

NEUTRON DIFFUSION IN
ABSORBING MEDIA

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ABSTRACT

Thermal neutron diffusion lengths have been measured in aqueous absorbing solutions contained in a large tank adjacent to the thermal column of the London University Research Reactor. In addition to pure water at 22.3°C and 65°C , boric acid has been used at both of these temperatures, and solutions of cadmium sulphate and gadolinium nitrate at the lower temperature only. In each case measurements were made over a wide range of concentrations. Representation of each set of results as a power series in absorber concentration has enabled the coefficients up to the spectral distortion term to be determined. A value has also been obtained for the microscopic absorption cross-section of hydrogen.

The diffusion lengths measured in the two highest cadmium concentration solutions were found to violate the theoretical limit given by Corngold.

The experimental results have been compared with the predictions of three scattering models for water, obtained by the use of a computer program DIS, which solves the transport equation by the B_L method. The latest Haywood model has been found to be no better than the established Nelkin model. Results for the inadequate free gas model are also given.

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NOTATION

a	See Σ_{ao}
B^2	Geometric buckling
$D(E)$	Diffusion coefficient
E	Neutron energy
E_o	Initial neutron energy
$\hbar$	Planck's constant ($=h/2\pi$)
L	Diffusion length
M_n	Neutron mass
M_2	Second energy transfer moment weighted by Maxwellian distribution
N	Number density of absorber atoms
$\underline{r}$	Position vector
T	Moderator temperature (in eV unless otherwise stated)
t	Time
v	Neutron velocity
v_o	Velocity of 2200 m/sec
v_T	Velocity of neutron with energy T
κ, κ_o	Fundamental spatial eigenvalue ($\equiv L^{-1}$)
κ_j	j th spatial eigenvalue
λ, λ_o	Fundamental temporal eigenvalue
λ_j	j th temporal eigenvalue
μ	Cosine of scattering angle
$\overline{\mu}$	Average cosine of scattering angle

σ_b	Bound atom microscopic scattering cross-section
$\sigma(E)$	Microscopic absorption cross-section
σ_{ao}	Microscopic absorption cross-section at $v = v_o$
σ_{aH}	Microscopic absorption cross-section at $v = v_o$ for hydrogen
σ_{oo}	Microscopic absorption cross-section, Maxwellian averaged
$\sigma_s(E)$	Microscopic scattering cross-section
$\sigma_n(E_o \rightarrow E)$	nth Legendre moment of differential microscopic scattering cross-section
$\sigma(E_o, E, \mu)$	Microscopic cross-section for neutrons of energy E_o scattered through angle μ to energy E
$\Sigma_T(E)$	Macroscopic total cross-section
$\Sigma_a(E)$	Macroscopic absorption cross-section
Σ_{ao}, a	Macroscopic absorption cross-section at $v = v_o$
Σ_B	Macroscopic absorption cross-section at $v = v_o$ for boron
Σ_{oo}	Macroscopic absorption cross-section, Maxwellian averaged
$\Sigma_s(E)$	Macroscopic scattering cross-section

$\Sigma_s(E' \rightarrow E)$	Macroscopic differential scattering cross-section
$\Sigma_s(E' \rightarrow E, \mu_0)$	Macroscopic cross-section for neutrons of energy E' scattered through angle μ_0 to energy E
$\varphi(E)$	Neutron flux
$\varphi_n(E)$	n th Legendre moment of neutron flux

Other symbols are defined in the text.

CHAPTER 1

INTRODUCTION

A variety of present-day thermal reactors use water either as moderator, or coolant, or both. It is therefore of great importance that sufficient information on the interaction between neutrons and water molecules should be available to adequately predict the performance of such reactors. The information needed is the probability that a neutron on collision with an atom should exchange specified amounts of energy and momentum with the atom. The scattering medium to be considered in this work is light water.

A simple model of the scattering process is obtained by assuming that the atoms in the medium are free from any effects of chemical binding, and have a Maxwell-Boltzmann energy distribution. This, the so-called free gas approximation, has been found to be inadequate both in the determination of neutron spectra and in the calculation of integral quantities. It will, however, be used throughout this work in order to give an indication of the importance of chemical binding effects in water, and also to help keep in perspective the apparent inaccuracies of the more sophisticated models.

The water molecule is known to have internal vibrational modes at 0.205, 0.474 and 0.488 eV. In addition,

there is a broad band of hindered rotations at around 0.06 eV. A physical model of the molecule was therefore suggested by Nelkin (1960a) in which harmonic oscillations of 0.06, 0.205 and 0.481 eV are combined with free translation of the entire molecule. With the aid of several other assumptions to be discussed in Chapter 2, the differential scattering cross-sections may then be computed. While this model represents a considerable advance on the free gas model, in which chemical binding effects are neglected altogether, it is not to be expected that certain of the assumptions, notably the condensation of the broad hindered rotational frequency band to a single frequency of 0.06 eV, and the assumption of unhindered translations, will give completely satisfactory results.

In the absence of a complete theoretical treatment of the dynamics of a molecule in the liquid state, an entirely different approach has been developed by Egelstaff, in which the experimental data on differential neutron scattering are used to construct a function analogous to the phonon frequency distribution in a solid, which may then be used to furnish differential scattering cross-sections in regions both inside and outside the range of the original experiments. This method has been applied by Haywood (1966) to water, but although total scattering

cross-sections have been computed, it is not yet known how well the model is able to predict integral quantities. One of the aims of the present work is to compute such predictions and compare them with experimental results.

The most sensitive test of a scattering model is its ability to correctly predict differential scattering cross-sections with respect to both energy and scattering angle. Such stringent criteria are not, however, necessary from the point of view of reactor calculations, since the quantities of interest here are normally spectrum-averaged reaction rates. The practical test of a model is then its ability to correctly describe transport phenomena under conditions where the neutron spectrum may be severely distorted, and it is on this basis that the experiments to be described in the present work were designed.

Experimental parameters relating to thermal neutron diffusion may be obtained by one of two methods, the formal relationship between which will be considered later in this work. The first of these methods consists in injecting a pulse of neutrons into a block of moderator, and measuring the asymptotic decay constant of the neutron flux. From a series of such measurements using blocks of varying sizes (characterized by different geometric bucklings) the diffusion parameters may be derived. For small blocks the spectrum is distorted since the removal process - the leakage of neutrons from the block - is not

proportional to the neutron density. The diffusion coefficient rises with neutron energy, and the spectrum inside the block is consequently 'cooled' (Von Dardel and Sjostrand, 1954). For blocks which are sufficiently small for this effect to be clearly observed, however, extrapolation distance uncertainties complicate the determination of the corresponding bucklings. Williams (1962) has shown that the extrapolation distance itself is a function of buckling, although his calculations were not for realistic scattering models. This problem may be circumvented by a method suggested by Friedman (1962), in which a large moderating system with minimal buckling uncertainties is used, and the decay constant measured as a function of the concentration of added non- $^{1/v}$ absorber. This method has the additional advantage that the spectrum distorting mechanism is separated from the moderator itself, and different absorbers may be used to derive further scattering parameters. Later experimental work by Friedman (1964) using this method, however, gave a much lower value for M_2 than expected.

The second method of obtaining diffusion parameters is to measure the spatial relaxation length of thermal neutrons in a moderating assembly fed by a constant source of neutrons. Apart from the familiar diffusion length for the pure moderator, parameters analogous to those given by the first method may be obtained by poisoning

the moderator with varying concentrations of $1/v$ absorber. In a pulsed field addition of such absorber would not distort the spectrum, but in the stationary case, the effect of the flux gradient is that the mean energy of neutrons diffusing into a volume element is higher than the mean energy of neutrons lost by absorption - by analogy this is referred to as diffusion heating. In general, as will be seen later, this second method permits the observation of spectrum distortion effects over a greater range than does the simple pulsed experiment.

The primary aim of the present work is to obtain accurate experimental information on the variation of the spatial decay constant with concentration of aqueous solutions of highly-absorbing compounds, including measurements under conditions of severe spectral distortion. To this end, as wide a range as practicable of concentrations of each absorber has been used. Furthermore, the absorbers were chosen so as to give widely differing spectral effects on account of the differences between their absorption cross-sections. The absorbers selected were boron, which is a pure $1/v$ absorber, in boric acid, and two non- $1/v$ absorbers: cadmium in cadmium sulphate, and gadolinium in gadolinium nitrate. These two elements have Westcott "g" values of 1.33 and 0.89 respectively, in contrast to a $1/v$ absorber for which $g = 1.00$.

Stationary experiments on light water have previously been reported by Starr and Koppel (1962) and Baumann and Pellarin (1966) using $1/v$ poison at room temperature. Both of these groups used activation techniques, and measured diffusion lengths over a wide range of concentrations. The results of Starr and Koppel are particularly accurate, of the order of .2% in L. No errors were quoted by the second group, but the results of both groups are in good agreement. No measurements have been made over a considerable concentration range at higher temperatures. Miller (1961) has performed experiments on six very high concentration solutions of cadmium sulphate - his results are to within 3%. Similar errors were obtained by Russell et al (1962) who measured two solutions, each of cadmium sulphate and gadolinium nitrate. Of the four series of experiments planned for this work - boric acid, cadmium sulphate and gadolinium nitrate at room temperature, and boric acid at 65°C, only the first has been studied previously in any detail; it is included here in order to establish the method.

The extraction of diffusion parameters from the results of both pulsed and stationary experiments involves fitting polynomials of varying degrees to the experimental points. This procedure has been criticized by Koppel and Lopez (1962) on the grounds that the higher order coefficients have large uncertainties. They recommend the comparison

of data point by point. In the present work both of these methods will be used in comparing the experimental results with the predictions of the three scattering models described above.

In order to assess the capabilities of each model it is essential that no inaccuracies, such as would be caused by the use of diffusion theory, are introduced in the derivation of the spatial eigenvalue κ from the scattering law. The computer program DIS, written by the present author, which is used to perform this operation has therefore been based on rigorous transport theory, and is able to take into account anisotropic scattering up to the third Legendre moment.

The Boltzmann equation for both the time and space-dependent cases gives rise to a series of eigenvalues λ_j and κ_j respectively, of which the fundamentals λ_0 and κ_0 govern the asymptotic measurements in the two experiments. Corngold et al (1963, 1964a) have shown that for the pulsed case a number of discrete eigenvalues exists up to a limit given by $(v\Sigma_T)_{\min}$, beyond which there is a continuum. Similar behaviour is exhibited by the κ_j ; the limit here is $(\Sigma_T)_{\min}$. Pulsed experiments have however been performed in both beryllium and graphite in which values of λ_0 well in excess of the predicted limit have been observed. Corngold and Michael (1964b) have suggested

that this arises from insufficient waiting times, so that the fundamental mode is not fully established. Values of κ_0 greater than the appropriate limit have been measured by Baumann and Pellarin (1966) for borated heavy water, and by Miller (1961) in light water solutions of cadmium sulphate. In the latter case, Corngold and Michael again consider the establishment of an asymptotic mode doubtful.

A subsidiary aim of the present experiments is to attempt to measure values of κ_0 greater than the limit for light water.

CHAPTER 2

NEUTRON SCATTERING THEORY

In this chapter a review will be given of the three neutron scattering models which are to be considered in the present work. These are the free gas, Nelkin and Haywood models.

The free gas and Nelkin models are almost entirely theoretical models of the neutron scattering process, drawing only one and two parameters, respectively, directly from neutron scattering experiments. The Haywood model, however, represents an attempt to fit a large volume of neutron scattering data to a theory which may then be used to predict the differential scattering cross-sections for situations both inside and outside the range of the experiments.

The present chapter contains the theory on which is based the first half of the FORTRAN IV program DIS (Johnson, unpublished), which establishes the scattering matrices $\sigma_n(E_o \rightarrow E)$ for whichever scattering model is required. The second half of the program, the determination of the spatial eigenvalue, will be dealt with in Chapter 3.

Some of the background material in this chapter has been taken from the recent book edited by Egelstaff (1965). Much of the coding for the Nelkin and free gas models (Sections 2.3 and 2.4) and also for the angular integration

(Section 2.6) has been adapted from the General Atomics program KERNEL, written by Federighi and Goldman (1962).

2.1 General

The interaction of fast neutrons with the atoms of a moderator may be described simply in terms of the laws of elastic collisions. For thermal neutrons (i.e. neutrons with energies below about 1 eV), however, the situation is complicated by two factors: first, the chemical binding between the moderator atoms, and second, the thermal motion of these atoms. The latter may be taken into account by treating the moderator as a gas of monatomic atoms - this is the so-called "free gas" model. As will be demonstrated later, however, this does not give satisfactory results, and it is apparent that allowance must be made for chemical binding. The theoretical prediction of scattering cross-sections therefore requires a full knowledge of the dynamics of atomic motions, although even with this information it is necessary to make several approximations in order to obtain manageable expressions.

2.2 Van Hove Correlation Functions

The link between the dynamics of the scattering system and the scattered neutron intensity is provided by the time-dependent correlation functions introduced by Van

Hove (1954). In the case of X-ray diffraction, the intensity of scattered radiation is given by the spatial Fourier transform of a function called the static pair distribution function, denoted by $g(\underline{r})$, which describes the average distribution of neighbouring atoms at any instant around a particular scatterer. For an observable scattering event, the energy transferred between the X-ray photon and the sample will be large compared with the binding energy of the atoms in the sample; this implies a short interaction time while the scattering event is actually taking place. The result of this is that the X-rays "see" an instantaneous picture of the scattering assembly. Thermal neutrons, however, may transfer energies of the same order as their incident energies; this corresponds to much larger interaction times, during which the motions of the scattering atoms are relevant. Van Hove therefore generalised the static pair distribution function to the following correlation functions, defined here for simplicity for a system in which all atoms are equivalent, as

$$G(\underline{r}, t) = \sum_{\underline{n}} \overline{\delta(\underline{r} + \underline{R}_0(0) - \underline{R}_{\underline{n}}(t))} \quad 2.2.1a$$

$$\text{and} \quad G_s(\underline{r}, t) = \overline{\delta(\underline{r} + \underline{R}_0(0) - \underline{R}_0(t))} \quad 2.2.1b$$

where $\underline{R}_{\underline{n}}(t)$ and $\underline{R}_0(t)$ are the position vectors of the n th atom and the zeroth (arbitrary) atom at time t , respectively,

the bar denotes a statistical average at temperature T , and the summation is over all the atoms n .

These functions may be classically defined as follows: Given a particle at the origin at time zero, $G_s(\underline{r}, t)$ gives the probability of finding that particle at position $\underline{r}$ after time t has elapsed, and $G(\underline{r}, t)$ the probability of finding any particle at position $\underline{r}$ and time t . From the definitions it follows that

$$G_s(\underline{r}, 0) = \delta(\underline{r}) \quad 2.2.2a$$

$$\text{and } G(\underline{r}, 0) = \delta(\underline{r}) + g(\underline{r}) \quad 2.2.2b$$

and also, for large times

$$G_s(\underline{r}, \infty) = 0 \quad 2.2.3a$$

$$G(\underline{r}, \infty) = \rho \quad 2.2.3b$$

where ρ is the macroscopic density of the system.

The intensity of scattered neutrons is now found by taking the four-dimensional space-time Fourier transform of one or other of these functions - $G(\underline{r}, t)$ gives rise to the coherent and $G_s(\underline{r}, t)$ to the incoherent cross-section. In the case of water the scattering is almost entirely incoherent since the coherent cross-section is given by the sum of the weighted amplitudes rather than intensities (e.g. Evans, 1955), and the singlet scattering length being negative it happens that singlet and triplet scattering almost completely cancel. We will therefore consider only $G_s(\underline{r}, t)$.

We obtain

$$\sigma(E_0, E, \mu) = \frac{\sigma_b}{8\pi^2} \sqrt{\frac{E}{E_0}} \iint e^{i(\underline{\kappa} \cdot \underline{r} - \omega t)} G_s(\underline{r}, t) d\underline{r} dt \quad 2.2.4$$

where the units employed are such that $\hbar = k$ (Boltzmann's constant) = 1, σ_b is the bound atom cross-section and $\underline{\kappa}$ and ω are respectively the momentum and energy transferred to the scatterer, so that $\frac{\kappa^2}{2} = E + E_0 - 2\mu\sqrt{EE_0}$. If we now take only the spatial transform of G_s , we obtain a function called the intermediate scattering function, $\chi(\underline{\kappa}, t)$, which is the time transform of $\sigma(E_0, E, \mu)$;

$$\sigma(E_0, E, \mu) = \frac{\sigma_b}{8\pi^2} \sqrt{\frac{E}{E_0}} \int e^{-i(E-E_0)t} \chi(\underline{\kappa}, t) dt. \quad 2.2.5$$

This function was first introduced by Zemach and Glauber (1956), who showed that expressions for χ may be obtained quantum-mechanically by representing the scattering of neutrons by protons by the Fermi (1936) pseudo-potential approximation in which the strong short-range proton-neutron interaction is approximated by a delta-function pseudo-potential. This will be satisfactory providing the resulting cross-sections are normalized to give the correct free-atom scattering cross-section. This potential may then be treated in the first order Born approximation, to give for the incoherent scattering

$$\chi(\underline{\kappa}, t) = \sum_a p_a(T) \overline{\langle \phi_a | \exp[-i \underline{\kappa} \cdot \underline{R}_0(0)] \exp[i \underline{\kappa} \cdot \underline{R}_0(t)] | \phi_a \rangle} \quad 2.2.6$$

where ϕ_a is the wave function for the initial state of the molecule, and $p_a(T)$ is a Boltzmann weighting factor for the population of state a at temperature T .

2.3 Nelkin Model

We now consider the model proposed by Nelkin (1960a), to describe the dynamics of the water molecule. For harmonically oscillating systems χ may be evaluated exactly from equation 2.2.6 without difficulty. Nelkin therefore proposed that the dynamical motions of protons in water should be described by four harmonic oscillations, together with a translational movement of the molecule as a whole, and further, that there should be no interaction between any of these modes. The intermediate scattering function may then be written as the product of the χ 's corresponding to each mode.

The vibrational energy levels in water have been determined by Herzberg (1950) to be .205, .474, and .488 eV. It is assumed that these levels are not significantly affected by the hindrance of neighbouring molecules in the liquid state. This assumption cannot, however, apply to rotation since this involves movement of the whole H_2O molecule. The energy appropriate to such motions covers a broad band with its peak at .06 eV; Nelkin represents this by a harmonic torsional oscillation mode of small amplitude at precisely this energy. Apart from the

asymptotic free atom scattering cross-section for protons, this figure is the only parameter in the present model to be taken directly from neutron scattering data. The translational movement of the molecule is assumed to be unhindered by neighbouring molecules.

A further approximation is also necessary: equation 2.2.6 involves an average over molecular orientations with respect to the incident neutron direction. It was assumed for convenience by Krieger and Nelkin (1957) that a reasonable approximation to the average of the product of the χ 's is given by the product of the averages. This assumption has been shown by Randolph (1961) to be satisfactory for the case of the angular distribution of neutrons scattered from methane.

In terms of these approximations we now write, using a self-evident notation:

$$\chi = \chi_T \chi_r \chi_{v_1} \chi_{v_2} \chi_{v_3} \quad 2.3.1$$

For free translations of the entire H_2O molecule

$$\chi_T(u, t) = \exp\left\{-\frac{u^2}{2M} (it + Tt^2)\right\} \quad 2.3.2$$

where $M = 18$ is the mass of the molecule (all masses are in terms of the mass of the neutron).

Now for an isotropic harmonic oscillator with angular frequency ω

$$\chi(\kappa, t) = \exp \left\{ \frac{\kappa^2}{m} \frac{1}{2\omega} [(n+1)(e^{-i\omega t} - 1) + n(e^{i\omega t} - 1)] \right\} \quad 2.3.3$$

$$\text{where } n = [e^{\omega/T} - 1]^{-1} \quad 2.3.4$$

is the average occupation number of the oscillator. The remaining χ 's are each to be treated in this fashion. Thus equations 2.3.3 and 2.3.4, with $m = m_r$ and $\omega = \omega_r$ ($= .06$ eV) gives the expression for χ_r . Krieger and Nelkin (1957) have considered the choice of an effective mass m_r for rotation. It is obvious that in a single event the effective rotational mass of a proton as seen by a neutron depends strongly on the direction of approach relative to the molecular orientation. This has given rise to the Sachs-Teller (1941) mass-tensor concept. Krieger and Nelkin have shown that $m_r = 2.32$ gives the best fit to the full mass-tensor approximation in the case of water vapour (free rotator). This value is therefore adopted in the present case for torsional oscillations. This assumption depends on the bond angle H-O-H being the same in both liquid and vapour states.

For all the vibrational levels $\omega_i \gg T$ and we may take $n_i = 0$ (i.e. the vibrations are not excited at ordinary temperatures). Further, the two highest levels are so close together that they may be assumed to be degenerate. We have therefore for the vibrational intermediate scattering function

$$\chi_{v1}\chi_{v2}\chi_{v3} = \exp \left\{ \frac{\kappa^2}{2m_v} \left[\frac{1}{3\omega_1} (e^{-i\omega_1 t} - 1) + \frac{2}{3\omega_2} (e^{-i\omega_2 t} - 1) \right] \right\}$$

2.3.5

where $\omega_1 = .205$ eV and $\omega_2 = .481$ eV. The appropriate mass m_v is given by the consideration that for high energy transfers (i.e. short times) the scattering cross-section should be that for free protons, i.e.

$$\lim_{t \rightarrow 0} \chi(\kappa, t) = \exp \left\{ -\frac{\kappa^2}{2} it \right\} \quad 2.3.6$$

Expanding the exponents of equations 2.3.2, 2.3.3, and 2.3.5 in powers of t and comparing with equation 2.3.6 we find

$$1 = \frac{1}{M} + \frac{1}{m_r} + \frac{1}{m_v} \quad 2.3.7$$

from which $m_v = 1.95$.

By inserting equations 2.3.2, 2.3.3 (with $m = m_r$ and $\omega = \omega_r$) and 2.3.5 into 2.3.1 we may now obtain an expression for $\chi(\kappa, t)$. However, the time transform 2.2.5 which leads to the scattering cross-section cannot be carried out explicitly, and so it is necessary to further approximate by treating only one mode exactly for a particular energy transfer, the others being considered in either the high or low energy transfer limit (corresponding to short and long collision times respectively). Thus for small t equation 2.3.3 reduces to

$$\chi_r = \exp \left\{ -\frac{\kappa^2}{2m_r} (it + \overline{E}_r t^2) \right\} \quad 2.3.8$$

$$\text{where } \bar{E}_r = \frac{\frac{1}{2} \omega_r}{e^{\omega_r/T} - 1} + \frac{1}{2} \quad 2.3.9.$$

is the average energy of rotations at temperature T.

Also, for energy transfers less than the vibrational frequencies, only elastic interactions with the vibrational modes will occur; equation 2.3.5 then reduces to a Debye-Waller factor

$$\chi_{v1} \chi_{v2} \chi_{v3} = \exp \left\{ - \frac{\kappa^2}{2m_v} \left(\frac{1}{3\omega_1} + \frac{2}{3\omega_2} \right) \right\} \quad 2.3.10$$

We now define three regions of energy transfer:

$\Delta E < 0.205$ eV, $0.205 \leq \Delta E < 0.481$ eV, and $\Delta E \geq 0.481$ eV.

(Nelkin originally defined regions in terms of the incident neutron energy; it was pointed out by Federighi and Goldman (1963) that the energy transfer is a more logical parameter.)

In the first of these regions we assume free translations and only elastic vibrational interactions, and treat the rotations properly. The resulting χ is then given by the product of equations 2.3.2, 2.3.3 and 2.3.10. The time transform of this χ may be performed analytically, and the resulting expression for the cross-section is

$$\begin{aligned} \sigma(E_o, E, \mu) = & \frac{\sigma_b}{4\pi} \left(\frac{E}{E_o} \right)^{\frac{1}{2}} \left(\frac{m}{2\pi E \kappa^2} \right)^{\frac{1}{2}} \exp \left(- \frac{\kappa^2}{2A} \right) \times \\ & \times \sum_{n=-\infty}^{\infty} \exp \left(- \frac{n\omega}{2T} \right) I_n \left(\frac{\kappa^2}{2B} \right) \exp \left[- \frac{m}{2E \kappa^2} \left(E - E_o - n\omega + \frac{\kappa^2}{2m} \right)^2 \right] \end{aligned}$$

$$\begin{aligned}
 \text{where } \left(\begin{aligned}
 m &= M = 18 \\
 \overline{E} &= T \\
 A^{-1} &= \alpha_v^{-1} + [m_r \omega_r \tanh(\omega_r/2T)]^{-1} \\
 \alpha_v^{-1} &= \frac{1}{m_v} \left(\frac{1}{3\omega_1} + \frac{2}{3\omega_2} \right) \\
 B &= m_r \omega_r \sinh(\omega_r/2T) \\
 \omega &= \omega_r
 \end{aligned} \right. \quad 2.3.12
 \end{aligned}$$

For energy transfer between .205 and .481 eV, rotations are treated in the high energy form. The .205 eV vibration is treated exactly, and the .481 eV level in the elastic limit as in the first region. Again an expression in the form of equation 2.3.11 is obtained; the parameters are now given by

$$\begin{aligned}
 \left(\begin{aligned}
 1/m &= 1/M + 1/m_r \\
 \overline{E} &= \overline{E}_1 = \frac{T m_r + \overline{E}_r M}{m_r + M} \\
 A &= \alpha_v \\
 B &= 3m_v \omega_1 \sinh(\omega_1/2T) \\
 \omega &= \omega_1 (= .205 \text{ eV}).
 \end{aligned} \right. \quad 2.3.13
 \end{aligned}$$

Finally for energy transfers greater than .481 eV, χ_{v1} is also treated in the high energy limit; only the .481 eV vibrations are treated exactly. Once more the final expression is in the form of 2.3.11; the parameters

are

$$\begin{aligned}
 \left(\begin{aligned} 1/m &= 1/M + 1/m_r + 1/3m_v \\ \bar{E} &= m\left(\frac{T}{M} + \frac{\bar{E}_r}{m_r} + \frac{\bar{E}_1}{3m_v}\right) \\ A &= \frac{3}{2} m_v \omega_2 \\ B &= \frac{2}{3} m_v \omega_2 \sinh\left(\frac{\omega^2}{2T}\right) \\ \omega &= \omega_2 (= .481 \text{ eV}) \end{aligned} \right. \quad 2.3.14
 \end{aligned}$$

The model is now in a form suitable for computation. The scattering cross-section part of the program DIS is under the control of the subroutine SCATT (see flow diagram Figure 3). The parameters for all three regions are first computed (equations 2.3.12, 2.3.13 and 2.3.14) in the subroutine JPAM, and subroutine NELKIN is then entered. This causes equation 2.3.11 to be evaluated (in function ESS) at the energy points specified by the user, at several angles μ , with the appropriate choice of parameters. The final step is the integration over angle to give the Legendre moments of the scattering cross-section. This is described in Section 2.6.

2.4 Free Gas Model

All chemical binding effects are ignored in this model; the protons are considered to interact with each other only by elastic collisions, so that they are in a state of thermodynamic equilibrium. The intermediate

scattering function for this situation is then given by equation 2.3.2, and taking the time transform of this we find

$$\sigma(E_0, E, \mu) = \frac{\sigma_b}{4\pi} \left(\frac{E}{E_0}\right)^{\frac{1}{2}} \left(\frac{m}{2\pi T \kappa^2}\right)^{\frac{1}{2}} \exp \left\{ -\frac{m}{2T \kappa^2} \left(E - E_0 + \frac{\kappa^2}{2m}\right)^2 \right\}$$

2.4.1

When the free gas option has been selected, this expression rather than that given by equation 2.3.11 is evaluated in the function ESS.

2.5 Haywood Model

Neutron scattering cross-sections have been measured by Haywood and Thorson (1962, 1964) using the phased-rotor time of flight selector at Chalk River (Egelstaff et al, 1961b), in particular for water at room temperature. The data are obtained as a function of the three parameters E_0 , E and μ . It is possible to condense this large amount of information into a function $S(\alpha, \beta)$, which Egelstaff (1961a) first introduced as the scattering law, and which is a function of only two parameters, the momentum and energy transfers. This may be done because of the detailed balance condition:

$$M(E) \sigma(E \rightarrow E', \mu) = M(E') \sigma(E' \rightarrow E, \mu) \quad 2.5.1$$

where $M(E) = E \exp(-E/T)$ is the Maxwellian.

S is defined by

$$\sigma(E_o, E, \mu) = \frac{1}{T} \frac{\sigma_b}{4\pi} \sqrt{\frac{E}{E_o}} e^{-\beta/2} S(\alpha, \beta) \quad 2.5.2$$

$$\text{where} \quad \alpha = \frac{E_o + E - 2\mu \sqrt{EE_o}}{AT} \quad 2.5.3$$

$$\text{and} \quad \beta = \frac{E - E_o}{T}.$$

(A is the mass of the principal scatterer in terms of the neutron mass.) It is only possible to measure $S(\alpha, \beta)$ over limited ranges of α and β (e.g. the energy transfers accessible are only those up to about $6T$) and therefore some method of both extrapolation and interpolation is necessary.

$S(\alpha, \beta)$ is related to the correlation function $G_s(\underline{r}, t)$ by a double Fourier space-time transform. If this function could be evaluated by transforming the data, the scattering law could be determined for any required α and β . In practice, however, this is not a useful technique owing to the incomplete range of the experimental data.

Egelstaff (1961a, 1963) has shown that if the experimental results are plotted as $\log (S/\alpha)$ against α , and extrapolated to $\alpha=0$, a generalised frequency distribution

$$p(\beta) = \lim_{\alpha \rightarrow 0} \left(\frac{S}{\alpha} \right) \beta^2 \quad 2.5.4$$

may be obtained. This equation relates only to the "self" or incoherent part of the scattering law - for low α interference effects may become important, although as was seen

in Section 2.2, in the case of water these effects should be small. Anharmonic effects may also be troublesome in this region. For these reasons any sharp change in gradient for small α is disregarded in performing the extrapolation.

The properties of $p(\beta)$ are conveniently considered in terms of a normalised frequency function

$$\rho(\beta) = \frac{2}{\beta} \sinh\left(\frac{\beta}{2}\right) p(\beta) \quad 2.5.5$$

which has unit area, and is analogous to the phonon frequency distribution in a solid.

Egelstaff and Schofield (1962) have shown how $S(\alpha, \beta)$ may now be reconstructed from the measured $p(\beta)$, providing it is assumed that anharmonic effects may be neglected. This corresponds to the assumption that the self-correlation function $G_s(\underline{r}, t)$ is a Gaussian function of position, with a time-dependent width $w(t)$; this approximation was first made by Vineyard (1958). Under this assumption

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp\{-\alpha w(t) + i\beta t\} dt \quad 2.5.6$$

where

$$w(t) = \int_0^{\infty} \frac{p(\beta)}{\beta^2} \left\{ \cosh \frac{\beta}{2} - \cos \beta t \right\} dt \quad 2.5.7$$

It can be shown that in the case of a fluid, where $w(t) \approx |t|$ at large times, $\tilde{w}(t)$ is the time transform of $p(\beta)$.

For small α , the width function is divided into two parts

$$w(t) = w_d(t) + w_b(t) \quad 2.5.8$$

where $w_d(t)$ represents diffusive motion of the molecule as a whole, and $w_b(t)$ the bounded motion of an atom relative to the molecule - this is finite at large times.

The time transforms of the second time derivatives of these functions yield corresponding functions $p_d(\beta)$ and $p_b(\beta)$. If the corresponding scattering laws are written as $S_d(\alpha, \beta)$ and $S_b(\alpha, \beta)$, we find

$$S(\alpha, \beta) = \int_{-\infty}^{\infty} S_d(\alpha, \beta') S_b(\alpha, \beta - \beta') d\beta' \quad 2.5.9$$

$S_b(\alpha, \beta)$ is then evaluated by means of a multi-phonon expansion involving a Debye-Waller factor $\exp(-\alpha\lambda)$, where the coefficient λ is given by

$$\lambda = 2 \int_0^{\infty} \frac{p_b(\beta)}{\beta^2} \cosh \frac{\beta}{2} d\beta. \quad 2.5.10$$

$S_d(\alpha, \beta)$ is computed by choosing a relatively simple form for the function $w_d(t)$ which satisfies the constraints, and $S(\alpha, \beta)$ evaluated by the convolution equation 2.5.9.

For large α the phonon expansion is unsatisfactory - too many terms are required - and a steepest descents method is used. This gives rise to an expansion in inverse powers of α , in contrast to the phonon expansion which is in ascending powers of α .

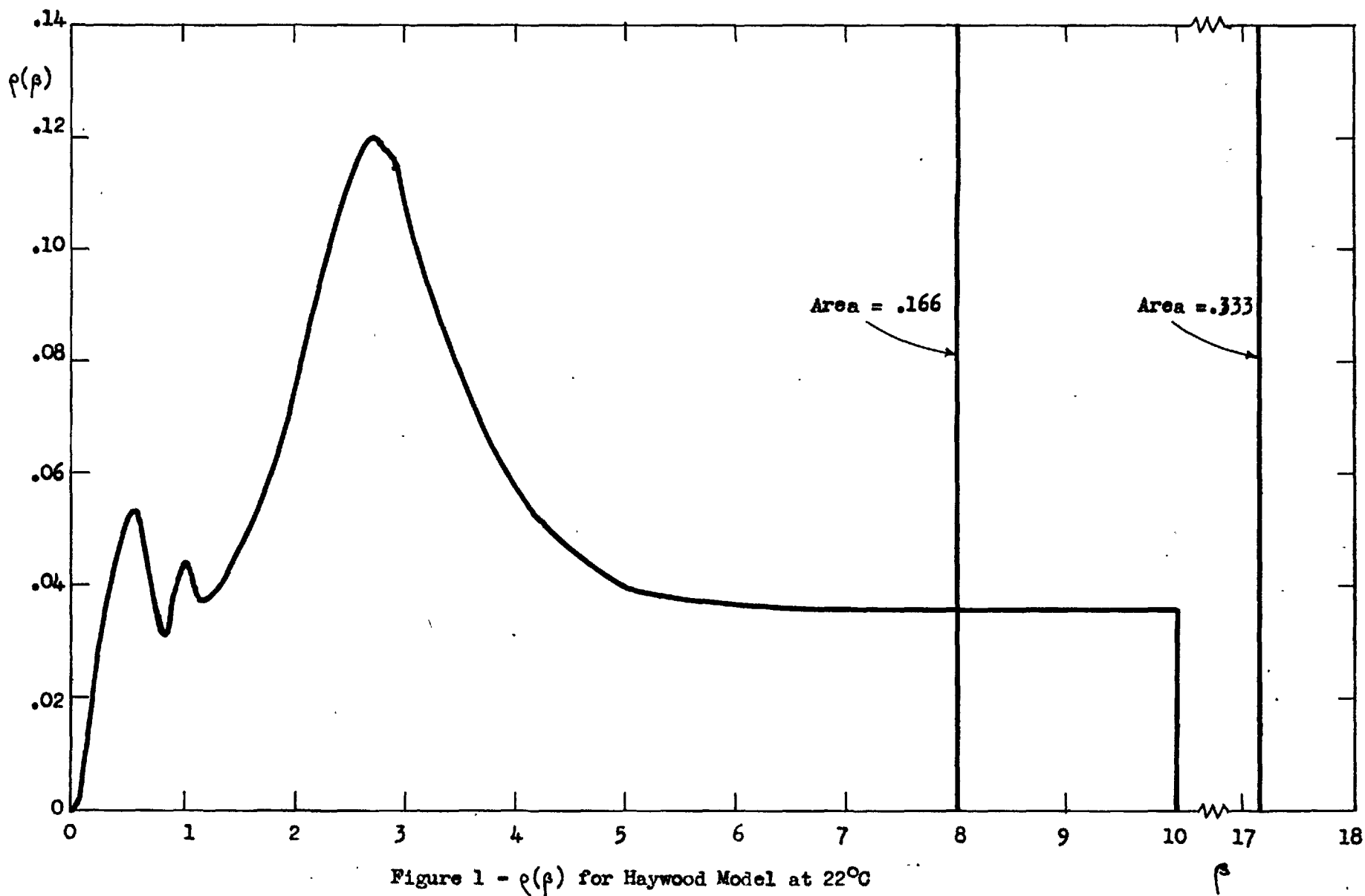
A computer program LEAP has been written by McLatchie

(unpublished; but see Macdougall, 1962) to evaluate $S(\alpha, \beta)$ from a given $p(\beta)$ by this method. An iterative procedure is adopted as follows: an initial $p(\beta)$ is obtained by extrapolating the experimental data as described above, and fed into LEAP. The $S(\alpha, \beta)$ output is then compared with the experimental values, and from the discrepancies corrections to $p(\beta)$ are deduced. The new $p(\beta)$ is again input to LEAP, and the procedure repeated until satisfactory agreement with experiment is obtained.

The output of LEAP also contains a Debye-Waller coefficient given by 2.5.10, which gives a δ -function contribution to $S(\alpha, \beta)$ of the form $e^{-\lambda\alpha} \delta(\beta)$.

The vibrational modes of the water molecule considered in Section 1.3 are taken as δ -functions in $\rho(\beta)$, each with an effective area A (so that the contribution to $\rho(\beta)$ for the vibration at β_i is $A_i \delta(\beta - \beta_i)$). A computer program ADDELTA, also written by McLatchie, and run together with LEAP, convolves up to 50 δ -functions with the experimental $p(\beta)$ to give the complete $S(\alpha, \beta)$. From equations 2.5.5 and 2.5.10 we see that the Debye-Waller coefficient output from LEAP has to be increased by $A_i \frac{\coth \frac{\beta_i}{2}}{\beta_i}$ for each of the δ -functions.

Haywood (1966) has derived a $\rho(\beta)$ for water at 22°C by the above method. This is shown in Figure 1; it is the basis of the Haywood model used in the present work.



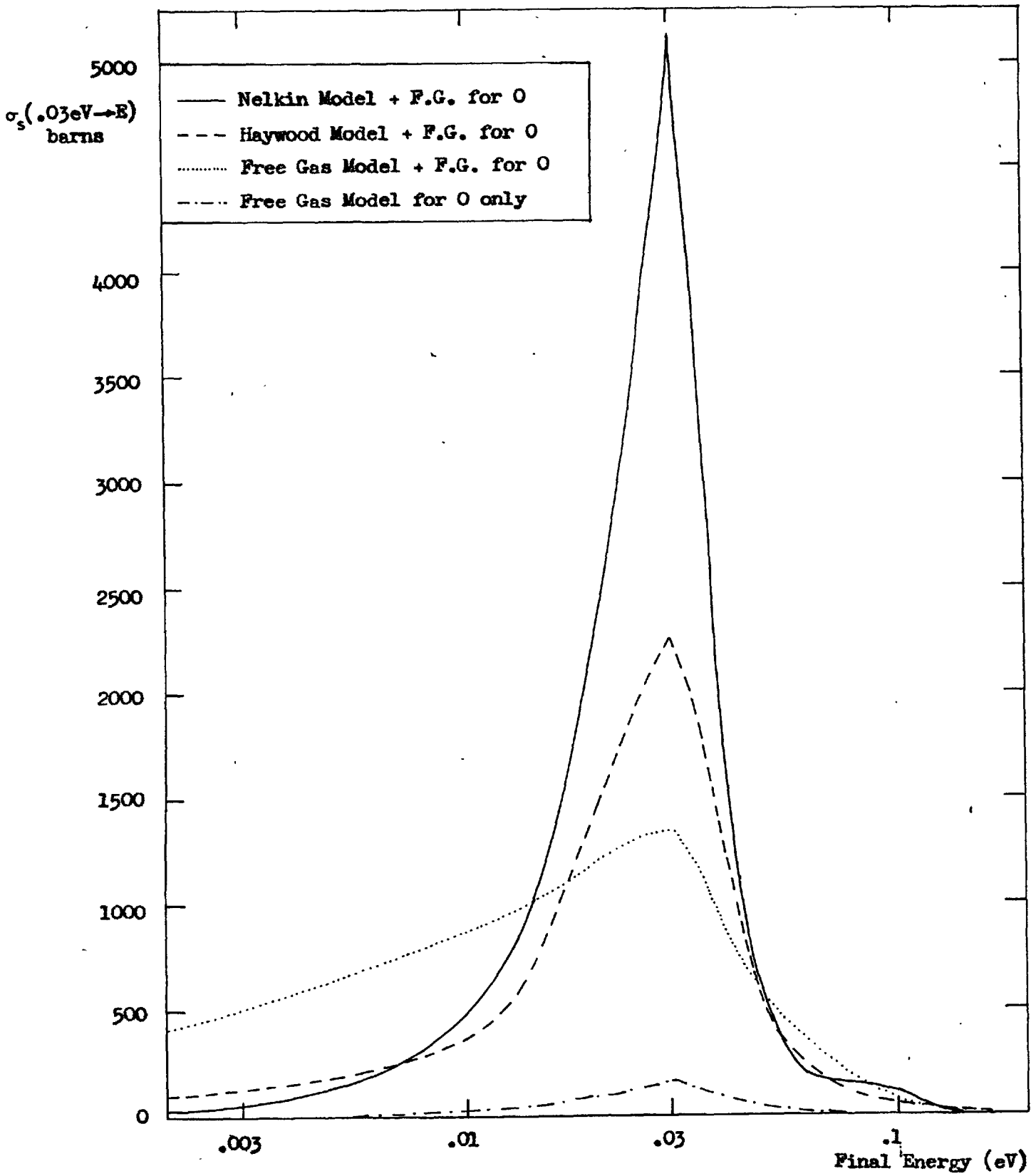


Figure 2 - Inelastic Scattering Cross-section at .03 eV for Water Models

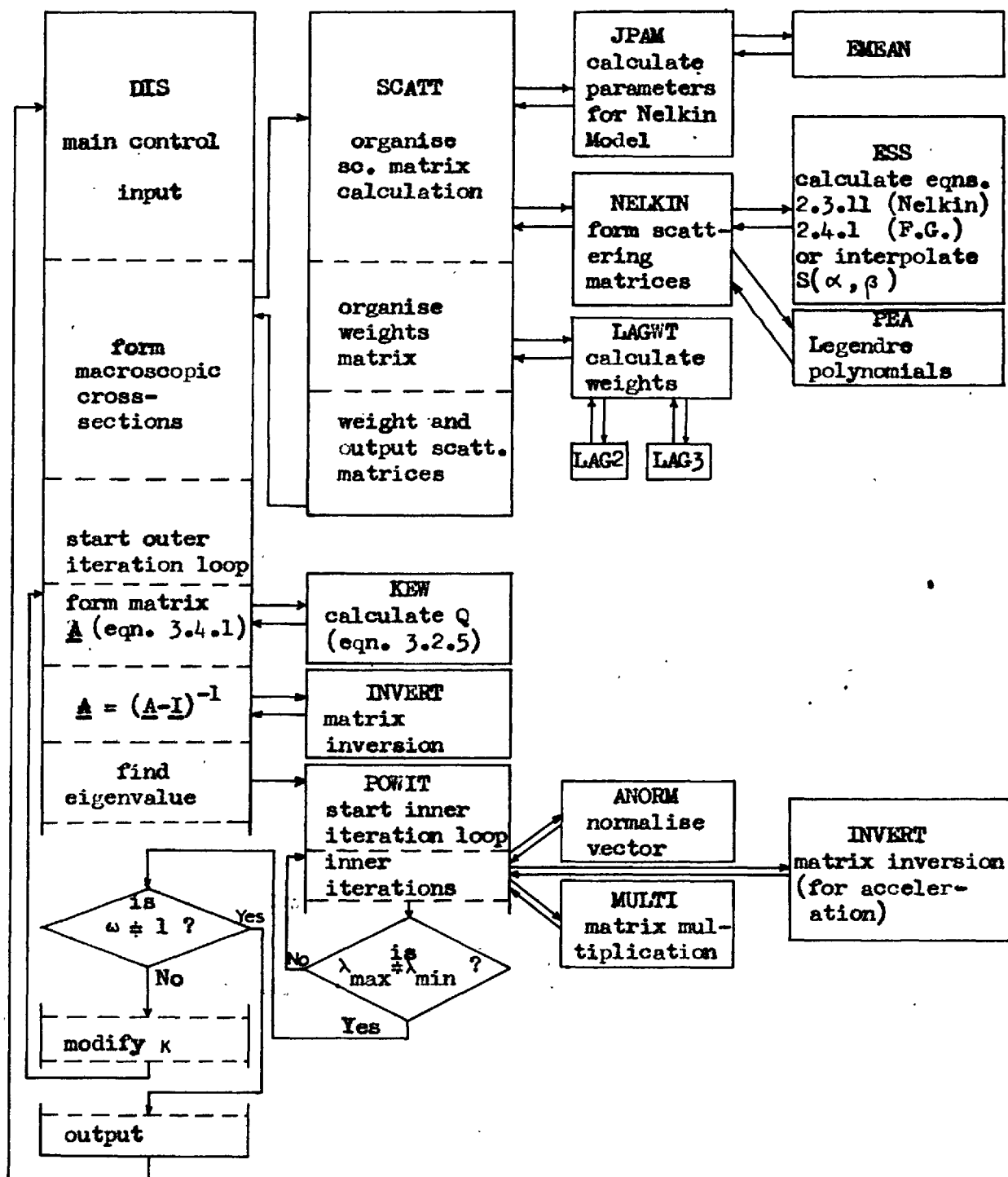


Figure 3 - Flow Diagram for DIS

The measured values only extend up to about $\beta = 6$; from there he has extrapolated smoothly up to $\beta = 10$, so that the total area under the curve is .5. This is because the relative areas of the vibrational and rotational parts of the spectrum are optimized during the iterative process described above, and in the present model this ratio has been taken as unity. .5 is therefore also allocated to the vibrations: as the upper vibration consists of two modes (taken to be degenerate) it is allocated area .333, and the lower vibration .167.

It will be seen that there are two small peaks at $\beta \approx 0.5$ and 1.0 . These are almost certainly due to translational movements of the molecule as a whole, while the broader peak at $\beta \approx 2.7$ is due to hindered rotations of the molecule. These hypotheses were checked by Larsson and Dahlborg (1962), who determined $\rho(\beta)$ for light and heavy water. They found that the broader peak in D_2O was indeed shifted by a factor of $\sqrt{2}$, as it should be if the hydrogen atom is the dynamic unit, whereas the smaller peaks at low β were unmoved, the theoretical shift of $\sqrt{18/20}$ being outside the resolution of their measurements.

The Haywood $\rho(\beta)$ was fed into a HARTRAN version of LEAP and ADDELT and run on the London University ATLAS computer by A.J.H.Goddard. S and $S \exp(\beta/2)$ matrices of 33α points $\times$ 35β points were output on cards, but in a

mixed form unsuitable for input to DIS. The matrices were therefore fed into a short FORTRAN IV program ATOB2, written by A.J.H.Goddard and modified by the present author. This was run on the Imperial College IBM 7090 computer; its function is to process the mixed matrices from LEAP into a format suitable for input to DIS, using the $S.\exp(\beta/2)$ input option. The Debye-Waller coefficient from equation 2.5.10 (output by LEAP) was .720036; adding on the contribution from the δ -functions at $\beta = 8.0$ and 17.3 increased this value to .760036.

For the scattering law input option in DIS, the subroutine ESS, instead of evaluating equation 2.3.11 as for the Nelkin model, interpolates the $S.\exp(\beta/2)$ matrix linearly in α and β . All values of α and β required by the calculation must lie within the range of the scattering law matrix - if this is not so a diagnostic will be printed and the run terminated. The addition of the Debye-Waller elastic scattering contribution will be described in Section 2.8.

2.6 Angular Integration

The Legendre moments of the scattering kernel are given by

$$\sigma_n(E_o, E) = 2\pi \int_{-1}^1 \sigma(E_o, E, \mu) P_n(\mu) d\mu \quad 2.6.1$$

where $P_n(\mu)$ is the Legendre polynomial of order n . The numerical evaluation of these integrals is complicated by

the fact that $d\sigma/d\mu$ changes sign at approximately

$$\mu = \mu_0 = \left(\frac{m+1}{2}\right) \sqrt{\frac{E}{E_0}} - \left(\frac{m-1}{2}\right) \sqrt{\frac{E_0}{E}} \quad 2.6.2$$

(for the Haywood model and for $E = E_0$, μ_0 is taken as zero). The range of integration is therefore broken into two parts

$$\int_{-1}^1 d\mu \rightarrow \int_{-1}^{\mu_0} d\mu + \int_{\mu_0}^1 d\mu \quad 2.6.3$$

Each integral is then evaluated by double Gaussian quadrature. (The angles and weights for quadrature of order 15 are built into the program; for other orders they must be supplied as data.)

In the case of the Nelkin and free gas models, equations 2.3.11 and 2.4.1 contain a singularity for $E = E_0$. We therefore subtract this from the integrand of the diagonal elements:

$$\begin{aligned} \int_{-1}^1 \sigma(E, E, \mu) P_n(\mu) d\mu &= \int_{-1}^1 \left\{ \sigma(E, E, \mu) - \frac{\sigma_b}{4\pi} \sqrt{\frac{m}{2\pi T n^2}} \right\} P_n(\mu) d\mu \\ &+ \frac{\sigma_b}{4\pi} \sqrt{\frac{m}{2\pi T}} \int_{-1}^1 \frac{P_n(\mu)}{n} d\mu \end{aligned} \quad 2.6.4$$

where the integral in the final term is analytic.

It is necessary to compute the entire scattering matrix; the downscattering elements only are determined, and the upscattering elements calculated from the detailed balance relationship:

$$M(E_o) \sigma_n(E_o, E) = M(E) \sigma_n(E, E_o) , \quad 2.6.5$$

where $M(E) = E \exp (-E/T)$ is the Maxwellian distribution.

2.7 Scattering from Carbon and Oxygen

In the preceding sections we have been concerned with neutron scattering by protons. This is at the same time the most important contribution, since the scattering cross-section for hydrogen is much greater than those for carbon and oxygen, and also the most difficult, since the proton and neutron have very nearly the same mass. It is, however, necessary to make some allowance for the scattering from other elements, and for this reason a facility has been provided in DIS to compute if required free gas scattering matrices for carbon and/or oxygen in the manner described in Sections 2.4 and 2.6, and to add these to the matrices already obtained for hydrogen according to the molecular composition specified in the data.

2.8 Energy Integration

It will be shown in the next chapter that the scattering information required for the computation of the spatial eigenvalue is the total scattering cross-section

$$\sigma_s(E) = \int_0^{\infty} \sigma_o(E \rightarrow E') dE' \quad 2.8.1$$

and also the differential scattering cross-section, which

occurs in integrals of the form

$$I_n(E) = \int_0^{\infty} \sigma_n(E' \rightarrow E) \varphi_n(E') dE' \quad 2.8.2$$

where $\varphi_n(E)$ is the n th Legendre moment of the flux. Note that 2.8.1 involves an integration over final energies, and 2.8.2 over initial energies.

The Lagrangian method of numerical integration (after Besant, 1965) is used for both integrations. For a total number of abscissae $N \geq 4$, in order to evaluate the area between x_j and x_{j+1} we fit a cubic through the four points x_{j-1} , x_j , x_{j+1} and x_{j+2} , and use this to calculate the value of the function at $\frac{1}{2}(x_j + x_{j+1})$. The area between x_j and x_{j+1} is then evaluated using Simpson's rule. For the first and last intervals in the range we use the cubics calculated for the second and penultimate intervals, respectively. If $N=3$ we fit a quadratic between the three points, use it to compute the value of the function at the mid-points $\frac{1}{2}(x_1 + x_2)$ and $\frac{1}{2}(x_2 + x_3)$, and apply Simpson's rule twice. For $N=2$ we use a simple trapezoidal rule.

Reference to Figure 2 shows that the differential cross-section undergoes a first-order discontinuity for $E=E'$. The integrals 2.8.1 and 2.8.2 are therefore split into two parts: $E' \leq E$ and $E' \geq E$. It is necessary to compute a matrix of weights where the typical element w_{ij} is the weight appropriate to the j th energy point for an

integration at E_i . Thus

$$\sigma_s(E_i) = \sum_{j=1}^{NE} w_{ij} \sigma_o(E_i \rightarrow E_j) \quad 2.8.3.$$

$$\text{and } I_n(E_i) = \sum_{j=1}^{NE} w_{ij} \sigma_n(E_j \rightarrow E_i) \varphi_n(E_j) \quad 2.8.4$$

where NE is the total number of energy points.

The formation of the weights matrix is under the control of subroutine SCATT; the weights themselves are computed in subroutines LAGWT, LAG3 and LAG2.

The total scattering cross-section is next formed according to equation 2.8.3, and the differential cross-sections are then weighted as in equation 2.8.4.

For the Haywood model, the Debye-Waller coefficient gives a δ -function contribution to $\sigma_n(E \rightarrow E')$ of the form

$$C_n(E) \delta(E-E') = \frac{\sigma_b}{2} \int_{-1}^1 e^{-\alpha \lambda} P_n(\mu) d\mu \delta(E-E') \quad 2.8.5$$

For $E=E'$, $\alpha = \frac{2E(1-\mu)}{AT}$, and the integrals involved are $B \int_{-1}^1 e^{c\mu} \mu^n d\mu$ where B and c are independent of μ . These integrals are readily evaluated analytically.

Because the $C_n(E)$ give a δ -function contribution, only after the weighting of equations 2.8.3 and 2.8.4 has been carried out is $C_o(E)$ added to $\sigma_s(E)$ and $C_n(E)$ to the diagonal elements of $\sigma_n(E \rightarrow E')$.

Typical curves of $\sigma_0(E \rightarrow E')$ are plotted in Figure 2; in comparing these it should be remembered that the Debye-Waller contribution to the elastic scattering for the Haywood model has not been included.

The scattering information has now been established. The time taken on the IBM 7090 to compute scattering matrices for a 32-point energy mesh is about 5 minutes for the Nelkin model, and about $1\frac{1}{2}$ minutes for either the free gas or Haywood models. It depends only slightly on how many Legendre moments are called for (the highest allowed are the $\sigma_3(E \rightarrow E')$). In order to save machine time for future runs where the same scattering cross-sections will be required, an option is available to punch on cards the weighted scattering matrices and the total cross-section, in such a format that they may be read in as data on subsequent runs. Options are also available to print weighted and/or unweighted matrices. Yet another option causes the unweighted matrices, the weights matrix and the $C_n(E)$ (if applicable) to be punched on cards in format suitable for input to the power series program TAPIO, described in Chapter 4.

Control is now returned to the main program, where the microscopic cross-sections are multiplied by number densities in readiness for the next stage.

CHAPTER 3

TRANSPORT THEORY DETERMINATION OF THE SPATIAL EIGENVALUE

Having assembled the relevant macroscopic cross-sections, the next stage in the determination of the spatial eigenvalue by the program DIS is the solution of the transport equation. Since we shall in this work be considering highly absorbing media, where diffusion theory will not be expected to apply, a rigorous transport theory method has been used, and will be described in the present chapter. The basic theory, given in the first three sections of this chapter, has been taken from Honeck (1962).

3.1 Formulation of the Problem

The stationary one-dimensional Boltzmann equation for a source-free semi-infinite medium defined for $z > 0$, and bounded by a plane source of neutrons at $z = 0$, is

$$\left[\mu \frac{\partial}{\partial z} + \Sigma_T(E) \right] \varphi(z, E, \mu) = \int_{-1}^1 d\mu' \int_0^{\infty} dE' \Sigma_s(E' \rightarrow E, \mu_0) \varphi(z, E', \mu')$$

3.1.1

where μ_0 is the cosine of the angle between μ and μ' , and $\Sigma_T(E)$ is the total macroscopic cross-section at energy E . We consider solutions of the form

$$\varphi(z, E, \mu) = \varphi(E, \mu) e^{-\kappa z} \quad 3.1.2$$

for $z > 0$, where in diffusion theory κ is the inverse of the diffusion length. Implicit in the separation of variables in equation 3.1.2 is the fact that the neutron spectrum does not change with position; i.e. neutrons of all energies have the same relaxation length $1/\kappa$.

Substituting equation 3.1.2 into 3.1.1 we obtain

$$[\Sigma_T(E) - \kappa \mu] \varphi(E, \mu) = \int_{-1}^1 d\mu' \int_0^\infty dE' \Sigma_s(E' \rightarrow E, \mu_0) \varphi(E', \mu) \quad 3.1.3$$

Inspection of equation 3.1.3 shows that since the R.H.S. and φ are essentially positive, the square bracket on the L.H.S. must be positive also. Thus, in order that the eigenvalue κ exist at all,

$$\kappa < \Sigma_T(E) \quad \text{for all } E. \quad 3.1.4$$

The physical reason for this is that the neutron flux for energy E cannot decay more rapidly than $\exp(-\Sigma_T(E).z)$.

There are two basic methods by which the eigenvalues of equation 3.1.3 may be obtained. The first is by expanding the flux and κ^2 as power series in the absorber concentration, and the second is by a direct P_L or B_L analysis. We shall be considering power series methods in the next chapter; for the present we confine the discussion to the direct method, on which the program DIS is based.

3.2 The B_L Method

The flux and scattering kernel of equation 3.1.3 are each expanded in a series of Legendre polynomials:

$$\varphi(E, \mu) = \sum_{n=0}^{\infty} \frac{(2n+1)}{2} \varphi_n(E) P_n(\mu) \quad 3.2.1$$

$$\text{and } \Sigma_s(E' \rightarrow E, \mu_o) = \sum_{n=0}^{\infty} \frac{(2n+1)}{2} \Sigma_n(E' \rightarrow E) P_n(\mu_o). \quad 3.2.2$$

Substituting equations 3.2.1 and 3.2.2 into equation 3.1.3 and using the addition theorem for Legendre polynomials we have

$$\begin{aligned} [\Sigma_T(E) - \kappa \mu] \sum_{n=0}^{\infty} \frac{(2n+1)}{2} \varphi_n(E) P_n(\mu) &= \\ &= \sum_{n=0}^{\infty} \frac{(2n+1)}{2} \int_0^{\infty} dE' \Sigma_n(E' \rightarrow E) \varphi_n(E') P_n(\mu) \end{aligned} \quad 3.2.3$$

In the P_L method, the next step is to replace the $\mu P_n(\mu)$ terms on the L.H.S. of equation 3.2.3 with terms containing $P_{n+1}(\mu)$ and $P_{n-1}(\mu)$, before multiplying the whole equation through by $P_\ell(\mu)$ and integrating over μ . In the present method, however, which for a given order L normally gives more accurate results (since (e.g. Beckurts and Wirtz, 1964) for isotropic scattering a B_0 calculation gives the complete solution, whereas several terms are still required with the P_L method), equation 3.2.3 is divided by the term in square brackets and then multiplied by $P_\ell(\mu)$ and integrated over μ :-

$$\varphi_{\ell}^{(E)} = \sum_{n=0}^{\infty} \frac{(2n+1)}{2} \int_{-1}^1 \frac{d\mu P_{\ell}(\mu) P_n(\mu)}{[\Sigma_T(E) - \kappa \mu]} \cdot \int_0^{\infty} dE' \Sigma_n(E' \rightarrow E) \varphi_n(E') \quad 3.2.4$$

$$\text{If we define } Q_{\ell n}(E, \kappa) = \frac{(2n+1)}{2} \int_{-1}^1 \frac{d\mu P_{\ell}(\mu) P_n(\mu)}{[\Sigma_T(E) - \kappa \mu]} \quad 3.2.5$$

equation 3.2.4 may be written

$$\varphi_{\ell}^{(E)} = \sum_{n=0}^L Q_{\ell n}(E, \kappa) \int_0^{\infty} dE' \Sigma_n(E' \rightarrow E) \varphi_n(E') \quad 3.2.6$$

where the infinite sum has now been replaced by a sum over $L+1$ terms, where L is the highest order of anisotropy allowed for in the expansions 3.2.1 and 3.2.2. The eigenvalue κ which is to be determined is implicit in equation 3.2.6 and cannot therefore be obtained by standard eigenvalue techniques directly. We therefore solve the following equation

$$\omega \varphi_{\ell}^{(E_i)} = \sum_{n=0}^L Q_{\ell n}(E_i, \kappa) \sum_{j=1}^{NE} w_{ij} \Sigma_n(E_j \rightarrow E_i) \varphi_n(E_j) \quad 3.2.7$$

in which we have approximated the energy integral by a finite sum over NE energy points, for ω at various fixed values of κ . The procedure is as follows: an initial value of κ which is expected to be close to the true value is supplied as input to the program, and using this value of κ the eigenvalue ω that is closest to unity is obtained by inverse iteration. On the basis of this ω ,

the value of κ is modified and a new eigenvalue ω obtained. The process is repeated until ω is sufficiently close to unity.

3.3 Computation of the Function Q

We now consider how the function defined by equation 3.2.5 may be evaluated. The product of two Legendre polynomials always yields either an even or an odd function. We may therefore reduce the range of integration in equation 3.2.5 to $\int_0^1 d\mu$. We obtain

$$Q_{\ell n}(E_i, \kappa) = \frac{2n+1}{\Sigma(E_i)} \int_0^1 d\mu \frac{P_\ell(\mu)P_n(\mu)}{1-c^2\mu^2} \times \begin{cases} (1, & \text{if } \ell+n \text{ is even} \\ c\mu, & \text{if } \ell+n \text{ is odd} \end{cases}$$

3.3.1

where $c \equiv c(E_i) = \kappa / \Sigma_T(E_i)$. The integrals are of the form

$$I_m = \int_0^1 d\mu \frac{\mu^m}{1-c^2\mu^2} \quad 3.3.2$$

where m is always even. It can readily be shown that

$$I_0 = \frac{1}{2c} \ln \left(\frac{1+c}{1-c} \right) \quad 3.3.3$$

$$\text{and that} \quad c^2 I_m = I_{m-2} - \frac{1}{m-1} \quad 3.3.4$$

for $m \geq 2$. The evaluation of Q takes place according to this scheme in subroutine KEW; the product $P_\ell(\mu)P_n(\mu)$ is expressed as a power series in μ , and $Q_{\ell n}(E_i, \kappa)$ computed as a sum of the I_m .

3.4 Inverse Iteration Method

We rewrite equation 3.2.7 in the form

$$\underline{A} \underline{\Phi} = \omega \underline{\Phi} \quad 3.4.1$$

where $\underline{\Phi}$ is a vector containing $NE \times (L+1)$ elements of the form $\varphi_{\ell}(E_i)$, arranged in blocks of constant ℓ . For the first iteration $\underline{\Phi}$ may either contain the final values from some previously run similar case or guess values supplied by the program as

$$\varphi_{\ell}(E) = \frac{M(E)}{2\ell+1} \quad 3.4.2$$

The square matrix $\underline{A}$ contains elements of the form

$$Q_{\ell n}(E_i, \kappa) w_{ij} \Sigma(E_j - E_i).$$

The method of power iteration which we shall use converges to give the largest eigenvalue of a matrix. In this case we require the eigenvalue nearest to unity, so we subtract unit matrix $\underline{I}$ from either side of equation 3.4.1 and multiply both sides by $(\underline{A} - \underline{I})^{-1}$, to give

$$\underline{B} \underline{\Phi} = \lambda \underline{\Phi} \quad 3.4.3$$

where $\underline{B} = (\underline{A} - \underline{I})^{-1}$ and $\lambda = \frac{1}{\omega - 1}$. The largest eigenvalue of equation 3.4.3 will then be that corresponding to the ω nearest to unity. The method of solving equation 3.4.3 by power iteration is described in Appendix 1. The computations take place under the control of the subroutine POWIT; subroutine ANORM normalises the vector at each iteration to make its largest element unity, and subroutine

MULTI performs matrix multiplication. See Figure 3. The largest and smallest values of the ratio of eigenvector elements for successive iterates (which should converge to the largest eigenvalue λ_1) are printed out at each iteration, as well as the ratio corresponding to the largest element of the vector, which is the value ultimately taken for λ_1 .

If the value of ω is too close to the true value, ω will be very close to unity, and λ will be so large that incorrect results are produced for that particular (outer) iteration. To avoid this, if $\omega - 1 < .0005$, instead of subtracting unit matrix from both sides of equation 3.4.1 at the start of the next iteration, $1.001 \mathbf{I}$ is subtracted, giving $\lambda = \frac{1}{\omega - 1.001}$. This value of λ will not be large enough to cause any trouble.

3.5 Acceleration Techniques

Two methods of speeding up the power iteration process are provided which may be selected at the discretion of the user. In both cases the straightforward method of Appendix 1 is used until a fairly good value of λ (λ_g say) has been obtained (in practice the criterion that the difference between the largest and smallest ratios of the corresponding elements of successive iterates should be less than 20% of the largest ratio has proved satisfactory), and in any case for the first three inner iterations.

In the first method, due to Wielandt (1944), we then operate on equation 3.4.3 to give

$$(\underline{B} - \underline{I} \lambda_g)^{-1} \underline{\Phi} = \frac{1}{\lambda - \lambda_g} \underline{\Phi} \quad 3.5.1$$

This defines a new eigenvalue problem where providing λ_g is sufficiently close to the true λ convergence will be speeded up. This method has been found to work very well, although because it involves a time consuming matrix inversion it is only allowed to operate after at least three inner iterations.

The second method is that of Erdelyi (1965). As before, the normal method is used until reasonable convergence has been obtained. The values of λ obtained during the next three iterations are stored (ℓ_1, ℓ_2, ℓ_3), and after these have been performed the following parameters are evaluated:

$$\begin{aligned} r &= \frac{\ell_3 - \ell_2}{\ell_2 - \ell_1} &) \\ \lambda_2 &= \frac{r}{1-r} (\ell_2 - r\ell_1) &) \\ q &= \frac{m-1}{m-r} \lambda_2 &) \end{aligned} \quad 3.5.2$$

where m is the factor (supplied as data; possible values are 2, 3 or 4) by which the ratio of the first to second eigenvalues is reduced. Subsequent iterations are now performed with

$$(\underline{B} - q\underline{I}) \underline{\Phi} = (\lambda_g - q) \underline{\Phi} \quad 3.5.3$$

which should accelerate convergence.

In practice it has been found that since at least six iterations are necessary before enough information has been assembled to apply equation 3.5.3, this method has little or no chance to create any effect, as convergence will normally have been achieved by this time anyway.

3.6 Modification of κ

When the inner convergence criterion has been satisfied, the value of ω is printed out. If this is sufficiently close to unity (in practice a criterion of $2 \cdot 10^{-6}$ for $|\omega - 1|$ has been used) control passes to the output stage. If this is not so a new value of κ must be derived. It was originally intended to do this by a method similar to that used by Wood (1965), which consists in applying Newton's method of finding a root to the equation

$$\omega(\kappa) - 1 = 0 \quad . \quad 3.6.1$$

For this we need to know $d\omega/d\kappa$, which is obtained by differentiating equation 3.4.1 with respect to κ and multiplying by $\underline{A}^{-1}$ to give

$$\frac{d\omega}{d\kappa} = \frac{\omega \left[\underline{A}^{-1} \frac{d\underline{A}}{d\kappa} \underline{\Phi} \right]_s}{[\underline{\Phi}]_s} \quad 3.6.2$$

where $[\underline{\Phi}]_s$ is the largest element of the vector $\underline{\Phi}$. The improved value κ' is then given by

$$\kappa' = \kappa - \frac{(\omega - 1)}{d\omega/d\kappa} \quad 3.6.3$$

The subroutine KEW was extended so that $dQ/d\kappa$ could be calculated, and the method tried, but it was found to suffer from two disadvantages: first, it involved a time-consuming matrix inversion, and second, it was necessary to provide computer storage for both the matrices A and B, which would have involved either restricting problems to B_1 complexity or storing the matrix A on tape while the inner iterations were being performed. It was therefore decided to try a simpler method, as follows: after the first iteration, the value of κ is modified according to an empirical rule

$$\kappa' = \kappa + \left(\frac{10}{\kappa} + 1.5 - 3\kappa\right)(1-\omega) \quad 3.6.4$$

and after subsequent iterations a straight line is fitted through the two sets of (ω, κ) so far produced which have values of ω closest to unity, and solved for $\omega=1$ to give κ' . This method has been found to work well, converging at the rate of at least one order of magnitude in $|\omega-1|$ per iteration, and is therefore the one incorporated in the present version of the program.

If the condition expressed by equation 3.1.4 should be violated, the program would produce false results, since it would be required to calculate the logarithm of a negative number (equation 3.3.3). A device has therefore been built into the program to determine $(\Sigma_T(E_i))_{\min}$

for each problem and to ensure that μ never exceeds this value.

CHAPTER 4

POWER SERIES EXPANSIONS

In this chapter we shall consider how κ^2 may be represented by a power series in absorber concentration, and how this is related to a similar technique in the analysis of pulsed experiments.

The coefficients of such an expansion may be derived theoretically from a set of direct determinations of κ by the method described in the previous chapter. Alternatively, they may be obtained directly by a power series solution of the transport equation, making the assumption that anisotropic scattering is elastic. Both of these methods will be discussed.

An analytic method will also be considered, in which the flux is represented by a series of Laguerre polynomials. Although this method differs from the two mentioned above in that it cannot be used to predict the expansion coefficients directly, under certain assumptions it shows how these coefficients are related to intrinsic properties of the moderator.

4.1 General

It can readily be shown (e.g. Glasstone and Edlund (1952)) that for simple diffusion theory where the absorption is small

$$\kappa^2 = \frac{1}{L^2} = \frac{\int \Sigma_a(E) \varphi(E) dE}{\int D(E) \varphi(E) dE} \quad 4.1.1$$

where L is the diffusion length. The distortion of the neutron spectrum by the added absorber should only affect κ^2 in the second order, and we may therefore write

$$\kappa^2 = N \frac{\int \sigma(E) M(E) dE}{\int D(E) M(E) dE} + O(N^2) \quad 4.1.2$$

where $M(E)$ is the Maxwellian, which is the spectrum in the non-absorbing case, and N is the number density of absorber atoms with microscopic absorption cross-section $\sigma(E)$.

For $1/v$ absorption, we shall consider the expansion

$$\kappa^2 = \alpha_1 a - \alpha_2 a^2 + \alpha_3 a^3 - \dots \quad 4.1.3$$

where a ($\equiv \Sigma_{a0}$) is the 2200 m/sec. macroscopic absorption cross-section, and for non- $1/v$ absorbers, where we have to include the $1/v$ effect of the hydrogen in water,

$$\kappa^2 = \beta_0 + \beta_1 N - \beta_2 N^2 + \beta_3 N^3 - \dots \quad 4.1.4$$

4.2 Relation between Pulsed and Stationary Experiments

In the die-away experiment a pulse of fast neutrons is injected into a moderating assembly. After sufficient time has elapsed only the fundamental spatial harmonic remains, and this decays exponentially with a time constant

λ . For a $1/v$ absorbing moderator, λ may be expressed as

$$\lambda = \lambda_0 + D_0 B^2 - C B^4 + F B^6 + \dots \quad 4.2.1$$

$$\text{where } \lambda_0 = v_0 \Sigma_{a0} \quad 4.2.2$$

and B^2 is the geometric buckling of the system.

The formal relation between this and the stationary experiments can be seen by referring to the transport equation 3.1.3. The square bracket on the L.H.S. is $\Sigma_T(E) - \kappa\mu$ for the stationary experiment; for the pulsed problem, it would be $\Sigma_T(E) - \frac{\lambda}{v} + iB\mu$. Thus the stationary experiment is equivalent to the pulsed one with

$$\lambda = 0 \quad 4.2.3$$

$$\text{and } B = i\kappa. \quad 4.2.4$$

Using equations 4.2.2, 4.2.3, and 4.2.4 we can relate the coefficients of the expansion 4.2.1 to those of 4.1.3 for $1/v$ poisoned experiments. The expressions are (Honeck 1962))

$$\begin{aligned} \alpha_1 &= v_0/D_0 &) \\ \alpha_2 &= v_0^2 C/D_0^3 &) \\ \alpha_3 &= v_0^3 (2C^2 - D_0 F)/D_0^5 &) \end{aligned} \quad 4.2.5$$

$$\begin{aligned} D_0 &= v_0/\alpha_1 &) \\ C &= v_0 \alpha_2/\alpha_1^3 &) \\ F &= v_0 (2\alpha_2^2 - \alpha_1 \alpha_3)/\alpha_1^5 &) \end{aligned} \quad 4.2.6$$

If pulsed experiments are performed in a non- $1/v$

absorbing medium, it is convenient to make the expansion (Calame, 1964b)

$$\lambda = \lambda_{00} + B^2 \lambda_{20} + B^4 \lambda_{40} + N(\lambda_{01} + B^2 \lambda_{21}) + N^2(\lambda_{02} + B^2 \lambda_{22}) + \dots$$

4.2.7

The coefficients of equation 4.1.4 have been expressed by the present author in terms of the coefficients of this expansion by

$$\begin{aligned} \beta_0 &= \frac{\lambda_{00}}{\lambda_{20}} + \frac{\lambda_{00}^2 \lambda_{40}}{\lambda_{20}^3} \\ \beta_1 &= \frac{\lambda_{01}}{\lambda_{20}} - \frac{\lambda_{21} \lambda_{00}}{\lambda_{20}^2} + \frac{2 \lambda_{00} \lambda_{40} \lambda_{01}}{\lambda_{20}^3} - \frac{3 \lambda_{00}^2 \lambda_{40} \lambda_{21}}{\lambda_{20}^4} \end{aligned}$$

4.2.8

(The expression for β_2 involves higher order terms than are given in the expansion 4.2.7 and is not considered worthwhile.)

4.3 Evaluation of Coefficients from Point Theoretical Values

In this, the most accurate of the theoretical methods to be presented for evaluating the α 's and β 's, a series of about ten runs are made for a given absorber, temperature and scattering model using the program DIS, varying the concentration of absorber between each run. The values of κ^2 obtained are then fitted to the appropriate expansion

4.1.3 or 4.1.4 by the least squares method.

A computer program POLY has been written in FORTRAN IV by the present author, incorporating a generalised subroutine PARFIT to perform the least squares fitting. This subroutine has been used elsewhere in this work, and the theory of it is given in Appendix 2. Two, three and four parameter fits are made, and a procedure used whereby points may be successively omitted from the high concentration end. The results will be presented in Chapters 6 and 7, where comparisons with experiment will also be made.

4.4 Power Series Solution of the Transport Equation

This method, which is taken from Honeck (1962), applies to $1/v$ absorbers. We write equation 3.1.3 as

$$[\Sigma_a(E) - \kappa \mu] \varphi(E, \mu) = S\varphi(E, \mu) \quad 4.4.1$$

$$\text{where } S\varphi(E, \mu) \equiv \int_{-1}^1 d\mu' \int_0^\infty dE' \Sigma_s(E' \rightarrow E, \mu_0) \varphi(E', \mu') - \Sigma_s(E) \varphi(E, \mu)$$

4.4.2

Consider the solution of equation 4.4.1 by expressing κ^2 as a power series

$$\kappa^2 = \sum_{j=1}^{\infty} (-1)^{j+1} \alpha_j a^j \quad 4.4.3$$

The flux is expanded as a power series in a , and also in Legendre polynomials in μ . A factor κ is extracted from each successive Legendre coefficient; the reason for this

will appear later.

$$\varphi(E, \mu) = \sum_{\ell=0}^{\infty} \sum_{m=0}^{\infty} \frac{(2m+1)}{2} \varphi_{m\ell}(E) P_m(\mu) \kappa^m a^\ell. \quad 4.4.4$$

The scattering kernel is also expanded in Legendre polynomials:

$$\Sigma_s(E' \rightarrow E, \mu_0) = \sum_{i=0}^{\infty} \frac{(2i+1)}{2} \Sigma_{si}(E' \rightarrow E) P_i(\mu_0) \quad 4.4.5$$

Insert expansions 4.4.4 and 4.4.5 into equation 4.4.1, multiply by $P_n(\mu)$ and integrate over μ . We obtain

$$\begin{aligned} & ag(E) \kappa^n \sum_{\ell} (\varphi_{n\ell}(E) a^\ell) - \sum_{\ell} \left[\frac{n}{(2n+1)} a^\ell \kappa^n \varphi_{n-1,\ell}(E) + \right. \\ & \quad \left. + \frac{(n+1)}{(2n+1)} a^\ell \kappa^{n+2} \varphi_{n+1,\ell}(E) \right] \\ & = \int_0^\infty dE' \Sigma_{sn}(E' \rightarrow E) \kappa^n \sum_{\ell} (\varphi_{n\ell}(E') a^\ell) - \Sigma_s(E) \varphi_{n\ell}(E) \kappa^n a^\ell \end{aligned} \quad 4.4.6$$

where $g(E) = \Sigma_a(E)/a$.

The significance of the factor κ^m in the expansion 4.4.4 now becomes apparent: we can cancel κ^n from equation 4.4.6 so that the eigenvalue appears only as κ^2 . Substituting equation 4.4.3 and equating coefficients of a^ℓ we obtain

$$\begin{aligned} g(E) \varphi_{n,\ell-1}(E) - \frac{n}{(2n+1)} \varphi_{n-1,\ell}(E) - \frac{(n+1)}{(2n+1)} \sum_{j=0}^{\ell-1} (-1)^{\ell-j+1} \\ \alpha_{\ell-j} \varphi_{n+1,j}(E) + \Sigma_s(E) [1 - b_n(E)] \varphi_{n\ell}(E) = S_n \varphi_{n\ell} \end{aligned} \quad 4.4.7$$

$$\text{where } b_n(E) = \frac{\Sigma_{sn}(E)}{\Sigma_s(E)} \quad 4.4.8$$

$$\text{and } S_n \varphi_{n\ell}(E) = \int_0^\infty dE' \Sigma_{sn}(E' \rightarrow E) \varphi_{n\ell}(E') - \Sigma_{sn}(E) \varphi_{n\ell}(E) \quad 4.4.9$$

We now proceed to solve the set of coupled integral equations 4.4.7. The general procedure has been described by Nelkin (1960b) for the similar case of the decay of a neutron pulse. The problem is considerably simplified if we assume, after Nelkin and Honeck, that anisotropic scattering is elastic. This will be exact in the constant cross-sections case. Under this approximation the terms $S_n \varphi_{n\ell}$ vanish for $n > 0$, and only the integral equations for $n=0$ need be solved.

There are two constraints on the system. The first is that of neutron conservation, which requires that

$$\int_0^\infty S_n \varphi_{n\ell} dE = 0 \quad \text{for all } n, \ell \quad 4.4.10$$

The second is the requirement of detailed balance, which is applicable to the thermodynamic equilibrium between neutrons and non-absorbing moderators:

$$\Sigma_s(E \rightarrow E') M(E) = \Sigma_s(E' \rightarrow E) M(E') \quad 4.4.11$$

In order to simplify the analysis, we define the following quantities (suppressing the arguments E):

$$D_1 = \frac{1}{3\Sigma_s(1-b_1)} \quad 4.4.12$$

$$D_2 = \frac{2}{15\Sigma_s^2(1-b_1)(1-b_2)} \quad 4.4.13$$

$$D_3 = \frac{2}{35 \Sigma_s^3 (1-b_1)(1-b_2)(1-b_3)} \quad 4.4.14$$

$$A_1 = g - \alpha_1 D_1 \quad 4.4.15$$

$$A_2 = 3D_1 (gD_1 - \frac{2}{3} \alpha_1 D_2) \quad 4.4.16$$

$$A_3 = 5D_2 (gD_2 - \frac{3}{5} \alpha_1 D_3) \quad 4.4.17$$

The equations 4.4.7 are solved recursively starting from the $n=0$, $\ell=0$ equation. The only integral equations which have to be solved are

$$A_1 M = S_0 \varphi_{01} \quad 4.4.18$$

$$\text{and } A_1 \varphi_{01} + (\alpha_2 D_1 + \alpha_1 A_2) M = S_0 \varphi_{02} \quad 4.4.19$$

for φ_{01} and φ_{02} . The coefficients α_j of expansion 4.4.3 are found to be given by

$$\overline{D}\alpha_1 = \int dE g M \quad 4.4.20$$

$$\overline{D}\alpha_2 = - \int dE (A_1 \varphi_{01} + \alpha_1 A_2 M) \quad 4.4.21$$

$$\begin{aligned} \text{and } \overline{D}\alpha_3 = \int dE \{ & A_1 \varphi_{02} + (\alpha_2 D_1 + \alpha_1 A_2) \varphi_{01} + \\ & + (-\alpha_2 A_2 - \alpha_1 \frac{A_2^2}{D_1} + 2\alpha_1 \alpha_2 D_1 D_2 + \alpha_1^2 A_3) M \} \end{aligned} \quad 4.4.22$$

$$\text{where } \overline{D} = \int dE D_1 M \quad 4.4.23$$

A FORTRAN IV program TAPIO has been written by the present author to evaluate these coefficients. The differential scattering cross-sections, weights for integration (and Debye-Wallter factors if appropriate) are obtained on cards in correct format from the program DIS, by selecting

the appropriate option, and input as data to TAPIO.

Besides the coefficients α_j , and their pulsed equivalents derived from the relations 4.2.6, TAPIO outputs in passing the total microscopic scattering and transport cross-sections and the diffusion coefficient, all as functions of energy, and also the Maxwellian averaged diffusion coefficient, defined by

$$\bar{D}_M = \frac{\int \frac{D \, dE}{3\Sigma_s(1-b_1)}}{\int M \, dE} \quad 4.4.24$$

The results will be presented in Chapter 6.

4.5 Laguerre Polynomials Method - $1/v$ Absorption

This method differs from the previous two in two respects; first, it is analytic rather than numeric, and second, it is based not on the transport but on the diffusion equation. The method has been used by Friedman (1962 and 1964) for the pulsed non- $1/v$ case, and a similar technique for the stationary case is given here.

We consider first the case of $1/v$ absorption. The diffusion equation is

$$[\Sigma_a(E) - \kappa^2 D(E)]\phi(E) = \int_0^\infty dE' [\Sigma_s(E' \rightarrow E)\phi(E') - \Sigma_s(E \rightarrow E')\phi(E)] \quad 4.5.1$$

Following Kazarnovsky et al (1958), the flux is represented by a Maxwellian, with characteristic temperature equal to that of the moderator, modified by a correction involving

a set of Laguerre polynomials of the order unity. The polynomials of order unity are chosen because their normalization involves the Maxwellian xe^{-x} . The functions $L_i^{(1)}(E/T)M(E)$ are in fact eigenfunctions of the heavy gas thermalization operator. We adopt the normalization

$$\int_0^{\infty} \frac{E}{T^2} e^{-E/T} L_i^{(1)}\left(\frac{E}{T}\right) L_j^{(1)}\left(\frac{E}{T}\right) dE = \delta_{ij} \quad 4.5.2$$

and henceforth omit the superscript (1).

We write for the flux

$$\varphi(E) = M(E) \sum_{j=0}^L c_j L_j(E/T) \quad 4.5.3$$

Substituting this expansion into equation 4.5.1, and integrating over energy, we obtain using the normalization equation 4.5.2

$$\sum_{j=0}^L c_j \left[\Sigma_{ao} v_o \frac{\sqrt{\pi}}{2v_T} w_{ij} - \kappa^2 D_{ij} + \gamma_{ij} \right] = 0 \quad i=0 \dots L \quad 4.5.4$$

$$\text{where} \quad w_{ij} = \frac{2v_T}{\sqrt{\pi}} \left(\frac{1}{v}\right)_{ij} \quad 4.5.5$$

$$v_T = (2T/M_n)^{\frac{1}{2}} \quad 4.5.6$$

$$\gamma_{ij} = \int_0^{\infty} \int_0^{\infty} dE' dE M(E) \Sigma_s(E \rightarrow E') \left[L_j\left(\frac{E}{T}\right) - L_j\left(\frac{E'}{T}\right) \right] L_i\left(\frac{E}{T}\right) \quad 4.5.7$$

$$f_{ij} = \int_0^{\infty} dE L_i\left(\frac{E}{T}\right) L_j\left(\frac{E}{T}\right) M(E) f(E) \quad 4.5.8$$

and T is the moderator temperature. The detailed balance condition 4.4.11 has been assumed. We note that

$$\gamma_{11} = N_H M_2 / 4 \quad 4.5.9$$

$$\text{where } M_2 = \frac{1}{T^2} \int_0^\infty \int_0^\infty dE' dE (E-E')^2 M(E) \sigma_s(E \rightarrow E') \quad 4.5.10$$

is the mean squared energy exchanged per collision, and N_H is the number density of scattering atoms, and also $\gamma_{ij} = \gamma_{ji}$, and $\gamma_{io} = 0$.

In order that the system of equations 4.5.4 may have a non-trivial solution for the c_j , the determinant of the coefficients must be zero. Writing

$$\kappa^2 = \alpha_1 \Sigma_{ao} - \alpha_2 \Sigma_{ao}^2 \quad 4.5.11$$

$$\text{we obtain } \left| \Sigma_{ao} v_o \frac{\sqrt{\pi}}{2v_T} w_{ij} - D_{ij} (\alpha_1 \Sigma_{ao} - \alpha_2 \Sigma_{ao}^2) + \gamma_{ij} \right| = 0 \quad 4.5.12$$

and on retaining only terms in Σ_{ao} we find

$$\alpha_1 = \frac{\sqrt{\pi}}{2v_T} \frac{v_o}{D_{oo}} \quad 4.5.13$$

To obtain α_2 we substitute equation 4.5.13 into equation 4.5.12, giving

$$\left| \Sigma_{ao} v_o \frac{\sqrt{\pi}}{2v_T} t_{ij} + \alpha_2 D_{ij} \Sigma_{ao}^2 + \gamma_{ij} \right| = 0 \quad 4.5.14$$

$$\text{where we have written } t_{ij} = D_{oo} w_{ij} - D_{ij} \quad 4.5.15$$

If we assume that the off-diagonal terms in the matrix γ are small compared with the diagonal ones (this is true for the heavy gas model) we obtain, retaining terms to

$$\Sigma_{ao}^2, \quad \alpha_2 = \frac{\pi}{4v_T^2} \frac{v_o^2}{D_{oo}^3} \sum_{j=1}^L \frac{t_{oj}^2}{\gamma_{jj}} \quad 4.5.16$$

$$\text{Now } D_{oo} = \frac{\sqrt{\pi}}{2v_T} D_o \quad 4.5.17$$

where D_o is the coefficient of B^2 in the pulsed experiment expansion 4.2.1. If we now assume $D(E)$ is proportional to v , we obtain

$$t_{o1} = \frac{1}{\sqrt{2}} \frac{\sqrt{\pi}}{2v_T} D_o \quad 4.5.18$$

and taking the case of $L=1$, and using the relation 4.2.6 we find for the diffusion cooling coefficient

$$C = \frac{\sqrt{\pi} D_o^2}{v_T M_2 N_H} \quad 4.5.19$$

This is identical to the expression obtained by Nelkin (1958) using the variational principle, where the flux was represented by a Maxwellian with characteristic temperature T_n taken as the variational parameter. In fact it can easily be shown (e.g. Beckurts and Wirtz, 1964) that the $L=1$ approximation is exactly equivalent to the representation of the perturbed spectrum by a Maxwellian distribution with a shifted temperature.

Using equation 4.5.19 we can determine M_2 from experimental determinations of α_1 and α_2 (and hence D_o and C). The errors introduced in the analysis (mainly from the use of diffusion theory and the assumption $D(E) \propto v$) can be checked by using values of D_o and C obtained from the method of Section 4.3 and comparing the value of M_2 obtained with the known M_2 of the appropriate model. This will be

discussed in Chapter 6.

4.6 Laguerre Polynomials Method - non- $1/v$ Absorption

For the hydrogen absorption of water,

$$\Sigma_a(E) = \frac{\Sigma_{a0} v_0'}{v} = \frac{\lambda_0}{v} \quad 4.6.1$$

The diffusion equation for N atoms/c.c. of added non- $1/v$ absorber, with microscopic absorption cross-section $\sigma(E)$,

is

$$\left[\frac{\lambda_0}{v} + N\sigma(E) - \kappa^2 D(E) \right] \varphi(E) = \int_0^\infty dE' [\Sigma_s(E' \rightarrow E) \varphi(E') - \Sigma_s(E \rightarrow E') \varphi(E)] \quad 4.6.2$$

We now write for the flux

$$\varphi(E) = M(E) \sum_{j=0}^L (c_j^{(0)} + N c_j^{(1)} + N^2 c_j^{(2)}) L_j\left(\frac{E}{T}\right) \quad 4.6.3$$

Substituting equation 4.6.3 into equation 4.6.2, multiplying by $L_i\left(\frac{E}{T}\right)$ and integrating over energy gives

$$\sum_{j=0}^L (c_j^{(0)} + N c_j^{(1)} + N^2 c_j^{(2)}) \left[\lambda_0 \frac{\sqrt{\pi}}{2v_T} w_{ij} + N \sigma_{ij} + \gamma_{ij} - \kappa^2 D_{ij} \right] = 0 \quad i = 0 \dots L \quad 4.6.4$$

We write

$$\kappa^2 = \beta_0 + \beta_1 N + \beta_2 N^2 \quad 4.6.5$$

The coefficient β_0 is just κ^2 for pure water. Now the second (diffusion heating) term in the expansion 4.5.11 is only about 1% of the first for pure water at room temperature. We therefore only consider the leading term; from

equation 4.5.13 we obtain

$$\beta_0 = \frac{\sqrt{\pi}}{2v_T} \frac{\lambda_0}{D_{00}} \quad 4.6.6$$

Substituting equations 4.6.6 and 4.6.5 into equation 4.6.4, we obtain, using the relation 4.5.15,

$$\begin{aligned} \sum_{j=0}^L (c_j^{(0)} + N c_j^{(1)} + N^2 c_j^{(2)}) \left[\frac{\lambda_0}{D_{00}} \frac{\sqrt{\pi}}{2v_T} t_{ij} + N \sigma_{ij} + \gamma_{ij} - D_{ij} (\beta_1 N - \beta_2 N^2) \right] = \\ = 0 \quad i = 0 \dots L \quad 4.6.7 \end{aligned}$$

We now look at the various powers of N in the system of equations 4.6.7. For the zeroth power we find

$$\begin{aligned} \sum_{j=0}^L c_j^{(0)} \left(\frac{\lambda_0}{D_{00}} \frac{\sqrt{\pi}}{2v_T} t_{ij} + \gamma_{ij} \right) = 0, \quad 4.6.8 \\ i = 0 \dots L \end{aligned}$$

which gives rise to the determinant 4.5.12 if $0(\lambda_0^2)$ is neglected.

For terms in N

$$\begin{aligned} \sum_{j=0}^L c_j^{(0)} (\sigma_{ij} - D_{ij} \beta_1) + \sum_{j=0}^L c_j^{(1)} \left(\frac{\lambda_0}{D_{00}} \frac{\sqrt{\pi}}{2v_T} t_{ij} + \gamma_{ij} \right) = 0, \\ i = 0 \dots L \quad 4.6.9 \end{aligned}$$

Multiplying equation 4.6.9 by $c_i^{(0)}$ and summing all the i equations, the terms with $c_j^{(1)}$ disappear because of equation 4.6.8, and we obtain

$$\beta_1 = \frac{\sum_{i,j=0}^L c_i^{(0)} c_j^{(0)} \sigma_{ij}}{\sum_{i,j=0}^L c_i^{(0)} c_j^{(0)} D_{ij}} \quad 4.6.10$$

β_1 thus depends only on the $c_j^{(0)}$ coefficients of the flux expansion, which we now need to determine. Solving the system of equations 4.6.8, and assuming, as in the previous section, that the matrix γ is diagonal, we find

$$c_j^{(0)} = - \frac{\sqrt{\pi}}{2v_T} \frac{\lambda_o}{D_{oo}} \frac{t_{oj}}{\gamma_{jj}} c_o^{(0)} \quad (j>0) \quad 4.6.11$$

Substituting this in equation 4.6.10 and neglecting $O(\lambda_o^2)$ we find

$$\beta_1 = \frac{\sigma_{oo}}{D_{oo}} + 2 \frac{\sqrt{\pi}}{2v_T} \frac{\lambda_o}{D_{oo}} \sum_{j=1}^L \frac{t_{oj} u_{oj}}{\gamma_{jj}} \quad 4.6.12$$

$$\text{where } u_{ij} = \sigma_{oo} D_{ij} - \sigma_{ij} D_{oo} \quad 4.6.13$$

For the N^2 terms of equations 4.6.7 we have

$$\sum_{j=0}^L c_j^{(0)} \beta_2 D_{ij} + \sum_{j=0}^L c_j^{(1)} (\sigma_{ij} - D_{ij} \beta_1) + \sum_{j=0}^L c_j^{(2)} \left(\frac{\lambda_o}{D_{oo}} \frac{\sqrt{\pi}}{2v_T} t_{ij} + \gamma_{ij} \right) = 0$$

$$i = 0 \dots L \quad 4.6.14$$

Multiplying through by $c_i^{(0)}$ and summing all i equations, we find using equation 4.6.8 that the final sum disappears as before, to give

$$\beta_2 = \frac{- \sum_{i,j=0}^L c_i^{(0)} c_j^{(1)} (\sigma_{ij} - D_{ij} \beta_1)}{\sum_{i,j=0}^L c_i^{(0)} c_j^{(0)} D_{ij}} \quad 4.6.15$$

so that we now need to determine the $c_j^{(1)}$. This can be done by substituting the expression 4.6.12 for β_1 into equation 4.6.9. The expressions are complicated so we

quote only the final result for $L = 1$

$$\beta_2 = \frac{u_{o1}^2}{D_{oo}^3 \gamma_{11}} \left\{ 1 + 2 \frac{\sqrt{\pi}}{2v_T} \frac{\lambda_o}{D_{oo}} \frac{t_{o1}}{\gamma_{11}} \left(\frac{D_{o1}}{D_{oo}} - \frac{u_{11}}{u_{o1}} \right) \right\} \quad 4.6.16$$

If we neglect terms in λ_o but retain off-diagonal elements in the matrix γ (to linear terms in $\frac{\gamma_{ij}}{\gamma_{ii}}$, $i \neq j$) and take $L = 3$ we find

$$\begin{aligned} \beta_2 = & \frac{u_{o1}^2}{D_{oo}^3 \gamma_{11}} \left(1 - 2 \frac{u_{o2}}{u_{o1}} \frac{\gamma_{12}}{\gamma_{22}} - 2 \frac{u_{o3}}{u_{o1}} \frac{\gamma_{13}}{\gamma_{33}} \right) \\ & + \frac{u_{o2}^2}{D_{oo}^3 \gamma_{22}} \left(1 - 2 \frac{u_{o3}}{u_{o2}} \frac{\gamma_{23}}{\gamma_{33}} \right) + \frac{u_{o3}^2}{D_{oo}^3 \gamma_{33}} \end{aligned} \quad 4.6.17$$

The expressions derived here are in quite a similar form to those of Friedman for the pulsed case. In fact the formulae for α_1 , α_2 and β_1 could have been obtained directly from Friedman's results using the relations 4.2.5 and 4.2.8.

The results of this section will be discussed further in Chapter 7.

CHAPTER 5

EXPERIMENTAL DETERMINATION OF THE SPATIAL
EIGENVALUE

An account is given in this chapter of a series of experiments in which the relaxation length of thermal neutrons was measured for several aqueous solutions. Pure water at 22.3°C and 65°C was first measured, followed by nine different concentrations of boric acid at the lower and ten at the upper temperature. Then followed ten concentrations of cadmium sulphate and finally ten of gadolinium nitrate, all at the lower temperature.

The emphasis in these experiments was the investigation of neutron transport phenomena under conditions where the neutron spectrum which would obtain in a non-absorbing medium would be severely distorted, and to this end the maximum concentration of each absorber was as high as was practicable. The absorbers were chosen to have the most varied effect on the neutron spectrum on account of the differences between their cross-sections.

The principle underlying the experimental design was the elimination as far as possible of corrections to be applied in the extraction of the spatial eigenvalue from the experimental data. Corrections for finite tank dimensions were particularly minimised since these involve diffusion approximations.

5.1 Description of Apparatus

The experiments to be described were performed by placing a tank inside a cave formed by concrete shielding blocks adjacent to the thermal column horizontal facility of the London University Research Reactor, CONSORT II, and measuring the neutron flux distribution in the tank along a line radial to the core. The tank contained successively pure water, and varying concentrations of boric acid, cadmium sulphate and gadolinium nitrate solutions. The various units of the apparatus are shown in Figure 4, where for clarity they have been spaced apart.

The thermal column of CONSORT is of hollow design; between the core and the access face are 4" of water and between 30" and 41" of graphite, depending on the exact path. The face itself is $37\frac{1}{4}$ " square; the nominal thermal flux at its centre is 10^9 n/cm²/sec at full power (100 kW), and the fast flux is 5% of this value.

In order to improve the fast/thermal ratio, and also to reduce harmonics, a graphite extension stack 14" thick and $36\frac{1}{4}$ " square was placed close to the access face, and in line with it. Cadmium sheet of thickness .020" was wrapped around this stack, which was mounted on a frame made of 1" square cross-section aluminium tube ("Speedframe"- Dexion Ltd.).

To correct for the effect of fast neutrons entering

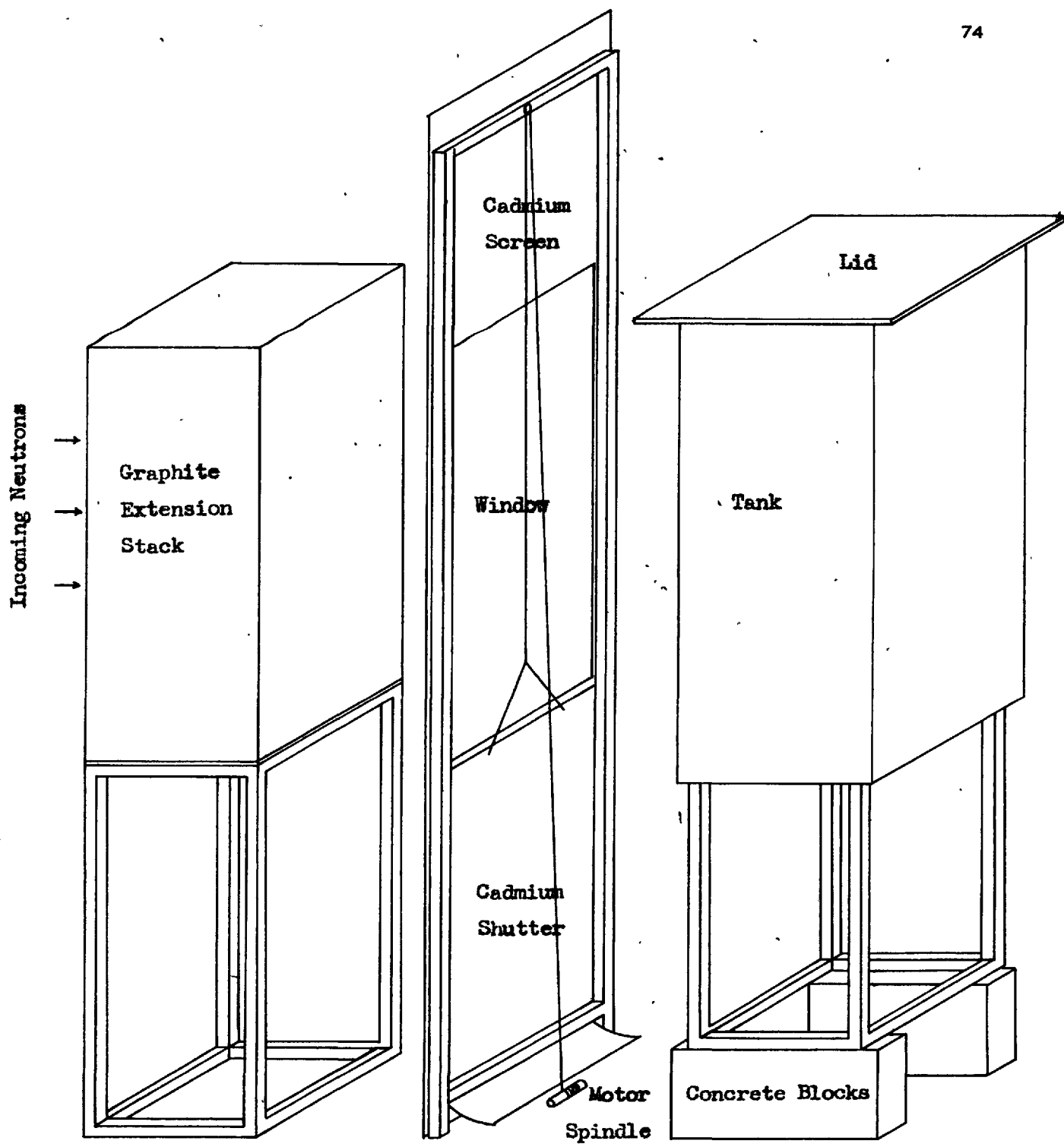
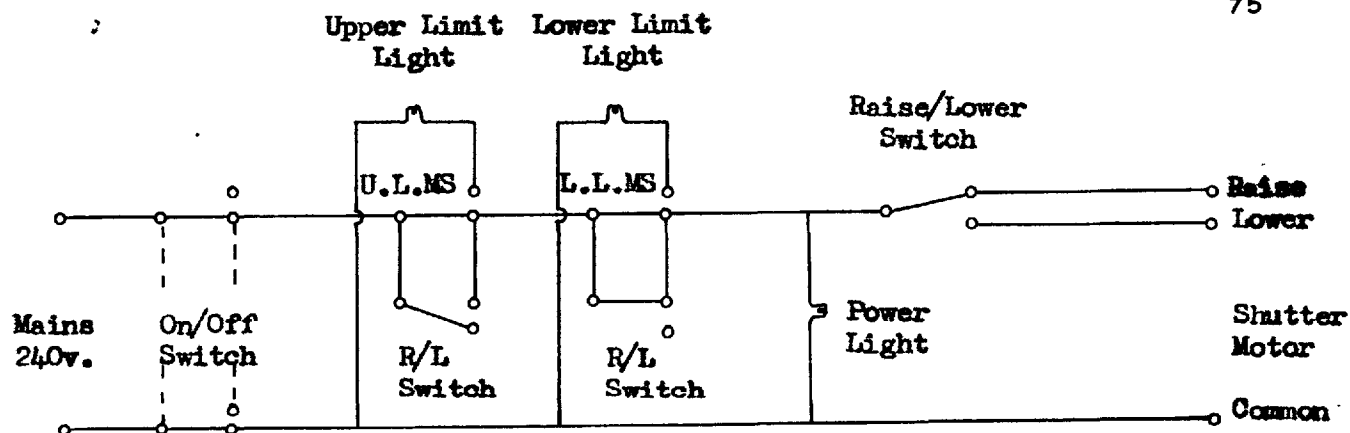
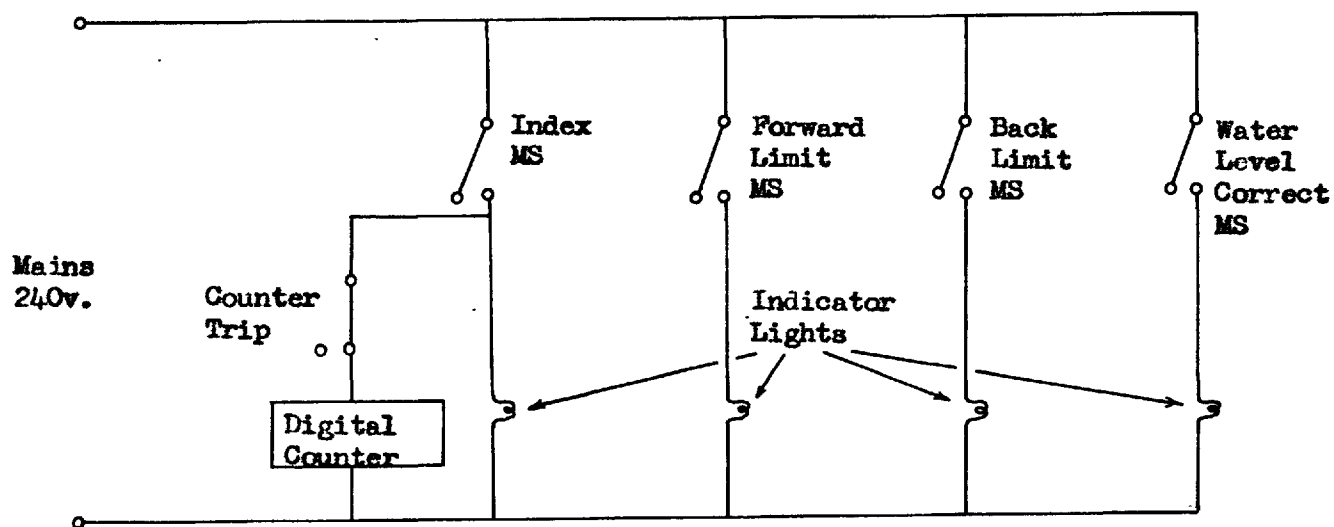


Figure 4 - Layout of Apparatus



(a) Shutter Control Circuit



(b) Information Circuits

Figure 5 - Control Circuits

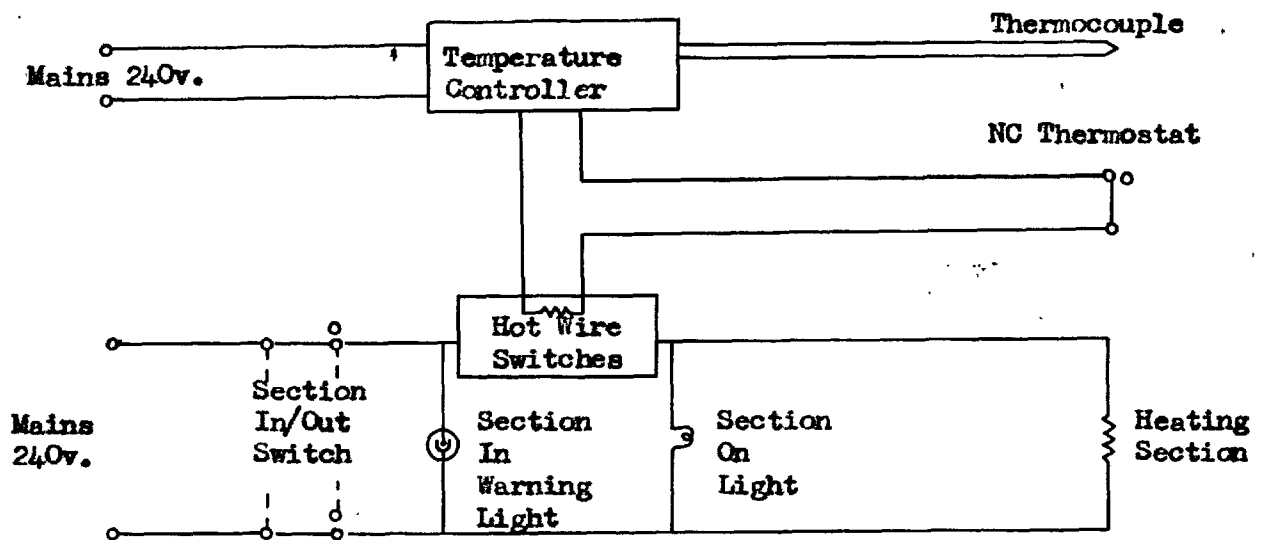


Figure 6 - Heating Circuits
(only one of three heating sections shown)



Plate 1 - Tank and Support Frame, with Lid in Position

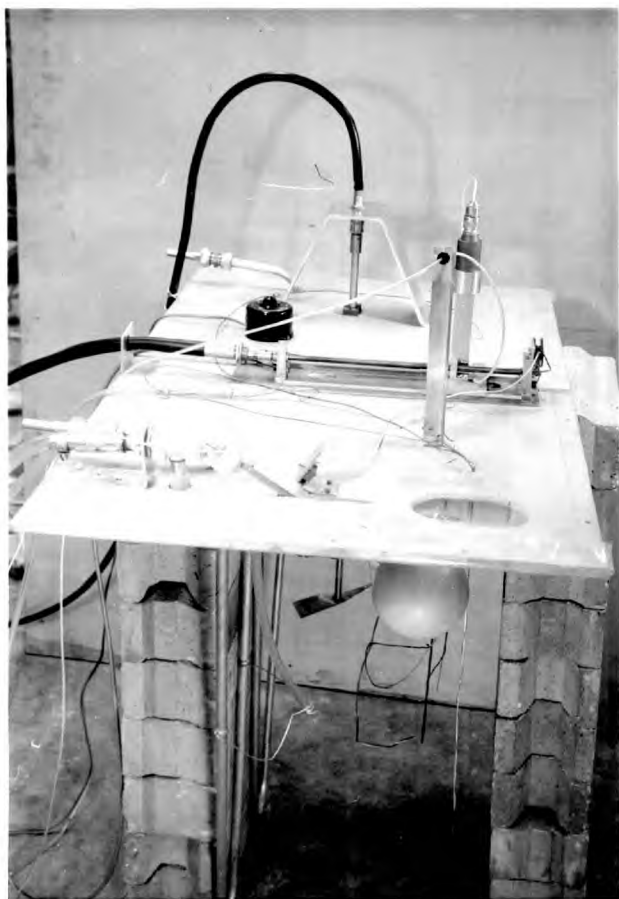


Plate 2

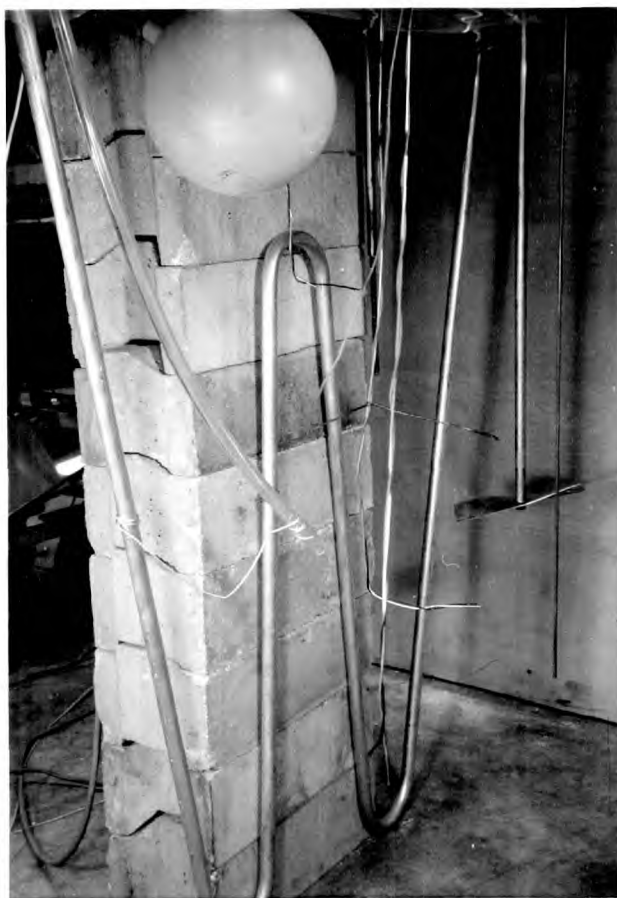


Plate 3

Two views of lid (temporarily supported on concrete blocks) showing fission chamber, stirrer, cooling coil and thermocouples.



Plate 4 - View down into cave with tank removed, showing shutter in open position. Graphite extension stack and monitor counter may be seen through the window.

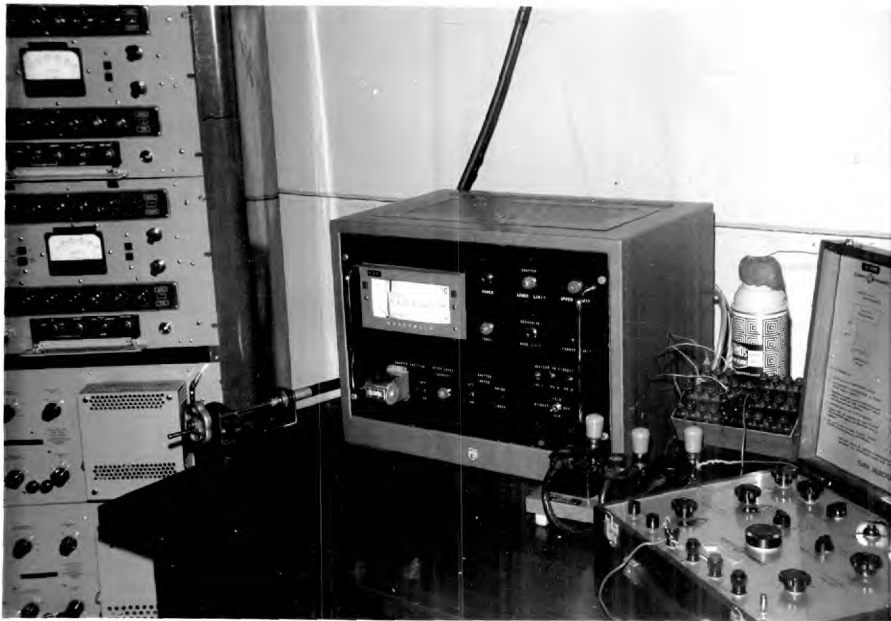


Plate 5 - Control Desk.

the tank it was necessary to provide a cadmium shutter which could be operated by remote control from outside the cave. A framework 7'4" high x 3'6" wide was therefore constructed from Speedframe, to the sides, top and bottom of which were fixed sheets of .020" cadmium sandwiched between $\frac{1}{16}$ " aluminium sheets, to form a screen in such a manner that only a window 37 $\frac{1}{2}$ " square in line with the thermal column and extension stack remained clear. A shutter 40" square was constructed from a similar cadmium-aluminium "sandwich", which could be raised to completely close the window, and lowered to open it. The shutter was guided between two lengths of aluminium angle on either side secured to the uprights of the framework. Because the lower edge of the access face (and therefore also of the window) was only 33" above floor level, however, the lower 12" of the shutter consisted only of cadmium without the aluminium on either side of it, and in the fully open position the lower 9" of shutter were guided in a smooth curve into a horizontal position (see Figure 4, in which the shutter is shown in the open position). In fact after the boric acid runs, for reasons which will be discussed later, most of the aluminium was removed from the shutter, leaving only a few inches at the top and sides to support the cadmium.

The shutter was supported by a steel wire attached to its top, which passed over an aluminium pulley fixed to the

centre of the upper cross-member of the frame, and from there to the spindle of a reversible motor situated on the floor behind (i.e. further from the core) the shutter. This motor was of the single phase permanent capacitor type (Fractional H.P. Motors Ltd.), and operating at 10 r.p.m. could raise or lower the shutter in one minute. Micro-switches were provided at the upper and lower limit positions of the shutter, and actuated by a bracket fixed to one side of the shutter. The actuation of a microswitch produced two effects: first, a light came on at the control panel indicating that the shutter had reached the desired position, and second, the supply to the motor was automatically cut off and could only be restored by reversing the direction of the motor. The wiring diagram for the motor control is shown in Figure 5.

In order to monitor possible fluctuations in reactor power, a BF_3 counter (20th Century Electronics Ltd., type 5EB40/13) was fixed to the framework of the shutter unit. A position a few inches from one corner of the extension stack was found to give suitable count rates.

The third unit consisted of a tank welded from $1/4"$ aluminium plate, alloy type S1C, and 38" wide x 15" long x 40" deep, which contained the solution to a depth of $37\frac{1}{2}"$. This size of tank was chosen so as to match the dimensions of the thermal column, and so that uncertainties in boundary effects would be negligible. At the top of

the tank the plate was bent out on all sides to form a 4" flange, to which was bolted a $\frac{1}{2}$ " thick Sindanyo lid. ("Sindanyo" is a hard asbestos-type material manufactured by Messrs. Bingley, Son and Follitt.) The underside of the lid was covered with polythene.

The neutron detector was a U^{235} fission chamber (20th Century Electronics Ltd., type FC16) of $\frac{1}{8}$ " diameter, having an active length of 20 cm. This was suspended vertically from a hollow aluminium block above the lid, 1" square cross-section x $4\frac{5}{8}$ " high, through which the counter passed, being located by rubber washers at the top and bottom of the block. Below the block, the counter passed through a slot in the lid and into the tank, its length being such that the mid-point of the active region was at the mid height of the solution. To avoid earth loops, the top of the counter was insulated from the block by a layer of polythene. The lead from the top of the fission chamber was passed through a clamp on the platform and from there through a fixed piece of aluminium angle, in order that it should not tend to tilt the fission chamber in any position.

The block was mounted on a travelling aluminium platform, which sat on parallel flat and inverted 'V' runners. The runners, which were covered with thin strips of polytetrafluoroethylene, were fixed to a $\frac{1}{8}$ " aluminium base-

plate which was screwed on to the lid. The block and platform unit also contained a fixed aluminium nut, through which passed a 14" length of $\frac{3}{8}$ " B.S.F. studding. By rotating this screw (which was located at the back end of the tank and was free running at the front) the block and platform, carrying the fission chamber, could be caused to move along the runners in a direction radial to the core. A flexible drive (S.S.White Dental Manufacturing Co., Ltd., type EX 876), 30' long, was connected to the end of the screw at the back of the tank, and passed out of the cave to the control desk, from where it could be turned by means of a handle.

Accurate positioning of the fission chamber was effected by means of an indexing microswitch which was actuated by a cam on the screw. The operation of this microswitch caused a light on the control panel to go on (see Figure 5), and also operated an electromagnetic digital counter which displayed the counter position relative to some datum in units of the screw pitch. Because of the backlash of the flexible drive, when the counter was moved to a new position (which was always approached from the forward, or core, side) the criterion was adopted that the index light, having just gone on, should remain so when the handle was released. A trip switch was therefore provided for the digital counter, so that while this adjustment was being made, which sometimes required several attempts, it

should not continue counting, thereby registering a false position. It was considered that reproducibility with this method was to within about 5° of rotation of the screw, which since the pitch was .127 cm. represents an uncertainty in the position of the fission chamber of .002 cm. Microswitches were also provided to indicate by lights on the control panel when the forward and back limits of the mechanism movement had been reached.

A stirrer was provided, which consisted of an aluminium propeller dipping into the solution to a depth of 16", and located by a Tufnol bush on the lid and an aluminium bridge 7" above it. Positioned well to the side and rear of the tank, it could have only a negligible effect on the measured flux. It was connected by a short length of flexible drive to a motor underneath the tank, of the same type as that used for the shutter, but operating at 400 r.p.m. The direction of rotation could be controlled from the control panel.

The rear, sides and base of the tank were covered with heating blankets (Isopad Ltd., type IP-3, heating density 300 watts/ft²). These heaters were wired in three sections: two 2.5 kW sections for the rear and sides, and one 900 watt section for the base. The power for each section was drawn from a 13 amp single phase point in the reactor hall. Each section could be switched out from the control desk, but they were all under the

control of a Honeywell "Servotronic" 0-200°C visual display temperature controller. This was actuated by a chromel-alumel thermocouple leading from inside the tank, and controlled three "Sunvic" hot wire vacuum switches, which only closed the supply circuits to each of the heating sections when the temperature as measured by the thermocouple was below that set on the controller. The circuit between the controller and the vacuum switches was passed through a normally closed safety thermostat in the tank, which was set to operate at 78°C. The heating circuits are shown in Figure 6.

Four further chromel-alumel thermocouples were distributed at various points in the tank and led out of the cave to the control desk where they were connected through a double-pole switch to an ice junction and a potentiometer (W.G.Pye, Ltd.). The junctions were spot-welded and the parts submerged in the tank coated with Araldite. The thermocouples entered the tank through small holes drilled in the lid, and were twisted around aluminium wire to keep them in position. The thermocouple nearest to the fission chamber approached the plane of movement of the chamber from a perpendicular (sideways) direction, and came no closer than 4" from the topmost part of the active region, thus causing no perturbation.

1" thick fibreglass insulation panels (Newalls Insulation Ltd.) were strapped outside the heating blankets

on the sides and rear face of the tank; the front face (which was not heated) was insulated by creating an air gap between the tank face and a $1/16$ " aluminium sheet held close to it but prevented from touching it by several small pieces of Tufnol glued on to the face. Underneath the base heating blanket was a $1/2$ " thick Sindanyo sheet. The weight of the tank was taken by 1 " wide x $1/4$ " thick Sindanyo strips on top of this sheet around its perimeter. The sheet was bolted to a Speedframe frame the sides of which rested on 8 " high concrete blocks on the floor of the cave.

In early laboratory tests the insulation described was found to be so effective (e.g. the temperature fell by $.6^{\circ}\text{C}/\text{hour}$ around 40°C) that the need for a means of cooling the water in the tank became evident. A $5/8$ " aluminium tube (wall thickness $.075$ "), shaped in the form of a W, through which cold water could be passed, was therefore passed through the lid into the tank. The length of tube submerged in the solution was about $8'$; the 'W' was in a plane very close to the back wall of the tank, where it could not cause any perturbation in the measured neutron flux.

It was found that the temperature could be controlled much more accurately by interrogating the four thermocouples with the potentiometer, and then either switching in one or more sections of heaters (by changing the set

temperature on the controller) or turning on the cooling water, as appropriate, than by using the controller directly. In this way the temperature in the tank was controlled to better than $1/4^{\circ}\text{C}$, and by frequent use of the stirrer the differences in temperature between any two thermocouples was always less than this figure.

Around the back and sides of the tank, outside the insulation panels, and also underneath the base Sindanyo sheet, cadmium sheets .020" thick were strapped. It was not possible to position the cadmium inside the insulation on account of its low melting point (321°C) - the temperature of the heating blankets might have reached this figure. However, the error introduced by the resulting uncertainty in extrapolation distance is negligible - from simple diffusion theory (in the highly poisoned cases where diffusion theory may not be valid leakage corrections are negligible anyway) the spatial eigenvalue κ is related to the inverse relaxation length γ by

$$\kappa^2 = \gamma^2 - 2\left(\frac{\pi}{a}\right)^2 \quad 5.1.1$$

where a is the extrapolated dimension in both vertical and horizontal directions (assumed equal). An error of $1\frac{1}{2}\%$ in a , which represents the maximum uncertainty, gives rise therefore to an error of .05% in κ^2 for the worst case (that of pure water). This is negligible.

The tank was filled with demineralised water through a plastic ball-valve of familiar domestic design secured to the lid. A lever microswitch was positioned in such a way that it was actuated by the ball arm when the water level in the tank had reached its correct height, and a light indicating this went on at the control panel.

A drip tray was provided underneath the tank, with an outlet to the drainage channel, in case any of the possibly active solution should leak from the tank. A 2 mm thick lead curtain was suspended from the back of the tank, to provide a shield against possible soft γ -ray emission from the solution if access had to be gained to the back of the cave soon after the reactor had shut down.

The pulses from the fission chamber and BF_3 counter were fed into parallel counting channels. Each channel contained a Dynatron 1430A head amplifier and linear pulse amplifier, and an Ekco N530G discriminator/scaler-timer unit, from which the E.H.T. supplies were also taken. Both head amplifiers were inside the cave; the remainder of the counting equipment was mounted in a rack alongside the control desk. The mains supplies for both amplifiers were taken from an Advance Component constant voltage transformer and two MacMichael low pass filters; supplies to the Ekco units were directly from mains.

Both counters are essentially $1/v$ detectors at thermal neutron energies, although the fission chamber has a slight resonance at .3 eV.

5.2 Dead Time of Counters

As will be seen later in this chapter, the difference technique employed necessitated high counting rates in order to obtain low statistical errors for each point, while completing a run in a reasonable time. It was therefore necessary to know the dead time of each counting system, so that a correction for this could be applied. Accordingly, a series of preliminary experiments was conducted in which counting rates were measured with the fission chamber at five different positions in the tank, for each of seven reactor powers ranging from 1 kW to 100 kW. The monitor counting rates were also recorded at the same time. Assuming that the reactor power is proportional to the true counting rate, we may write

$$c_t = Ap \quad 5.2.1$$

$$\text{and} \quad c_o = \frac{Ap}{1+Ap\tau} \quad 5.2.2$$

where p is the reactor power in kW

A is constant for a particular position of the fission chamber

τ is the dead time of the counter

c_t is the true counting rate

and c_o is the observed counting rate.

A FORTRAN IV programme MORFIT was written by the present author incorporating the subroutine PARFIT (Appendix 2) to fit the experimental values of c_o against the

reactor power p by optimizing the values of A and τ .

The results for three of the fission chamber positions (characterized by the maximum observed counts per second at 100 kW) and for the BF_3 counter are given in the following table:

Counter	Max c_0 (sec. ⁻¹)	τ (μsec)
F.C.	42000	5.87 ± 0.05
F.C.	17000	5.90 ± 0.16
F.C.	6000	7.00 ± 1.13
BF_3	23000	8.14 ± 0.08

For the pure water and boric acid series, therefore, the value $5.87 \mu\text{sec.}$ was adopted for the dead time of the fission chamber. It was decided to keep the counting rate of the fission chamber in the main experiment to less than 5000 counts/sec., since at this figure the dead time correction is 3%, and any uncertainties in dead time produce negligible errors. To do this it was necessary to use two or three reactor powers for each of the lower poison concentration runs, the power being increased in stages when the fission chamber had reached such a position that the limit of 5000 counts/sec. imposed would not be exceeded.

In order to take into account fluctuations in reactor power each fission chamber counting rate measured was nor-

malized by dividing by the monitor counting rate taken over the same period. After each power change, the last point measured at the old power was repeated, and the ratio of the counting rates of the fission chamber at the old and new powers (each of these measurements being made to a statistical accuracy of .1%) used to normalize the observed points at the new power. This normalization was effected by assuming the value 8.14 μ sec. for the monitor dead time at the lowest reactor power, and deducing an effective monitor dead time for each successive power on the basis of the fission chamber ratio mentioned above. The monitor dead time at the new power is given by

$$\tau_2 = \frac{c_{t2} - c_{o2}}{c_{t2} c_{o2}} \quad 5.2.3$$

$$\text{where } c_{t2} = R c_{t1} \quad 5.2.4$$

where c_{t1} , c_{t2} are the true monitor counting rates at the first and second powers respectively, R is the fission chamber ratio of true counting rates at new/old powers, and c_{o2} is the monitor observed counting rate at the second power.

A short FORTRAN IV program MOR3 was written by the present author to compute effective monitor dead times for each power change. They lay typically within the range 8-10 μ sec. The only way in which the artificiality of these computed dead times could affect the results is in

the magnitude of the power fluctuations, but since these were normally less than 1%, the error here is negligible.

This procedure gave good results but as well as being cumbersome it suffered from three disadvantages:

- (i) in order to achieve statistical accuracies of .1% it was necessary to count over rather long periods for the point immediately before a power change,
- (ii) even with these accuracies there still remained an error of .2% in the normalization, and
- (iii) changes in reactor power would be inconvenient for other users of the reactor.

For these reasons it was decided to adopt a new procedure for the cadmium and gadolinium runs. The dead time experiments were repeated with greater accuracy. Eight reactor powers between 1 kW and 100 kW were used, and measurements made for six positions of the fission chamber in the tank at each power. For the two positions of highest counting rate the statistical ^{error} accuracy at all powers was less than .3%. These results were input to the program MORFIT and on the basis of the fitted dead time for each set of observations an estimate of the relative reactor power was calculated. These estimates showed systematic variations of up to 1% from the nominal reactor powers. MORFIT was then modified so that for each position of the fission chamber the following procedure

was adopted: mean values for the relative powers obtained from the above estimates were used as the abscissae (p in equations 5.2.1 and 5.2.2) for the appropriate set of monitor observations, and from the fitted dead time better estimates of the true relative reactor powers obtained. These were then used as abscissae for the fission chamber results, and the dead time for this calculated. This fit was repeated dropping first one and then two points from the high counting rate end. The procedure was then repeated for each of the six fission chamber positions.

The observations for a typical monitor set and for the highest counting rate position of the fission chamber are shown in Figure 7, where the excellence of the fit can be seen. The results for the two highest counting rate positions of the fission chamber were

Position	Max c_o (sec^{-1})	τ (μsec)
1	36000	$5.86 \pm .07$
1	27000	$6.09 \pm .04$
1	17000	$6.12 \pm .09$
2	25000	$5.86 \pm .20$
2	18000	$6.40 \pm .19$
2	11000	$7.10 \pm .25$

and the best monitor fit gave $\tau = 8.955 \pm .020 \mu\text{sec}$.

The value adopted for the fission chamber was $6.00 \mu\text{sec}$.

With these more accurate results, the cadmium and gadolinium runs were performed with the reactor at full power (100 kW), the highest fission chamber counting rate measured being 40000 counts/sec., for which the dead time correction was 25%. However, after this correction had been applied, such points were found to lie within .3% of the fitted exponential.

5.3 Other Preliminary Experiments

The thermocouples used were chromel-alumel type T1-T2 (Honeywell Ltd.). A set of maker's calibration figures were available, and these were checked at 20° and 62.3°C by heating the actual thermocouples used in an oil bath with a standard thermometer. At 20°C exact agreement was obtained with the tabulated figure (.80 mV; the cold junction being in ice) for all four thermocouples; at 62.3°C, where the value interpolated from the tables was 2.54 mV, the readings of the four thermocouples, corrected for the cooling of the oil bath during the three minutes required for making the measurements, were 2.54, 2.55, 2.55 and 2.54 mV. Since the oil bath was not lagged, and the temperature fell by 1°C (.04 mV) during the measurements, these results were taken to confirm the tabulated values, which were therefore used for all four thermocouples.

To check whether the Sindanyo used for the lid might

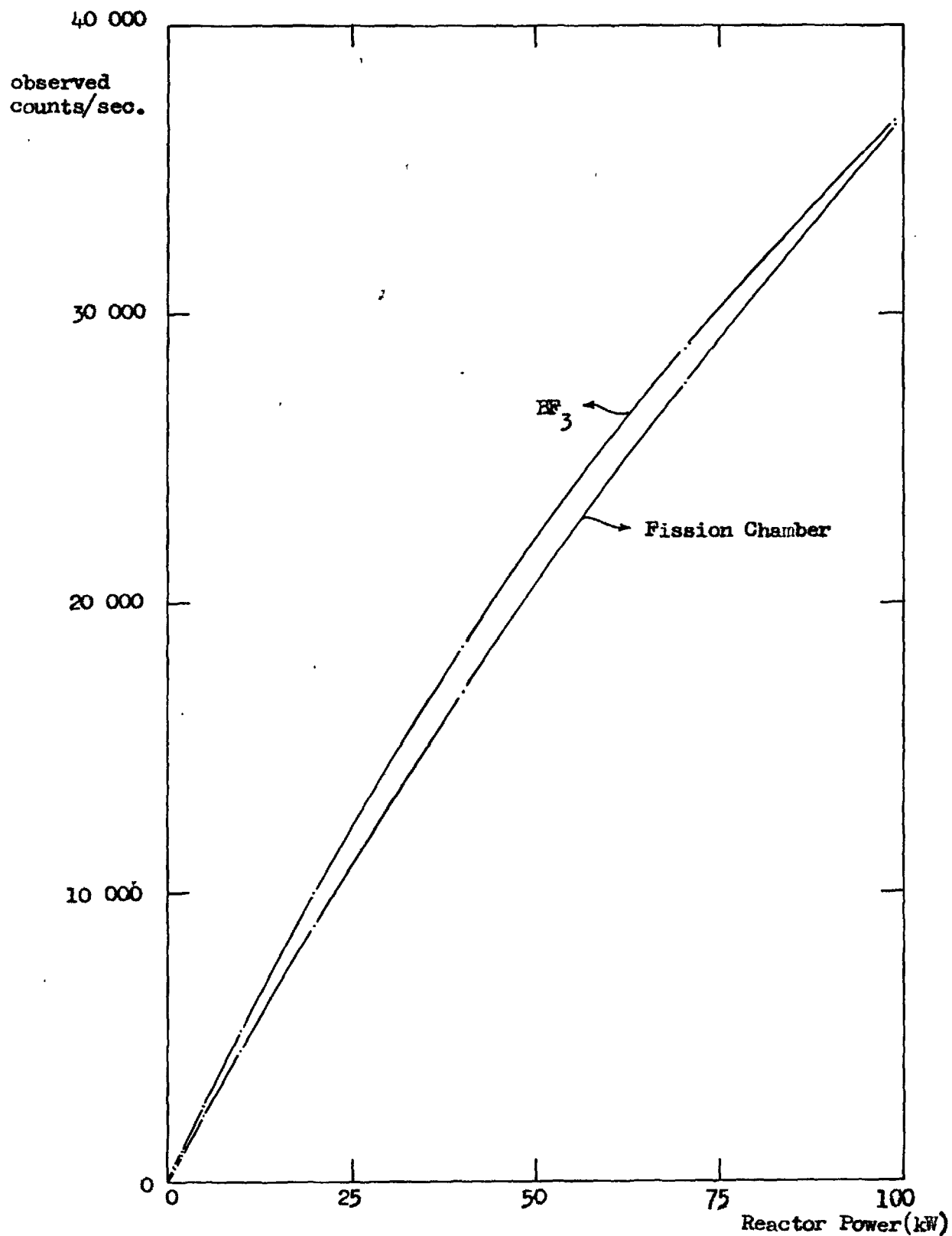


Figure 7 - Determination of Counter Dead Times
(observed points shown on fitted lines)

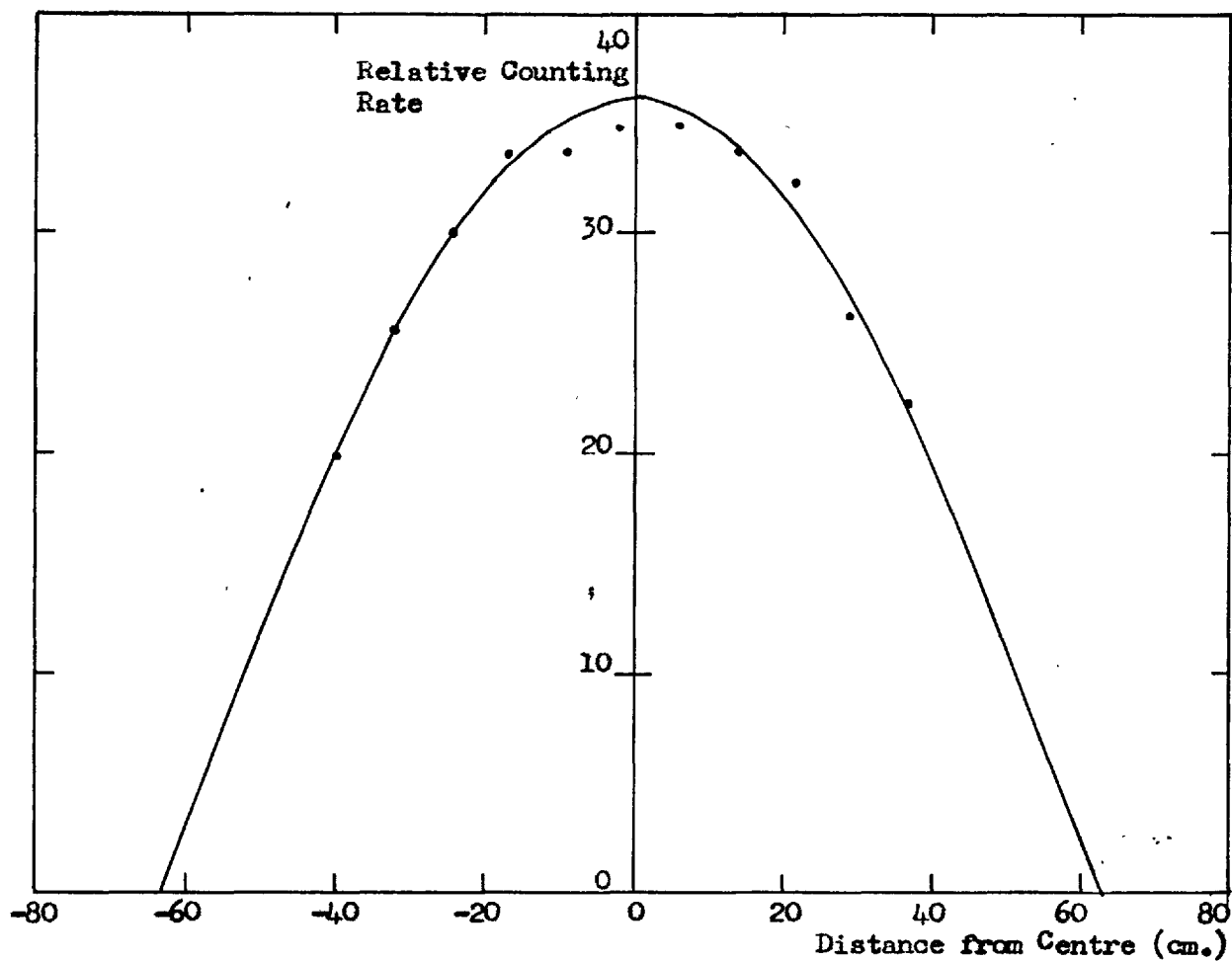


Figure 8 - Flux Distribution in Vertical Plane
across Tank Face

(observed points ($\pm 1\%$) shown on fitted cosine)

have buckled owing to the effects of heating and moisture, thereby causing the base plate of the mechanism to buckle also, a 12" steel ruler was placed with its edge on the base plate, parallel to the screw. Over 10" the gap between the edge of the ruler and the base plate was less than $1/4$ mm. at the centre. No dip at all was detected in the runners. These measurements were first made before the apparatus was installed at the reactor, and repeated both after the boric acid runs, and after the final gadolinium runs, with similar results.

In order to test the harmonic content of the neutron flux entering the tank, a sheet of polythene was suspended along the front face of the tank, and fixed to it were 25 gold foils .002" thick and $7/32$ " diameter, arranged at intervals in the form of a cross. The foils were irradiated with the shutter open and counted in a Geiger-Müller system, corrections being applied for weight fluctuations of individual foils, and also for the gold decay (decay constant = $.0107 \text{ hour}^{-1}$). The relative counting rates in the vertical plane are shown in Figure 8, together with a cosine fitted to the points by the program COSFIT, written by the present author and incorporating the subroutine PARFIT (Appendix 2). It can be seen that the points show little evidence of harmonic contributions, the discrepancies between the points and the fitted curve being attributed to creasing of the polythene sheet, since

its positioning from above with the tank very close to the shutter was very awkward. The extrapolated dimensions obtained were 126 cm. for the vertical and 154 cm. for the horizontal planes. These are larger than the dimensions of the stack, window and solution (95 cm. square), and the reason for this is the 7" air gap between the window and the tank face, which results from the tank flange having to clear the shutter wire. A more useful experiment would have been to determine the lateral distributions inside the tank: this would show how the cadmium at the sides and base of the tank influences the extrapolated dimensions of the flux within it. However, since the buckling only alters the value of κ^2 by a maximum of 1.7%, this more tedious experiment was not considered worthwhile.

5.4 Conduct of Experiments

The principle of the experiment is a negative source technique, whereby the neutron flux which is expected to decay exponentially with distance is given by the difference between the counting rates at any position with the shutter open and closed. This is the flux which would be measured directly due to a source of neutrons at the shutter, with a strength at any point equal to the rate at which neutrons are absorbed at that point in the present experiment. By using a material for the shutter such as cadmium, which

has a very high absorption cross-section in the thermal energy range compared with that for fast neutrons, the effect of the latter should be almost entirely eliminated.

The method has the added advantage that neutrons entering the tank from any direction other than through the window are automatically subtracted out. It is clearly advantageous in the elimination of any spurious possibly time-dependent interference to take counts with the shutter open and closed at each point in turn, rather than do a complete traverse with it open followed by a traverse with it closed. This was possible in the present case owing to the short time (one minute) taken for the shutter position to be changed.

The experiments were therefore conducted in the following manner. While the reactor was being brought to power, the temperature of the tank was stabilised with the stirrer on. When the reactor power was steady the stirrer was switched off, and counting started with the fission chamber in the position nearest to the front face of the tank, and the shutter open. When this count was completed the shutter was lowered; while this was taking place the stirrer was switched on again, and also, if necessary, the heaters. The stirrer and heaters were switched off at least 15 seconds before counting started, in order that conditions in the tank should be stable. At the end of the shutter closed count the

fission chamber was moved further from the front face by the method described in Section 5.1. The readings of the scalers were then recorded; the operations were conducted in this order so that any vibrations of the fission chamber after moving it to a new position would be completely damped out before counting began. In this second position counts were recorded with the shutter first closed and then open; in this way the shutter had only to be moved once for each position of the counter. The procedure was repeated for about 20 such positions. Where time allowed, a previous point was repeated to check reproducibility.

The minimum counting time was 100 seconds; this was increased if necessary so that at least 150000 counts were recorded for the fission chamber with the shutter open, the aim being that the difference counting rate at each position should have statistical standard deviation of less than $\frac{1}{2}\%$.

The procedure for controlling the temperature of the tank was described in Section 5.1; it was found that a short burst from the heaters while the shutter was in motion was sufficient to compensate for heat losses in the 65°C runs; for the lower temperature runs only very rare heating was necessary.

For each concentration of the boric acid solutions, a sample was taken immediately before the low temperature

run, and further samples taken immediately before and after the 65°C run. For the non- $1/v$ poisons, for which only 22.3°C runs were performed, two samples were taken for each concentration either before or after the run. The poison concentration was increased by adding crystals of the poison through a funnel placed temporarily in the slot of the lid. Samples were also taken through this slot using a 4' long pipette with a rubber bulb attached to its top. The water level occasionally needed to be adjusted to compensate for evaporation losses and the extraction of samples. This was done at the same time as poison was added, by passing demineralized water through the ball valve until the indicator light on the control panel showed the water level to be correct.

In every case the tank was stirred for at least one hour between adding poison and taking a sample or starting a flux traverse.

5.5 Analysis of Experiments

The raw data were fed directly into a FORTRAN IV program PERSE, written by the present author. The observations were first corrected for dead time, provision being made for up to three different reactor powers with different monitor dead times, and the standard deviation of each observation calculated. The monitor counting rate with the shutter closed was about 30% lower than with it

open owing to the extinction of neutrons reflected back from the tank in the former position. The ratio of the average monitor counting rate for shutter open/closed was therefore calculated for each region of constant power, and the shutter closed monitor data increased by this factor. The fission chamber measurements were then corrected for power fluctuations by dividing by the appropriate individual monitor counting rate and multiplying by the average monitor counting rate. The difference flux at each position was then formed by subtracting the shutter closed from the shutter open corrected fission chamber counting rates, and the standard deviations also computed. The subroutine PARFIT (Appendix 2) was then used to fit the difference flux to the expression

$$\varphi(z) = Ae^{-\gamma z} \quad 5.5.1$$

where z is the distance of each counting position from an arbitrary zero, by optimizing A and γ . No correction for the tank being finite in the z direction was necessary; the furthest point from the front face at which measurements were made was at 20 cms., and the end correction here is less than .001%. The diffusion length was finally calculated from equation 5.1.1, using the value for the buckling fed into the program as data (.0023). Provision was made for the fit to be repeated with points successively omitted from either or both ends.

Two runs were performed with pure water at 22.3°C , one before and one after the pure water 65°C run, to check the reproducibility of the equipment. Figure 9 shows the corrected shutter open and closed fission chamber counting rates, as well as the difference, for the second of these runs. It can be seen that the fast flux is about 5% of the total flux near to the front face of the tank (the first position at which measurements were taken was about $1/4$ " from the inside of the front face). Figure 9 also shows that the flux due to the fast neutrons decays much less rapidly than does the difference flux - this is expected since the slowing down length for fission neutrons in water is about double the thermal diffusion length.

It was noticed that at distances of 15-20 cm. from the front face (for pure water at 22.3°C), the difference flux showed a tendency to be greater than would be obtained from a pure exponential decay. This was attributed to fast neutrons being absorbed by the shutter. In order to explain the departure from a pure exponential it was necessary to increase the shutter closed flux by about 5% for distances greater than 15 cm.; from the known values of the total cross-sections of cadmium and aluminium the shutter was calculated to remove about 7% of the neutrons in the MeV range, in good agreement with this figure. However, increasing the shutter closed flux by either 2

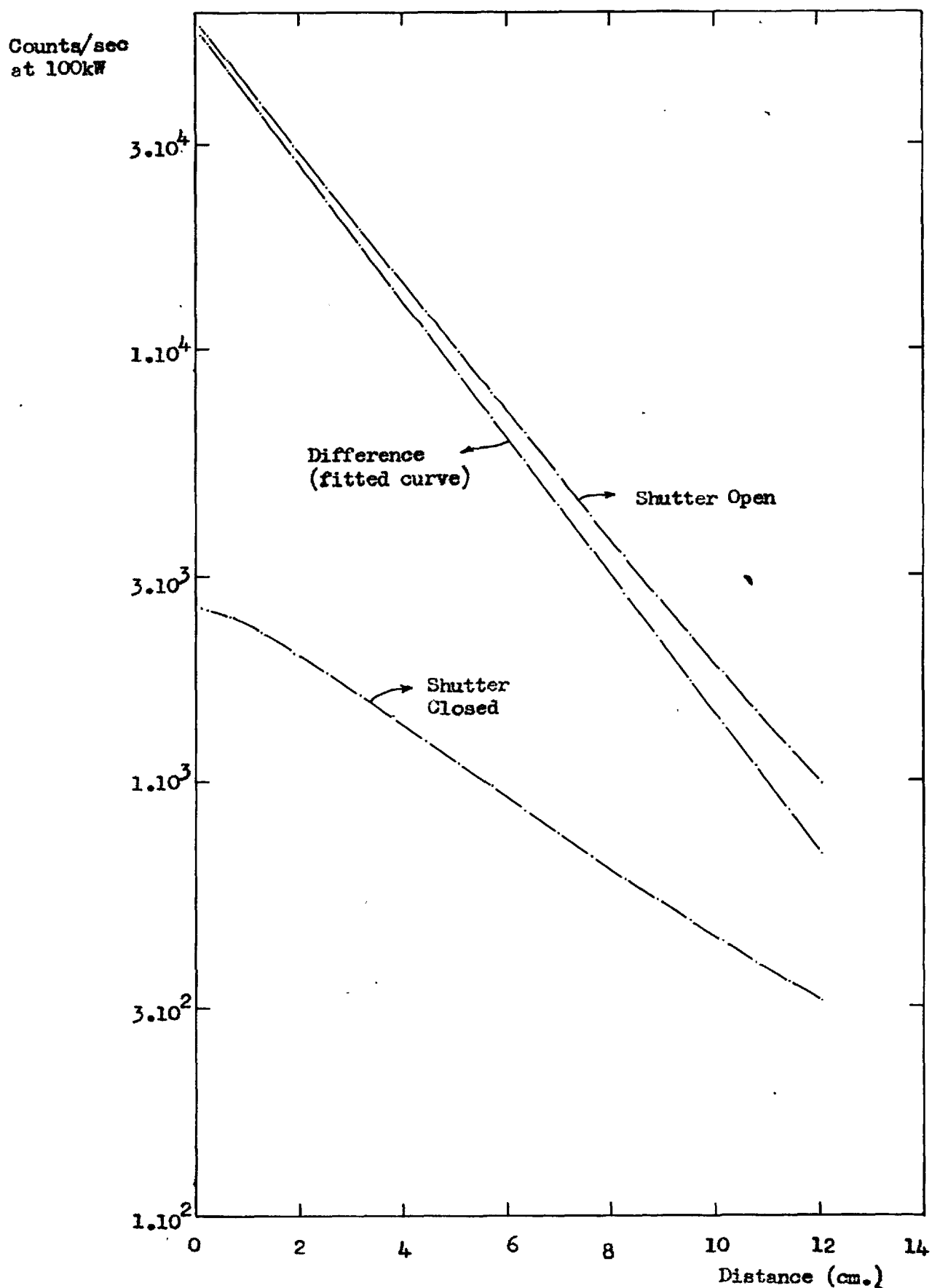


Figure 9 - Measured and Difference Fluxes in Pure Water at 22.3°C

Counts/sec
at 100 kW

$3 \cdot 10^4$

$1 \cdot 10^4$

$3 \cdot 10^3$

$1 \cdot 10^3$

$3 \cdot 10^2$

$1 \cdot 10^2$

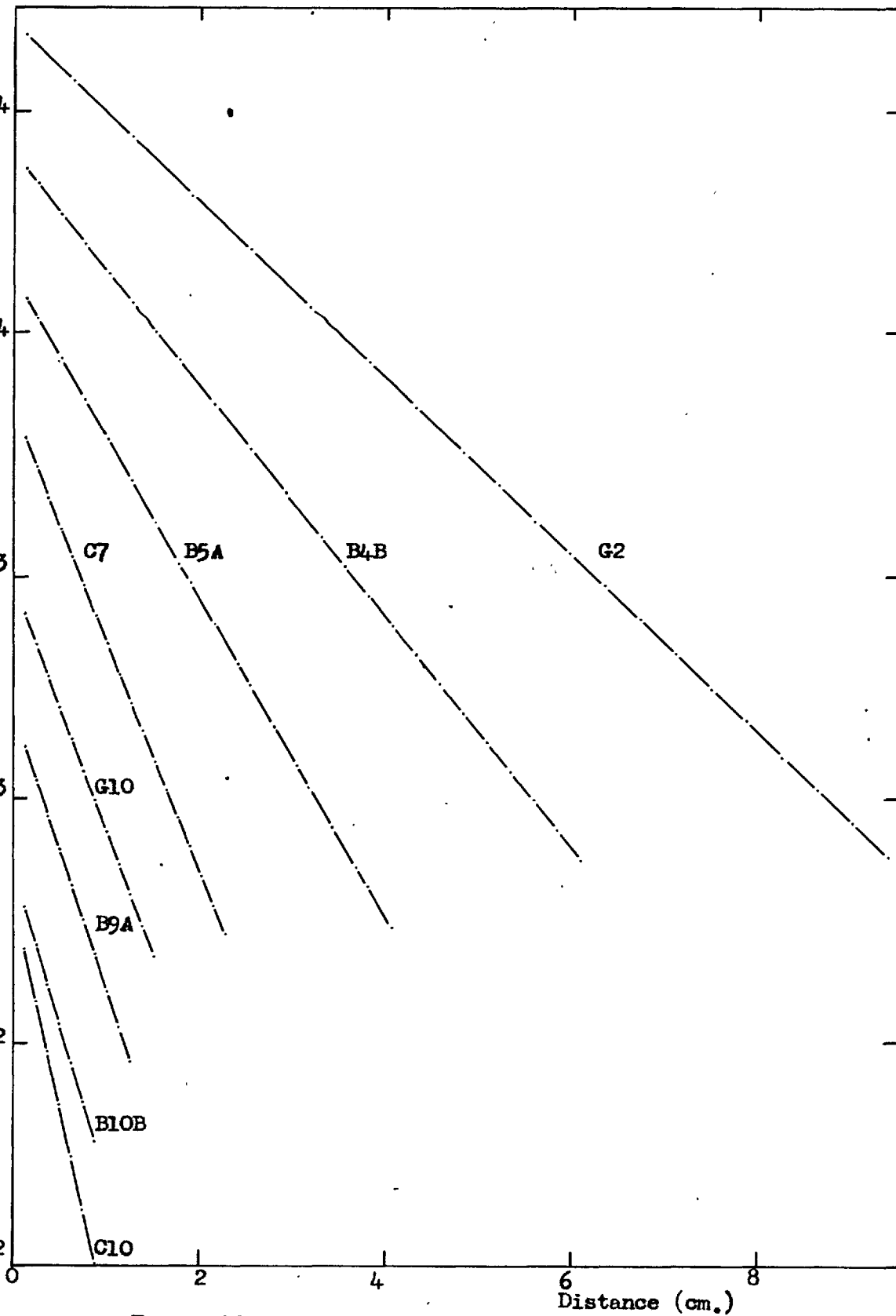


Figure 10 - Typical Difference Flux Plots

or 5% was found to give worse fits for points in the 0-12 cm. range, although the diffusion length was only changed by a maximum of .4% for the pure water case. It was therefore concluded that the effect was primarily due to neutrons of MeV energies. The criterion was adopted that all points whose shutter closed flux was more than one-third of the shutter open flux should be eliminated. No tendency was then found for the points at the end of a fit to deviate from an exponential. Since the effect of the aluminium in the shutter was calculated to be about twice that of the cadmium, after the boric acid runs most of the aluminium was removed from the shutter, leaving only a few inches around the top and sides to support the cadmium.

In all cases, the points at the high flux end of a traverse lay close to the fitted exponential; no evidence of any boundary effects was found. For the cadmium and gadolinium runs, to prevent these points from being unduly weighted in the least squares fit (100 second counts were taken and the counting rate was very high, PERSE was modified to add an error due to the dead time uncertainty (taken as 1%) to the error from counting statistics.

Figure 10 shows some typical difference flux plots. The runs are characterized by a letter, which is the initial letter of boron, cadmium or gadolinium, and a number

referring to the particular concentration. The final A or B in the boron runs signifies low and high temperature respectively. Included in Figure 10 are the plots for the most concentrated solutions of each series.

The results will be given in detail in Section 5.8.

5.6 Composition of Solutions

All of the samples were analysed by the Analytical Services Laboratory of Imperial College. The results will be tabulated in Section 5.8.

Boric acid was determined by converting it to a strong acid with mannitol, and then titrating against sodium hydroxide. The accuracy quoted for the first eight solutions was .5%, and for solutions 9 and 10, 1%. The spread of the concentrations of different samples of nominally the same solution was about 1% for the first seven solutions, a barely significant increase in concentration for the higher temperature run due to evaporation losses being detected. The values adopted were the concentration of the first sample for the lower temperature run, and the mean of the second and third for the 65°C run. For the eighth solution there was an inexplicably large scatter: the three samples yielded 32.2, 35.0 and 33.4 gm./litre of H_3BO_3 respectively. These were averaged to give 33.5 ± 1.0 gm./litre for both runs. For the ninth solution four samples were

taken since the concentration of this solution was very close to the solubility limit for boric acid at room temperature. The values obtained were 45.6, 46.4, 47.7 and 45.8 gm./litre; these were averaged to give 46.4 ± 0.5 gm./litre. The tenth solution exceeded the solubility at room temperature and only a 65°C run was performed. Samples taken before and after the run gave 71.0 and 67.6 gm./litre respectively; the average value 69.3 ± 1.4 gm./litre was adopted.

Cadmium was determined by titration against EDTA, using Erichrome Black T as indicator. There were no solubility problems with cadmium sulphate and the titrations were performed to an accuracy of .2%. The mean of the values obtained for the two samples taken for each solution (which never differed by more than .5%) was adopted.

Gadolinium was determined by the same method as was used for cadmium. Most of the titrations were again to an accuracy of .2%, the earlier solutions being less accurate on account of their very low concentrations.

For the boric acid and cadmium sulphate solutions the effect of reducing the number density of water molecules by the addition of solute was considered. No data for boric acid were available, and so the specific gravity of three boric acid solutions made up in 100 ml. flasks

was measured. The results were

$\% \text{H}_3\text{BO}_3$ (gm./100 ml. of solution)	Specific Gravity $\frac{23}{4}^\circ \text{C}$
1.7190	1.00433
3.0407	1.00921
4.2905	1.01333

These data show that for solution 9, the number density of water molecules has been decreased by about 3%.

For cadmium sulphate data were taken from Hodgman (1957). In this case the effect was much less; for solution 7 (the worst case) there was only a reduction of .18% in the number of water molecules.

The manner in which these corrections were applied will be discussed in the next two chapters.

The choice of the microscopic absorption cross-section for the boron used presented some difficulty. Debus et al (1963) have measured σ_{ao} for several samples of boron; their results suggest that there are two distinct naturally occurring forms of boron containing slightly different properties of B^{10} , the isotope responsible for the high absorption. The cross-section of the first and more common group (ANL-BNL Standard) is 759 barns, while that of the second (Harwell-Fontenay) is 771 barns. While the origin of the Harwell stock is

unknown, the Fontenay sample is thought to originate from Turkey. The suppliers of the boric acid used in the present experiments (British Drug Houses Ltd.) considered that this was almost certainly of Californian origin. Jones (1966) has measured the absorption cross-section of boron known to have been mined in California, and finds it to be 760.1 barns, indicating that it is of the first group mentioned. The accepted value for this group of 759 barns was therefore used for the boron cross-section of the present batch.

5.7 Errors

PARFIT produces an estimate of the standard deviations of the fitted parameters by the method described in Appendix 3. This will include errors due to counting statistics and also random errors in counter positioning. It will not include any systematic error in the screw pitch, which has been estimated at .05% in linear dimensions. (The expansion of the brass screw would be .07% for the 65°C runs if the screw were at the temperature of the water in the tank. However, it is themally insulated by the Sindanyo lid, and so will not reach this temperature. No expansion correction has therefore been applied.) There is also a buckling error which has been estimated at .025% in diffusion length for pure water,

decreasing with addition of poison. The errors quoted in Section 5.8 have been derived from a combination of these sources.

5.8 Results

(i) Pure water.

Two independent runs were performed at 22.3°C . The diffusion lengths obtained were $2.767 \pm .004$ and $2.755 \pm .004$ cm. The value of $2.761 \pm .004$ cm. was therefore adopted.

At 65°C the result was $L = 2.971 \pm .005$ cm.

(ii) Boric acid at 22.3°C

Solution No.	$L(\text{cm.})$	$\text{g/l H}_3\text{BO}_3$	$\Sigma_B(\text{cm}^{-1})$
1	$2.383 \pm .004$	$.996 \pm .005$	.007364
2	$2.139 \pm .003$	$1.99 \pm .01$	.01471
3	$1.866 \pm .002$	$3.56 \pm .02$	.02632
4	$1.621 \pm .002$	$5.68 \pm .03$	.04200
5	$1.269 \pm .002$	$11.40 \pm .06$	.08429
6	$1.094 \pm .002$	$17.10 \pm .08$	.1264
7	$.9738 \pm .0015$	$22.9 \pm .1$	.1693
8	$.8278 \pm .0019$	33.5 ± 1.0	.2477
9	$.7348 \pm .0017$	$46.4 \pm .5$	.3431

Σ_B is the macroscopic absorption cross-section of boron atoms for neutrons with velocity 2200 m/sec.

(iii) Boric acid at 65°C

Solution No.	L(cm.)	g/L H_3BO_3	$\Sigma_B (cm^{-1})$
1	2.563 $\pm$.004	1.000 $\pm$.005	.007266
2	2.305 $\pm$.003	2.00 $\pm$.01	.01453
3	2.014 $\pm$.002	3.59 $\pm$.02	.02609
4	1.754 $\pm$.003	5.69 $\pm$.03	.04135
5	1.351 $\pm$.002	11.50 $\pm$.06	.08357
6	1.170 $\pm$.003	17.20 $\pm$.08	.1250
7	1.0325 $\pm$.0013	22.85 $\pm$.10	.1660
8	.8793 $\pm$.0023	33.5 $\pm$ 1.0	.2434
9	.7755 $\pm$.0020	46.4 $\pm$.5	.3372
10	.6581 $\pm$.0021	69.3 $\pm$ 1.4	.5036

The concentrations are, given in gm./l as determined in the laboratory at 23°C. The column headed Σ_B gives the macroscopic boron cross-section corrected for the reduction in solution density due to the higher temperature.

(iv) Cadmium sulphate at 22.3°C.

Solution No.	L(cm.)	Concn. (g/l)	$N_{Cd} \times 10^{-19} (cm^{-3})$
1	2.408 $\pm$.003	.774 $\pm$.001	.1817
2	2.138 $\pm$.003	1.732 $\pm$.004	.4066
3	1.859 $\pm$.003	3.251 $\pm$.005	.7634
4	1.566 $\pm$.002	5.772 $\pm$.010	1.3554
5	1.234 $\pm$.002	10.90 $\pm$.02	2.559
6	1.0251 $\pm$.0014	17.07 $\pm$.03	4.008
7	.8843 $\pm$.0014	23.72 $\pm$.04	5.571
8	.7165 $\pm$.0019	37.82 $\pm$.08	8.880
9	.5983 $\pm$.0019	55.52 $\pm$.10	13.037
10	.4925 $\pm$.0029	82.15 $\pm$.13	19.29

The third column gives the concentration in g/l of $3\text{CdSO}_4 \cdot 8\text{H}_2\text{O}$.

A small (constant) quantity of boron (8.7 ± 0.9 p.p.m.B) was detected in the cadmium solutions spectrographically and determined by titration using mannitol. It is inconceivable that this amount could have remained behind in the tank after the boric acid runs through incomplete washing; it is thought that boron was adsorbed on to the walls of the tank and subsequently re-dissolved in the cadmium solutions. Similar trouble has been experienced elsewhere (Besant and Goddard, 1966).

(v) Gadolinium nitrate at 22.3°C

Solution No.	L(cm.)	Concn.(g/l)	$N_{\text{Gd}} \times 10^{-19} (\text{cm}^{-3})$
1	2.487 $\pm$.004	.0992 $\pm$.0050	.01324
2	2.301 $\pm$.006	.169 $\pm$.003	.02255
3	2.044 $\pm$.004	.335 $\pm$.001	.04470
4	1.658 $\pm$.002	.725 $\pm$.004	.09674
5	1.458 $\pm$.002	1.062 $\pm$.002	.1417
6	1.279 $\pm$.002	1.546 $\pm$.003	.2063
7	1.0995 $\pm$.004	2.370 $\pm$.005	.3163
8	.9775 $\pm$.0018	3.251 $\pm$.006	.4338
9	.8937 $\pm$.0018	4.155 $\pm$.008	.5544
10	.8222 $\pm$.0015	5.405 $\pm$.010	.7212

The concentration in the third column refers to the hydrated salt $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$. No trace of either boron or cadmium was detected in the gadolinium solutions.

CHAPTER 6

ANALYSIS AND DISCUSSION OF RESULTS - PART I

The scattering cross-sections and spectra computed by the methods described earlier are first presented and discussed. The remainder of this chapter is concerned with the reduction of the data given in Chapter 5 for $1/v$ absorption to familiar diffusion parameters, and the comparison of the results both with the experimental results of other workers, and also with the present theoretical values.

Results for non- $1/v$ absorption will be presented in Chapter 7.

6.1 Theoretical Scattering Results

Results for the differential scattering cross-sections for the three models under consideration have already been given in Figure 2, in which large discrepancies between the models are apparent. The total scattering cross-sections are presented in Figure 11, and compared with experimental points taken from the compilation by Hughes and Harvey (1955). The effect of the Debye-Waller contribution to the Haywood cross-section, which could not be represented in Figure 2, is about 75% at .001 eV, falling to 25% at .06 eV and 5% at .6eV.

It is clear that the free gas model leads to gross inaccuracies; over much of the thermal range the cross-section which it predicts is too small. This is to be expected since the factor of four increase in cross-section at low energies only applies to a proton bound in a heavy molecule. The increase in the free gas cross-section at very low energies is a result of the Doppler effect.

Over much of the region of importance the values predicted by the Nelkin and Haywood models are lower and higher, respectively, than the experimental values. As would be expected, the discrepancy is most noticeable at the energies of the rotational modes, where the main difference between the models lies. Rather surprisingly the much cruder treatment of the Nelkin model gives marginally better results; however, shifting the frequency ω_1 of the Nelkin rotational delta-function from .06 eV ($\beta = 2.38$ at 22°C) to .07 eV ($\beta = 2.77$) increases the cross-section at .03 eV from 99.7 to 104.0 barns (the Haywood cross-section at this energy is 109.0 barns). Reference to the Haywood (Figure 1) and Larsson and Dahlborg (1962) derived frequency distributions indicates that even a wider choice of ω_1 would be equally plausible.

The discrepancies between the Nelkin and Haywood models appear more marked when the influence of anisotropic

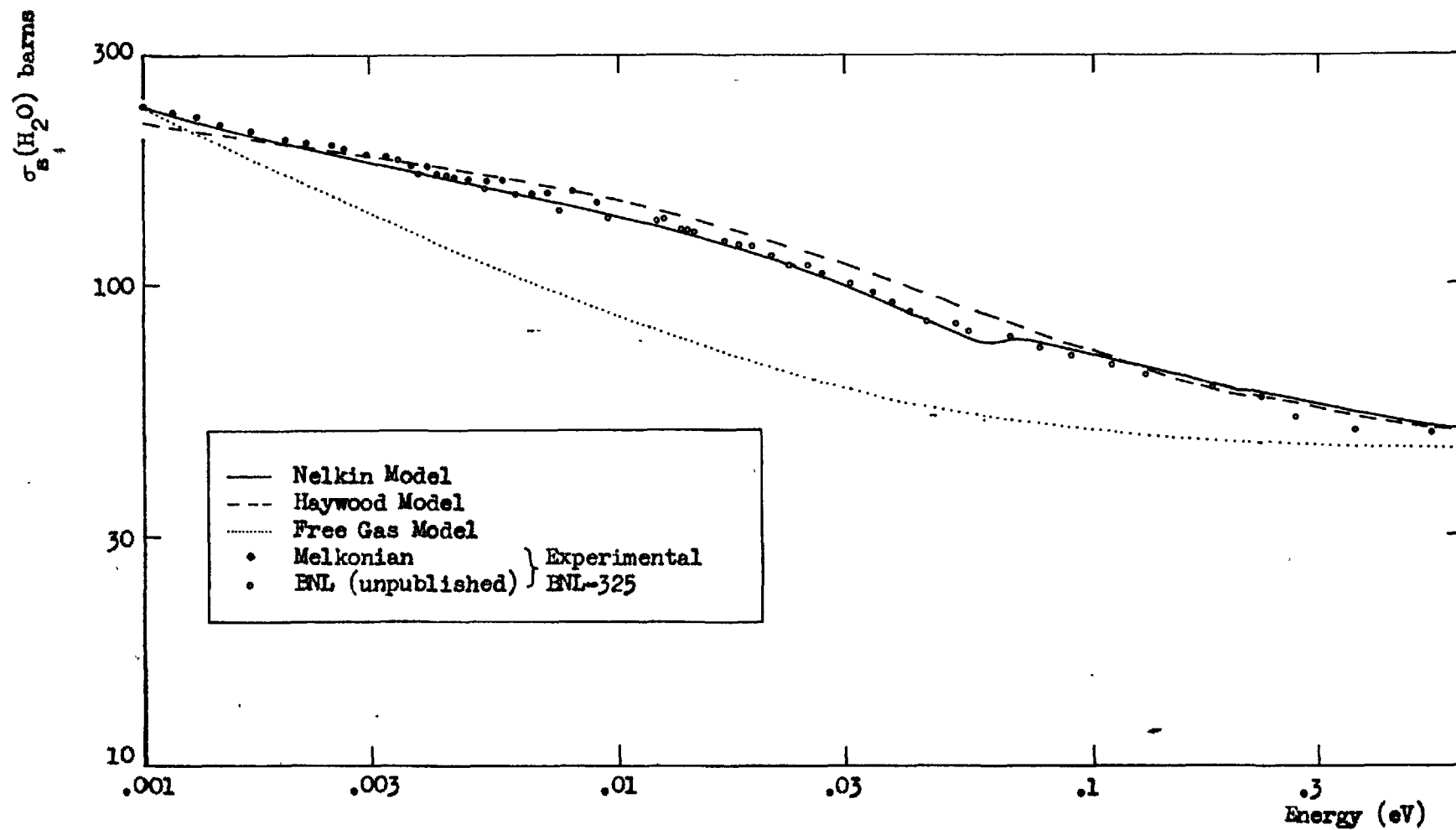


Figure 11 - Scattering Cross-Section of Water at 22.3°C

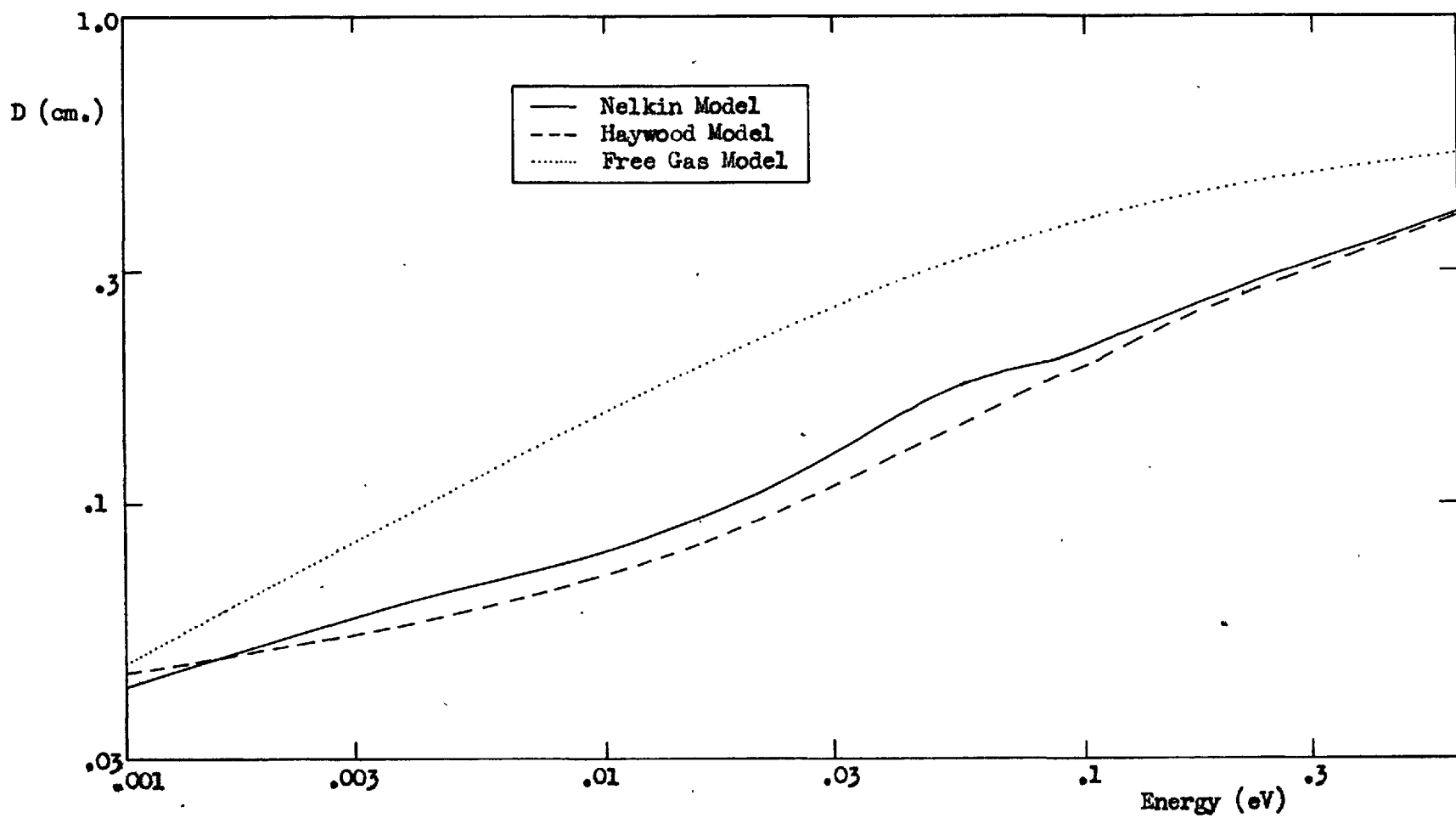


Figure 12 - Diffusion Coefficient of Water at 22.3°C

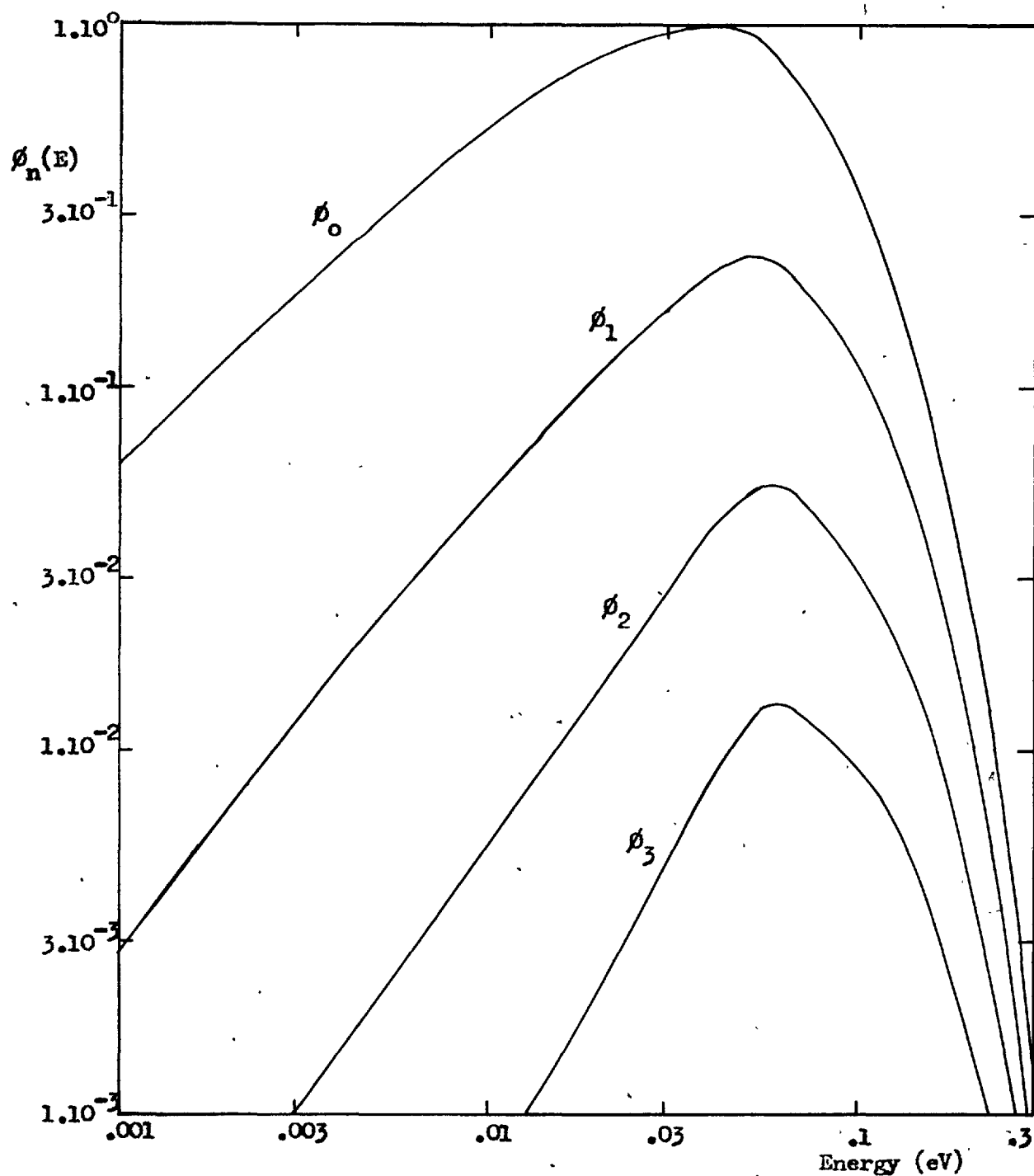


Figure 13 - Legendre Components of Neutron Flux in Boric Acid Solution
(Nelkin model at 22.3°C; Absorption 6.97 b/H atom)

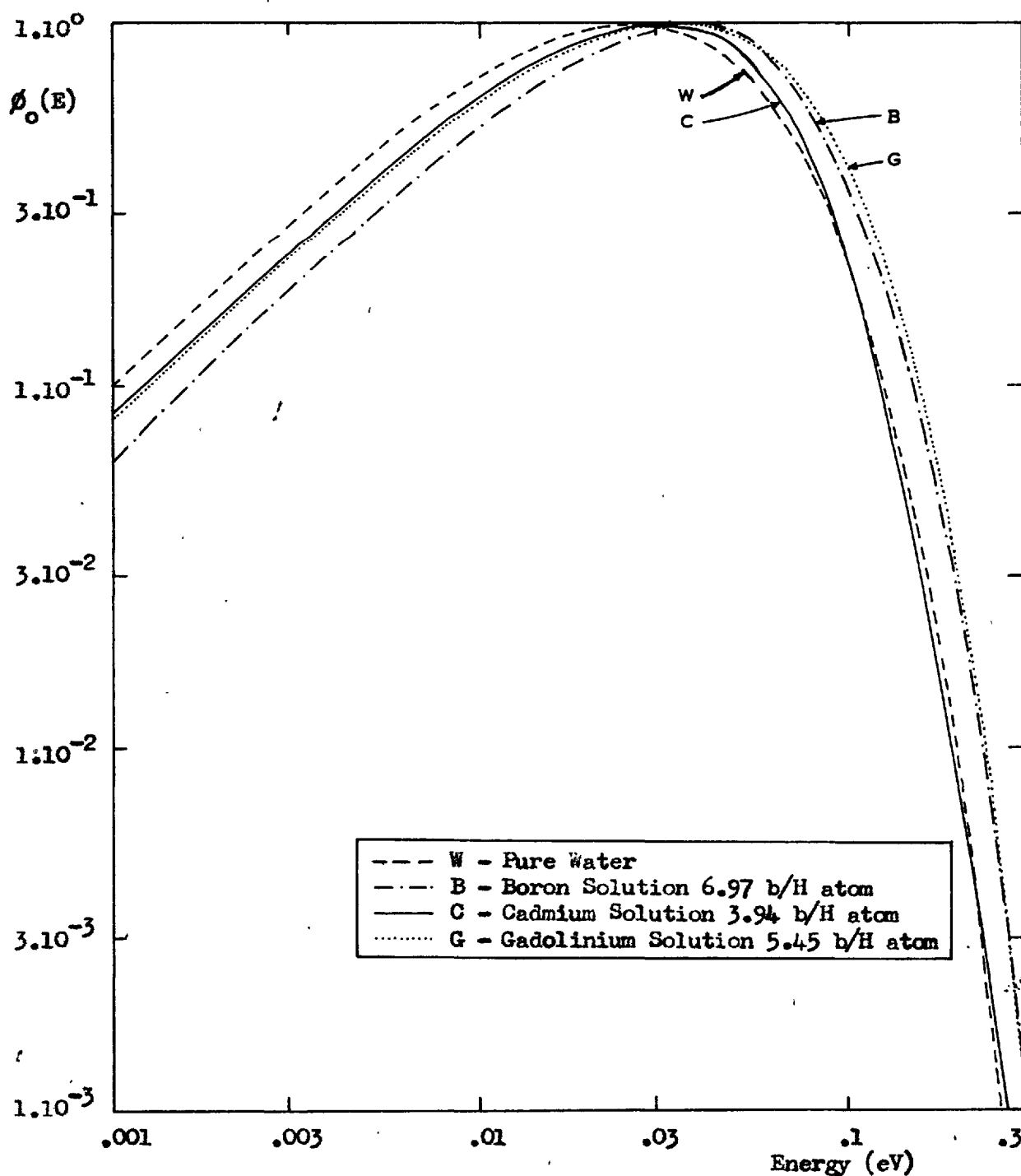


Figure 14 - Neutron Spectra in Absorbing Solutions
(Nelkin Model at 22.3°C)

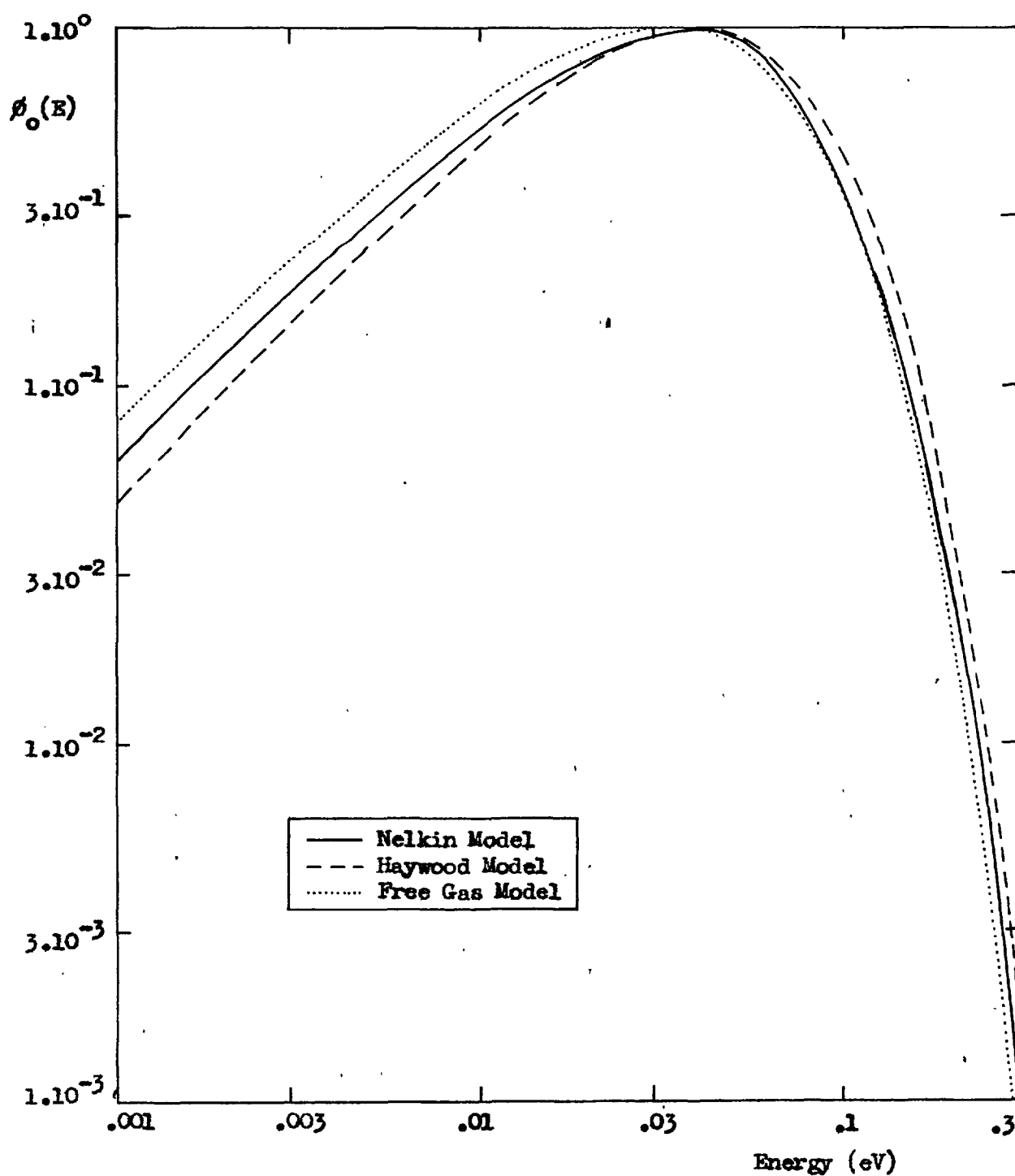


Figure 15 - Neutron Spectra in Boric Acid Solution
(Temperature 22.3°C; Absorption 6.97 b/H atom)

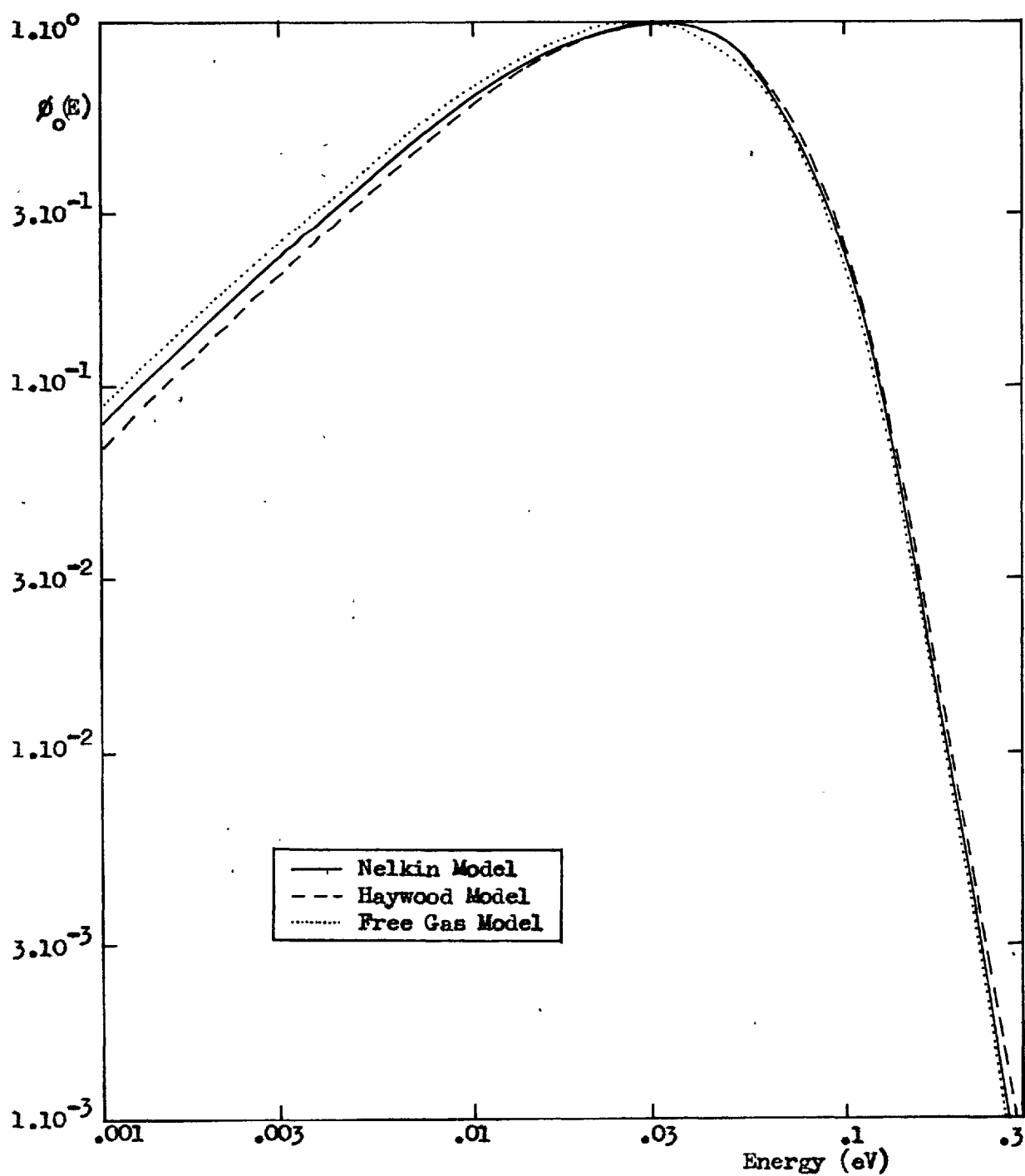


Figure 16 - Neutron Spectra in Cadmium Solution
(Temperature 22.3°C ; Absorption 3.94 b/H atom)

scattering is taken into consideration. This can be seen in Figure 12, where the diffusion coefficient (defined by equation 4.4.12) is given for the various models. The results for the Maxwellian average diffusion coefficient $\bar{D}_M$ (equation 4.4.24) are:

Model	$\bar{D}_M$	
	T=22.3°C	T=65°C
Nelkin	.1524	.1624
Haywood	.1321	-
Free Gas	.2841	.2887

The coding of the scattering part of DIS has been checked by making the following comparisons. The Nelkin differential cross-sections have been compared with the compilation of Federighi and Goldman (1963), and the total cross-sections with Besant (1965). The program PIXSE (Macdougall, 1963) has been used to produce Haywood differential cross-sections (to order zero) and Debye-Waller contributions to the total cross-section, which have also been compared with the values from DIS. In all cases, the general agreement was better than .5%.

6.2 Theoretical Neutron Spectra

The first four Legendre components of the flux in a highly concentrated boric acid solution are shown in Figure 13. It is seen that at low neutron energies the

flux is almost isotropic, while at higher energies it is strongly directional. This arises because the higher moments of the scattering kernel increase with energy relative to the zero order moment.

Figure 14 shows how the scalar flux given by the Nelkin model is affected by the cross-section of added absorber. In particular, the cadmium spectrum, although harder than the pure water spectrum for lower energies where the cadmium cross-section is $\sim 1/v$, becomes softer around .18 eV in the region of the cadmium resonance. Conversely, the spectrum for gadolinium hardens more than does that for boric acid around .1 eV, where the gadolinium cross-section falls off sharply with energy.

Figures 15 and 16 compare the spectra predicted by the various models for boric acid and cadmium solutions. It is seen that the free gas model gives results similar to those of the more sophisticated models; this is not unreasonable since in an infinite, non-absorbing medium the neutron spectrum would be a pure Maxwellian - independent of the dynamics of the scattering process.

It was not possible to compare any of these theoretical spectra with experimental values, since the latter are normally obtained in systems where the supply of neutrons is from a high energy source. In the present case, the transport equation 3.1.1, upon which the program DIS

is based, contains no source term; the spectrum obtained is such that its shape is self-perpetuating in space, under the particular conditions of absorption and scattering specified. Since the presence of a source term influences the entire thermal spectrum, comparison with measured spectra would be inappropriate. It is, however, known (e.g. Beyster, 1962) that spectra predicted by the Nelkin model are too soft, and it may then be that the harder Haywood spectra will give better agreement with experiment. Further work is needed on this point.

In the experiments described in this work, a negative source technique involving a cadmium shutter was used, and the effective source is therefore given by the reaction rate in the shutter. Since the shutter will be virtually 'black' to thermal neutrons, while absorbing only a few percent. of the fast neutrons, the effective source will be very close in shape to the equilibrium spectrum calculated by DIS, and it is therefore the 'no-source' method which is in this case appropriate to the comparison of theory with experiment.

6.3 Comparison of $1/v$ Experiments with Theory

Before comparing the experimental values of the spatial eigenvalue with theory, it is necessary to apply the correction mentioned in Section 5.6, to allow for the reduction in the number density of moderator molecules

on addition of solute. It was stated that this effect was about 3% for solution 9. Boric acid, however, contains hydrogen; the dissociation constant is very small and for this purpose this hydrogen may be considered to have the same cross-section as that in water. The net reduction in hydrogen concentration is then only 1%.

From diffusion theory

$$\kappa^2 = 3\Sigma_a \Sigma_s (1-\bar{\mu})(\dots) \quad 6.3.1$$

where the second bracket contains dimensionless terms. For each solution Σ_a may easily be calculated knowing the amount of hydrogen and boron present, but in order to reduce all observations to the same Σ_s , κ^2 is given by

$$\kappa^2 = \kappa_o^2 / f \quad 6.3.2$$

where κ_o^2 is the observed value, and f is defined as the ratio of the hydrogen number density in a particular solution to that in pure water. The value of f varied from 1.00 for pure water to .99 for the most concentrated solution, and was assumed to be independent of temperature.

The program POLY was modified to perform this operation, before fitting κ^2 to a power series in Σ_a . Since the least squares fitting method used weights each κ^2 point according to the reciprocal of the square of its standard deviation, the uncertainties in concentration were converted to an effective uncertainty in κ^2 , and

combined with the errors in L quoted in the tables in Section 5.8.

The results are shown in Figures 17 and 18, which give κ^2 as a function of Σ_{ao} . Also shown in Figure 17 are the experimental results of Starr and Koppel (1962) who used indium foils to determine spatial decay constants as a function of boric acid concentration at 21°C in a large tank situated on top of the Brookhaven Graphite Reactor. The agreement is seen to be excellent. Further recent results by Baumann and Pellarin (1966) over a similar range of concentrations have not been plotted since they were not available in tabular form, but the agreement here appears equally good.

It is seen that while the free gas model gives very poor results, the Nelkin and Haywood predictions are nicely bisected by the experimental points. This is in line with the scattering cross-sections shown in Figure 11, where again over a considerable range of energies the experimental points lie between the predictions of the two theories. From simple diffusion theory (equation 6.3.1) the slope of the κ^2 vs. Σ_{ao} plot is proportional to Σ_s - hence the differences observed here.

If the series expansion 4.1.3 is divided by a we obtain

$$\kappa^2/a = \alpha_1 - \alpha_2 a + \alpha_3 a^2 \quad 6.3.3$$

and plotting κ^2/a vs. a gives a graph which demonstrates with much greater sensitivity the effect of diffusion heating. This has been done in Figures 19 and 20, where the downward trend of the points with increasing absorption shows clearly this effect.

The results of fitting polynomials to both theoretical and experimental points will be given in the next section.

6.4 Power Series Coefficients

The coefficients of the power series expansion 4.1.3 have been obtained theoretically both by the 'direct' method (denoted by D) of Section 4.3, and also by the 'series' method (S) of Section 4.4, and the results are given in Table 6.1. The best values of the coefficients for the former method were obtained by selecting those from fits which showed no systematic deviations of the points from the fitted line. B_1 calculations ($L = 1$ in the table) were performed for all scattering models, and B_3 calculations ($L = 3$) for the Nelkin model only, since these are very time-consuming. Approximate confidence limits are given, based both on the statistical deviations given by POLY, and also on the observed variation of the coefficient between different fits.

Each theoretical point was obtained by running the program DIS for a particular scattering model and temperature, and a known amount of $1/v$ absorption. The absorp-

tion for the experimental points, however, is complicated by the presence of the hydrogen in water. In order to determine a value for the hydrogen absorption cross-section, therefore, the experimental results were fitted to the expression

$$\kappa^2 = \alpha_0 + \alpha_1 a - \alpha_2 a^2 + \alpha_3 a^3 \quad 6.4.1$$

rather than the expansion 4.1.3. $a \equiv \Sigma_{a0}$ was taken as $\Sigma_B + \Sigma_H$ where Σ_H is the macroscopic hydrogen absorption cross-section. The number density of hydrogen atoms was corrected for addition of boric acid in the manner described in the previous section, and the microscopic hydrogen absorption cross-section at .0253 eV, σ_{aH} , taken initially as .325 barns. Fits were made to both three (omitting the final term) and four coefficients of equation 6.4.1, and from the fitted value of α_0 an improved estimate of σ_{aH} obtained from the relation

$$\sigma_{aH} = .325 + \frac{\alpha_0}{N_H \alpha_1} \text{ barns} \quad 6.4.2$$

The three-parameter fits obtained by using the new values of σ_{aH} are shown in Figures 19 and 20.

For the experimental results, the effect on the fitted coefficients of dropping successive high poison points from both three- and four-parameter fits is shown in Figures 21-23. (All results, both experimental and theoretical, are presented in the form of the pulsed

expansion coefficients obtained from the relations 4.2.6, rather than the less familiar stationary coefficients α_j .) It is seen that there is no significant trend in the values of any of the coefficients as the number of points is decreased. In general four-parameter fits gave higher empirical variances. The values taken for D_0 , C and σ_{aH} are therefore those from the three-parameter fit retaining all the experimental points; the values for F (with standard deviations $>100\%$) are those from the four-parameter fit with all points included. The experimental results are

$T=22.3^\circ\text{C}:-$

$$\begin{aligned} D_0 &= (35620 \pm 230) \text{ cm}^2 \text{ sec}^{-1} \\ C &= (2800 \pm 260) \text{ cm}^4 \text{ sec}^{-1} \\ F &= (370 \pm 410) \text{ cm}^6 \text{ sec}^{-1} \\ \sigma_{aH} &= (321.9 \pm 2.1) \text{ mb} \end{aligned}$$

$T=65^\circ\text{C}:-$

$$\begin{aligned} D_0 &= (41340 \pm 390) \text{ cm}^2 \text{ sec}^{-1} \\ C &= (2470 \pm 400) \text{ cm}^4 \text{ sec}^{-1} \\ F &= (360 \pm 540) \text{ cm}^6 \text{ sec}^{-1} \\ \sigma_{aH} &= (326.5 \pm 3.8) \text{ mb} \end{aligned}$$

Combining the two results for σ_{aH} gives

$$\sigma_{aH} = (324 \pm 3) \text{ mb.}$$

The difficulty in extracting a value for F can be seen from Figures 19 and 20; the final term of equation 6.4.1, from which F is obtained, implies a curvature in these graphs which is not easy to detect. To illustrate this the two-parameter fits to equation 4.1.3 are shown for both Nelkin and Haywood models: the difference between these and the exact theoretical curves is considerably less than the errors of the experimental points. In Figure 24 the experimental results for 22.3°C (above) have been used to calculate the contributions of the second and third terms of the expansion 4.1.3 to κ^2 . It is seen that the cubed term only represents 1% of the value of κ^2 for the highest poison concentration, which is barely detectable. While this figure is very uncertain owing to the high relative error in F , it is clear from these considerations that coefficients of higher order than C are elusive.

Two tables now follow; in the first of these diffusion parameters obtained by the present author using theoretical methods described in this work are given, and in the second the present experimental results for $1/v$ absorption are compared with those obtained by other workers using both pulsed (denoted by P) and stationary (S) methods.

Table 6.1 - Theoretical diffusion parameters for water

Model	T °C	Method	L	D_0 $\text{cm}^2 \text{sec}^{-1}$	C $\text{cm}^4 \text{sec}^{-1}$	F $\text{cm}^6 \text{sec}^{-1}$
F.G.	22.3	D	1	70113 ± 2	1612 ± 10	410 ± 10
Haywood	22.3	D	1	33045 ± 5	3700 ± 50	400 ± 30
Nelkin	22.3	D	1	38050 ± 5	3030 ± 50	160 ± 40
Nelkin	22.3	D	3	38344 ± 4	3170 ± 20	280 ± 40
F.G.	22.3	S	3	71087	38805	578800
Haywood	22.3	S	3	33041	3716	179420
Nelkin	22.3	S	3	38139	2949	43
F.G.	65	D	1	76169 ± 2	1860 ± 10	460 ± 10
Nelkin	65	D	1	43347 ± 2	3340 ± 15	200 ± 10
F.G.	65	S	3	77214	2897	724
Nelkin	65	S	3	43424	3199	117

Table 6.2 Experimental diffusion parameters for water

Reference	T °C	Method	D $\text{cm}^2 \text{sec}^{-1}$	C $\text{cm}^4 \text{sec}^{-1}$	σ_{aH} mb
Von Dardel (1954)	22	P	36340 ± 750	7300 ± 1500	333 ± 3
Antonov (1955)	23	P	35000 ± 1000	4000 ± 1000	329 ± 10
Dio (1958)	22	P	35450 ± 600	3700 ± 700	328 ± 6
Kuchle (1960)	22	P	35400 ± 700	4200 ± 800	326 ± 6
Bretscher (1962)	26	P	36985 ± 1630	5021 ± 2490	322 ± 17
Lopez (1962)	26.7	P	37503 ± 366	5116 ± 776	325.3 ± 1.6
Starr (1962)	21	S	35850 ± 100	2900 ± 350	326.9 ± 1.6
Bretscher (1963)	24	S	37135 ± 1196	2836 ± 598	323 ± 32
Present	22.3	S	35620 ± 230	2800 ± 260	321.9 ± 2.1
Present	65	S	41340 ± 390	2470 ± 400	326.5 ± 3.8

6.5 Discussion

A 32-point energy mesh was used for all the theoretical calculations, with the exception of those by the 'direct' B_3 method. Core storage limitations forced the number of mesh points here to be reduced to 28. However, two B_1 runs using this 28-point mesh produced values of κ differing by only .2% and .01% for pure water and for the highest concentration of $^{1/v}$ absorber, respectively, from those obtained with the 32-point mesh.

The series method is seen to give good results for both D_0 and C (apart from the free gas model in the latter case), indicating that the assumption of elastic anisotropic scattering is reasonable as far as the second Legendre moment of the scattering kernel. There is a significant rise with temperature in all three parameters.

The 22.3°C experimental value obtained for D_0 is in excellent agreement with the values obtained by other experimenters using both pulsed and stationary methods, apart from the results of Lopez and Beyster (1962). Correcting their value for D_0 to 22°C gives $(36900 \pm 370) \text{ cm}^2 \text{ sec}^{-1}$, which is still significantly different from the more accurate of the other results. Beckurts (1962) has suggested that the discrepancy is due to the different shapes of the containers used - Lopez and Beyster used nearly cubical systems, and Kuchle flat cylinders.

The value of C obtained agrees very well with that of Starr and Koppel (1962), but the values obtained from pulsed experiments, although less accurate, are somewhat higher. The reason for the greater inaccuracy is that for large B^2 , where the effect of diffusion cooling is most noticeable, extrapolation distance uncertainties become important. For the largest value of B^2 ($.59 \text{ cm}^{-2}$) used by Lopez and Beyster, estimation of extrapolation distance by two different techniques altered this value by 1.2%. By contrast, in the present experiments the largest value of κ^2 ($= -B^2$) measured was 1.85 cm^{-2} . From Figure 25 it can be seen that much greater curvature is apparent over the range of the stationary experiments than over that of the pulsed.

For the microscopic hydrogen absorption cross-section, the values obtained by both methods are in very good agreement although they are rather lower than the presently recommended value of $(332 \pm 2) \text{ mb}$ given by Stehn et al (1964).

A detailed comparison of the room temperature diffusion length of pure water has not been given, but it is worth mentioning that the value of $(2.761 \pm .004) \text{ cm}$. obtained is in excellent agreement with the average value $(2.767 \pm .008) \text{ cm}$. of several stationary measurements quoted by Beckurts (1962).

6.6 Determination of M_2

It was shown in Section 4.5, by the use of diffusion theory and the assumption that $D(E) \propto v$, that the coefficients D_0 and C may be used (equation 4.5.19) to derive a value for M_2 , the second energy transfer moment weighted by the Maxwellian distribution. The error in the derivation of C introduced by using diffusion rather than transport theory is known (e.g. Calame, 1964a) to be about 10%, to which must be added a further error of about 5% for the assumption that $D(E) \propto v$. The error may also be estimated directly by applying this procedure to the theoretical parameters. The results for the direct method are

Model	T °C	L	M_2 barns/ H atom
F.G.	22.3	1	367.0
Haywood	22.3	1	35.5
Nelkin	22.3	1	57.4
Nelkin	22.3	3	55.8
F.G.	65	1	358.0
Nelkin	65	1	64.3

The known M_2 for the Nelkin model at 23°C is (Friedman, 1964) 52.1 b/H atom, showing that the error introduced in the procedure of Section 4.5 is not greater than 10%.

Adding an error of this magnitude to the errors in D_0 and C , we therefore obtain for the stationary experimental results

Reference	T °C	M_2 barns/H atom
Starr (1962)	21	53 ± 12
Present	22.3	55 ± 11
Present	65	79 ± 21

These results are in good agreement with the Nelkin model, but in rather poorer agreement with the Haywood model. The high value for the free gas model is expected, since the effective mass of a proton as seen by a neutron will increase with chemical binding and the average energy exchanged in a collision will be greater if binding effects are ignored. This fact is also evident from the much flatter inelastic scattering cross-section for the free gas model shown in Figure 2.

Friedman (1964) has obtained a value for M_2 of (12.6 ± 1.6) b/H atom for water at 23°C , from measurements of the decay constant in solutions of non- $1/v$ absorbers. Since this would imply a cooling coefficient (assuming an average experimental D_0) of about $12000 \text{ cm}^4 \text{ sec}^{-1}$, this result must be viewed with considerable suspicion.

6.7 Non-Stationary Diffusion Length Experiments

The analogy between conventional pulsed and stationary experiments has been stressed at several points in this work, and the results presented in the previous section demonstrate the agreement between diffusion parameters obtained by these means. . From the relations given in Section 4.2 the experimental results from both methods may be represented in one plot, and this has been done in Figure 25, where the results from the present stationary experiment, and from the pulsed measurements of Lopez and Beyster (1962) are shown. The three-parameter fit to all the present experimental points has been extrapolated back to the third (pulsed) quadrant, and the agreement is very good.

Until recently it was thought (e.g. Honeck, 1962) that the second quadrant of Figure 25 was inaccessible to the experimenter, on account of the non-zero inherent absorption of the moderator. However Joest and Memmert (1964) have pointed out that if the source of neutrons in a stationary experiment can be made to decay with time constant λ , this is then equivalent to a negative poisoning with an effective cross-section of magnitude λ/v . Subsequent to this remark, a series of experiments has been carried out by Arai and Kuchle (1965), in which a pulse of neutrons was injected into a graphite block, and

the diffusion length measured in an adjacent tank of water. A necessary condition for this experiment to succeed is that the initial pulse should decay faster in the water than in the graphite, which then acts as a decaying source of neutrons. This is easily accomplished on account of the much greater absorption cross-section for water than for graphite. After a few milliseconds waiting time, the spatial relaxation length in the water was found to be the same for each channel. Three points were obtained by varying the size of the graphite block and hence λ . These results have also been plotted in Figure 25, and are seen to lie on the curve fitted to the present experimental points.

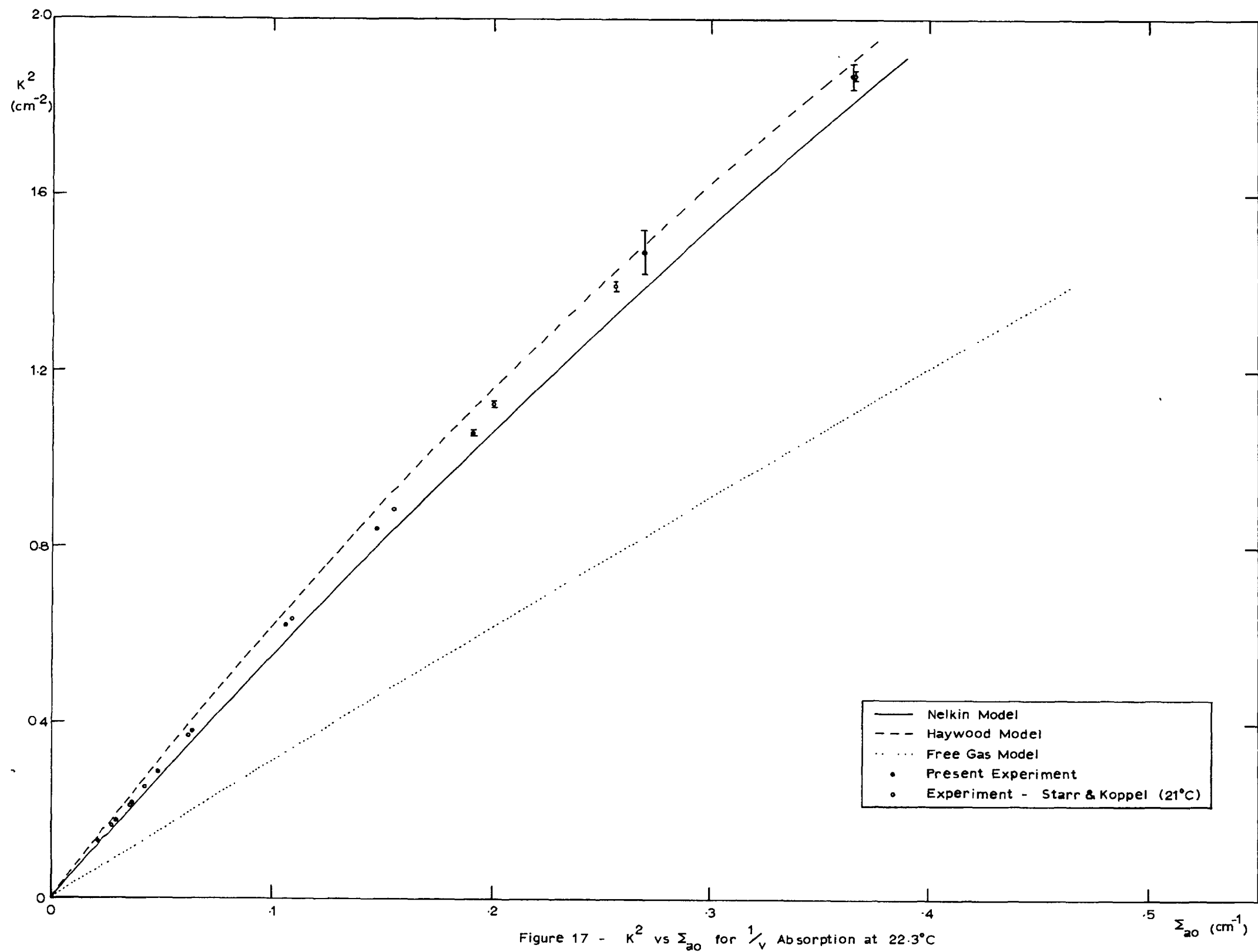
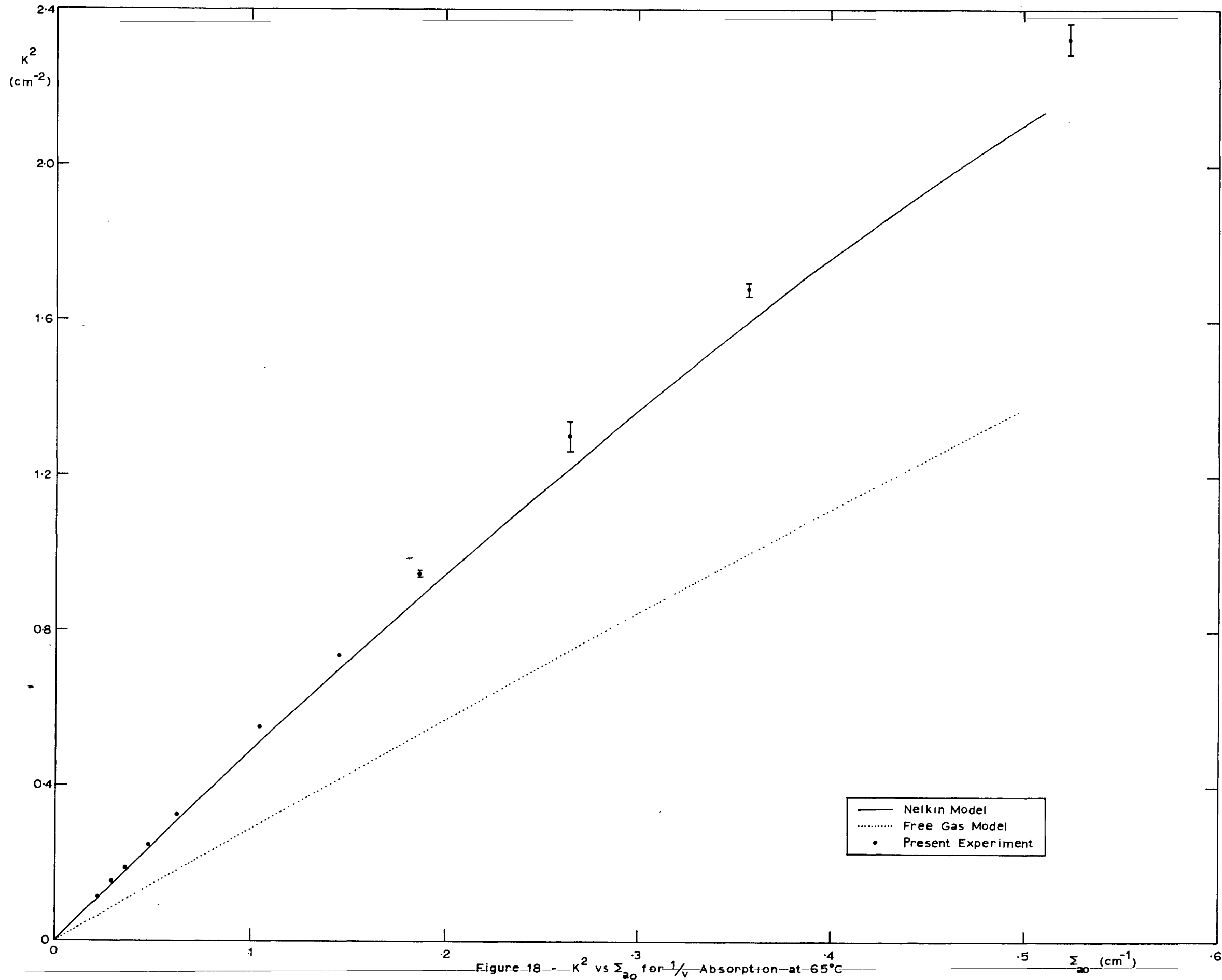


Figure 17 - K^2 vs Σ_{ao} for $1/v$ Absorption at 22.3°C

Figure 18 -- K^2 vs Σ_{∞} for $1/V$ Absorption at 65°C

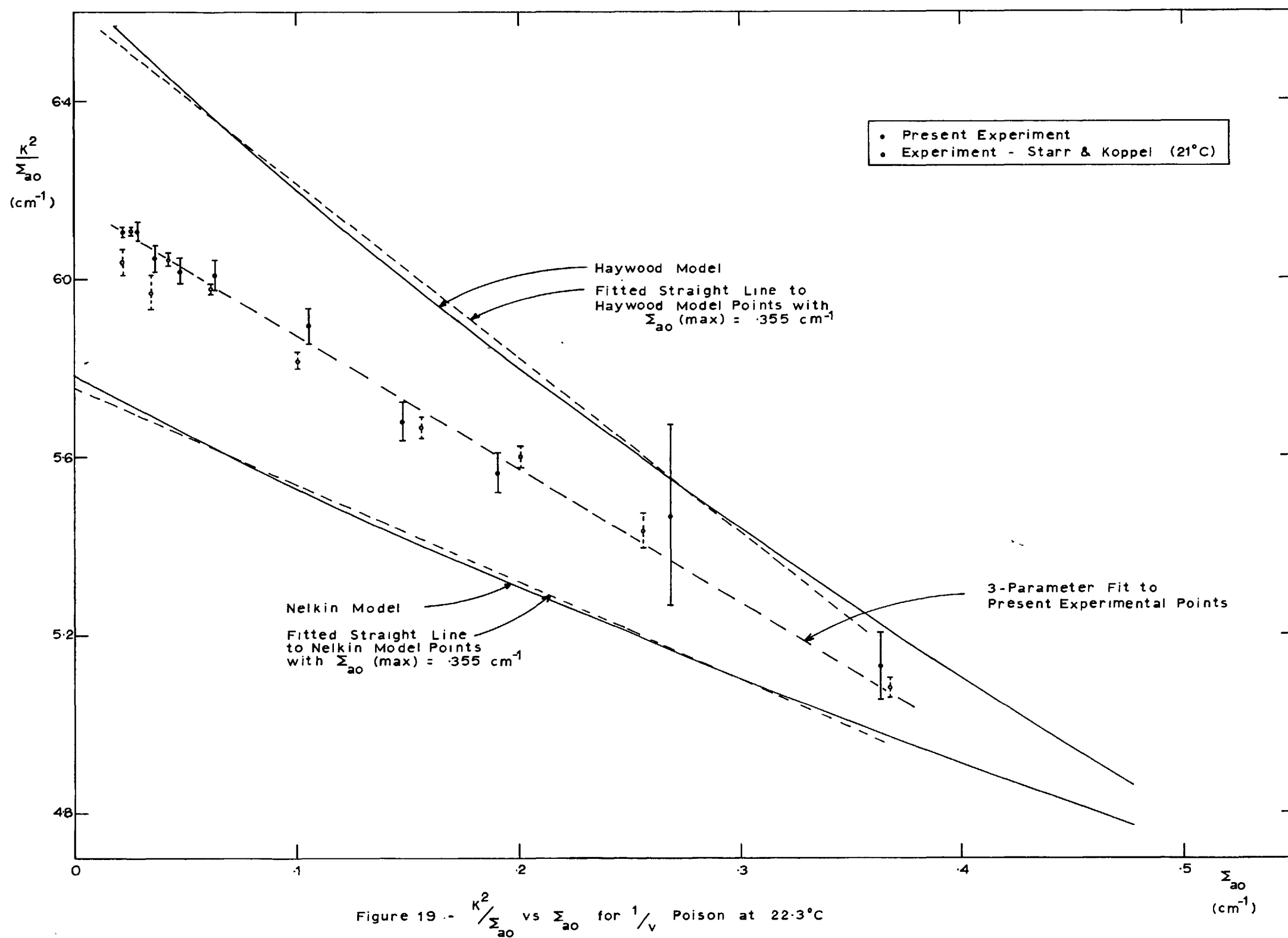


Figure 19.- $\frac{K^2}{\Sigma_{ao}}$ vs Σ_{ao} for $1/v$ Poison at 22.3°C

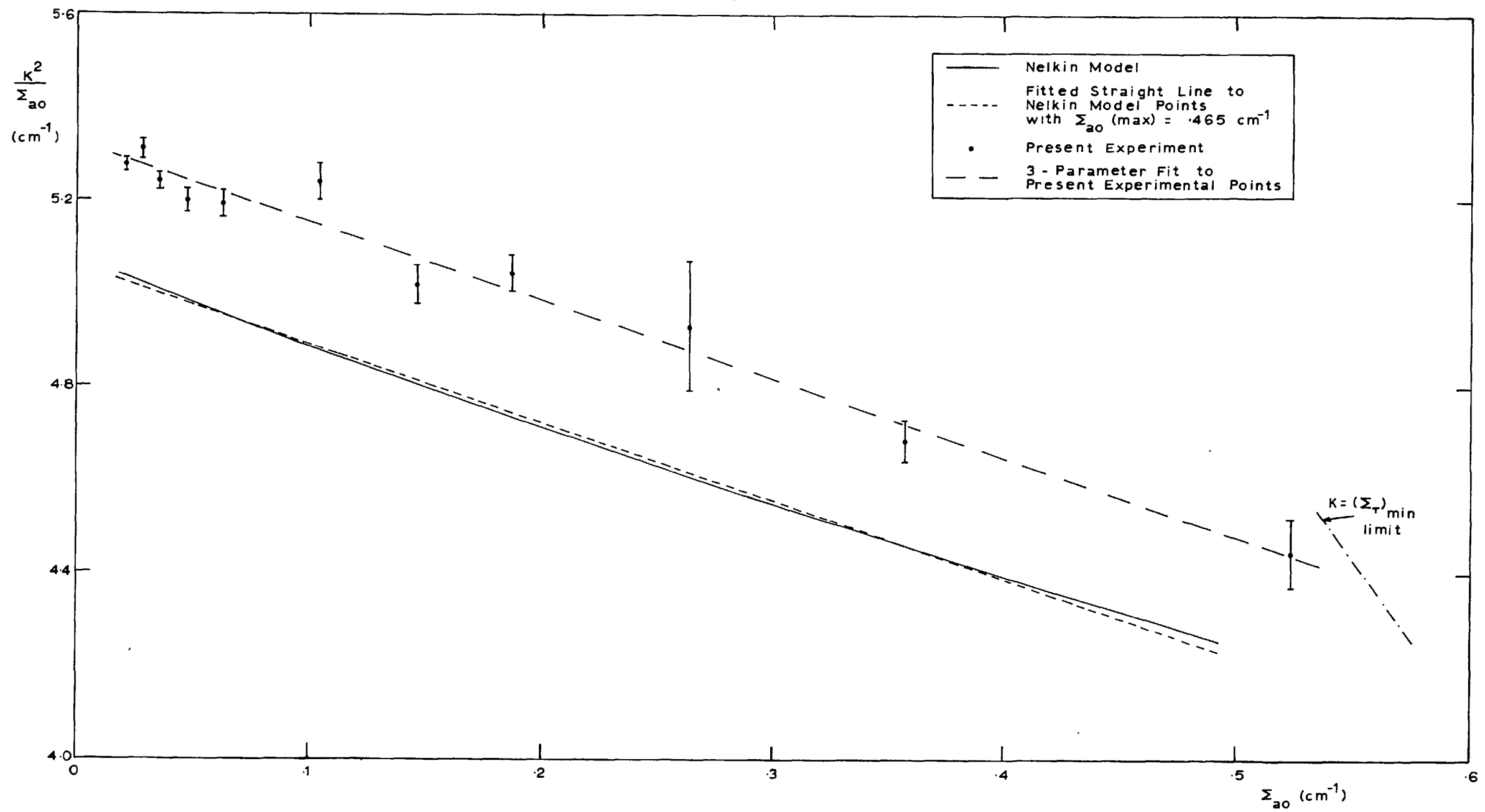


Figure 20 - $\frac{K^2}{\Sigma_{ao}}$ vs Σ_{ao} for $1/v$ Poison at 65°C

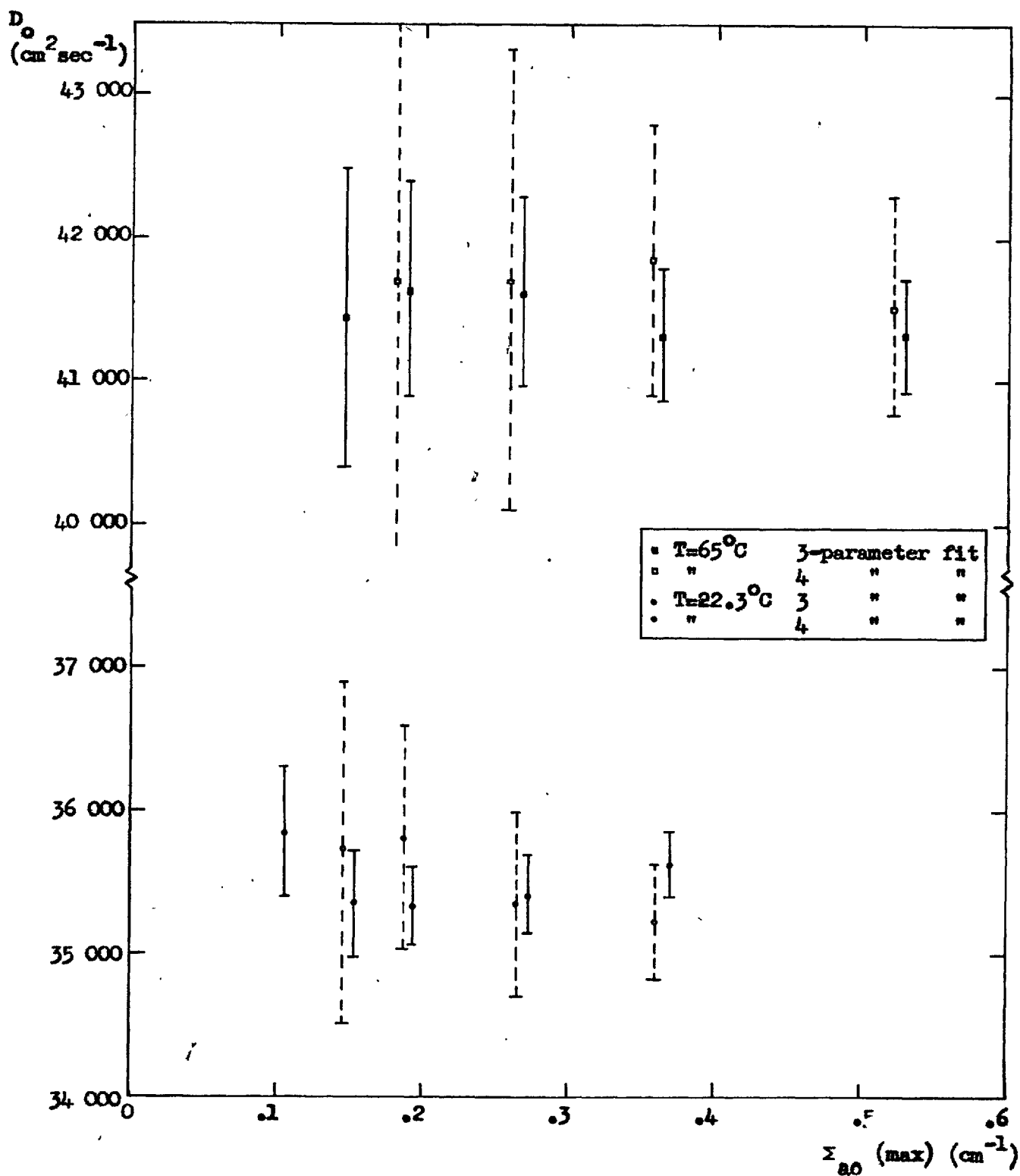
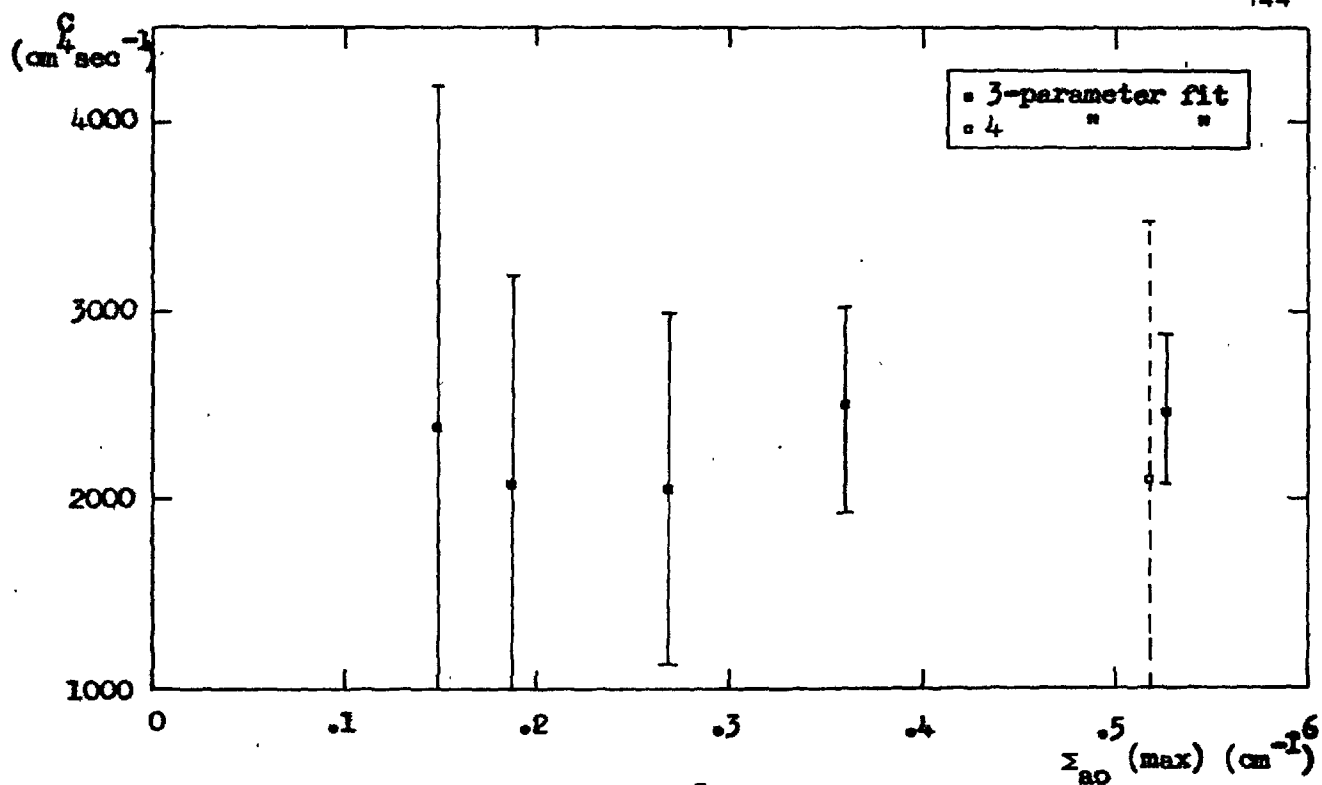
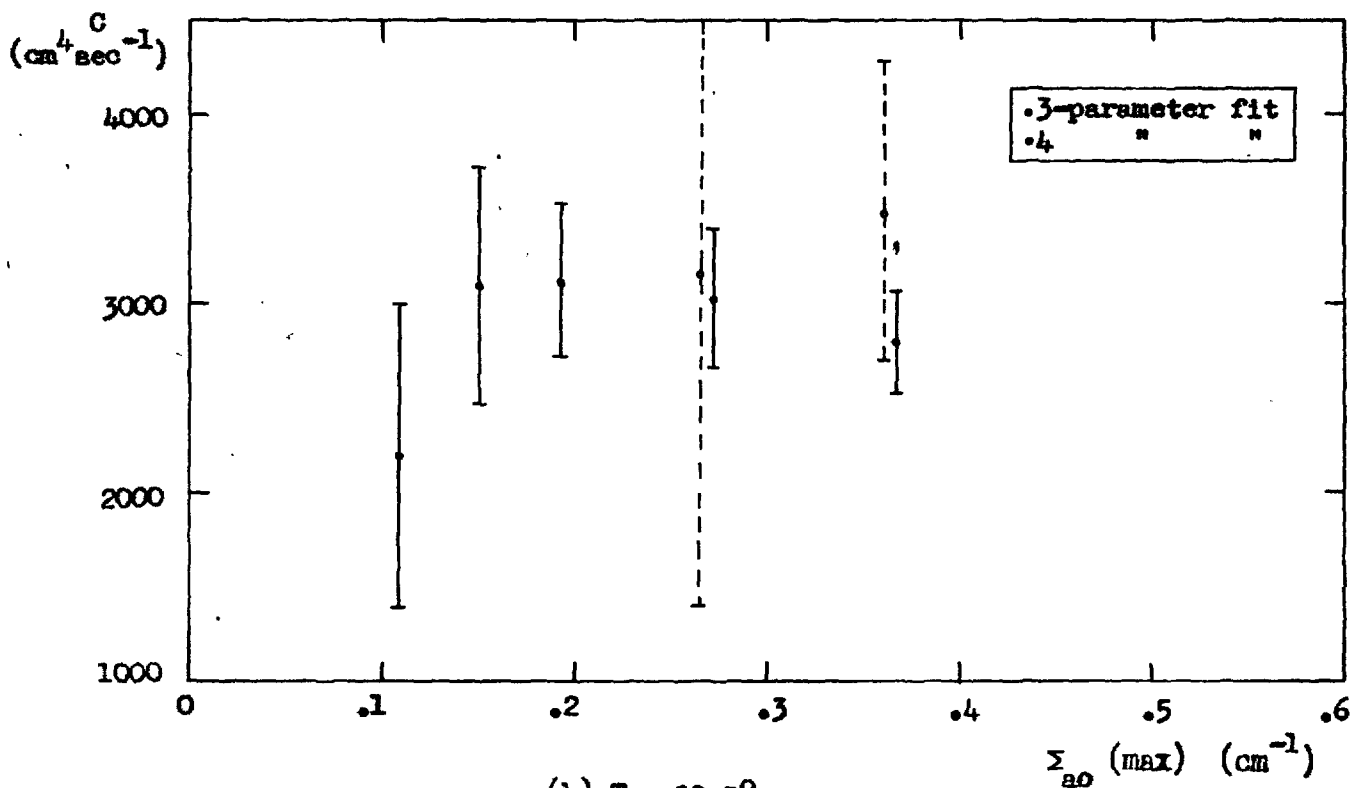


Figure 21 - Effect of Dropping High Poisson Points on Fitted Value of D_o

(a) $T = 65^\circ\text{C}$ (b) $T = 22.3^\circ\text{C}$ Figure 22 - Effect of Dropping High Poison Points on Fitted Value of C

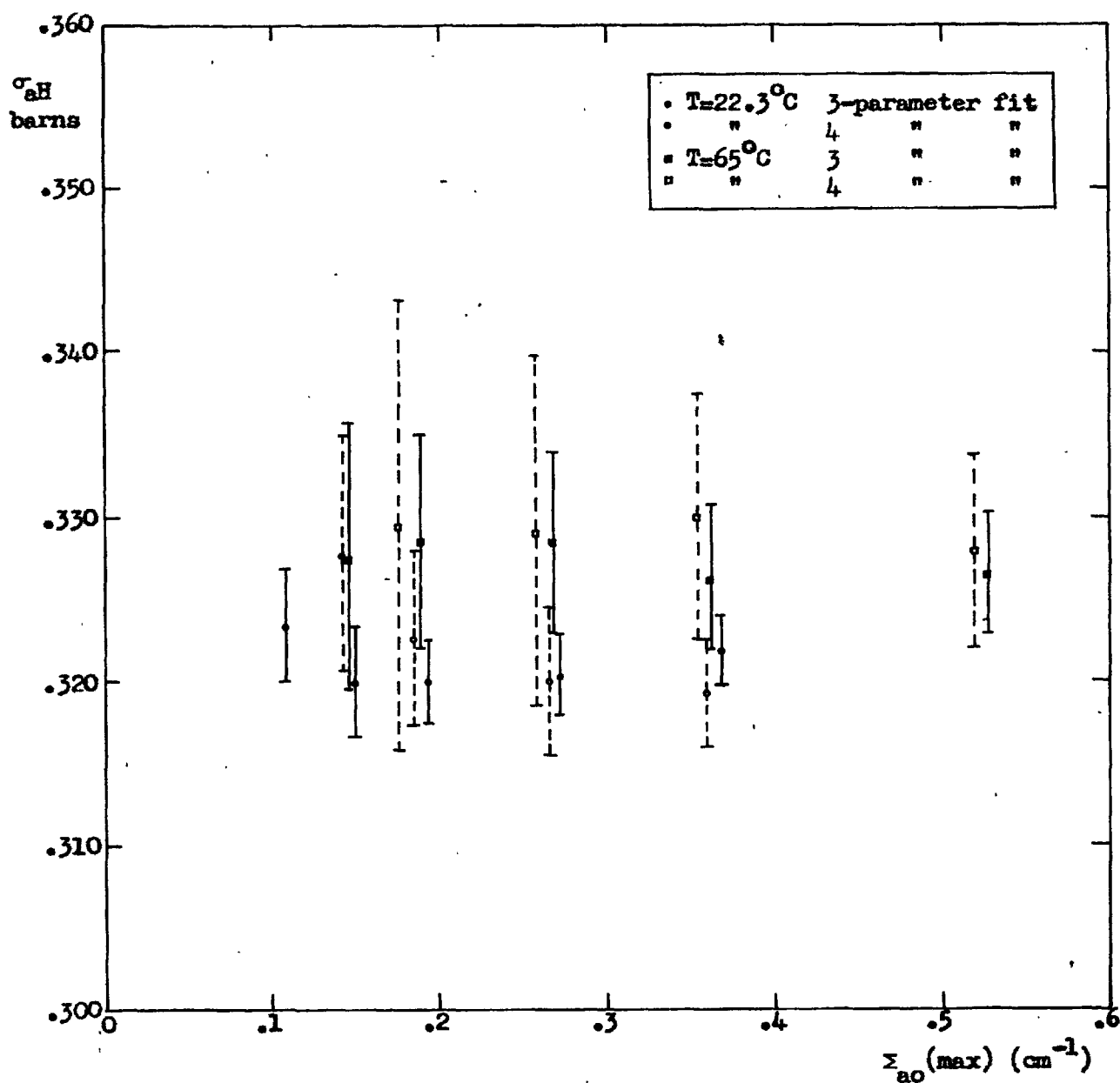


Figure 23 - Effect of Dropping High Poison Points on Fitted Value of σ_{aH}

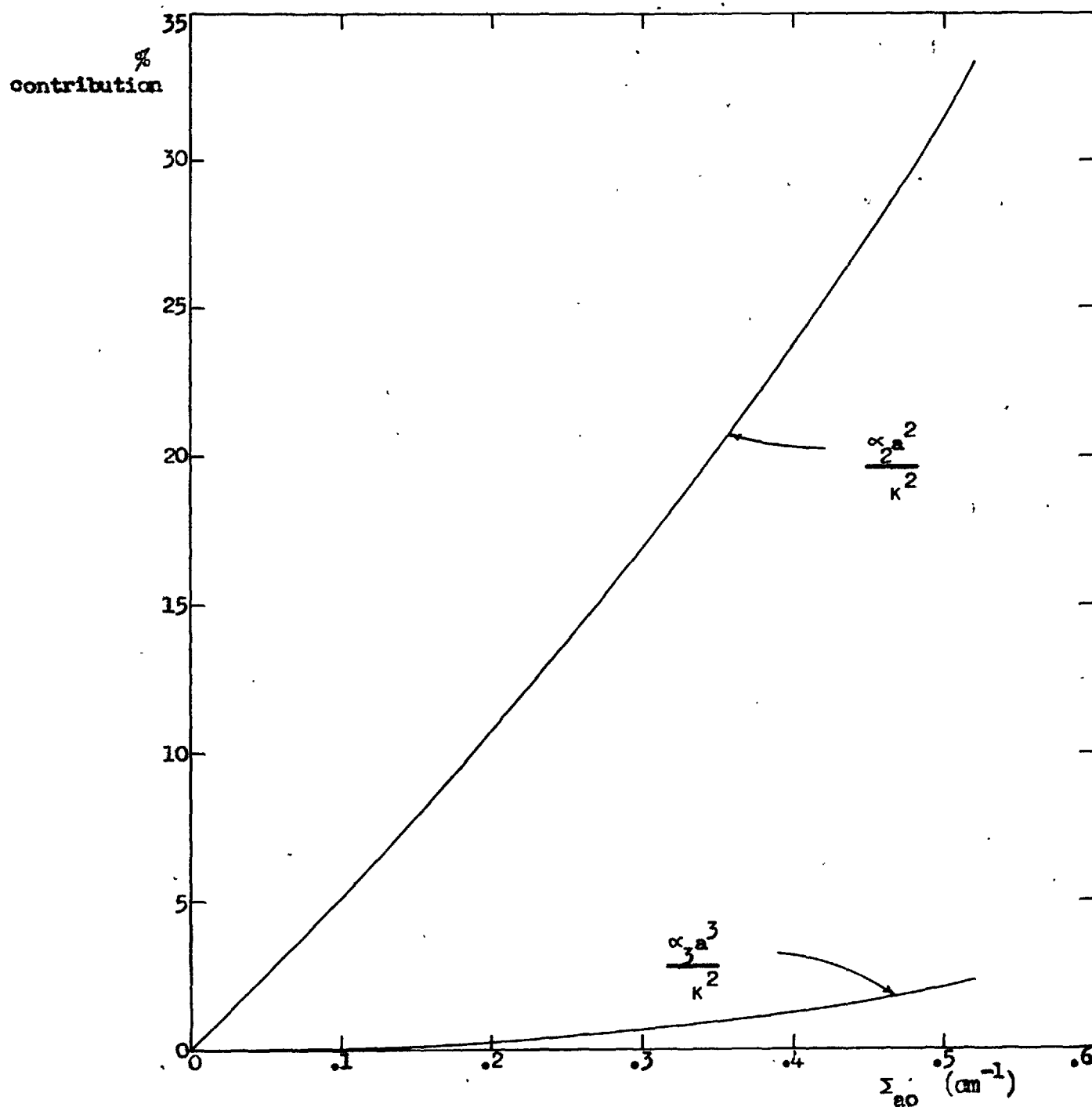


Figure 24 - Percentage contributions of a^2 and a^3 Terms to κ^2 at 22.3°C

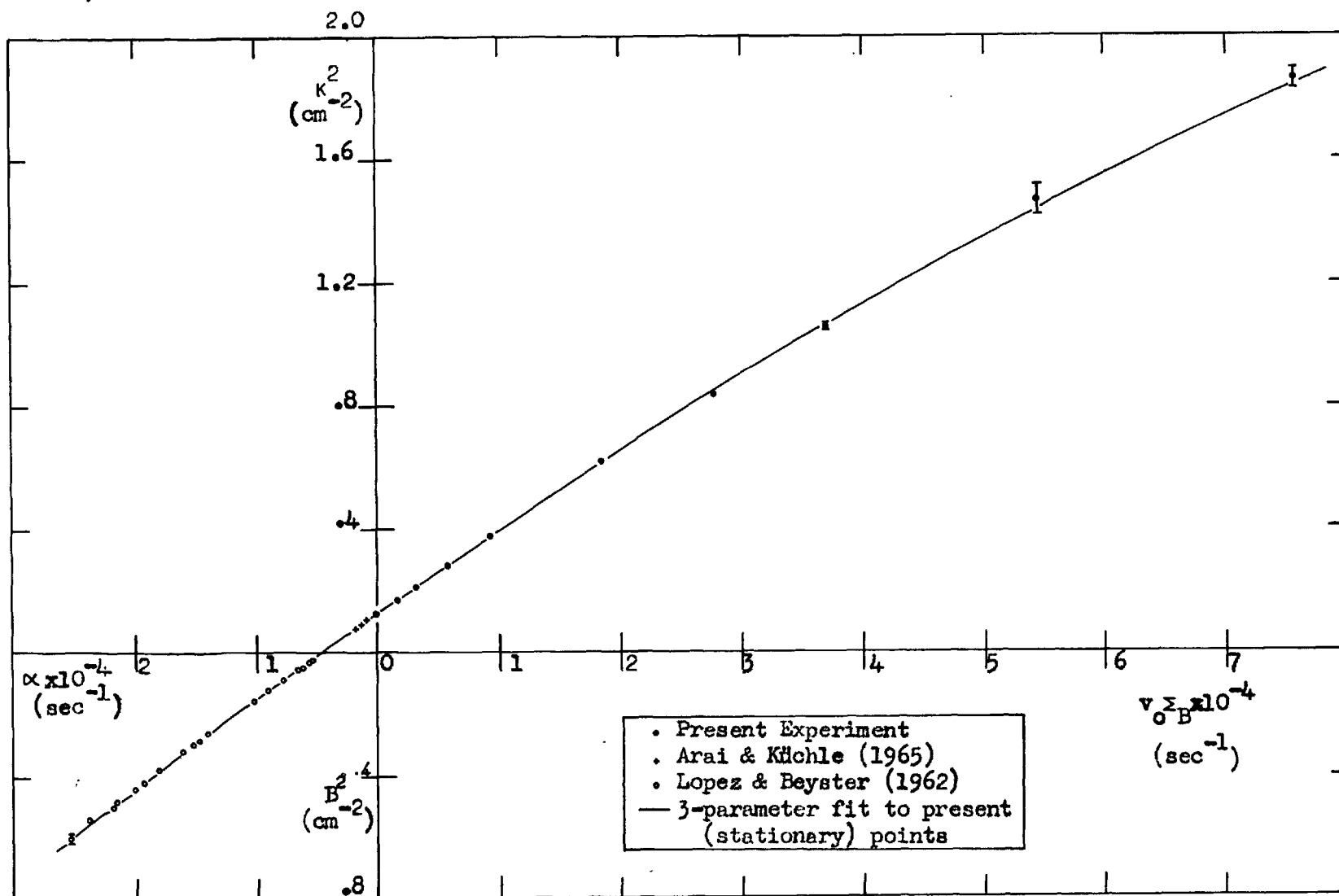


Figure 25 - Determination of Diffusion Parameters of Water

CHAPTER 7

ANALYSIS AND DISCUSSION OF RESULTS - PART II

In this chapter experimental and theoretical data for aqueous solutions of cadmium sulphate and gadolinium nitrate are analysed and compared.

7.1 Comparison of non- $1/v$ Experiments with Theory

An account was given in Section 6.3 of the manner in which a correction was applied to the boric acid results in order to take into account the reduction in the hydrogen number density on addition of poison. A similar correction was applied to the cadmium sulphate results on the basis of the specific gravity data obtained from Hodgman (1957). As mentioned in Section 5.6, the maximum correction was .18%. No correction was applied for gadolinium since the concentrations were much lower.

In Figure 26 the corrected κ^2 are plotted against Σ_{00} , where the latter quantity is obtained from the product of the non- $1/v$ absorber number density and its microscopic absorption cross-section averaged over a Maxwellian distribution, σ_{00} . A short FORTRAN IV program, MACE, was written by the present author to calculate σ_{00} using a 32-point energy mesh and the Lagrangian weights generated by DIS. The values obtained were 2986 and 35684 barns for cadmium and gadolinium respectively, at 22.3°C.

The advantage of this choice of abscissa is that for low poison concentrations, where spectral distortion effects are small, the gradients are the same for both absorbers. This can be seen in Figure 26, the slight increase in κ^2 for the cadmium points as a result of the small quantity of boron present in the solutions being too small to show up on this scale.

The predictions of the three scattering models are also plotted in Figure 26. The hydrogen microscopic absorption cross-section for the gadolinium solutions has been taken as .332 barns, in accordance with the recommendations of Stehn et al (1964). The presence of boron in the cadmium solutions was taken into account by adopting a value of .337 barns for σ_{aH} in this case.

Rearrangement of the series expansion 4.1.4 gives

$$\frac{\kappa^2 - \beta_o}{\Sigma_{oo}} = \frac{\beta_1}{\sigma_{oo}} - \frac{\beta_2}{\sigma_{oo}^2} \Sigma_{oo} + O(\Sigma_{oo}^2) \quad 7.1.1$$

Plotting $(\kappa^2 - \beta_o) / \Sigma_{oo}$ vs. Σ_{oo} therefore yields a graph whose slope demonstrates the effect of diffusion heating, in the same way as was done for $1/v$ absorption in Figures 19 and 20. This is shown in Figure 27, where in fact a slight diffusion cooling effect is seen to occur in the case of cadmium.

As was the case with $1/v$ absorption, the experimental points generally lie between the predictions of the Haywood

and Nelkin models, the free gas model being totally inadequate. However, it appears that the experimental cadmium points lie closer to the Nelkin values, while the curvature of the gadolinium points in Figure 26 (i.e. the gradient in Figure 27) is better represented by the Haywood model. In the former case spectral distortion effects are far less conspicuous : in the diffusion approximation these depend on the difference between the absorption cross-section and the diffusion coefficient, and this difference is relatively small because of the cadmium resonance. For gadolinium, however, the difference is much greater (in the theory of Section 4.6, the ratio $-u_{01}/\sigma_{00} D_{00}$, which would be zero for $\sigma_a \propto v$ is .184 for cadmium and .831 for gadolinium), and it is therefore suggested that while the Nelkin model gives a better scattering cross-section than does the Haywood model, the latter describes spectral effects more accurately.

The intercepts in Figure 27 are a measure of the quantity β_1/σ_{00} ; β_1 contains a contribution (see equation 4.6.12) depending on the product of non- $1/v$ and $1/v$ absorber concentrations. It is this term, which may be thought of as representing the diffusion heating effect in pure water, which is responsible for the gadolinium intercept being below that of cadmium for both of the theoretical models. In the experimental case this is surprisingly

reversed; this is attributed both to uncertainties in the fitted lines, and also to possible small errors in the absorption cross-section as read from Hughes and Harvey (1955). The boron content of the cadmium solutions has little noticeable effect.

In Figures 26 and 27 the theoretical upper bound on the value of κ considered in Chapter 1 is shown. It is seen that the two highest cadmium concentration points are well in excess of the limit. This point will be discussed further in Chapter 8.

7.2 Power Series Coefficients

Three- and four-parameter fits of both experimental and theoretical points to the expansion 4.1.4 have been performed using the program POLY. The procedure adopted for the theoretical parameters was similar to that described in Section 6.4. Figures 28-30 show the effect of dropping high poison points on the fitted experimental parameters; while no significant trend is observed in the case of gadolinium, each of the cadmium coefficients does appear to show a systematic variation. Consideration of Figure 27 and of the error bars in Figures 28-30, however, leads to the belief that this is spurious. As in the $1/v$ cases, therefore, the experimental parameters adopted are those appropriate to three parameter fits of all the experimental points. The results are given in the following table.

Table 7.1 - Non- $^{1/v}$ coefficients for water at 22.3°C

Model	Element	L	β_0 (cm $^{-2}$)	$\beta_1 \times 10^{19}$ (cm)	$\beta_2 \times 10^{38}$ (cm ^{4})
F.G.	Cd	1	.070429 $\pm$ 1	.10784 $\pm$ 1	-.00116 $\pm$ 4
Haywood	Cd	1	.147242 $\pm$ 1	.22408 $\pm$ 1	-.00017 $\pm$ 1
Nelkin	Cd	1	.128702 $\pm$ 3	.19530 $\pm$ 4	-.00059 $\pm$ 10
Nelkin	Cd	3	.129101 $\pm$ 1	.19501 $\pm$ 1	-.00029 $\pm$ 1
Expt.	Cd	-	.1356 $\pm$ 5	.202 $\pm$ 2	-.00022 $\pm$ 13
F.G.	Gd	1	.069385 $\pm$ 2	1.2648 $\pm$ 1	.140 $\pm$ 4
Haywood	Gd	1	.145097 $\pm$ 7	2.6000 $\pm$ 5	1.05 $\pm$ 3
Nelkin	Gd	1	.126790 $\pm$ 2	2.2902 $\pm$ 3	.618 $\pm$ 8
Nelkin	Gd	3	.127190 $\pm$ 2	2.2984 $\pm$ 3	.660 $\pm$ 10
Expt.	Gd	-	.1315 $\pm$ 2	2.49 $\pm$ 2	.86 $\pm$ 3

(The errors given are appropriate to the last digit, in each case.) as has been done in Table 7.1 Taking $\sigma_{aH} = .337$, as opposed to .332 barns in the case of cadmium alters the theoretical values for β_1 by about .015% and those for β_2 by about 3%. The β_3 have not been given in Table 7.1, since for both absorbers they are very small and difficult to determine, both experimentally and theoretically. Approximate values for $\beta_3 \times 10^{57}$ in units of cm ^{7} are as follows: the Nelkin model gives .08 for gadolinium and -.000012 for cadmium; the Haywood model .18 and 0 $\pm$.000002. The only acceptable experimental value (with about 100% uncertainty) is -.000025 for cadmium, in reasonable agreement with the Nelkin value.

7.3 Application of Laguerre Polynomial Technique

In order to examine the feasibility of extracting thermalization parameters from experimental points by the Laguerre polynomials method, we shall first carry out some consistency checks using theoretical results. The parameters β_0 , β_1 and β_2 will be calculated by the method of Section 4.6, using values for the γ_{ij} given by Friedman (1964) for the Nelkin model, Friedman's values for the σ_{oj} , and the value for D_0 given in Table 6.1 obtained by fitting a polynomial to the Nelkin model (B_1) poison points. The values obtained for the parameters will then be compared with those quoted in Table 7.1 for B_1 calculations with the Nelkin model.

β_0 :- This is merely the value of κ^2 for pure water (contaminated with boron in the case of cadmium). From equation 4.6.6 we obtain values for β_0 of .1280 and .1300 cm^{-2} for the two cases. Making a correction for the diffusion heating effect in water (equation 4.5.16 with $L=1$) these become .1265 and .1285, in good agreement with the values .1268 and .1287 given in Table 7.1.

β_1 :- From equation 4.6.12 with $L=1$, we find for $\beta_1 \times 10^{19}$ the values .1933 and 2.293 cm for cadmium and gadolinium respectively. The agreement with the values taken from Table 7.1 is 1% in the former case and even closer for

gadolinium. Considering that the values from Table 7.1 depend on the cross-sections as read from the 'Barn Book', and those obtained by the method of Section 4.6 depend on Friedman's tabulation of the σ_{oj} , this agreement is apparently very good. However, it should be noted that the Laguerre polynomials approximation and the truncation to $L=1$ only affect the second term of equation 4.6.12, which contributes only .6% for cadmium and 2.9% for gadolinium to the values of β_1 .

β_2 - Taking first equation 4.6.16, in the derivation of which L has been taken as 1, we obtain for $\beta_2 \times 10^{38}$, .000231 and .727 cm^4 for cadmium and gadolinium respectively. The agreement with the direct parameters (Table 7.1) is poor especially in the case of cadmium where the sign is wrong. Since the term coupling $1/v$ and non- $1/v$ absorption (the term in brackets in equation 4.6.16) contributes only .6 and 6%, respectively, we consider whether equation 4.6.17, in which $1/v$ absorption has been neglected but higher order Laguerre polynomials included, gives better agreement. Since the γ_{ij} are positive it is apparent that in order to obtain a negative β_2 the off-diagonal elements of the matrix γ must play an important part. However the term $2 \frac{u_{o2}}{u_{o1}} \frac{\gamma_{12}}{\gamma_{22}}$ only gives a correction of 15% for cadmium and 6% for gadolinium; further terms in u_{oj}/u_{o1} are even smaller. Inclusion of the $\frac{u_{o2}^2}{D_{oo}^3 \gamma_{22}}$

term, however, more than doubles the value of β_2 for cadmium; with gadolinium the increase is about 20%.

The final results including the terms mentioned are $.00045 \text{ cm}^4$ for cadmium and $.80 \text{ cm}^4$ for gadolinium.

While the latter result is at least of the same order as the direct value ($.62 \text{ cm}^4$), the result for cadmium is now even worse than that given by equation 4.6.16.

With this extremely slow convergence it is apparent that any attempt to derive elements of the γ matrix is doomed.

Friedman (1964), in the analysis of his non- $1/v$ poisoned pulsed system results, retains only the equivalent of

the $\frac{u_{01}^2}{D_{00}^3 \gamma_{11}}$ term. It is then easy to see why the value

for M_2 which he obtained is in such marked disagreement with other estimates.

It is considered unlikely that the use of diffusion theory is responsible for the failure of this method; it will be recalled that the value of M_2 obtained from the $1/v$ results (Section 6.6) through the use of Laguerre polynomials was in good agreement with the Nelkin value.

The difficulty lies in the representation of the flux, which is strongly distorted by the presence of non- $1/v$ absorption, by a Maxwellian multiplied by a polynomial correction. In order to illustrate the problem, the ratio of the scalar flux to a Maxwellian is plotted in

Figure 31 for each of the three absorbers used. Over the important energy range 0-.20 eV, the $1/v$ poison plot is reasonably linear, indicating that a truncation of the polynomial series at $L=1$ (equivalent to the neutron temperature concept) will give good results. In the case of gadolinium, however, at least quadratic terms will be required, while for cadmium the influence of cubic terms is manifest. We therefore conclude that the Laguerre polynomials technique is only practicable in the case of $1/v$ -absorbing systems.

In the discussion of the coefficient β_1 it was seen that the coupling term of equation 4.6.12 is small compared with the leading term. The experimental values for β_1 may then be used to obtain a value for σ_{oo} , and hence an estimate of the Westcott 'g' factor defined by

$$g = \frac{2v_T}{\sqrt{\pi}} \frac{\sigma_{oo}}{v_o \sigma_{ao}} \quad 7.3.1$$

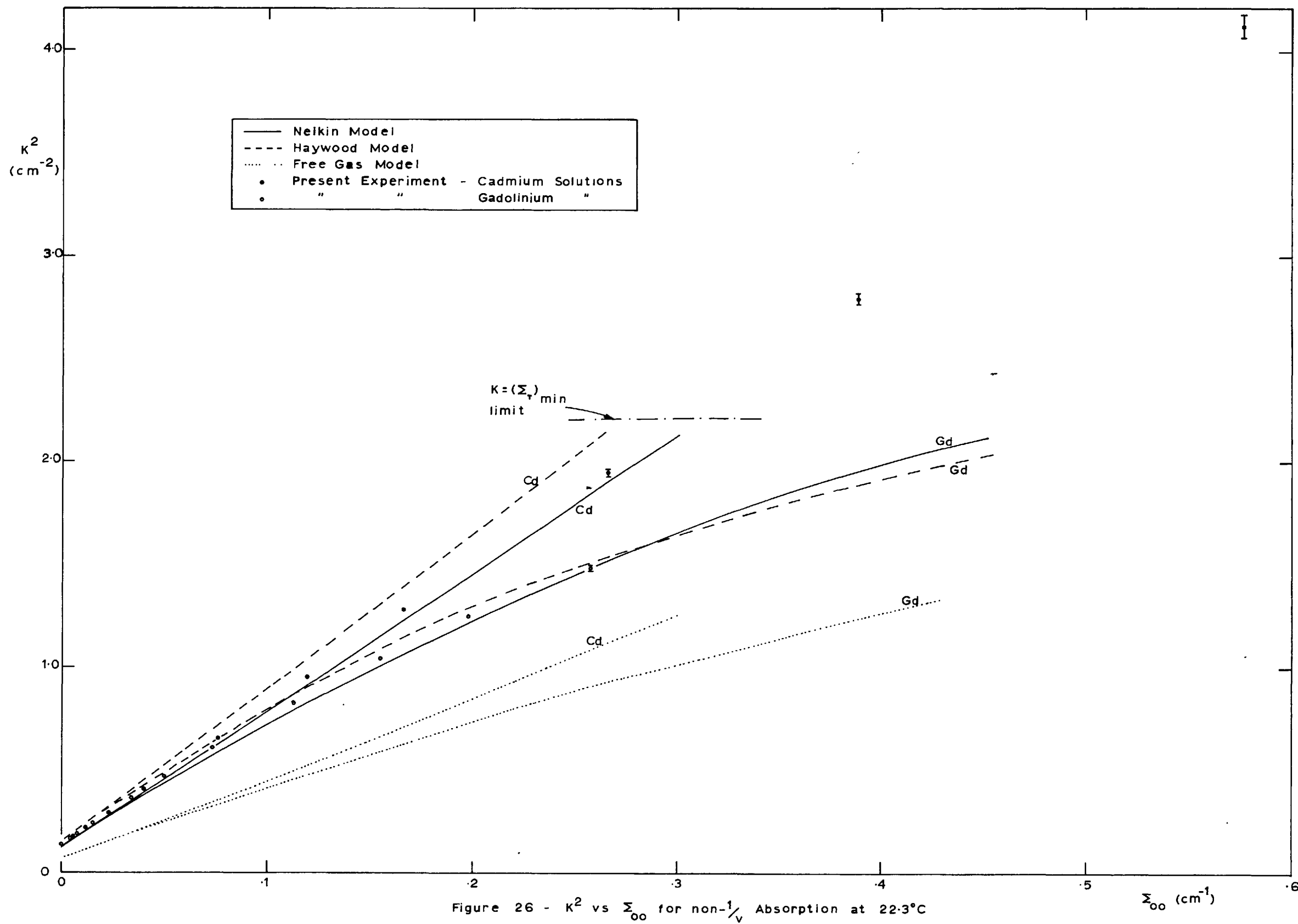
(σ_{ao} is the 2200 m/sec absorption cross-section).

Retaining only the leading term of equation 4.6.12 we find

$$g = \frac{\beta_1 D_o}{v_o \sigma_{ao}} \quad 7.3.2$$

For D_o we take the value $35620 \text{ cm}^2 \text{ sec}^{-1}$ obtained from the boric acid experiments. The values of σ_{ao} , 2452 for cadmium and 46220 barns for gadolinium, are taken from Beckurts and Wirtz (1964). g is then found to be

$1.334 \pm .041$ for cadmium and $0.872 \pm .025$ for gadolinium, in excellent agreement with the values of 1.338 and .888 given by Westcott.



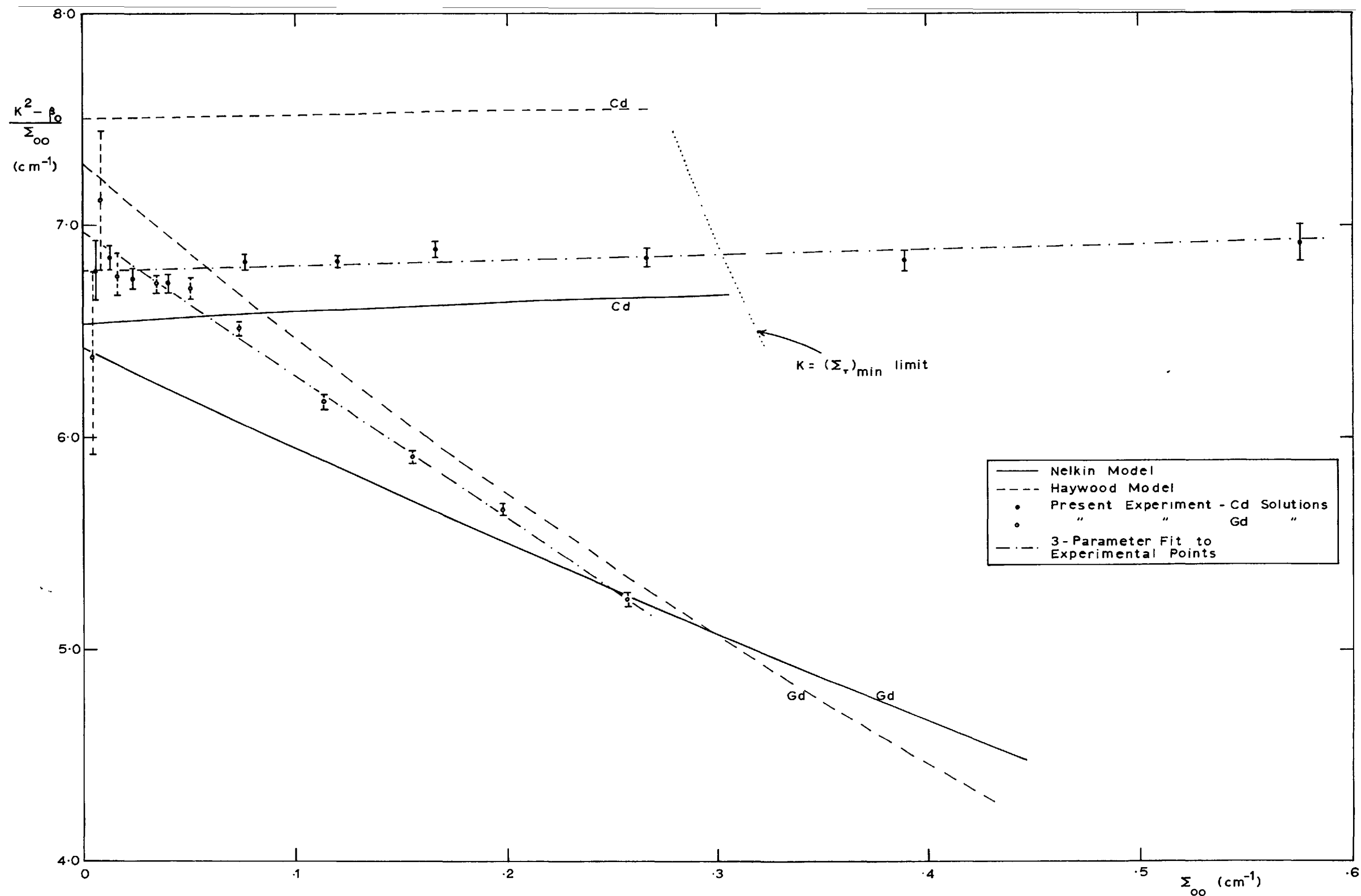


Figure 27 - $\frac{K^2 - \beta_0}{\Sigma_{00}}$ vs Σ_{00} for non- $\frac{1}{v}$ Absorption at 22.3°C

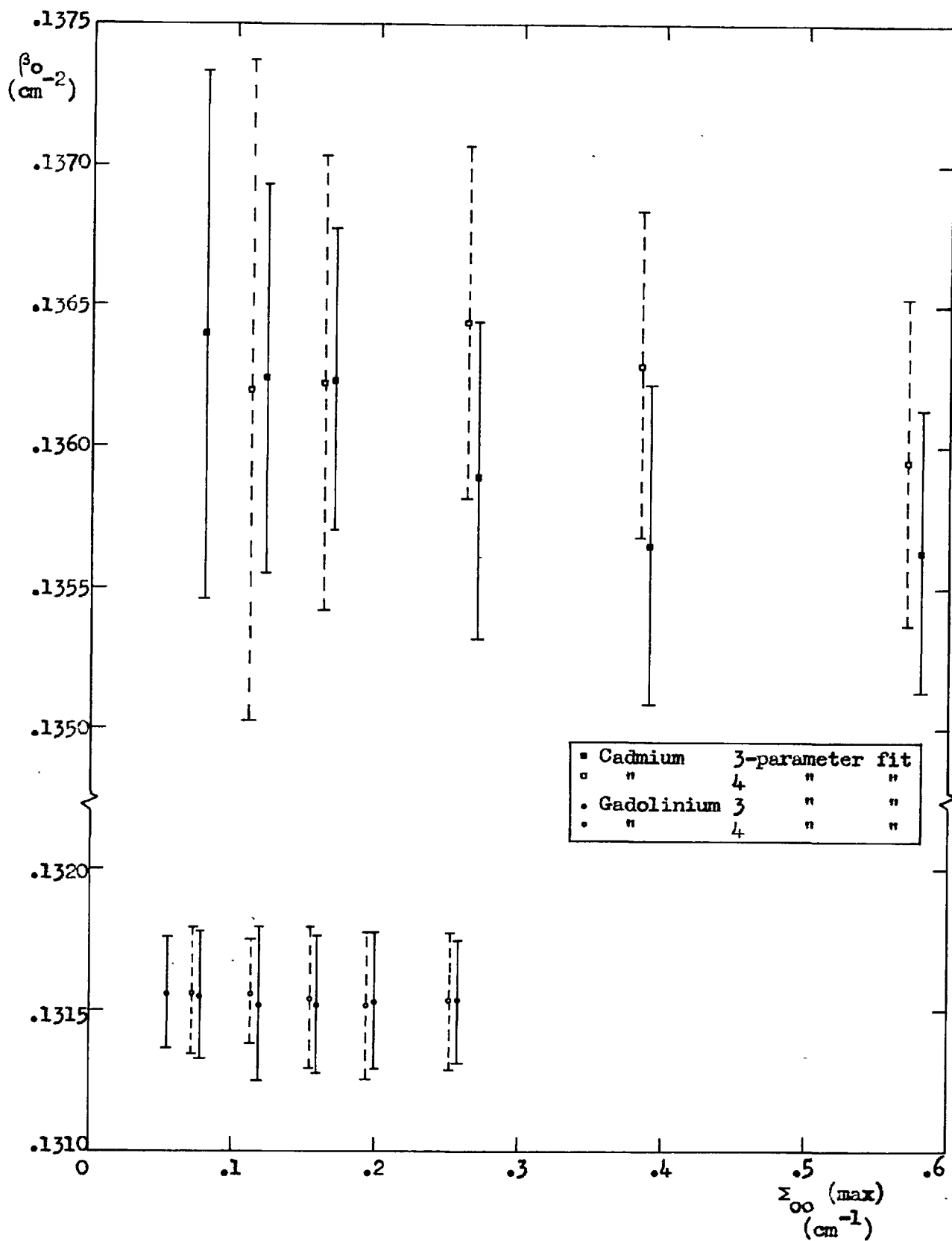


Figure 28 - Effect of Dropping High Poison Points on Fitted Value of β_0

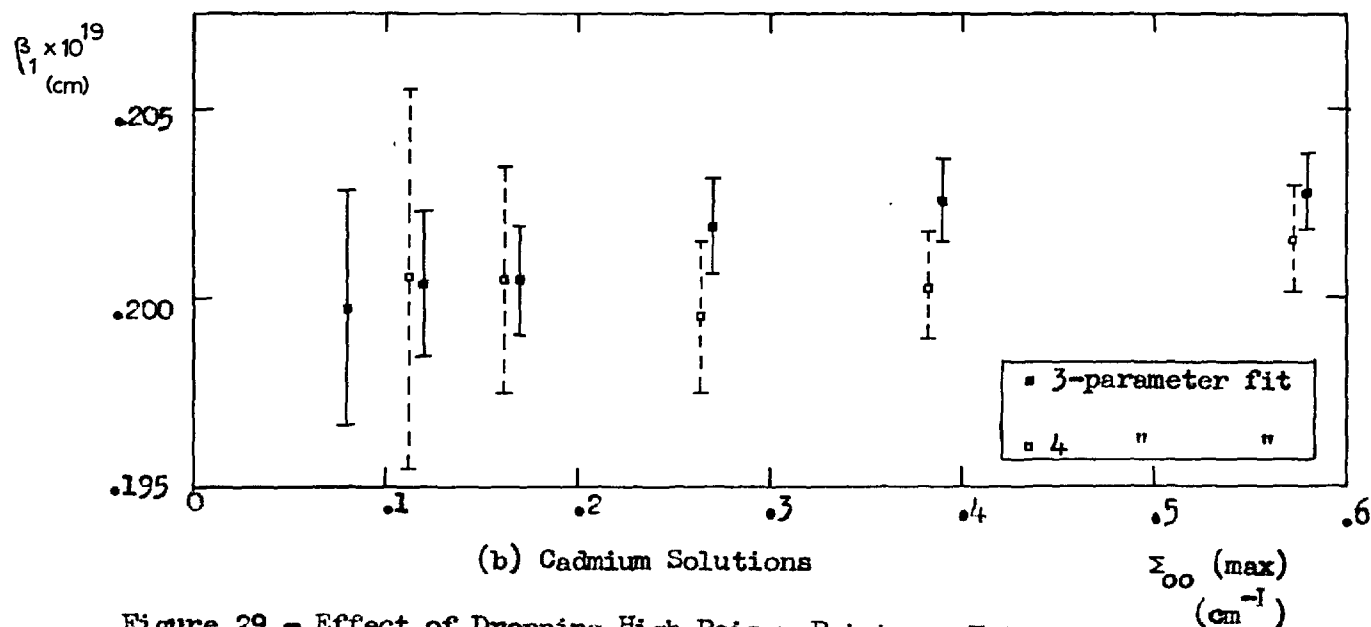
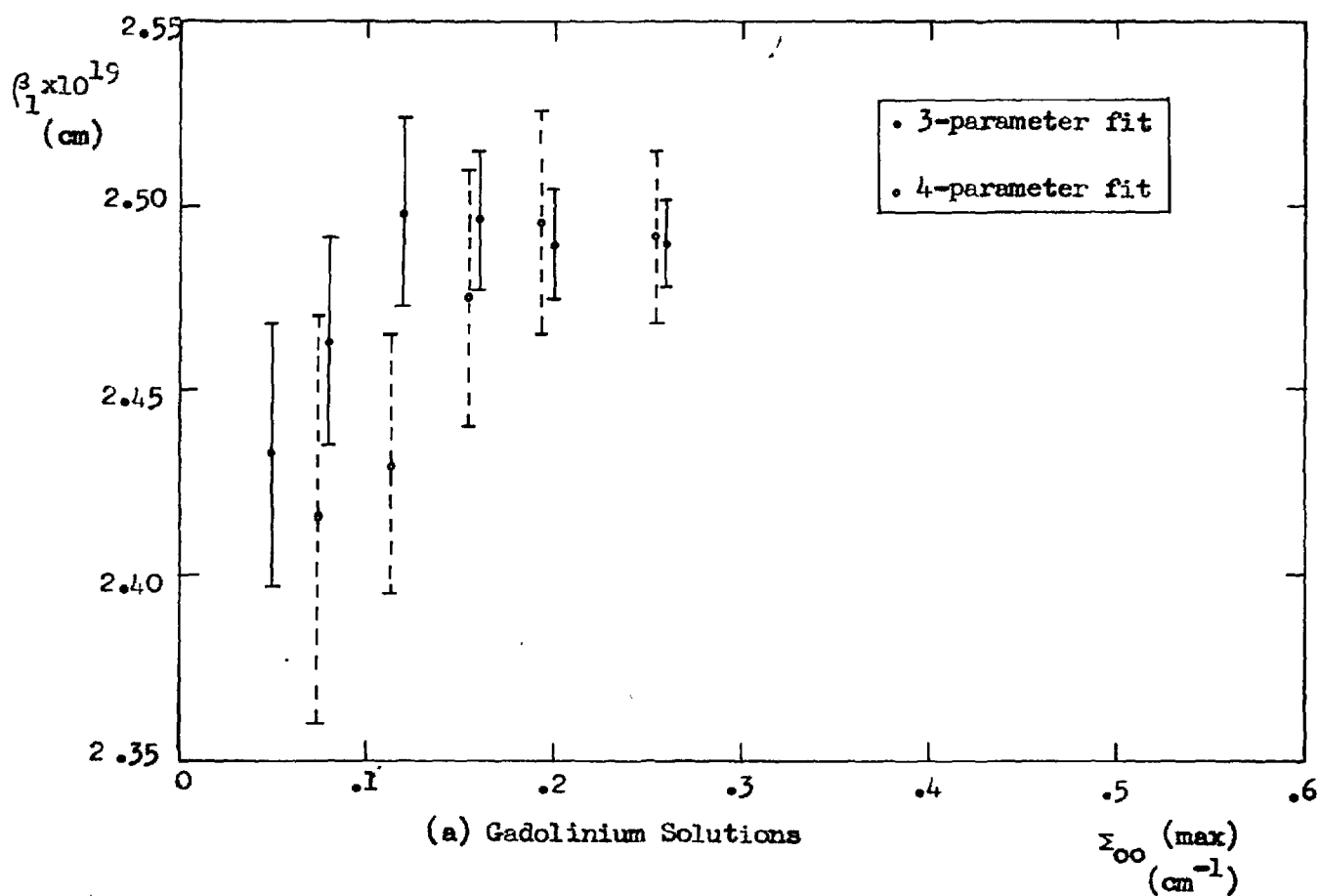


Figure 29 - Effect of Dropping High Poison Points on Fitted Value of β_1

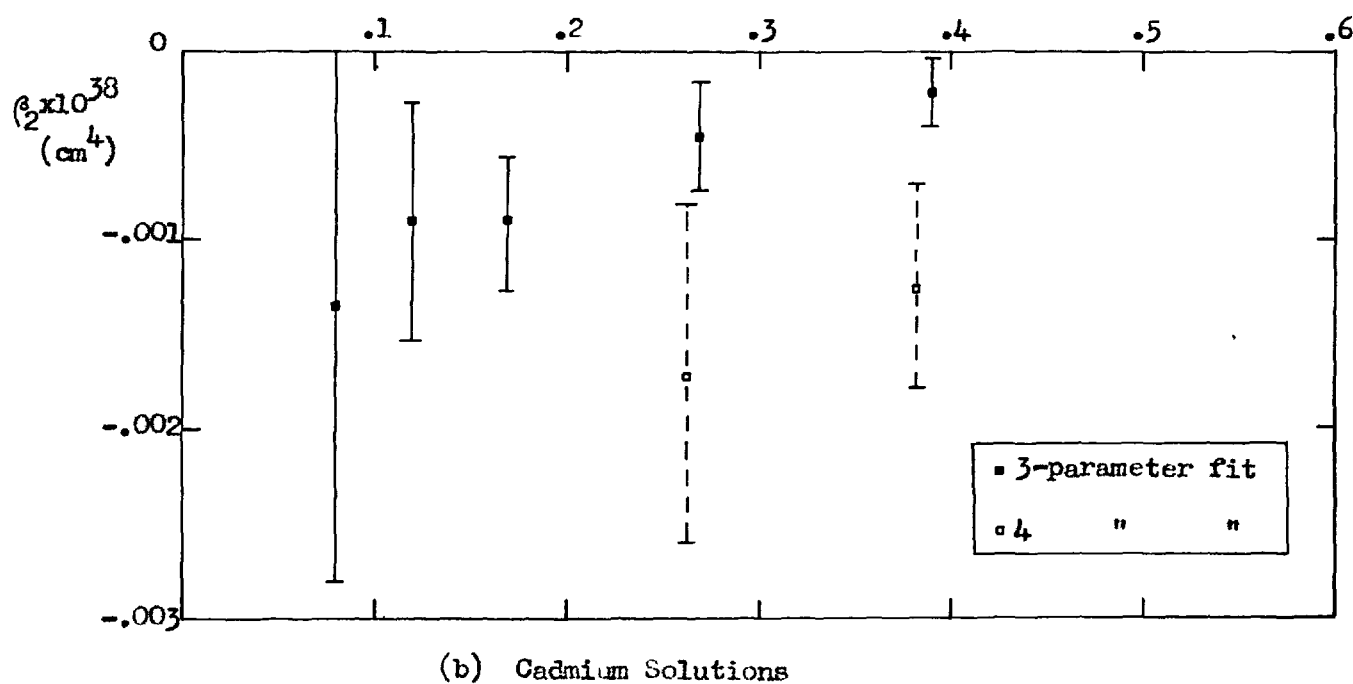
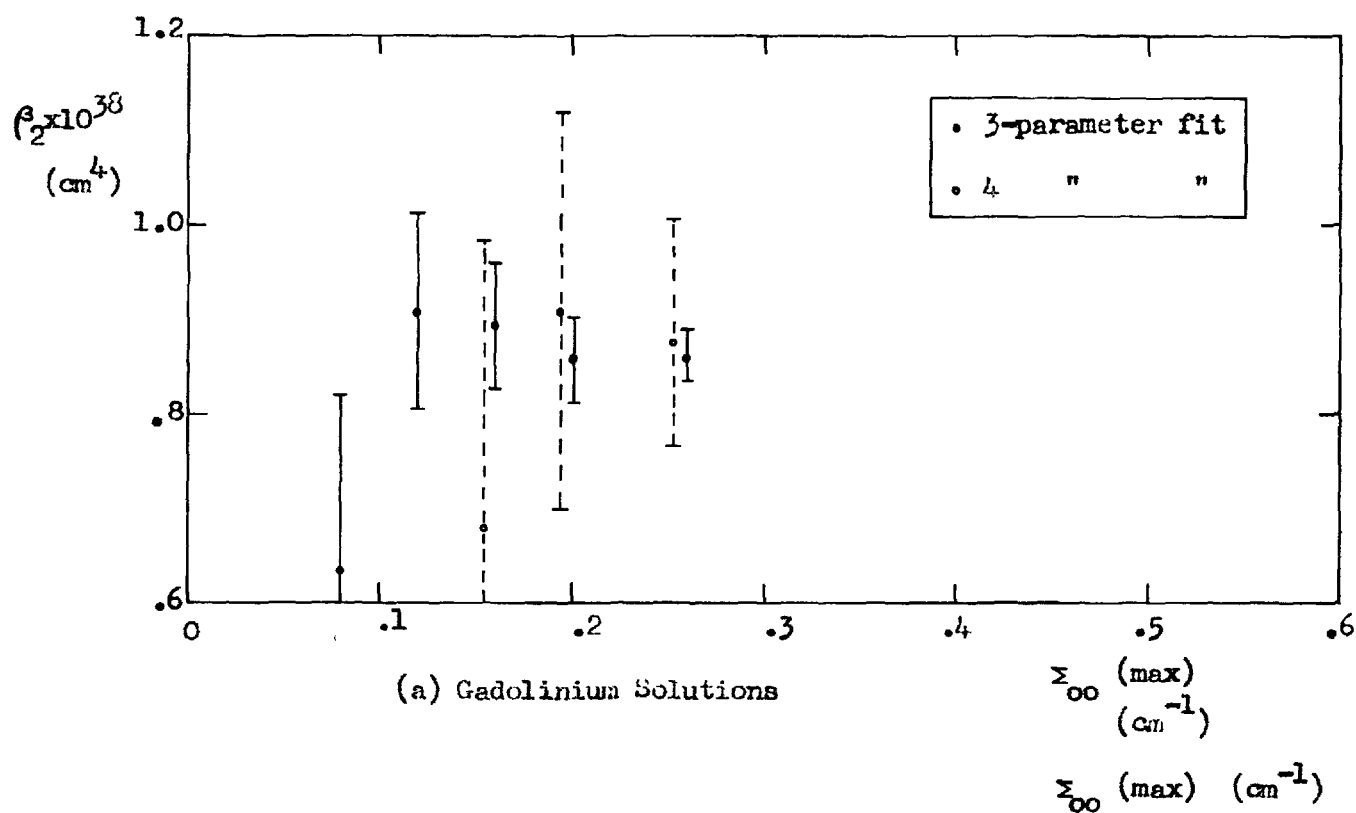


Figure 30 - Effect of Dropping High Poison Points on Fitted Value of β_2

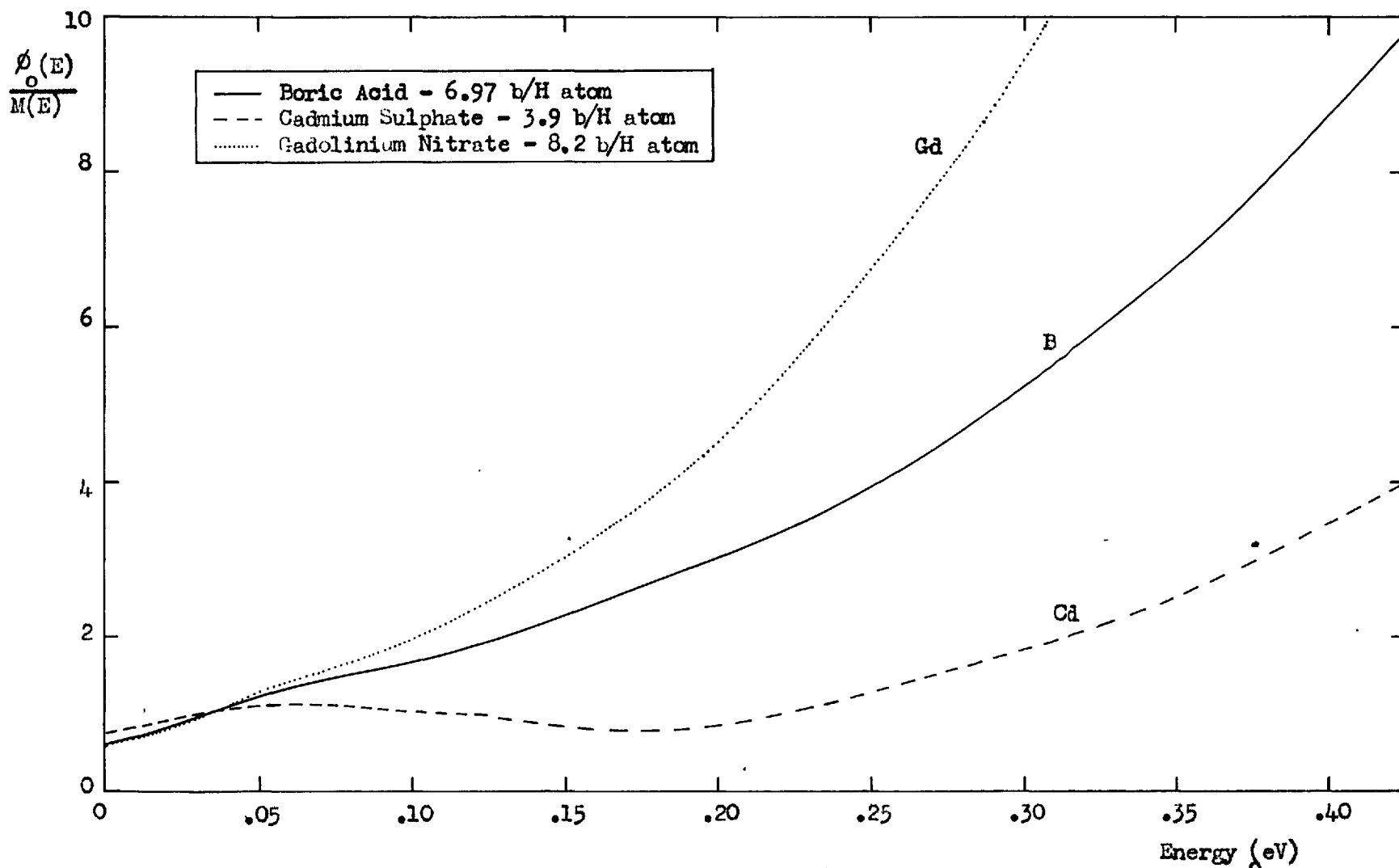


Figure 31 - Ratio Scalar to Maxwellian Flux for Aqueous Solutions at 22.3°C
(Nelkin Model)

CHAPTER 8

CONCLUSIONS

Thermal neutron relaxation lengths have been measured for a large number of absorbing aqueous solutions. The experimental technique used has yielded results with an accuracy of better than $1/3\%$ in all but one case, although larger errors have arisen in the chemical analyses of some of the solutions. The method has been proven by the excellent agreement of the low temperature boric acid results with those of Starr and Koppel (1962). The use of cadmium and gadolinium as absorbers has enabled the observation of contrasting spectral effects; in the former case the resonance absorption produces a slight net cooling of the spectrum, while the absorption in the latter case gives rise to a spectrum heating more marked than that found for a $1/v$ absorber.

It is necessary to consider the effect of the perturbation to the neutron flux caused by the presence of the fission chamber. For the smallest relaxation length measured, the flux decays by a factor of 1.9 in a distance equal to the diameter of the fission chamber. Whatever the perturbation, however, for any position it must be possible to define an effective measuring point, not necessarily coincident with the centre of the counter, at

which the same counting rate would be measured if there were no perturbation. Since the flux gradients in the neighbourhood of the counter will not change as the counter is moved (providing it is sufficiently far from boundaries) owing to the use of a plane source, the position of this effective measuring point relative to the counter will be constant also. The decay being of a pure exponential nature, the relaxation length measured will thus be the same as if no perturbation at all were caused.

The experimental results have been compared with the predictions of three scattering models, the latter being obtained from a rigorous transport theory computer program, DIS. Both series coefficients and point-by-point comparisons have been made, and it is felt that both of these methods serve a useful purpose; in particular the former method enables the comparison of pulsed and stationary experiments. Consideration of polynomial fits involving terms of higher order than the diffusion heating term indicates that even over such a wide range of concentrations as was used in the present experiments, the influence of such terms is swamped by normal experimental errors.

The already known complete inadequacy of the free gas model has been confirmed; the inclusion of chemical binding effects in the two other models gives much better

agreement with experiment. Rather surprisingly, however, the Haywood model, with a much more advanced treatment of the hindered rotations of the water molecule than that given by the Nelkin model, does not give improved agreement. There are indications from the gadolinium results that it is better able to follow spectral effects than is the Nelkin model, but the latter gives a superior total cross-section. The extrapolation of S/α to $\alpha=0$, from which the Haywood frequency distribution $\rho(\beta)$ is derived, is complicated by multiple scattering and interference effects; also the generation of $S(\alpha, \beta)$ from $\rho(\beta)$ involves the harmonic approximation. Another difficulty is that the data extend only up to $\beta=6$; above this the sole criterion is that the normalization condition be satisfied. Recent experiments by Kirouac et al (1966), using a linear electron accelerator and incident neutron energies of 0.83, 1.0, and 1.8 eV, should help to define $\rho(\beta)$ for higher β .

Koppel and Young (1964) have reported an extension of the Nelkin model in which the anisotropic character of the vibrational modes is taken into consideration. Slightly improved agreement with measured total cross-section values around 0.04 - 0.1 eV is obtained, and, surprisingly, considerable improvements below 0.01 eV, where the Nelkin model was thought to be inaccurate because

of the assumption of free translations - an assumption retained by Koppel and Young. More work is needed to elucidate the reason for this. The value of D_0 predicted by this model is 4% lower than that given by the Nelkin model, bringing it closer to the experimental value. Somewhat improved spectra are also obtained.

Recently, McMurtry, Russell and Brugger (1966) have suggested a new model of the scattering process in which water is supposed to exist as a mixture of 10% of free water molecules, together with 45% each of two kinds of aggregate molecules held together by hydrogen bonding, the one having a mass of 75 and the other 150. All molecules exhibit internal vibrations at 0.20, 0.45 and 0.46 eV, the higher two being taken as degenerate, as in the Nelkin and Haywood models. Each aggregate has a set of about four translational modes distributed in the region $0 < E \leq 0.025$ eV, and a similar number of rotational modes in the range $0.025 < E \leq 0.125$ eV. The many parameters involved are optimized to give agreement both with double-differential scattering cross-section data obtained using a velocity selector, and also with the total cross-sections given in the 'Barn Book'. As expected, therefore, the final form of the model gives much better agreement with these quantities than does the Nelkin model. Small discrepancies occur for small α and β values, where the long time behaviour of the aggregates

(assumed to be free from interactions in the model) becomes important.

It will be of great interest to compare the predictions of the McMurry-Russell model with the experimental results reported in this work. It will also be interesting to compute spectra for both this model and the latest Haywood model (used in this work), for pure and poisoned systems fed by a constant source of high-energy neutrons, and to compare the results with experiment. In the present work source-free spectra have been evaluated; since such spectra have not been observed for poisoned systems no comparison with experiment was possible.

Since we are at present far from a complete theoretical understanding of the complex behaviour in a liquid, it is felt that the future scattering model for water will be an extension of the present Haywood model, based on a wider range of experimental data and consideration of anharmonic effects. Until this is available, however, the Nelkin model (with the modifications of Koppel and Young) must be considered as good as any for the representation of integral effects, although more extensive testing of the McMurry-Russell model may well demonstrate the latter's superiority.

In the Introduction a theoretical limit on the value of κ was discussed, and one of the aims of this work was given as attempting to measure values in excess of this

limit. For the boric acid runs, this objective was not achieved, although the final 65°C point was very close to the limit, as can be seen in Figure 20. Diffusion heating prevented very high κ values with the gadolinium concentrations employed, but the two highest cadmium concentrations (see Figures 26 and 27) are well in excess of this limit. The flux distribution for the higher of these is given in Figure 10. Although the range of observation is small, there is no evidence to suggest departure from a pure exponential decay.

In the pulsed case the limiting value of λ , $(v\Sigma_T)_{\min}$, occurs at $v = 0$. Ghatak et al (1965) have pointed out that since there are relatively few neutrons with very low energies, the effect of these may in practice be ignored. They suggest a new limit for crystalline moderators, which occurs at the Bragg peak. Corngold (1965) has criticized this argument on the grounds that one cannot arbitrarily decide to cut off the neutron spectrum at any particular energy; the Boltzmann equation refers to a full range of energies. Where the limit has apparently been exceeded, the eigenvalues form a continuous spectrum, whose amplitude determines the rate of decay.

It is instructive to consider the problem from both of these points of view. In stationary experiments, the limit $(\Sigma_T)_{\min}$ occurs at $v = \infty$. However, if one considers

only neutrons with energies up to .4 eV, the limiting value of κ shown in Figures 26 and 27 more than doubles, increasing from 1.49 to 3.07 cm^{-1} , well above the highest observed value of 2.03 cm^{-1} . It is apparent from Figure 16 that very few neutrons are thereby excluded, and it is unlikely that these neutrons should seriously affect the majority. Nevertheless, in this situation the flux will not be asymptotic - there will be a gradual build-up of higher energy neutrons as the distance from the source is increased. Mathematically, there is no discrete eigenvalue, but the amplitude of the continuum eigenvalue spectrum will be strongly peaked around the value obtained by ignoring high energy neutrons, and it is this value that the experimenter will obtain as a pseudo-discrete eigenvalue.

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APPENDIX 1

DETERMINATION OF THE LARGEST EIGENVALUE OF A MATRIX BY
POWER ITERATION

Given an $n \times n$ matrix $\underline{B}$, we have to find the largest eigenvalue and the corresponding eigenvector of the equation

$$\underline{B} \underline{x}_i = \lambda_i \underline{x}_i \quad \text{A.1.1}$$

Suppose that the eigenvalues λ_i are arranged in descending modulus, i.e.

$$|\lambda_1| > |\lambda_2| \geq |\lambda_3| \geq \dots \quad \text{A.1.2}$$

The problem is then to determine λ_1 and $\underline{x}_1$, the corresponding eigenvector. The method has been described by Hildebrand (1952). We start with a guess vector $\underline{u}_0$, chosen so as to be as close as possible to the expected $\underline{x}_1$. (Note that the subscript on $\underline{u}$ refers to the number of iterations performed; those on λ and $\underline{x}$ refer to a particular solution of equation A.1.1.) In the case of a non-symmetric real matrix whose eigenvalues are real and distinct, the vectors $\underline{x}_i$ are linearly independent. We may then express $\underline{u}_0$ as

$$\underline{u}_0 = \sum_{i=1}^n a_i \underline{x}_i \quad \text{A.1.3}$$

$\underline{u}_0$ is then inserted into the L.H.S. of equation A.1.1, giving

$$\begin{aligned}\underline{A} \underline{u}_0 &= \underline{A} \sum_{i=1}^n a_i \underline{x}_i = \sum_{i=1}^n a_i \lambda_i \underline{x}_i \\ &= \ell_1 \underline{u}_1 \quad (\text{say})\end{aligned}\quad \text{A.1.4}$$

where ℓ_1 is chosen so as to make the largest element of $\underline{u}_1$ unity.

If we now insert $\underline{u}_1$ into the L.H.S. of equation A.1.1 we obtain

$$\underline{A} \underline{u}_1 = \sum_{i=1}^n a_i \lambda_i^2 \underline{x}_i \quad \text{A.1.5}$$

and from this a next approximation $\underline{u}_2$ with largest element unity is derived as before. After p such iterations, we have

$$\underline{u}_p = \frac{1}{\ell_1 \ell_2 \dots \ell_p} \sum_{i=1}^n a_i \lambda_i^p \underline{x}_i \quad \text{A.1.6}$$

For the s th - the largest - elements of $\underline{u}_p$ and $\underline{u}_{p-1}$ we obtain

$$\frac{u_{p,s}}{u_{p-1,s}} = 1 = \frac{1}{\ell_p} \left\{ \frac{\sum_{i=1}^n a_i \lambda_i^p x_{i,s}}{\sum_{i=1}^n a_i \lambda_i^{p-1} x_{i,s}} \right\} \quad \text{A.1.7}$$

$$\therefore \ell_p = \lambda_1 \left\{ \frac{1 + \sum_{i=2}^n b_i \left(\frac{\lambda_i}{\lambda_1}\right)^p}{1 + \sum_{i=2}^n b_i \left(\frac{\lambda_i}{\lambda_1}\right)^{p-1}} \right\} \quad \text{A.1.8}$$

where $b_i = \frac{a_i x_{i,s}}{a_1 x_{1,s}}$.

From equation A.1.2, $\left(\frac{\lambda_i}{\lambda_1}\right)^p \rightarrow 0$ as $p \rightarrow \infty$, and so

λ_1 , the largest eigenvalue. It can be seen from equation A.1.8 that convergence is improved both if the initial guess of $\underline{u}_0$ is a good one, so that the $b_i (i>1)$ are small, and also if the ratio $\left| \frac{\lambda_1}{\lambda_2} \right|$ is large.

Furthermore,

$$\underline{u}_p = \frac{\underline{x}_1 + \sum_{i=2}^n b_i \left(\frac{\lambda_i}{\lambda_1} \right)^p \underline{x}_1}{x_{1,s} + \sum_{i=2}^n b_i \left(\frac{\lambda_i}{\lambda_1} \right)^p x_{i,s}} \quad \text{A.1.9}$$

so that if $\underline{x}_1$ is chosen such that its largest element $x_{1,s}$ is unity, then $\underline{u}_p \rightarrow \underline{x}_1$ for $p \rightarrow \infty$.

APPENDIX 2

PARFIT

PARFIT is a FORTRAN IV subroutine written by the present author, and designed to fit a set of up to 50 experimental observations to a theoretical expression by the method of least squares. This expression may contain 2, 3 or 4 parameters, the values of which are optimized to give the closest fit to the experimental points.

The basis of the procedure is an expression of the theoretical expression in a Taylor series in the parameters to be fitted about the initial guess values of these parameters, keeping first order terms only. The condition that the sum of squares of vertical deviations be a minimum with respect to changes in each parameter, yields a set of linear equations in the parameter increments. The solution of this set enables the initial guesses to be modified by these increments; the process is then repeated until all increments are smaller than convergence criteria specified by the user. PARFIT also calculates standard deviations on the final values of the parameters, and the empirical variance of the observed points from the fitted curve. A subroutine FUNCT must be provided which gives the theoretical expression, and its partial derivatives with respect to each of the parameters.

Consider a function y of several parameters $p_1, p_2 \dots$ and an independent variable x . Values y_i of y have been determined at several points x_i .

The function is expanded in a Taylor series in the parameters about some initial guess values $p_{10}, p_{20} \dots$

$$y(x; p_1, p_2 \dots) = y(x; p_{10}, p_{20} \dots) + \left(\frac{\partial y}{\partial p_1} \right)_o \delta p_1 + \left(\frac{\partial y}{\partial p_2} \right)_o \delta p_2 + \dots$$

A.2.1

The method is to determine the δp_j such that the sum of squares of the deviations of the L.H.S. of this equation from the observed values y_i is a minimum.

The sum of squares is given by

$$S = \sum_i \frac{1}{\sigma_i^2} \{ y_i - y(x_i; p_{10}, p_{20} \dots) - \left(\frac{\partial y}{\partial p_1} \right)_o \delta p_1 - \left(\frac{\partial y}{\partial p_2} \right)_o \delta p_2 - \dots \}^2$$

A.2.2

where σ_i is the standard deviation of observation i .

Equation A.2.2 may be written

$$S = \sum_i (a_i - b_i \beta - c_i \gamma - d_i \delta - e_i \epsilon)^2 \quad \text{A.2.3}$$

where

$$\begin{cases} a_i = \frac{1}{\sigma_i} \{ y_i - y(x_i; p_{10}, p_{20} \dots) \} \\ b_i = \frac{1}{\sigma_i} \left(\frac{\partial y}{\partial p_1} \right)_o \\ c_i = \frac{1}{\sigma_i} \left(\frac{\partial y}{\partial p_2} \right)_o \\ \vdots \\ \vdots \end{cases} \quad \begin{cases} \beta = \delta p_1 \\ \gamma = \delta p_2 \\ \vdots \\ \vdots \end{cases}$$

For the sum of squares to be a minimum, $\frac{\partial S}{\partial \beta} = \frac{\partial S}{\partial \gamma} = \dots = 0$, from which we obtain a system of N linear equations in the δp_j ($j=1, \dots, N$):

$$\left\{ \begin{array}{l} \beta \Sigma b_i^2 + \gamma \Sigma b_i c_i + \delta \Sigma b_i d_i + \epsilon \Sigma b_i e_i = \Sigma a_i b_i \\ \beta \Sigma b_i c_i + \gamma \Sigma c_i^2 + \delta \Sigma c_i d_i + \epsilon \Sigma c_i e_i = \Sigma a_i c_i \\ \vdots \end{array} \right. \quad \text{A.2.4}$$

The set A.2.4 is solved by standard determinantal methods to give the δp_j , and the parameters modified

$$p_j = p_{j0} + \delta p_j \quad j=1, \dots, N \quad \text{A.2.5}$$

The procedure is repeated until all the δp_j are less than specified convergence criteria. When this has been achieved, the summations Σa_i^2 , $\Sigma a_i b_i$ etc. are all repeated using the final values of the p_j , in order to evaluate the standard deviations.

The empirical variance of the fit is defined as

$$EV = \frac{\Sigma a_i^2}{M - N} \quad \text{A.2.6}$$

where M = number of experimental points

and N = number of parameters fitted.

In order to apportion this error between the various parameters the following expression is used (Topping, 1955):

$$\frac{S_\beta^2}{D_{\beta\beta}} = \frac{S_Y^2}{D_{YY}} = \dots = \frac{E.V}{\Delta} \quad \text{A.2.7}$$

where Δ is the determinant of the coefficients of the L.H.S.

of the set A.2.4, and $D_{\beta\beta}$ is the complementary minor of Δ formed by omitting the row and column in which Σb_i^2 appears. $S_\beta, S_Y \dots$ are the standard deviations on the parameters $p_1, p_2 \dots$