometry.
III. EQUIPMENT

The main components of the apparatus used in the experimental work are

A. Neutron generator.
B. Pulsing system.
C. 400-channel analyser.
D. Timer system.
E. Neutron detection system.
F. Spherical containers.
G. Shielding facility and detector mount.

The experimental arrangement of these components was as indicated by the schematic diagram shown in Figure 3.1. The individual components are described below. The generator control and pulsing console, 400-channel analyser, timer system and monitor detector are shown in Figure 3.2.

A. Neutron Generator

Fast neutrons were produced by the reaction of an accelerated positive ion beam (deutrons or protons) on a tritium target according to the following reaction:

\[ \text{D}^2 + \text{T}^3 \rightarrow \text{n}^1 + \text{He}^4 + 17.6 \text{ Mev.} \]

The ion beam was produced by the Texas Nuclear Corporation Model 9400 Neutron Generator whose operation depends on the production, extraction and acceleration of ions. The major
Figure 3.1. Arrangement of experimental equipment
Figure 3.2. Neutron generator and pulsing console, monitor scaler timer system, 400-channel analyzer, and accessory equipment
components of this generator are shown pictorially in Figure 3.3 and schematically in 3.4. Positive ions are produced in a radio-frequency type ion source and are extracted by applying a potential across the ion source bottle. The ions are focused by a gap lens situated directly after the exit canal of the ion source base. The ions leave the gap lens and enter the field of the accelerating tube where they are accelerated through a potential of 150 Kv. After leaving the accelerating tube, the ions drift through a potential free region (drift tube) until they strike the target. A vacuum is maintained through the entire system to minimize scattering of the ion beam.

The ion source used in the generator is a radio frequency type which is capable of producing an ion beam current in excess of one milliamper. The current is composed approximately of 90% singly ionized atomic ions and 10% molecular ions. Hydrogen (or deuterium) gas is allowed to flow into the pyrex ion bottle by means of a palladium leak. The gas from the leak enters through a hole in the ion source base. An r-f field (approximately 60 Mc/sec.) applied to the two excitor rings causes intense ionization of the hydrogen gas. The positive ions in the discharge are forced towards the exit canal by applying a positive potential across the bottle. A magnetic field whose lines of force are in the direction of the long axis of the bottle
Figure 3.3. Neutron generator
Figure 3.4. Major components of neutron generator
is produced by a solenoid coil and serves in restricting the electron paths to the center portion of the bottle and causes them to spiral. The spiraling motion increases the ionization probability in the region of the exit canal.

The target used with the 9400 consists of tritium absorbed onto a thin layer of titanium approximately 2 to 3 mg/cm² thick. The layer has been evaporated onto a 0.01 inch thick copper disc. The active area of the disc is 1.0 inch in diameter.

B. Pulsing System

The Texas Neutron Generator is equipped with a dual pulsing system which essentially eliminates any residual beam between pulses. This system is composed of pre-acceleration and post-acceleration systems operating simultaneously. The post and pre-acceleration systems are similar in the electrostatic deflection of the beam. After leaving the ion source, the beam is deflected by the pre-acceleration system. The post-acceleration system deflects the beam in the drift-tube section after it has been accelerated.

By using the dual pulsing system, an ion beam current is supplied to the target with the following specifications:

1. Pulse repetition rates over the continuous range from $10^{-10}$ pps, or continuous beam operation.
2. Pulses 1 μ sec. to $10^4$ μ sec. duration; duty cycle not to exceed 90 percent.
3. Pulse rise and decay times of approximately 0.5 μ sec.
4. Peak pulse currents variable from 0 to 1 ma.
5. Residual beam between pulses approximately 0.0006 percent of the peak pulse current.

The above specifications were obtained from the Texas Nuclear Corporation instruction manual for pulsing systems (50).

C. 400-Channel Analyzer

The neutron flux as a function of time was recorded by the RIDL Model 34-12B transistorized 400-channel analyzer made by the Radiation Instrument Development Laboratory. The time analyzer was designed with a channel selector to provide 50, 100, 200 or 400 total channels with any desired duration. In its time mode it operates as though it were a large number of single channel analyzers. Each address channel becomes the equivalent of one single channel analyzer, with gross counting of input pulses through a controlled time period in each of the sequentially assigned channels. At the end of the time interval, the address is advanced to the next sequential channel. This operating cycle is advanced through the analyzer with a repetition rate determined by the product of the channel width and the total number of channels selected.
This means that the total time between pulses is constant, with any error in time divided by the number of channels that have been selected. The minimum cycle time for this operation is 10 $\mu$ sec. per channel.

There is provision for the temporary storage of one count during address advance, thus providing an effective zero dead time when the probability of two or more pulses within the 10 $\mu$ sec. time interval is small.

D. Timer System

For the operation of the channel analyzer in its time mode, an accessory time base control was used. This system is a Radiation Instrument Development Laboratory Model 88-901 Timer System. It consists of a set of two single size modular units installed in a Designer Series Model 29-1 instrument case and power supply. These units are a Model 54-6 Time Base Generator and a Model 52-9 Time Mode System Controller.

The Model 54-6 Time Base Generator furnishes pulses to the analyzer to provide channel advance. The timing provided for a channel width is adjustable from 12.5 $\mu$ sec. to 800 $\mu$ sec. The analyzer dead time is held to a constant 12.5 $\mu$ sec. for the shorter channel widths. A normal automatic cycle consists of a period of dwell time, the same period of dwell in the second channel, a channel advance, etc. This is repeated until the number of channels selected as a subgroup
in the analyzer has been used; then the analyzer signals completion of one sequence and the Model 54-6 may stop automatically or it may repeat the sequence, depending on settings of its controls.

The Model 52-9 provides an optional automatic programming control for Model 34-12 and Model 54-6 combination. It permits the analyzer, operating in the time mode, to be recycled through a preset number of store cycles and then to be transferred to a read cycle for automatic readout or printout.

E. Neutron Detection System

The components of this system are two BF₃ proportional counters, a preamplifier, a linear amplifier and a Radiation Instrument Development Laboratory scaler.

The first detector serves as a transverse detector inside the sphere. This detector is a Miniature Model Mn i, produced by the N. Wood Counter Laboratories. It is one-fourth inch in diameter and one inch long. Its active length is about 2.2 cm. The filling gas is BF₃ with 96 percent enrichment in B¹⁰. The gas pressure is only known to lie between 20 and 60 centimeters of mercury. For protection against water, the detector was sealed inside a long lucite light pipe to be held by a clamp mounted on a vertically graduated aluminum holder.
The second BF\textsubscript{3} neutron detector was used as a monitor for normalization purposes. The detector is about one-half inch in diameter and four inches long. The active length is about one inch. The detector was placed at a fixed position inside the shielded tank and coupled to a Radiation Instrument Development Laboratory scaler.

At intervals during the series of measurements, the detector and the time analyser were checked by a Chi-square test for randomness and reproducibility.

**F. Spherical Containers**

The spherical geometry was made possible by placing water inside round bottom pyrex flasks. Each flask was chosen to have the narrowest possible neck so that it would approximate a sphere when filled up to the neck with distilled water. The degree of sphericity of each flask was checked by comparing its average radius, as determined by volumetric methods, with the radius along the neck axis measured from the center to the water level. The two values were found to agree up to the first decimal.

The flasks used have the following average radii:

<table>
<thead>
<tr>
<th>Flask number</th>
<th>Average radius</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>17.025 ± 0.005 cm.</td>
</tr>
<tr>
<td>2</td>
<td>14.412 ± 0.005 cm.</td>
</tr>
<tr>
<td>3</td>
<td>10.830 ± 0.004 cm.</td>
</tr>
<tr>
<td>4</td>
<td>8.968 ± 0.003 cm.</td>
</tr>
<tr>
<td></td>
<td>Diameter (cm)</td>
</tr>
<tr>
<td>---</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>5</td>
<td>7.735 ± 0.003 cm.</td>
</tr>
<tr>
<td>6</td>
<td>6.348 ± 0.004 cm.</td>
</tr>
<tr>
<td>7</td>
<td>4.863 ± 0.001 cm.</td>
</tr>
<tr>
<td>8</td>
<td>4.196 ± 0.0005 cm.</td>
</tr>
<tr>
<td>9</td>
<td>3.553 ± 0.0003 cm.</td>
</tr>
</tbody>
</table>

The spheres were held in position by placing them on cork rings lined with 20 mils cadmium to minimize scattering by the cork material.

G. Shielding Facility and Detector Mount

Figure 3.5 is a photograph of the rectangular tank enclosure used to prevent room-return neutrons from entering the water system. The tank is supported by a wooden frame 17.5 x 17.5 x 19.0 inch and lined from inside to the outside with a 40-mil cadmium sheet followed by a paraffin layer 1.25 inch thick, 20-mil cadmium layer then an outermost layer of 2-inch thick plexiglass. An opening just large enough for the target assembly was left in the middle of the side facing the drift tube. The top of the assembly was covered by similar layers which could be removed to insert the sphere and allow the positioning of the traverse detector at a given source-sphere distance.

The detector holder consists of an aluminum sliding bridge, a graduated vertical stand and a plastic sliding clamp for holding the detector. The bridge slides on alu-
Figure 3.5. Shielding facility and detector mount
minum rails in a direction parallel to the axis of the drift tube. The vertical stand is positioned at the middle of the sliding bridge and supports the detector clamp that can be moved in a vertical direction. Thus the motion of the detector is restricted to an axial and vertical direction with respect to the source.
IV. EXPERIMENTAL PROCEDURE

To measure the flux distribution in a sphere along its Z-direction (the neck axis) it was necessary to locate the effective center of the detector. This was done by sliding the detector into a cadmium sleeve with a transverse slit 0.2 cm wide. The position of the slit on the detector was recorded and the detector was then placed inside a paraffin block in a neutron field from Po-Be source. The count rate at the given slit position was taken and the procedure was repeated until the whole length of the detector was surveyed. The differential curve obtained by this method and corrected for the background is shown in Figure 4.1 together with the corresponding integral curve. This integral curve represents the total count rate as a function of the Cd-uncovered length of the detector.

The differential curve is seen to exhibit an asymmetric Gaussian distribution. The position at the maximum of this distribution corresponds to the effective center of the detector; while the full width at half-maximum was considered as the effective length of the detector.

The average radius of each sphere was determined according to the following procedure. The sphere was cleaned with chromic-sulphuric acid mixture and rinsed with distilled water, alcohol, and then ether. After evaporating
Figure 4.1. Determination of the effective center of the BF$_3$ detector
Differential Curve

Integral Curve
the ether in a dry air current and allowing the sphere to reach equilibrium at room temperature, water was added quantitatively to a level at which the tangent of the meniscus coincides with the spherical continuation at the neck. The transferal of water was carried out using volumetric flasks and burettes standardized at 20°C. With the volume of water in the sphere known, the average radius could be calculated.

Each sphere containing light water was symmetrically bombarded at the equatorial plane by pulses of fast neutrons (see Figure 4.2). These neutrons were produced by the D-T reaction as described earlier in chapter III. The ion beam was constantly maintained at 600 microamps. Pulsing was carried out at a rate of 50 pulses per second, with a neutron pulse width of 100 microsecond.

The counts from the detector were recorded and stored in the 400-channel analyzer. One hundred channels were used each having a 25-microsecond channel width and a dead time of 12.5 microseconds. The monitor detector counts were recorded on the monitor scaler that served as a basis to normalize each run to a constant pulsed source exposure.

Measurements were taken at a detector position until the monitor registered a preassigned total counts. This ranged from 50,000 counts for the largest sphere to 120,000 counts for the smallest one. The monitor and the 400-channel
Figure 4.2. Experimental arrangement of neutron source, the spherical container and BF$_3$ detector.
analyzer were then switched to the off position at the same time. The data stored in the 400-channel were automatically printed out by means of an accessory IBM typewriter. The detector was moved vertically to a new position and the procedure repeated until the accessible part of the z-axis was surveyed. The data were then corrected for the dead time and background.
V. ANALYSIS OF DATA, RESULTS AND DISCUSSION

A. Parameters of Pulsed Neutron Experiments

1. Determination of the decay constants for large spheres

Consider \( M \) space points along the \( z \)-axis (see Figure 5.1). Then at time \( t_k \) (which corresponds to the midpoint of the \( k \)th time channel), one has \( M \) measurements of the neutron flux

\[
\phi(r_m, t_k) = \sum_{i=0}^{\infty} A_i(t_k) j_0(B_i r_m)
\]

(5.1)

where

\[
B_i^2 = \left[ \frac{(i + 1)\pi}{R + d(B_i g)} \right]^2, \quad i = 0, 1, 2, \ldots
\]

\[
B_0^2 = B_0^2 = \text{The geometric buckling.}
\]

Using only \( N \) terms in 5.1, one has \( M \) equations of conditions and \( N + 1 \) unknown coefficients to determine

\[
\phi(r_1, t_k) = A_0(t_k) j_0(B_0 r_1) + A_1(t_k) j_0(B_1 r_1) + \ldots
\]

\[
+ A_N(t_k) j_0(B_N r_1)
\]

\[
\phi(r_M, t_k) = A_0(t_k) j_0(B_0 r_M) + A_1(t_k) j_0(B_1 r_M) + \ldots
\]

\[
+ A_N(t_k) j_0(B_N r_M)
\]

(5.2)

or, using matrix notation,
Figure 5.1. Flux mapping along the z-axis of a sphere with $R_z = 14.46$ cm and an average radius of $14.412$ cm.
\[ x(t_k) = W \Delta(t_k) \tag{5.3} \]

where \( x \) is a \( M \)-dimensional column vector, \( \Delta \) is an \((N + 1)\)-dimensional column vector and \( W \) is an \( M \times (N + 1) \) matrix. By pre-multiplication of both sides of 5.3 by \((W^TW)^{-1}W^T\), one gets

\[ \Delta(t_k) = (W^TW)^{-1}W^T x(t_k) \tag{5.4} \]

The harmonic amplitudes of the four largest spheres were obtained from 5.4 by the method of least squares (32). Examples of these amplitudes are shown in Figure 5.2 and 5.3. For amplitudes higher than \( A_2 \), oscillations with time were observed. They were found to be independent of the truncation order of 5.2. The occurrence of these oscillations became significant for small spheres. The data in Table 5.1 illustrates this phenomenon for a sphere with an average radius of 8.968 cm. From this table one observes the fundamental amplitude decreasing with time in an exponential way free from any oscillation. The oscillations appear with the first harmonic amplitude and increase with increasing order of the harmonic.

There are two possible explanations for this unique feature of the spherical geometry. The first is the fact that the zeroth-order spherical Bessel functions have amplitudes that behave like \( 1/r \) and the count rate per unit length of a detector in a sphere is not symmetrically dis-
Figure 5.2. Relative decay of the fundamental mode and the higher harmonics along the z-axis of a sphere with $R_{av} = 17.025$ cm, $R_z = 16.42$ cm
$A_2(t)$, Arbitrary Units

The graph shows three sets of data points labeled $A_0(t)$, $A_1(t)$, and $A_2(t)$. The data points are plotted on a logarithmic scale for both the channel number and the arbitrary units, indicating a linear relationship on a logarithmic scale.
Figure 5.3. Relative decay of the fundamental and the higher harmonics along the $z$-axis of a sphere with $R_{av} = 14.42$ cm
Table 5.1. Oscillatory behavior of higher harmonics for a sphere with an average radius of 8.968 cm

<table>
<thead>
<tr>
<th>Channel number</th>
<th>$A_0(t)$</th>
<th>$A_1(t)$</th>
<th>$A_2(t)$</th>
<th>$A_3(t)$</th>
<th>$A_4(t)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>67622.69</td>
<td>-6493.60</td>
<td>1638.28</td>
<td>-112.21</td>
<td>-181.51</td>
</tr>
<tr>
<td>5</td>
<td>46423.29</td>
<td>-2718.35</td>
<td>396.64</td>
<td>85.40</td>
<td>-25.68</td>
</tr>
<tr>
<td>6</td>
<td>32480.63</td>
<td>-1653.73</td>
<td>513.06</td>
<td>40.33</td>
<td>-91.37</td>
</tr>
<tr>
<td>7</td>
<td>22510.83</td>
<td>-801.09</td>
<td>449.31</td>
<td>-139.06</td>
<td>58.84</td>
</tr>
<tr>
<td>8</td>
<td>15956.35</td>
<td>-290.89</td>
<td>288.42</td>
<td>-82.03</td>
<td>-45.41</td>
</tr>
<tr>
<td>9</td>
<td>11216.57</td>
<td>-82.61</td>
<td>104.78</td>
<td>-5.16</td>
<td>25.82</td>
</tr>
<tr>
<td>10</td>
<td>8119.30</td>
<td>-78.24</td>
<td>84.74</td>
<td>-23.30</td>
<td>-4.29</td>
</tr>
<tr>
<td>11</td>
<td>5736.41</td>
<td>-96.42</td>
<td>205.52</td>
<td>-95.96</td>
<td>5.28</td>
</tr>
<tr>
<td>12</td>
<td>4204.54</td>
<td>-5.86</td>
<td>43.13</td>
<td>-37.42</td>
<td>25.49</td>
</tr>
<tr>
<td>13</td>
<td>3033.79</td>
<td>-84.15</td>
<td>48.65</td>
<td>-12.21</td>
<td>1.09</td>
</tr>
<tr>
<td>14</td>
<td>2166.71</td>
<td>-24.69</td>
<td>21.98</td>
<td>-7.92</td>
<td>3.46</td>
</tr>
<tr>
<td>15</td>
<td>1538.44</td>
<td>-15.48</td>
<td>-2.81</td>
<td>9.98</td>
<td>-3.33</td>
</tr>
<tr>
<td>16</td>
<td>1133.28</td>
<td>-25.16</td>
<td>25.31</td>
<td>-9.82</td>
<td>-1.04</td>
</tr>
<tr>
<td>17</td>
<td>764.18</td>
<td>-23.82</td>
<td>31.06</td>
<td>-8.03</td>
<td>4.31</td>
</tr>
</tbody>
</table>

tributed about the effective center of the detector. As a result, the detector does not behave like a point detector and would be, in a sphere, more sensitive to any variation with time than it would be in any other geometry. For the sake of illustration consider a sphere with an extrapolated radius $R_{\text{ext}}$ and a slab with an extrapolated thickness $H_{\text{ext}}$. Consider two similar detectors placed in positions as shown in Figure 5.4. For detector number 2 there is a zero net contribution from the two equal shaded areas on both sides.
Figure 5.4.a. First and second harmonic in a sphere

Figure 5.4.b. First and second harmonic in a slab
of its effective center. On the other hand, the net contribution to detector number 1 is non-zero since the two areas are not equal. This implies, therefore, that detector number 2 at that position behaves like a point detector; while number 1 retains the response of its effective length.

The other possible explanation is the inefficiency of the expansion form given by 5.2. However, it will be found later that the method of this section gives fundamental decay constants compatible with both theoretical and experimental results obtained by other authors.

In either case, it should not be taken for granted that these oscillations are inherent in the character of the higher amplitudes and, accordingly, that they describe the traveling wave phenomenon discussed before. Rather, there is a belief that if the expansion 5.2 is proper, these oscillations are mere contaminations, the degree of which depends on the order of the amplitude and the size of the sphere. In support of this idea is the fact that it is possible to isolate a portion of the oscillating harmonic amplitude that decays exponentially with decay constant characteristic of the given harmonic.

The data for the harmonic amplitudes, or selected portions thereof, were then analyzed by means of the "Cornell Method" (10) assuming that, for the kth channel of width $\Delta t$:

$$A_i(t_k) = A \exp[-\lambda_i k(\Delta t)]$$  \hspace{1cm} (5.5)
where \( A \) is a constant and \( \lambda_i \) is the presumed decay constant of the \( i \)th harmonic. The method is summarized in that, for \( n \) channels such that \( n/3 = b \) is an integer, \( \lambda_i \) is given by

\[
\lambda_i = -\frac{1}{b\Delta t} \ln \left( \frac{\sum_{k=b+1}^{2b} |A_i(t_k)| - \sum_{k=2b+1}^{3b} |A_i(t_k)|}{\sum_{k=1}^{b} |A_i(t_k)| - \sum_{k=b+1}^{2b} |A_i(t_k)|} \right)
\]  
(2.6)

The variance of the fundamental decay constant was calculated by separately analyzing successive portions of the \( n \)-channel data utilizing four overlapping series of \( (n-3) \) channels each \( (1-(n-3), 2-(n-2) \) and \( 4-n) \) or seven overlapping series of \( (n-6) \) channels each \( (1-(n-6), \ldots, 7-n) \). This procedure takes into account the random distribution of the original counts about their best fit.

In several cases there were uncertainties in the radial buckling along the \( z \)-axis as a result of the uncertainty in the water level at the neck of the sphere. In these cases more than one run was made with varying water level. The points of each run were treated separately using the above procedure.

Values of \( \lambda_0 \) for the four largest spheres are listed in Table 5.2 together with both the average and the corresponding radial bucklings along the \( z \)-axis. It should be noted that the values of the fundamental decay constants are for the \( z \)-direction unless it is otherwise stated. In calculating the
Table 5.2. Fundamental decay constants for the largest spheres. The space points used in the analysis are indicated.

<table>
<thead>
<tr>
<th>$R_{av}$</th>
<th>$B_{g,av}^2$</th>
<th>$R_z$</th>
<th>$B_{g,z}^2$</th>
<th>$\lambda_0$ (sec.$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(cm)</td>
<td>(cm$^{-2}$)</td>
<td></td>
<td></td>
<td>5 space points</td>
</tr>
<tr>
<td>17.025</td>
<td>0.03270</td>
<td>16.420</td>
<td>0.0351</td>
<td>---</td>
</tr>
<tr>
<td>± 0.005</td>
<td>± 0.008</td>
<td></td>
<td></td>
<td>5910 (for $B_z^2$)</td>
</tr>
<tr>
<td>14.412</td>
<td>0.04508</td>
<td>14.460</td>
<td>0.04500</td>
<td>6452±207</td>
</tr>
<tr>
<td>± 0.005</td>
<td>± 0.006</td>
<td></td>
<td></td>
<td>6457±215</td>
</tr>
<tr>
<td>14.412</td>
<td>0.04508</td>
<td>14.400</td>
<td>0.04504</td>
<td>7825±157</td>
</tr>
<tr>
<td>± 0.005</td>
<td>± 0.05</td>
<td></td>
<td></td>
<td>7798±165</td>
</tr>
<tr>
<td>10.830</td>
<td>0.07900</td>
<td>10.790</td>
<td>0.07960</td>
<td>7798±165</td>
</tr>
<tr>
<td>± 0.004</td>
<td>± 0.05</td>
<td></td>
<td></td>
<td>7998±165</td>
</tr>
<tr>
<td>8.968</td>
<td>0.1137</td>
<td>8.97</td>
<td>0.1137</td>
<td>9069±240</td>
</tr>
<tr>
<td>± 0.003</td>
<td>± 0.03</td>
<td></td>
<td></td>
<td>9069±240</td>
</tr>
</tbody>
</table>

$^aR_{av}$ and $B_{g,av}^2$ are the average radius and the average buckling respectively.

$^bR_z$ is the radius measured along the z-axis and $B_{g,z}^2$ is the corresponding buckling.
the buckling, the $P_3$ extrapolation distances (24) were used.

The decay constants for the higher harmonics are given in Table 5.3. For the sake of comparison, the theoretical values were calculated from

$$\lambda_i = \lambda_0 + [(i + 1)^2-1]D_0 B^2_{g,z} - [(i + 1)^4-1]C B^4_{g,z} \tag{5.7}$$

where $B^2_{g,z}$ is the radial buckling along the z-axis, $D_0 = 38692 \text{ cm}^2 \text{ sec.}^{-1}$, $V_o \Sigma_a = 4876 \text{ sec.}^{-1}$ and $C = 3618 \text{ cm}^4 \text{ sec.}^{-1}$. These values are the $P_1$-$L_1$ values for the Nelkin's water (see Table 2.12). The results are also listed in Table 5.3 from which it is clear that both the theoretical and experimental results agree with each other within 9%.

2. **Determination of the decay constants for small spheres**

The method used here to separate the fundamental mode decay from the higher modes is based on the determination of the optimum source distance and the waiting time. The source distance is here defined as the distance between the target and the facing point on the surface of the sphere (see Figure 4.2). The ratio of this distance to the average radius of the given sphere will be referred to as the "normalized" source distance, $s$. Figure 5.5 is a plot of the ratio of the first and the second mode amplitude to the fundamental as a function of this distance. The plot is for a sphere with an average radius of 10.83 cm at the 8th time channel.
Table 5.3. Decay constants of higher harmonics along the z-axis of the corresponding sphere

<table>
<thead>
<tr>
<th>$B_0^2$ (cm$^{-2}$)</th>
<th>$\lambda_1$ (sec.$^{-1}$)</th>
<th>$\Delta\lambda_1$%</th>
<th>$\lambda_2$ (sec.$^{-1}$)</th>
<th>$\Delta\lambda_2$%</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0351</td>
<td>9425</td>
<td>10249</td>
<td>8.9</td>
<td>15324</td>
</tr>
<tr>
<td>0.0450</td>
<td>11120</td>
<td>11723</td>
<td>5.4</td>
<td>18660</td>
</tr>
<tr>
<td>0.0796</td>
<td>16020</td>
<td>16833</td>
<td>5.1</td>
<td>28500</td>
</tr>
<tr>
<td>0.1137</td>
<td>20936</td>
<td>21739</td>
<td>3.8</td>
<td>37813</td>
</tr>
</tbody>
</table>

According to this Figure, s is about 0.4, i.e., double the value reported for rectangular geometry (34). Data for other channels essentially gave similar results.

The waiting time is defined as that time required for a particular mode in a given sphere to decay to 1 percent of the fundamental amplitude. Use has been made of the data of the previous section to plot the waiting time $t_w$ versus sphere average radius $R_{av}$ in Figure 5.6. The curves were then interpolated to lower values of $t_w$. The Figure shows that for radii less than 6 cm the waiting time is practically zero. Since the first mode can be eliminated to an appreciable extent by placing the detector at half of the extrapolated radius, the waiting time would be mainly determined by the second mode amplitude.

The data obtained by the waiting time method were then fitted to the logarithmic difference of the counts in two
Figure 5.5. Relative harmonic content at the 8th time channel as a function of normalized source distance, s
Figure 5.6. Experimental minimum waiting time versus average radius, $R_{av}$
For $A_1$

For $A_2$

$t_w$ (Microseconds) vs. $R_{av}$ (Cm)
successive time channels. If the number of counts in a given channel is of the form
\[ N_n = A \exp(-\lambda_0 t_n) + B_G \quad , \tag{5.8} \]
where \( B_G \) is the background and assumed to be constant, then the difference between the \( n \)th channel and the \( (n + 1) \)th channel will not contain the background. The logarithm of the difference is
\[ \ln(N_n - N_{n+1}) = -\lambda_0 t_{n+1} + \ln(A(\exp(-\lambda_0 \Delta t)-1)) \quad (5.9) \]
The result is a straight line with a slope of \( \lambda_0 \).

The fundamental decay constants obtained by the above procedure for five small spheres are tabulated in Table 5.4 where \( B^2_\text{g,av} = B^2_\text{g,z} = B^2_\text{g} \). The large variance in this table is probably due to the poor counting statistics for small systems.

3. Determination of the diffusion parameters

The usual fit of the experimental points to the function
\[ \lambda_0 = f(B_\text{g}^2), \text{ i.e.,} \]
\[ \lambda_0 = V_0 \Sigma_{ao} + D_0 B^2_\text{g} - C B^4_\text{g} + O B^6 \quad (5.10) \]
is done here according to the following procedure (19): consider
Table 5.4. Fundamental decay constant for small spherical geometries

<table>
<thead>
<tr>
<th>Radius of the sphere, ( R ) (cm)</th>
<th>Buckling, ( B_g^2 ) (cm(^{-2}))</th>
<th>( \lambda_0 ) (sec(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.735 ± 0.003</td>
<td>0.1470</td>
<td>10633 ± 155</td>
</tr>
<tr>
<td>6.348 ± 0.004</td>
<td>0.2206</td>
<td>12996 ± 214</td>
</tr>
<tr>
<td>4.863 ± 0.001</td>
<td>0.3651</td>
<td>18239 ± 362</td>
</tr>
<tr>
<td>4.196 ± 0.0005</td>
<td>0.4810</td>
<td>22101 ± 460</td>
</tr>
<tr>
<td>3.553 ± 0.0003</td>
<td>0.6553</td>
<td>28050 ± 810</td>
</tr>
</tbody>
</table>

\( R = R_{av} = R_z \).

\[
\sum W_i \lambda_0 = V_0 \sum \lambda_0 + D_0 \sum W_i B_g^2 + C \sum W_i B_g^4
\]

\[
\sum W_i B_g^2 \lambda_0 = V_0 \sum \lambda_0 + D_0 \sum W_i B_g^2 + C \sum W_i B_g^4
\]

\[
\sum W_i B_g^4 \lambda_0 = V_0 \sum \lambda_0 + D_0 \sum W_i B_g^2 + C \sum W_i B_g^4
\]

where \( W_i \) is the inverse of the variance of \( \lambda_0 \) and \( \Sigma \) denotes the summation over all the experimental points \( \lambda_0 \) and the corresponding bucklings. The coefficient matrix, \( A \), is given by

\[
A = \begin{bmatrix}
\sum W_i & \sum W_i B_g^2 & \sum W_i B_g^4 \\
\sum W_i B_g^2 & \sum W_i B_g^4 & \sum W_i B_g^6 \\
\sum W_i B_g^4 & \sum W_i B_g^6 & \sum W_i B_g^8 
\end{bmatrix}
\]
The inverse elements $s_{lm}$ are obtained by taking the cofactor of $lm$ and dividing by the determinant $\det(A)$.

$$ s_{lm} = \frac{\text{cofactor of } lm}{\det(A)} $$

1, $m = 1, 2, 3$

The inverse matrix is denoted by

$$ A^{-1} = \begin{bmatrix}
    s_{11} & s_{12} & s_{13} \\
    s_{21} & s_{22} & s_{23} \\
    s_{31} & s_{32} & s_{33}
\end{bmatrix} \quad (5.13) $$

$$ V_0 \Sigma_{ao} = s_{11} \sum_i W_i \lambda_0 + s_{12} \sum_i W_i \lambda_0 B_g^2 + s_{13} \sum_i W_i \lambda_0 B_g^4 \quad (5.14) $$

$$ D_0 = s_{21} \sum_i W_i \lambda_0 + s_{22} \sum_i W_i \lambda_0 B_g^2 + s_{23} \sum_i W_i \lambda_0 B_g^4 \quad (5.15) $$

$$ C = s_{31} \sum_i W_i \lambda_0 + s_{32} \sum_i W_i \lambda_0 B_g^2 + s_{33} \sum_i W_i \lambda_0 B_g^4 \quad (5.16) $$

The corresponding standard deviations are given by:

$$ d(V_0 \Sigma_{ao}) = \left[ s_{11} \sum_i W_i e_i^2 / (N - 3) \right]^{\frac{1}{2}} \quad (5.17) $$

$$ d_{D_0} = \left[ s_{22} \sum_i W_i e_i^2 / (N - 3) \right]^{\frac{1}{2}} \quad (5.18) $$

$$ d_C = \left[ s_{33} \sum_i W_i e_i^2 / (N - 3) \right]^{\frac{1}{2}} \quad (5.19) $$

where $N$ is the number of points used in the least-squares fit
and \( e_i = \chi_0 \) (calculated) \(-\) \( \chi_0 \) (observed)

It was felt that the use of \( \dot{W}_i = 1 \) yields a more realistic set of parameters for these reasons. 1) Large geometries were not perfect spheres and the radius along the z-axis differs from the average radius. This difference was small for small geometries. 2) Large spheres had wide necks and the water levels at these necks were flat. Hence, there is an uncertainty in the effective center of the sphere. 3) A weighing factor equal to the inverse of the statistical variance of \( \chi_0 \) tends to weigh more heavily the experimental points of large geometries.

The procedure was applied to the data given in Table 5.1 and 5.4. For spheres with more than one run only values of \( \chi_0 \) that made the variance of \( C \) a minimum were retained. Table 5.5 summarizes the selected data for the best fit in the given range of buckling. Values of \( \chi_0 \) for spheres where there is a large difference between the average geometric buckling \( B_{g,av}^2 \) and the radial buckling \( B_{g,z}^2 \) along the z-axis are omitted from this table. For the rest \( B_{g,av}^2 = B_{g,z}^2 = B_g^2 \). The results of the fit for various ranges of bucklings are summarized in the following:

1. Fitting range from 0.045 to 0.365 cm\(^{-2}\).
   
   \[
   V_0 \Sigma a_0 = (4.734 \pm 0.036) \times 10^3 \text{ sec}^{-1}
   \]
   
   \[
   D_0 = (3.8892 \pm 0.0442) \times 10^4 \text{ cm}^2 \cdot \text{sec}^{-1}
   \]
Table 5.5. Experimental values of the fundamental decay constant that give the best fit in the indicated buckling range

<table>
<thead>
<tr>
<th>( B_g^2 (\text{cm}^{-2}) )</th>
<th>( \lambda_0 (\text{sec}^{-1}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.04504</td>
<td>6465</td>
</tr>
<tr>
<td>0.07960</td>
<td>7825</td>
</tr>
<tr>
<td>0.11370</td>
<td>9069</td>
</tr>
<tr>
<td>0.14700</td>
<td>10372</td>
</tr>
<tr>
<td>0.22060</td>
<td>13082</td>
</tr>
<tr>
<td>0.36510</td>
<td>18323</td>
</tr>
</tbody>
</table>

\( C = (4.590 \pm 1.034) \times 10^3 \text{ cm}^4 \cdot \text{sec}^{-1} \)

2. Fitting range from 0.0796 to 0.365 cm\(^{-2}\):

\( V_o \Sigma_{ao} = (4.767 \pm 0.068) \times 10^3 \text{ sec}^{-1} \)

\( D_o = (3.8570 \pm 0.0736) \times 10^4 \text{ cm}^2 \cdot \text{sec}^{-1} \)

\( C = (3.940 \pm 1.600) \times 10^3 \text{ cm}^4 \cdot \text{sec}^{-1} \)

3. Fitting range from 0.045 to 0.2206 cm\(^{-2}\):

\( V_o \Sigma_{ao} = (4.718 \pm 0.065) \times 10^3 \text{ sec}^{-1} \)

\( D_o = (3.9209 \pm 0.1110) \times 10^4 \text{ cm}^2 \cdot \text{sec}^{-1} \)

\( C = (5.840 \pm 4.039) \times 10^3 \text{ cm}^4 \cdot \text{sec}^{-1} \)

It is clear from these results that, at the time \( V_o \Sigma_{ao} \) and \( D_o \) are not sensitive to the range of fitting, \( C \) and its
associated standard error show a large variation. This reflects the usual inherent difficulty of determining the curvature of an unknown function defined by a small set of experimental points having finite standard deviation.

Values of the microscopic absorption cross section for hydrogen calculated on the basis of \( V_o = 220,000 \text{ cm. sec}^{-1} \) are as follows:

\[
\sigma_a^H = 321 \pm 2.46 \text{ mb} \quad \text{, fitting range: } 0.045-0.365 \text{ cm}^{-2}
\]

\[
\sigma_a^H = 323 \pm 4.7 \text{ mb} \quad \text{, fitting range: } 0.0796-0.365 \text{ cm}^{-2}
\]

The diffusion length \( L \) is deduced from the parameters by using the relation

\[
L^2 = -\frac{1}{B^2/g}
\]

where \( B^2/g \) is found for the stationary state by solving equation 5.10 with \( \lambda_0 = 0 \). The result is

\[
L^2 = \frac{V_o \Sigma_{ao}}{V_o \Sigma_{ao} C^2} (1 + \frac{V_o \Sigma_{ao} C}{D_o})^2
\]

(5.20)

When \( V_o \Sigma_{ao} C \ll D_o^2 \) equation 5.20 should yield values of \( L \) directly comparable to values measured by stationary methods. The values of \( L \) for two ranges of fitting are as follows:

\[
L = 2.887 \pm 0.024 \text{ cm} \quad \text{, fitting range: } 0.045 - 0.365 \text{ cm}^{-2}
\]

\[
L = 2.860 \pm 0.030 \text{ cm} \quad \text{, fitting range: } 0.0796 - 0.365 \text{ cm}^{-2}
\]
Table 5.6 shows a comparison between these results and those obtained by other investigators. The values of the diffusion cooling constant obtained here are lower than that obtained by Lopez and Beyster (34) and higher than that obtained by Dio (20). The relatively large standard deviation associated with the present values of C are expected on the basis of the inherent difficulty of determining the spherical parameters and hence the expected uncertainty of buckling.

The values of the other parameters are in fair agreement with those reported in Table 5.6.

The fit of the experimental data to the expression

$$\lambda_0 = V_o \Sigma_{ao} + D_o B_1^2 - C B_1^4$$  \hspace{1cm} (5.21)

was also tried. The values of $B_1^2$ were calculated according to equation 2.52 by using

$$D_o = 38570 \text{ cm}^2 \text{ sec}^{-1} \quad \text{(from the fit to } \lambda_0 = f(B_1^2))$$

$$M_2 = 3.34 \text{ cm}^{-1} \quad \text{(for Nelkin's water)}$$

Table 5.7 shows a comparison between the values of the parameters obtained from this fit and those from the previous one. This table shows:

1. Within the experimental errors, the values of $V_o \Sigma_{ao}$ and $D_o$ are essentially the same for the two fits.
2. There is improvement in the standard error of C.
3. The value of C from the new fit is as high as 1.5 times the value obtained from the other fit.
<table>
<thead>
<tr>
<th>Reference</th>
<th>$D_o$ (cm$^2$ sec$^{-1}$)</th>
<th>$C$ (cm$^4$ sec$^{-1}$)</th>
<th>$\sigma^H$ (mb)</th>
<th>$L$ (cm)</th>
<th>Range of $B^2_g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present work</td>
<td>38892 ± 442</td>
<td>4590 ± 1034</td>
<td>321 ± 2.46</td>
<td>2.887 ± 0.024</td>
<td>0.045 - 0.365</td>
</tr>
<tr>
<td>Present work</td>
<td>38570 ± 736</td>
<td>3940 ± 1600</td>
<td>323 ± 4.7</td>
<td>2.860 ± 0.03</td>
<td>0.0796 - 0.3651</td>
</tr>
<tr>
<td>Lopez and Beyster (34)</td>
<td>36700 ± 370</td>
<td>4852 ± 800</td>
<td>---</td>
<td>2.795 ± 0.016</td>
<td>0.014 - 0.018</td>
</tr>
<tr>
<td>Scott et al. (46)</td>
<td>38500 ± 800</td>
<td>---</td>
<td>320 ± 8.0</td>
<td>2.850 ± 0.050</td>
<td>0.006 - 0.018</td>
</tr>
<tr>
<td>Antonov et al. (1)</td>
<td>35000 ± 1000</td>
<td>4000 ± 1000</td>
<td>329 ± 10.0</td>
<td>2.700 ± 0.100</td>
<td>0.09 - 0.93</td>
</tr>
<tr>
<td>Dio (20)</td>
<td>35450 ± 600</td>
<td>3700 ± 700</td>
<td>328 ± 6.0</td>
<td>2.715 ± 0.06</td>
<td>0.093 - 0.87</td>
</tr>
</tbody>
</table>
Table 5.7. Comparison between the diffusion parameters obtained from the fit to $\lambda_0 = f(B_{1}^{2})$ and those from $\lambda_0 = f(B_{g}^{2})$ for the range of buckling from 0.0796 to 0.3651 cm$^{-2}$

<table>
<thead>
<tr>
<th>$\lambda_0 = f(B_{1}^{2})$</th>
<th>$\lambda_0 = f(B_{g}^{2})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_{0}\Sigma_{ao}$ (sec$^{-1}$)</td>
<td>4770 ± 68</td>
</tr>
<tr>
<td>$D_{0}$ (cm$^2$. sec$^{-1}$)</td>
<td>38493 ± 722</td>
</tr>
<tr>
<td>$C$ (cm$^4$. sec$^{-1}$)</td>
<td>5861 ± 1540</td>
</tr>
</tbody>
</table>

The speculation that follows from these observations is that if the experimental values of the decay constant previously reported are fitted to equation 5.21 a reduction in the uncertainty of the diffusion cooling constant might be achieved.

The question to be imposed at this point is, why the theoretical value of $C$ obtained from the same expression does not agree with the value of the fit. The explanation for this is the fact that in calculating $C$ its $g$ factor given according to equation 2.47 was taken to be 1.776. This value is for two Laguerre polynomials. If higher polynomials could have been taken into consideration, $g$ would have been equal to 2.004, as reported by Perez and Uhrig (42), and one would have obtained a theoretical value of $C$ more close to the value of the fit. Another reason for the discrepancy is the uncer-
Figure 5.7. Least squares fit to data of the fundamental decay constant in spherical geometry
\[ \lambda_0 = (4.734 \pm 0.036) \times 10^5 + (3.8892 \pm 0.0442) \times 10^4 \, B^2 - (4.590 \pm 1.034) \times 10^3 \, B^4 \]
tainty of $M_2$ and the low order of $P_N$ approximation used in deriving $B^{2}_{T}$. To correct for the last factor one should use a more realistic model of the Boltzmann's equation like the $P_3$ approximation.

The disadvantage of the new fit, therefore, is the strong link between theory and experiment and the fact that accurate theoretical predictions are prerequisite for the experiment. To put it more simply, the new fit requires accurate values of $M_2$ and this so far is obtained from theoretical models.

B. Extrapolation Distances of Pulsed Neutron Experiments

The effect of the uncertainty in the extrapolated end points on the diffusion parameters extracted from pulsed source experiments was emphasized in earlier work (24, 34) and it was clear that more accurate measurements, especially in spherical geometries, were required. Beckurts (2) stated this need quite explicitly. The differences reported between extrapolated end points obtained by flux plotting in pulsed and steady state experiments also called for further investigation. Early pulse measurements in water at about 20°C (8, 18) gave values in the range 0.4 - 0.46 cm which are well above the upper limits now indicated by steady state values (in the range 0.32 - 0.35 cm) (56). To help in resolving these inconsistencies further measurements have been
carried out according to the following procedure:

A computer program was written to perform the least squares fitting of the extrapolation distance data to equation 5.4. A trial value, $d^{(0)}$, of the extrapolation distance, $d$, was first found graphically from the flux plots. Values of $d$ in the range from $(d^{(0)} - 0.5)$ to $(d^{(0)} + 0.5)$ cm were tried with an increment of 0.05 cm. In each case the amplitudes ($A_i$), the corresponding flux at the various space points and the sum ($S$) of the squares of the residuals were computed. The value of $d$ corresponding to the minimum of the function $S = f(d)$ (see Figure 5.8) was as the best fitted value for the given increment $d$. This value was then taken for $d^{(0)}$ and the procedure repeated with smaller increment until the desired accuracy was reached.

To take into account the statistical fluctuation of $d$ with time, the method has been applied to data of five different spheres recorded at various times after the end of the fast neutron burst. The results of fitting to three harmonics are given in Table 5.8. A unit weighing factor was used in the analysis. The results generally show a relatively large standard deviation of $d$ at both short and long time after the initiation of the neutron pulse. In the former case the deviation could be accounted for by the insufficiency of the three harmonic fit and the random distribution of the count rate about the best fit. The deviation
Figure 5.8. Variation of the sum of the squares of the residuals with the trial value of the extrapolation distance for a sphere with $R_z = 14.46$ cm.
Table 5.8. Extrapolation distances for 3 harmonic fit

<table>
<thead>
<tr>
<th>Number of sphere</th>
<th>$R_x$ (cm)</th>
<th>Number of harmonics</th>
<th>Number of space points</th>
<th>Channel number</th>
<th>$A_0$</th>
<th>$A_1$</th>
<th>$A_2$</th>
<th>$S \times 10^{-4}$</th>
<th>$d$ (cm)</th>
</tr>
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<tbody>
<tr>
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<td>11</td>
<td>18</td>
<td>18179</td>
<td>-1481</td>
<td>212</td>
<td>13.94</td>
<td>.400</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>11</td>
<td>11</td>
<td>24</td>
<td>5340</td>
<td>-390</td>
<td>-75</td>
<td>9.98</td>
<td>.415</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>11</td>
<td>25</td>
<td>25</td>
<td>4126</td>
<td>93</td>
<td>-127</td>
<td>0.40</td>
<td>.380</td>
</tr>
<tr>
<td>2</td>
<td>14.46</td>
<td>3</td>
<td>11</td>
<td>14</td>
<td>22308</td>
<td>-717</td>
<td>-152</td>
<td>16.43</td>
<td>.291</td>
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<td></td>
<td>15</td>
<td>17894</td>
<td>-506</td>
<td>-158</td>
<td>6.27</td>
<td>.333</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>13934</td>
<td>-244</td>
<td>-82</td>
<td>4.65</td>
<td>.431</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>5642</td>
<td>-74</td>
<td>-74</td>
<td>3.08</td>
<td>.373</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>10.79</td>
<td>3</td>
<td>5</td>
<td>13</td>
<td>7070</td>
<td>62</td>
<td>-167</td>
<td>1.17</td>
<td>.365</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>5311</td>
<td>27</td>
<td>-11</td>
<td>1.37</td>
<td>.338</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>3833</td>
<td>172</td>
<td>-37</td>
<td>0.98</td>
<td>.347</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>2986</td>
<td>148</td>
<td>-8</td>
<td>1.20</td>
<td>.356</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>8.97</td>
<td>3</td>
<td>5</td>
<td>10</td>
<td>8083</td>
<td>21</td>
<td>36</td>
<td>.054</td>
<td>.340</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>5625</td>
<td>200</td>
<td>73</td>
<td>.36</td>
<td>.372</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>4124</td>
<td>201</td>
<td>-43</td>
<td>.016</td>
<td>.346</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>3015</td>
<td>-28</td>
<td>20</td>
<td>.005</td>
<td>.348</td>
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<tr>
<td></td>
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<td>2152</td>
<td>16</td>
<td>2.5</td>
<td>.023</td>
<td>.335</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>1496</td>
<td>92</td>
<td>-48</td>
<td>.003</td>
<td>.350</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>7.735</td>
<td>3</td>
<td>5</td>
<td>5</td>
<td>37153</td>
<td>-2030</td>
<td>-1436</td>
<td>2.44</td>
<td>.352</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>13900</td>
<td>-466</td>
<td>302</td>
<td>4.72</td>
<td>.305</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>11494</td>
<td>-1306</td>
<td>358</td>
<td>2.15</td>
<td>.370</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
at longer times would be mainly due to the statistical fluctuation of the count rate data.

To account for the effect of higher harmonics, the same data were fitted to an increasing number of modes until consistent results were obtained. These results are listed in Table 5.9. The standard deviation shown is mainly statistical in nature and does not account for the uncertainty in the value of the radius along the z-axis.

The boundary effect on d was not investigated. However, the space distribution taken for each sphere was such that points beyond a distance from the boundary of the order of one cm were excluded from the analysis. Hence, the boundary effect on the reported values is expected to be negligible.

Recently Walker et al. (56) measured d from flux distribution in pulsed cubical systems. They obtained values of

\[ 0.38 \pm 0.04 \text{ cm} \quad \text{for} \quad B^2_g = 0.087 \text{ cm}^{-2} \]

and

\[ 0.35 \pm 0.02 \text{ cm} \quad \text{for} \quad B^2_g = 0.25 \text{ cm}^{-2} \]

These values of d are in agreement with the present results.
Table 5.9. Extrapolation distances for water in spherical geometries at 22°C. The number of space points and harmonics used in the analysis are indicated.

<table>
<thead>
<tr>
<th>Number of sphere</th>
<th>$R_7$ (cm)</th>
<th>$R_{av}$ (cm)</th>
<th>$B_{g,2}^2$ (cm$^{-2}$)</th>
<th>Number of space points</th>
<th>Number of harmonics</th>
<th>$d(b_g^2)$ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>16.420</td>
<td>17.025</td>
<td>0.0349</td>
<td>11</td>
<td>5</td>
<td>0.392</td>
</tr>
<tr>
<td></td>
<td>± 0.08</td>
<td>± 0.005</td>
<td></td>
<td></td>
<td></td>
<td>± 0.05</td>
</tr>
<tr>
<td>2</td>
<td>14.460</td>
<td>14.412</td>
<td>0.0449</td>
<td>11</td>
<td>5</td>
<td>0.356</td>
</tr>
<tr>
<td></td>
<td>± 0.060</td>
<td>± 0.005</td>
<td></td>
<td></td>
<td></td>
<td>± 0.45</td>
</tr>
<tr>
<td>3</td>
<td>10.790</td>
<td>10.830</td>
<td>0.0794</td>
<td>5</td>
<td>4</td>
<td>0.352</td>
</tr>
<tr>
<td></td>
<td>± 0.050</td>
<td>± 0.004</td>
<td></td>
<td></td>
<td></td>
<td>± 0.012</td>
</tr>
<tr>
<td>4</td>
<td>8.970</td>
<td>8.968</td>
<td>0.1129</td>
<td>5</td>
<td>4</td>
<td>0.347</td>
</tr>
<tr>
<td></td>
<td>± 0.030</td>
<td>± 0.003</td>
<td></td>
<td></td>
<td></td>
<td>± 0.013</td>
</tr>
<tr>
<td>5</td>
<td>7.740</td>
<td>7.735</td>
<td>0.1510$^b$</td>
<td>5</td>
<td>3</td>
<td>0.338</td>
</tr>
<tr>
<td></td>
<td>± 0.060</td>
<td>± 0.003</td>
<td></td>
<td></td>
<td></td>
<td>± 0.033</td>
</tr>
</tbody>
</table>

$^a$The error does not include the uncertainty in the radius along the z-axis. It is mainly due to the statistical fluctuation of the value of $d$ with time.

$^b$This value was calculated using the value of $R_{av} = 7.735$ cm.
VI. SUMMARY

A study of neutron thermalization in a sphere of light water was carried out both theoretically and experimentally. The theoretical aspect of the problem was dealt with utilizing the \( P_1 \)-\( L_1 \) approximation. The space dependence was represented by the spherical Bessel functions of zeroth-order. The use of this representation made it possible to avoid the Fourier transform technique which strictly applies to infinite media. The study employed values of the thermalization parameter, \( M_2 \), corresponding to the scattering kernels of Mass-1, Mass-18, Brown & St. John and Nelkin. When the time decay constant, \( \lambda \), was plotted versus the spherical Bessel function variable, \( B^2 \), two limiting values of \( B^2 \) were obtained. One was \( B_{1,max}^2 \), below which all decay constants were real and the other one was \( B_{2,max}^2 \), beyond which no real decay constants existed.

The curve describing the fundamental time eigenvalue in the \( (\lambda, B^2) \) plane had a negative curvature at the origin for all values of \( M_2 \) investigated. The amplitude of the curvature increased with decreasing value of \( M_2 \). For Nelkin's water, the curvature was in agreement with the experimental results and in disagreement with the findings of Travelli and Calame (52) obtained from a 4-group treatment in the \( P_1 \) approximation. These authors found a positive
curvature for the Radkowsky kernel.

The sensitivity to $M_2$ was also observed in the limiting value, $\lambda_{\text{lim}}$, of the fundamental decay constant obtained graphically from the $(\lambda, B^2)$ plots. The values obtained were higher than the limit reported by Corngold and Michael (12) and lower than that obtained by Purohit et al. (45) for the Doppler corrected kernel. $\lambda_{\text{lim}}$ for the Mass-1 kernel was in excellent agreement with that obtained by Ohanian and Daitch (41).

The expansion of the fundamental eigenvalue, $\lambda_0$, in power series of $B^2$ was shown to have a radius of convergence that covers all the buckling ranges available to experiments. On the other hand, $\lambda_1$ (the first eigenvalue) had a very small radius of convergence and its expansion in power series of $B^2$ was shown to be questionable even for large geometries.

The "buckling-dependence" of the diffusion cooling coefficient in the $P_1$ approximation was suppressed by expanding the fundamental decay constant in power series of $B^2_T$, a slowly varying function of $B^2$. The difference between $B^2_T$ and $B^2$ was correlated to the difference between the $P_1$ and the diffusion approximation in the time dependent case. Values of the decay constant obtained from the function $\lambda_0 = f(B^2_T)$ differed by no more than fractions of percent from the corresponding values obtained from the original polynomial given by 2.37.

The thermalization time constant was obtained from the
limiting case of the transcendental quadratic equation in \( \lambda_{1} \), as \( B^2 = \Sigma_{\infty} = 0 \). Two cases were considered. A case in which the transport mean free path varied as \( \sqrt{E} \) and another in which \( \lambda_{tr} \) was constant. The values of \( t_{th} \) for the latter case were about 4 times as large as the corresponding values for the energy dependent case. The result for Nelkin's scattering kernel was in good agreement with the experimental result reported by Möller and Sjöstrand (37).

The concept of buckling for spherical water systems was investigated and found to be valid as long as the fundamental decay constant, \( K_0 \), is less than about twenty thousands times the thermalization parameter, \( M_2 \). This concept broke down for the mass-18 kernel at values of \( B^2 \) greater than 0.5 cm\(^{-2}\).

The neutron spectrum of the asymptotic distribution presented in this work exhibited the diffusion cooling phenomenon shown in the results reported by Clendenen (9) for the high order \( P_N \) approximation.

The space transient near the boundary and its effect on the extrapolation distance were studied. The transient effect was negligible beyond a distance from the boundary of the order of 0.5 cm. At the boundary the effect was determined by the neutron spectra.

A calculation of the extrapolation distances for spherical geometry showed that the Marshak's boundary condition used for the outer boundary is not suited for this type of con-
figuration and that it is desirable to work the problem in
the "equivalent" slab geometry that suited the indicated
boundary condition for the given order of the $P_N$ approxi-
mation.

Pulsing data for nine spheres with a buckling range of
0.0351 - 0.6553 cm$^{-2}$ were obtained and analyzed by the method
of least squares (32). The amplitudes of higher harmonics
showed with time an oscillatory behavior. A possible ex-
planation for this phenomenon was offered.

The data for the fundamental decay constant were fitted
to two functions. $\lambda_0 = f(g^2)$ and $\lambda_0 = f(I^2)$. Within the
experimental errors, values of the absorption cross section
and the diffusion coefficient $D_0$ obtained from the two fits
were almost identical. On the other hand, the diffusion
cooling coefficient $C$ obtained from the first fit was much
lower than that of the second fit. The new fit gave a rel-
atively improved standard deviation of $C$.

The extrapolation distances for five spheres were ob-
tained by a least squares fit to the flux distribution in
the pulsed experiments. The effect of higher harmonic con-
tamination on the magnitude of the extrapolation length and
the rule of the flux variation with time on the fitted values
were considered in the analysis.
VII. CONCLUSIONS

1. Based on a comparison between the theoretical considera-
tions of this work and that of Travelli and Calame (52), it
is concluded that the $P_1 L_1$ approximation, with a
realistic scattering kernel for light water, predicts
the time behavior in pulsed neutron experiments more
accurately than few-groups $P_1$ approximation and that
the diffusion cooling phenomenon is best exhibited in
the continuous energy representation as was anticipated
by Daitch and Ebeoglu (15).

2. In case of light water, the usual expansion of the fun-
damental decay constant is justifiable on the basis that
the value of buckling beyond which a travelling wave
phenomenon occurs is of the order of $6 \text{ cm}^{-2}$ (or even
higher (15)). This value covers all the experimental
ranges of interest. The same is not equally applicable
to the first eigenvalue, a matter that leads to doubting
the method of Purohit (43) in expanding this constant in
a power series of $B^2$.

3. Although the diffusion and the $P_1 L_1$ approximations might
be identical in the time-independent case, they are dif-
ferent from each other when the time dependence is re-
tained. This difference can be correlated with that one
between the geometric buckling and the transport buckling
4. The concept of buckling for a spherical geometry in the energy dependent case is valid as long as the asymptotic region is well established. This is not possible for small systems described by the Mass-18 gas kernel.

5. The sharp variation of the effective buckling and the effective average energy of neutrons in pulsed spheres is due to the diffusion cooling phenomenon in finite media of light water as a result of the energy dependence of the transport mean free path.

6. For reliable values of the extrapolation distances obtained from pulsed spheres one must take into account:
   i. The boundary effect due to the transient distribution. The effect of this transient is negligible for space points far from the boundary by distances of the order of 0.5 cm.
   ii. The statistical variation of the extrapolation distance with time. This could lead to large errors especially at long times after the initiation of the neutron pulse.
   iii. Effect of higher harmonics which could be serious for large geometries.

In addition, the number and distribution of the space points used in the analysis.

7. A BF$_3$ detector at a zero of a given higher harmonic might
behave as a point detector in multidimensional systems; while in a sphere it does not behave the same. This is because the count rate per unit length varies along the effective length in case of spherical geometry. As a result, a detector in a sphere is very sensitive to any fluctuation that might occur with time in the flux distribution. A unique feature that has been observed in pulsed spheres is the oscillatory behavior of the amplitudes of higher harmonics with time. The oscillations contribute to the difficulty in determining higher decay constants. Another inherent difficulty with these experiments is the determination of the effective center of the sphere. This results mainly from the neck effect. The water level at the neck is flat especially if the neck is wide. Hence, the upper and lower halves of the sphere are not symmetric. The experiments also depend strongly on the exact determination of the effective center of the detector. Any uncertainty in finding its location is reflected in the radial buckling. In general, a successful experiment in spherical geometry requires:

i. Exact location of the effective center.

ii. A small BF₃ detector.

iii. A sphere for which the average radius is very close to the radius as measured along the z-axis. This, in turn, depends on the degree of the sphericity of the body and the diameter of the neck.
VIII. LITERATURE CITED


IX. ACKNOWLEDGMENT

I wish to express my gratitude to Distinguished Professor of Engineering Dr. Glenn Murphy, Head of the Department of Nuclear Engineering, for his continuous interest, helpfulness and criticism during the progress of this work.
X. APPENDIX

The computation in the theoretical chapter employed values of the total scattering cross section and the average of the scattering cosine obtained by McMurry et al. (36). Table A.1 and A.2 show a comparison between values for the McMurry-Russell model and those obtained from different sources.

Table A.1. Comparison of total scattering cross sections.\textsuperscript{a}

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<thead>
<tr>
<th>E (ev)</th>
<th>Observed\textsuperscript{b}</th>
<th>MR\textsuperscript{c}</th>
<th>N\textsuperscript{d}</th>
<th>N-KY\textsuperscript{e}</th>
<th>KY\textsuperscript{e}</th>
<th>FG\textsuperscript{f}</th>
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<tr>
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<td>166</td>
<td>165</td>
<td>174</td>
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<td>143</td>
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<td>106</td>
<td>105</td>
<td>105</td>
<td>86.2</td>
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<td>83</td>
<td>82</td>
<td>77.7</td>
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</tr>
<tr>
<td>0.100</td>
<td>70.1</td>
<td>67.3</td>
<td>69.1</td>
<td>70</td>
<td>72</td>
<td>67.1</td>
</tr>
<tr>
<td>0.150</td>
<td>63.1</td>
<td>61.0</td>
<td>62.3</td>
<td>64</td>
<td>64</td>
<td>62.4</td>
</tr>
</tbody>
</table>

\textsuperscript{a}The data were compiled by McMurry et al. (36).

\textsuperscript{b}Data for E < 0.1 ev are from Hughes and Schwartz (28). For E > 0.1 ev, data are from Beyster et al. (3).

\textsuperscript{c}MR refers to calculations by McMurry, Russell and Brugger (36).

\textsuperscript{d}N refers to Nelkin's model.

\textsuperscript{e}N-KY and KY refer to calculations by Koppel and Young (31) who used the Nelkin model and a model that corrects for the vibrational anisotropy.

\textsuperscript{f}FG refers to the free gas model.
Table A.2. Comparison$^a$ of average scattering cosines ($\bar{\mu}$)

<table>
<thead>
<tr>
<th>E(eV)</th>
<th>Observed</th>
<th>MR$^a$</th>
<th>N</th>
<th>N-KY$^b$</th>
<th>KY$^b$</th>
<th>FG$^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.002</td>
<td>0.017</td>
<td>0.035</td>
<td>0.023</td>
<td>---</td>
<td>0.101</td>
<td></td>
</tr>
<tr>
<td>0.005</td>
<td>0.033</td>
<td>0.048</td>
<td>0.071</td>
<td>0.060</td>
<td>---</td>
<td>0.145</td>
</tr>
<tr>
<td>0.025</td>
<td>0.140</td>
<td>0.180</td>
<td>0.190</td>
<td>0.180</td>
<td>0.160</td>
<td>0.258</td>
</tr>
<tr>
<td>0.050</td>
<td>0.220</td>
<td>0.270</td>
<td>0.280</td>
<td>0.260</td>
<td>0.240</td>
<td>0.316</td>
</tr>
<tr>
<td>0.100</td>
<td>0.290</td>
<td>0.350</td>
<td>0.350</td>
<td>0.320</td>
<td>0.300</td>
<td>0.365</td>
</tr>
<tr>
<td>0.150</td>
<td>0.340</td>
<td>0.380</td>
<td>0.380</td>
<td>0.350</td>
<td>0.340</td>
<td>0.392</td>
</tr>
</tbody>
</table>

$^a$MR refers to McMurry, Russell, and Brugger (36) who compiled the data in this table.

$^b$N-KY and KY refer to calculations by Koppel and Young (31) who used the Nelkin model and a model that corrects for the vibrational anisotropy.

$^c$FG refers to the free gas model.

From Table A.1, it is seen that the $N$ and MR calculations agree well with the experimental data. The MR model uses higher effective masses, and this accounts for the $\bar{\mu}$ values being smaller than those of the $N$ model.

The absorption cross section used in calculating the decay constant was obtained by Gelbard and Davis (24) for the Radkowsky kernel. In the quoted paper

$$V_o \Sigma_{a0} = 4876 \text{ sec}^{-1}$$ (for the $P_3$ calculation).