1970

An experimental investigation of fission

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Recommended Citation
http://ro.uow.edu.au/theses/2084
AN EXPERIMENTAL INVESTIGATION

OF FISSION

J.W. Boldeman B.Sc. (Hons.)

May 1970
This thesis is submitted for the degree of Doctor of Philosophy to the Wollongong University College in the University of New South Wales, and has not been submitted for a higher degree to any other University or institution.

(JOHN W. BOLDEMAN)

May, 1970.
SUMMARY

Accurate measurements have been made of the average number of prompt neutrons, $\bar{v}_p$, emitted per fission for thermal neutron fission of U233, U235, Pu239 and Pu241 and spontaneous fission of Pu240 and Pu242. The large liquid scintillator method has been used and sources of error in this method have been closely examined. In addition, precise measurements have been made of the probability distribution of neutron emission for each of the six cases above and the spontaneous fission of Cf252. The data have been compared with the neutron emission distribution calculated from fragment kinetic energy data. It is noted that the existing discrepancy between experiment and calculation may be due to gamma ray competition.

The variation of $\bar{v}_p$ with compound excitation has been determined for neutron fission of U233 and U235. The $\bar{v}_p(E_n)$ dependence was found to be linear and the claim of fine structure in some previous experiments has not been substantiated. The variation of the correlated parameter, the average total kinetic energy of the fission fragments ($\bar{E}_K$) with compound excitation for neutron fission of U235 has also been examined. $\bar{E}_K$ was found to be constant in the range measured (0 - 1 MeV) and the lack of structure in $\bar{E}_K(E_n)$ confirmed the linear data for $\bar{v}_p(E_n)$. From an evaluation of the present and existing $\bar{v}_p$ data for U233, U235 and Pu239, it is observed that the $\bar{v}_p(E_n)$ dependence, although linear, is characterised by a change in slope at the pairing energy. The nature of the dependence has been explained in terms of the double
humped fission barrier with the adiabatic assumption of weak coupling of the collective saddle point energy to the nuclear degrees of freedom at scission. The change in the character of the $\tilde{\nu}_p (E_n)$ dependence between U233 and U235 is associated with the change in the relative heights of the two humps of the fission barrier. It is noted that gamma ray competition has a slight effect on the $\tilde{\nu}_p (E_n)$ dependence.

Measurements have been made of the average neutron emission from fission fragments of specific mass for values of the total fragment kinetic energy in the thermal neutron fission of U235. The well known sawtooth curve has been observed and the data confirm the most recent previous investigation.
ACKNOWLEDGMENTS

I would like to thank the Australian Atomic Energy Commission for the award of a scholarship which has allowed the research contained in this thesis to be undertaken.

Many members of the staff at the A.A.E.C. Research Establishment have assisted me during the course of this work. I wish to draw attention to the assistance of my colleagues, Mr. R.L. Walsh, Mr. N.N. Ajitanand and Dr. A.W. Dalton, who helped with several of the experiments. I would also like to acknowledge the co-operation of the HIFAR and MOATA operational staff, the willingness of the Accelerator technical staff to maintain at almost any hour the sometimes cantankerous Van de Graaff, and the technical expertise of the Instrumentation and Control Division who designed some of the electronics. In the analysis of the experimental data, the assistance of Mr. G. Trimble is gratefully acknowledged.

During the course of this study, I have appreciated the support and encouragement of my joint supervisors, Professor A. Keane and Dr. J.L. Symonds. Mr. W. Gemmell, Dr. J.R. Bird and Mr. A. Musgrove have always been prepared to discuss aspects of the research.

Finally, there is that little group who have perhaps the least satisfying task of all - the production of the thesis. I acknowledge the patience and competence of the typist, Miss G. Higgs, and the skill of Mr. D. Hyde and Mr. F. Baker in the preparation of the drawings. Lastly, my wife, in addition to her constant encouragement, proved a very satisfactory reference on matters of grammar.
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Chapter VIII FINAL COMMENTS

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CHAPTER I

1. INTRODUCTION

1.1 History

The nuclear fission process was first discovered by two German radio chemists, Hahn and Strassmann (1939a, 1939b) when they identified the presence of barium isotopes in neutron irradiated samples of uranium. This sensational discovery was immediately interpreted as the division of an excited uranium nucleus into two medium weight nuclei by Meitner and Frisch (1939) and these authors first coined the phrase 'fission'. Confirmation of this correct analysis followed with the identification of other medium weight nuclei in irradiated uranium samples. Meitner and Frisch (1939) from a consideration of the mass deficiencies of elements in the periodic table predicted a large energy release of approximately 200 MeV per fission. Soon afterwards, an energy release of this magnitude was observed by Frisch (1939) and Joliot (1939).

The asymmetric nature of the fission process was first demonstrated by Jentschke and Prankl (1939) from quantitative measurements of the ionisation of the two fragments in a gas chamber. They observed the presence of two energy groups - one situated at approximately 60 MeV energy and the other at 100 MeV. Detailed radiochemical investigations confirmed this by showing the presence in the mass yield curve of two mass groups centering around mass numbers 95 and 138. A consideration of the systematics of nuclear charge density indicated that the initial fission fragments would be neutron rich and unstable towards β decay. Together with
the large excitation energy involved, this suggested to Hahn and Strassmann (1939a) the real probability of neutron emission as a fragment de-excitation mode. Neutron emission was soon observed by several groups amongst whom Halban et al. (1939a, 1939b) and Anderson et al. (1939) might be mentioned.

Shortly after the identification of the main features of the fission process the classical liquid drop model was proposed by Bohr and Wheeler (1939) and independently by Frenkel (1939) to explain the phenomenon. The liquid drop model had some striking success, but failed in several notable aspects, e.g. the asymmetric mass division.

Since the earliest days a variety of different fission models have been proposed - some independent of the liquid drop model and others merely a sophistication of its assumptions. None can claim to have achieved unambiguous success and, at best, our knowledge of the fundamentals of the fission process can be described as fragmentary.

1.2 Fission Models

A complete description of the fission process could be obtained from a solution of the exact Hamiltonian of the nucleus

\[
H = \sum_{i=1}^{A} \frac{P_i^2}{2m} + \frac{1}{2} \sum_{i \neq j}^{A} V_{ij} + E.M. \tag{1.1}
\]

Here \(P_i\) is the momentum of the \(i\)th particle and \(V_{ij}\) is the exact potential of the interaction of the \(i\)th and \(j\)th particles. \(E.M.\) is a term to account for the existence of the electromagnetic field. The
complete solution of equation 1.1 is far too complex for existing mathematical methods even if a full understanding of nucleons and internuclear forces existed. It has been necessary in practice to reduce the degrees of freedom to a number which has been assumed to include the pertinent ones and to attempt a solution of the simplified problem. For example, the exact Hamiltonian in the liquid drop model becomes

$$H = V(\alpha) + T(\dot{\alpha})$$  \hspace{2cm} (1.2)

where $V(\alpha)$ is the potential energy of the analogous liquid drop in terms of a set of deformation parameters, $\alpha$, and $T(\dot{\alpha})$ is the kinetic energy as a function of the time derivative of the deformation variables. In the original version of the liquid drop model the Hamiltonian was approximated even further

$$H = V(\alpha)$$  \hspace{2cm} (1.3)

It is intended in Chapter 2 to discuss various fission models and it will be convenient to group them as follows:

2.1 Liquid Drop Model (Statics, Dynamics, Adiabatic Assumption)

2.2 Statistical Model.
1.3 References

Hahn O., and Strassmann F., (1939a), Naturwiss 27, 11.
Hahn O., and Strassmann F., (1939b), Naturwiss 27, 89.
Jentschke, W., and Frankl F. (1939), Naturwiss 27, 134.
Joliot F. (1939), Compt. Rend. 208, 341, 647.
2. REVIEW OF FISSION MODELS

2.1 Liquid Drop Model

The liquid drop model of fission (henceforth LDM) was first proposed by Bohr and Wheeler (1939) and, independently, by Frenkel (1939). In its original form, the LDM was concerned with the statics of deformed, idealised liquid drops and its principal objective was the understanding of the saddle point configuration. It soon became apparent that a full understanding of this configuration could not be achieved by the model in its simplest form. The realisation followed that the LDM described the average behaviour only of deformed nuclei, and corrections were required for the individual case. The introduction of shell effects has had a dramatic effect upon the understanding of the saddle point. A further difficulty was the inability of the model to account for the asymmetric mass division. This led to the consideration of the dynamics of the model and to assumptions concerning the relation of the saddle point to the scission stage. Out of these assumptions emerged the Adiabatic LDM and the Statistical Model. In the present review of the LDM, the original model will be treated in some detail as subsequent developments retain many elements of the first exposition.

The basic assumptions of the LDM as proposed by Bohr and Wheeler can be stated quite simply. The nucleus is assumed to be equivalent to a liquid drop in which the short range nuclear forces are idealised by the surface tension of the drop and the Coulomb repulsive
forces of the protons are included by assuming the drop to be uniformly charged throughout its volume. The feature of interest is the stability of the nucleus to deformations of various kinds and, in particular, we are interested in that critical deformation for which the nucleus is just on the verge of proceeding to fission. The drop will then possess a shape corresponding to unstable equilibrium: the work required to produce any infinitesimal displacement from the equilibrium configuration vanishes in the first order. If we consider arbitrary distortions of various kinds and plot the potential energy as a function of the parameter specifying the distortion, fig. 2.1, the potential barrier hindering fission will be observed to have similarities to a pass or saddle point between two potential valleys. The critical energy for fission i.e. the fission threshold $E_f$ is the saddle point with lowest potential energy of deformation. If we consider a small distortion of the liquid drop from its spherical shape, the distorted radius is conveniently parameterised as the sum of a series of Legendre Polynomials i.e.

$$R(\theta) = R \left[ 1 + a_0 + a_2 P_2(\cos \theta) + a_3 P_3(\cos \theta) + \ldots \right]$$  \hspace{1cm} 2.1

where the $a_n$ are the deformation parameters. Bohr and Wheeler show that the deformation has increased the surface and electrostatic energy to $E_{S+E}$, where

$$E_{S+E} = 4\pi (r_o A^{3/2} \Omega \left[ 1 + 2a_2^2/5 + 5a_3^2/7 + \ldots + (n-1)(n+2)a_n^2/2(2n+1)+\ldots \right]$$

$$+ 3(Ze)^2/5r_o A^{3/2} \left[ 1 - a_2^2/5 - 10a_3^2/49 - \ldots - 5(n-1)a_n^2/(2n+1)^2 \right]$$  \hspace{1cm} 2.2
Figure 2.1 Potential Barrier to Fission. Simple L.D.M.

Figure 2.2 Plot of calculated fission thresholds from simple L.D.M. Bohr and Wheeler (1939)
and where it has been assumed that the drop is composed of an incompressible fluid of volume \((\frac{4\pi}{3}) R^3 = (\frac{4\pi}{3}) r_o^3 A\) uniformly electrified to charge Ze and possessing a surface tension \(\sigma\). The coefficient of \(a_2^2\) in 2.2 is
\[
4\pi r_o^2 \sigma A^{2/3} \left\{ \left( \frac{2}{5} \right) \left( 1 - \frac{Z^2}{A} \left[ e^2/10(4\pi/3)r_o^3 \sigma \right] \right) \right\}
\]
As the ratio \(\left( \frac{Z^2}{A} \right)\) increases, a limiting value
\[
\left( \frac{Z^2}{A} \right)_{\text{limiting}} = 10 \left\{ \frac{4\pi}{3} \right\} r_o^3 \sigma / e^2 = 50.13
\]
is reached beyond which the nucleus is no longer stable with respect to deformation of the \(P_2\) type. Eqn. 2.4 predicts that all nuclei of \(Z \geq 120\) will be characterised by the absence of a classified barrier towards spontaneous fission. For nuclei where \(Z < 120\) the critical deformation energy \(E_f\), the fission barrier, can be written
\[
E_f = 4\pi r_o^2 \sigma A^{2/3} \left\{ \frac{Z^2/A}{(Z^2/A)_{\text{limiting}}} \right\}
\]
\[
= 4\pi r_o^2 \sigma A^{2/3} \text{f}(x)
\]
\(E_f\) can be determined provided the shape of the nucleus in the critical state is known. This is given by the solution of the well known equation for the form of a surface in equilibrium under the action of a surface tension \(\sigma\) and volume forces described by a potential \(\varphi\)
\[
K \sigma + \varphi = \text{constant}
\]
where \(K\) is the total normal curvature of the surface. Bohr and Wheeler evaluated \(E_f\) for three cases.
1. \( x = 0 \) i.e. no electro-static forces aiding fission. In this case

\[
E_f = 2.4\pi r_0^2 \left( \frac{A}{2} \right)^{2/3} - 4\pi r_0^2 A^{2/3}
\]

and therefore

\[
f(0) = 0.260
\]

2. When the electrostatic force is very small i.e. \( x \) is only slightly larger than zero. The critical shape will approximate that of two spheres in contact and Bohr and Wheeler show that the influence of the connecting neck is quite small. Then

\[
E_f = 2.4\pi r_0^2 \left( \frac{A}{2} \right)^{2/3} - 4\pi r_0^2 A^{2/3} + 2.3 \left( \frac{Ze}{2} \right)^2 / 5r_0 (A/2)^{1/3}
\]

\[
+ \left( \frac{Ze}{2} \right)^2 / 2r_0 \left( \frac{A}{2} \right)^{1/3} - 3(Ze)^2 / 5r_0 A^{1/3}
\]

and therefore

\[
f(x) = 0.260 - 0.215 x
\]

3. When \( x \) is only slightly less than 1. Here, only a small distortion from the spherical shape is required to reach the critical state. The potential energy of deformation will have its smallest values for deformation of the \( P_2 \) type and we can write from equation 2.2

\[
\Delta E_{S+E} = 4\pi r_0^2 A^{2/3} \left[ \frac{2\alpha_2}{5} + \frac{116\alpha_2}{105} + \frac{101\alpha_2}{35} + \frac{2\alpha_2}{35} + \alpha_4 \right]
\]

\[
- \frac{3(Ze)^2}{5r_0 A^{1/3}} \left[ \frac{\alpha_2}{5} + \frac{64\alpha_2}{105} + \frac{58\alpha_2}{35} + \frac{8\alpha_2}{35} + \frac{5\alpha_4}{27} \right]
\]
If the potential energy is minimised with respect to $\alpha_4$

$$\alpha_4 = -\left(\frac{243}{595}\right) \alpha_2^2$$ \hspace{1cm} 2.12

and with this value of $\alpha_4$ in 2.11 it can be shown that $f(x)$ for values of $\frac{2}{A}$ near the instability limit is given by

$$f(x) = \frac{98(1-x)^3}{135} - \frac{11368(1-x)^4}{34425} + \ldots$$ \hspace{1cm} 2.13

Fig. 2.2 is the plot from Bohr and Wheeler of $f(x)$ for various values of $x$, interpolating between the extreme values of $x$ in a reasonable way. The difficulties at this stage in the development of the LDM were the discrepancy between the predicted and experimental fission thresholds, and the inability of the model to account for the asymmetric mass distribution. Subsequent development of the model endeavoured to overcome these problems by the inclusion of additional deformation terms and by calculating $f(x)$ for a wider range of values of $x$.

The power series 2.13 was extended by Present and Knipp (1940) who also included the odd $P_3$ and $P_5$ terms. Present, Reines and Knipp (1946) then included such additional terms as were required, to enable them to determine the saddle point shape and reaction thresholds in the range $1.0 \geq x \geq 0.8$. However the next major advance was the machine calculations of Frankel and Metropolis (1947) for values of $x$ in the range 0.65 - 1.0. They considered axially symmetric distortion conveniently parameterised as follows

$$R'(\mu) = R_0 R(\mu)$$

$$R(\mu) = 1 + \alpha_1 P_1(\mu) + \alpha_2 P_2(\mu) + \ldots$$ \hspace{1cm} 2.14
where \( \mu \) is the cosine of the co-latitude angle.

\( P_i \) is the \( i \)th Legendre Polynomial.

\( R_0 \) is a scale factor determined by the following constant volume condition:

\[
\frac{1}{2} \int_{-1}^{1} (R')^3 \, d\mu = R_0^3 V = 1
\]

\[
V = \frac{1}{2} \int_{-1}^{1} R^3(\mu) \, d\mu
\]

The relative surface energy \( (B_s) \), defined as the ratio of the distorted to the undistorted surface (i.e. \( E_s = B_s E_s^o \)), is given by

\[
B_s = \frac{1}{2} V^{-2/3} \int_{-1}^{1} R \left[ R^2 + (1-\mu^2) \left( \frac{dR}{d\mu} \right)^2 \right]^{1/2} \, d\mu
\]

The relative Coulomb Energy \( (B_c) \), similarly defined, is given by

\[
B_c = \left( \frac{15}{16} \kappa^2 \right) \left[ v^{-5/3} \int \varphi \, d\tau \right]
\]

where \( \varphi = \int \frac{d\tau'}{|r-r'|} = \int_{-1}^{1} \, d\mu' \int_{0}^{R'(\mu')} r'^2 \, dr' \int_{0}^{2\pi} \frac{d\psi'}{|r-r'|} \)

The change in the total energy of the nucleus produced by the deformation is expressed by the quantity \( \xi \), where

\[
\xi = \frac{\Delta E}{E_s^o} = B_s - 1 + 2x(B_c - 1)
\]

Fig. 2.3 shows the fission thresholds calculated by Frankel and Metropolis for nuclei near uranium.
FIG. 2.3. Fission thresholds for nuclei near uranium. Curves for various Z values are given. Data from Frankel and Metropolis (1947).

FIG. 2.4. The energy of deformation corresponding to two spheres in contact as a function of the fractional volume V for various x values. From Frankel and Metropolis (1947).
An interesting feature of the paper by Frankel and Metropolis was their search for an explanation of the asymmetric mass division. They investigated the effect of the addition of various combinations of $P_1$ and $P_3$ to several symmetric expansions. However, they found no evidence that the influences producing the asymmetry of fission are represented in their model. Fig. 2.4 is a plot of the deformation energy of two spheres in contact as a function of the fractional volume $V$ for various $x$ values. The deformation energy has a minimum for symmetric division.

Following Frankel and Metropolis, it became apparent that the large scale deformations of the saddle point liquid drop were more accurately represented as a power series in deviation from the spheroidal shape rather than the spherical shape. A spheroid can be represented by a series

$$R(\Theta) = \frac{R_o}{\lambda} \left[ 1 + \sum_{n=2}^{\infty} \alpha_n P_n \right]$$

where $n$ is restricted to even values and $\lambda$ is a constant which maintains constancy of volume. The values of the coefficients $\alpha_n$ will vary with the eccentricity. Formulae have been developed by Nossoff (1956), Businaro and Gallone (1955), and Swiatecki (1956), (1956), (1958) and (1962), and may be found in these references. Of course, expressions of expansion about a spheroid must reduce to those about a sphere when the eccentricity is reduced to zero.
The most important work in recent years on equilibrium configurations of idealised charged liquid drops has been the definitive calculations of Cohen and Swiatecki (1963). Here, the configuration of the drop, as in previous publications, was parameterised in terms of an expansion of the radius vector in Legendre polynomials e.g. 2.19. The surface and electrostatic energies of the drop were calculated by numerical integrations and, for a given value of the fissionability parameter $x$, the total energy was made stationary with respect to small changes of all $\alpha_n$'s. The behaviour of the family of symmetric equilibrium shapes as a function of $x$ was traced out by starting with known members of the family e.g. $x = 1$ for a sphere and $x = 0$ for two spheres in contact and by decreasing or increasing the value of $x$ in small steps using the known shape as a starting point of the search at the new $x$ value. For each symmetric equilibrium shape, properties such as its surface, electrostatic and total energy, the moments of inertia about different axes and its quadrupole moment were determined. A detailed tabulation of these important properties for values of $x$ between 0.30 and 1.0 is given in Cohen and Swiatecki. It is interesting to compare in fig. 2.5 the calculated necked-in shapes for $x = 0.3, 0.4, 0.5$ and 0.6 with the cylinder like shapes for $x = 0.7, 0.8, 0.9$ and 1.0. In the first case, the overall length of the shapes increases with $x$; in the second, it decreases. The transition from one type of behaviour to the other is fairly rapid as shown in fig. 2.6 where the major and minor axes are plotted against $x$. The transition occurs at about $x = 0.67$ and is accompanied by a rapid change in the diameter of the
FIG. 2-5 Saddle point shapes for various values of $x$. For $x \leq 0.6$, the saddle point shapes have a strong necked-in appearance. For $x \geq 0.7$, the saddle point shapes are more cylindrical. From Cohen and Swiatecki (1963).

FIG. 2-6 The behaviour of the major and minor axes of saddle point shapes as a function of $x$. From Cohen and Swiatecki (1963).
neck connecting the two fragments. The importance of the change at $x = 0.67$ should be emphasised. 'Saddle point shapes for $x < 0.67$ have a large necked-in shape and are quite close to the scission configuration. In other words, the rupture of the nucleus has almost been accomplished at the saddle point. From the shape of the nucleus it is obvious that the coulomb field will make the descent from the saddle point shape to the scission configuration a very rapid process and consequently the saddle point shape will dominate the properties of the fragments. It is for $x < 0.67$ that the LDM has had its greatest success. For elements lighter than Ra226, Nix and Swiatecki (1965) have taken the static picture above, included dynamic considerations and obtained excellent agreement with experimental data for fragment kinetic energies, fragment masses and individual excitation energies. This development by Nix and Swiatecki will be referred to in slightly more detail later in this section.

For nuclei with $x > 0.67$, the liquid drop description of the saddle point shape, even if correct, is not adequate without an accurate description of the saddle point to scission stage. There is the problem of the asymmetric mass distribution. As for the saddle point configuration, the discrepancies between predicted fission thresholds and the experimentally observed values suggest that the model requires refinement. The observed thresholds are far less sensitive to $Z^2/A$ than the LDM would predict.

Let us consider firstly the problem with the fission barriers. The great increase in recent years of our knowledge of the systematics of fission has shown the important role that intrinsic nuclear states
and shell effects play in the fission process. Their effects are as follows.

1. The Anisotropy of the Fission Fragment Angular Distribution.

A Bohr (1956) first suggested that for a fissioning nucleus with excitation only slightly more than the fission threshold, the nucleus at the saddle point is cold with respect to internal excitation, all the energy is bound up in potential energy of deformation and the only nuclear states at the saddle point via which fission may proceed will be collective states similar to those of the heavy deformed nuclei near their ground states. The transition states will be characterised by the quantum numbers I and K where I is the total spin of the compound nucleus and K its projection on the symmetric axis. Fig. 2.7 is reproduced from Griffin (1965) and shows a comparison of the collective states for transition state nuclei and the stable heavy deformed nuclei. Depending upon the angular momentum of the projectile causing fission, the fragment angular distributions will be uniquely determined by the (I, K) quantum numbers of the transition channel. The excellent description of the observed angular distributions that the model achieves (see e.g. Lamphere, 1965) may be taken as a measure of its validity. There have been minor difficulties in recent years with the model in that the predicted magnitudes of the anisotropy for the very heavy nuclei have been significantly less than experiment. This problem has been accounted for in the recent developments referred to later in this section.
Fig. 2.7. Comparison of transition states at the fission barrier with the spectrum of low lying states for heavy deformed nuclei. From Griffin (1965).
2. **The Sawtooth Curve for the Mean Number of Neutrons Emitted from Individual Fission Fragments**

Numerous experiments, e.g. Milton and Fraser (1965), Apalin et al (1965), Maslin et al (1967) have shown the peculiar sawtooth characteristic of the variation of neutron emission with fragment mass. This has been attributed (see e.g. Terrell 1965, Vandenbosch 1964) to the stiffness to deformation of the fission fragments near the closed shells $N = 50$ and $N = 82$, $Z = 50$.

3. **Spontaneous Fission Isomers**

In recent years a large number of short lived spontaneously fissioning nuclei have been produced whose half lives have been far too short to attribute to ground state spontaneous fission (see e.g. Polikanov et al. 1962, Lark et al. 1969, and Vandenbosch and Wolfe 1969). The activities were attributed to the decay of isomeric states. It is difficult from first principles to understand how the spontaneous fission lifetime of an isomeric state could be so short, and yet resist gamma or alpha decay.

4. **Intermediate Structure**

Cross section studies of $^{237}$Np, (Paya et al. 1966) and $^{240}$Pu, (Migneco and Theobald 1968) in the resonance region have shown a remarkable pattern for the fission widths of low energy neutron resonances. For $^{240}$Pu, for example, most of the resonances from 1eV upwards have very small fission widths (much less than 1 meV). There appear however, at intervals of about 600 eV, groups of four or five resonances with fission widths of several meV or even tens of meV.
It is obvious then that an account of the fission process must include shell effects and intrinsic states. Myers and Swiatecki (1966) have presented a semi-empirical theory of nuclear masses and deformations which was based principally on the LDM but included a shell correction term. The two assumptions made in this derivation were

(1) The bumps in the deviation of the nuclear masses from the smooth liquid drop formula are associated with a bunching of energy levels in a spherical nuclear potential - the filling of a bunch corresponding to a closed shell configuration.

(2) The bunching, being associated with the spherical shape of the nuclear potential, will disappear for a sufficiently distorted configuration.

The calculated nuclear masses were in good agreement with experimental values for $N>20$ but the treatment was still unable to reproduce in detail the behaviour of the observed fission barriers. Table 2.1 is a comparison of the experimentally observed fission barriers with those calculated by Myers and Swiatecki and those from a pure liquid drop model. It will be observed that the calculated barriers for elements heavier than thorium are too low and decrease too rapidly with increasing $Z$. 
### Table 2.1
**Comparison of Fission Barriers**

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$x$</th>
<th>Experimental Barrier</th>
<th>L.D.M.</th>
<th>Myers &amp; Swiatecki</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl201</td>
<td>0.6761</td>
<td>22.5</td>
<td>17.438</td>
<td>22.418</td>
</tr>
<tr>
<td>Bi207</td>
<td>0.6914</td>
<td>20.6</td>
<td>14.924</td>
<td>22.145</td>
</tr>
<tr>
<td>Po210</td>
<td>0.6991</td>
<td>18.6</td>
<td>13.763</td>
<td>21.006</td>
</tr>
<tr>
<td>At213</td>
<td>0.7068</td>
<td>15.8</td>
<td>12.674</td>
<td>16.242</td>
</tr>
<tr>
<td>Th232</td>
<td>0.7410</td>
<td>5.95</td>
<td>8.642</td>
<td>5.183</td>
</tr>
<tr>
<td>U233</td>
<td>0.7620</td>
<td>5.49</td>
<td>6.652</td>
<td>4.288</td>
</tr>
<tr>
<td>U235</td>
<td>0.7597</td>
<td>5.75</td>
<td>6.858</td>
<td>4.246</td>
</tr>
<tr>
<td>U238</td>
<td>0.7566</td>
<td>5.80</td>
<td>7.148</td>
<td>4.154</td>
</tr>
<tr>
<td>Pu239</td>
<td>0.7775</td>
<td>5.48</td>
<td>5.432</td>
<td>3.599</td>
</tr>
<tr>
<td>Am241</td>
<td>0.7864</td>
<td>6.00</td>
<td>4.807</td>
<td>3.182</td>
</tr>
<tr>
<td>Cm244</td>
<td>0.7941</td>
<td>4.4</td>
<td>4.309</td>
<td>2.711</td>
</tr>
<tr>
<td>Cf250</td>
<td>0.8097</td>
<td>4.1</td>
<td>3.416</td>
<td>2.417</td>
</tr>
<tr>
<td>Fm254</td>
<td>0.8274</td>
<td>3.5</td>
<td>2.554</td>
<td>1.713</td>
</tr>
</tbody>
</table>
The most important advance in the theoretical description of the fission process in recent years has been brought about by Strutinsky (1967), (1968), (1969a), (1969b). In principle his method is similar to that of Myers and Swiatecki (1966) in that a shell correction term has been added to the usual liquid drop description. However the significant advance relates to a new definition of nuclear shells particularly for deformed nuclei. Nuclear shells were previously regarded as an effect of the degeneracy of the single particle states produced by the sphericity of the nuclear shape. The new definition regards them as a large scale non-uniformity in the distribution of the single particle states. In particular, a nuclear shell corresponds to a low density of single particle states near the Fermi energy. Consequently, shells are a characteristic not only of the spherically symmetric case but may be expected for any type of the average field and shape of the nucleus. Fig. 2.8 reproduced from Strutinsky (1968) is a qualitative picture of the distribution of the single particle states in the deformed nucleus. The circles are low density regions and correspond to shells. It will be noted that there is a regular occurrence of shells with increasing deformation. The specification of the magic properties of nuclei therefore requires not only the $N$ and $Z$ numbers but also the particular deformation. For example, the spherically magic nuclei have $Z$ values 50, 82, 114-120 and $N$ of 50, 82, 126, 184. For deformations of $0.2 - 0.3$ magic numbers are $Z, N = 60 - 64, 100, N = 150 - 152$. It is the closed shells at deformations $0.2 - 0.3$ and $0.5 - 0.6$ which dominate many features of the fission
Fig. 2-8. Qualitative picture of the distribution of single Particle states in a deformed nucleus. The low density regions (shells) are shown by circles. From Strutinsky (1968).
process. Fig. 2.9 shows the variation of the deformation energy of a heavy nucleus with increasing deformation. The potential energy curve has its first minimum at a deformation of approximately 0.2 to 0.3 and this corresponds to the ground state of the heavy deformed nuclei. It is the existence of this potential minimum at a deformation 0.2 - 0.3 which accounts for the stability of the very heavy nuclei against spontaneous fission. With increasing deformation the potential energy increases as in the simple L.D.M. but the shells at a deformation of 0.6 produce another shell minimum in the potential energy. The potential barrier hindering fission is characterised therefore by at least two humps rather than the single one as in the simple L.D.M. The existence of the potential minimum at a deformation of 0.6 and the intrinsic states therein have been used in explaining fission isomerism and intermediate structure. The collective states at the two fission humps are involved in the determination of the fragment angular distributions as in the original A. Bohr theory with minor modifications. In the present discussion it is the influence which these new ideas have on the fission barriers that is our immediate concern.

It can be shown in a qualitative way that the shell effects on the fission barriers reduce the dependence of the barrier heights on the terms of the L.D.M. With respect to the L.D.M. the ground states of the heavy nuclei are lowered by about 3 MeV and the heights of the barriers are increased by a few MeV above the L.D.M. average. This
Figure 2.9 Double Humped Fission Barrier From Strutinsky (1969)
has the effect of flattening the dependence on $Z^2/A$. Quantitative estimates have experienced difficulty in reproducing the experimental barriers, Krappe and Willie (1969). This is not surprising at the present time. Nilsson (1966) has shown that the calculated barrier heights depend strongly on the deformation dependence of the pairing force parameter.

It can be concluded that the fission process to the saddle point appears to be well understood despite some computational problems.

There remains the problem of the asymmetric mass distribution. This appears to be a feature of the saddle point to scission stage. The simple L.D.M. predicts a symmetrical shape at the saddle point and symmetric division at scission. It seems likely that the recent shell correction of the L.D.M. description of fission preserves symmetry at the saddle point. This cannot be stated categorically as quantitative calculations of the saddle point configuration with the new model are extremely difficult and are, for the most part, inconclusive. The starting point of any theoretical description of the saddle point to scission stage must of course be the saddle point itself and the collective modes of motion there deduced from the experimental data. Two extreme assumptions have been made concerning the coupling of these collective modes of motion to the nuclear degrees of freedom at scission. A scheme of fission models reproduced from Swiatecki (1965), fig. 2.10, demonstrates these alternative views. The assumptions are

(i) Weak coupling - Adiabatic Liquid Drop Model
Figure 2.10 Schematic representation of fission models
From Swiatecki (1965)
(ii) Strong coupling - Statistical Model.

In the later model as equilibrium exists at scission the saddle point to scission stage is relatively unimportant and the dynamics of the process do not require consideration. The reverse is true of the Adiabatic L.D.M.

Progress in understanding the dynamics of the saddle point to scission stage has been very poor. This is to be expected, in view of the severe computational difficulties and, in fact, the difficulty of even formulating the problem. What is required is a formulation and solution of the Hamiltonian of the system

\[ H(\alpha, \dot{\alpha}) = T(\alpha, \dot{\alpha}) + V(\alpha) \]

where \( \alpha \) denotes a set of shape parameters. Activity in recent years has been concerned with investigation under simple assumptions of the principles of the problem and the development of methods to assist in the entire problem. Hill (1958) for example has followed the history of a U235 nucleus to scission under the action of a 50 MeV excitation in a \( P_2 \) deformation mode. Kelson (1964) has provided a method for following the motion of an idealised liquid drop using the 'Wheeler Condition', Hill and Wheeler (1953). The liquid flow is regarded as a flow of circular layers of the fluid i.e. all points which are at one time on a plane perpendicular to the symmetry axis will continue to be on that plane. Nix and Swiatecki (1965) have idealised the liquid drop as two overlapping spheroids prior to fission. A comparison of the saddle point shapes of the two spheroid approximation with the calculated shapes from Cohen and Swiatecki (1963) showed that the approximation reproduced the more accurately calculated
configurations for \( x \leq 0.67 \). The two spheroid approximation has been very successful in accounting for many features of the fission process for the lighter heavy nuclei. Hasse et al (1967) have described the liquid surface in cylindrical co-ordinates and calculated the effective masses with respect to those co-ordinates. The kinetic energy takes the form

\[
T = \frac{1}{2} \sum_{ij} m_{ij} \dot{z}_i \dot{z}_j \tag{2.21}
\]

where \( z_i \) and \( z_j \) are the parameters specifying the nuclear shape in cylindrical co-ordinates.

Adiabatic features can be introduced into the model by the inclusion of collective restraints on the solutions of the complete many body Hamiltonian. Collective dynamics can be introduced through the cranking model which yields an expression for the associated mass parameter to replace those evaluated previously from the L.D.M. Damgaard et al (1969) have used the cranking model to investigate the effects of intrinsic structure on the effective masses and inertia parameters.

It is apparent from this discussion that the theoretical description of the saddle point to scission stage is still in its infancy. The most important requirement at the present time, since the model cannot be adequately worked out, is external data justifying the underlying assumption - namely weak coupling of the collective and nucleonic degrees of freedom.
2.2 The Statistical Model

The alternative assumption of strong coupling leads to the Statistical Model. The statistical theory of nuclear fission was first propounded by Fong (1953), (1956). It was proposed that fission is a slow process and that the time between the saddle point and scission is long compared with the time required for a nucleon to cross the nucleus many times. The observed features of fission then would be properties of the late stages of fission and would not be influenced by the saddle point configuration. It is assumed in the statistical model that statistical equilibrium exists at scission and that the probability of occurrence of a particular fission mode is proportional to the corresponding density of quantum states. These states are the excitation and translational states of the two nuclei. The total density of states \( Q(E_T) \) may be written

\[
Q(E_T) = \int_0^{E_T} \Omega(E) \omega(E_T - E) \, dE
\]

where \( \Omega(E) \) is the density of excitation states

\( \omega(E_T - E) \) is the density of momentum states

and \( E_T \) is the total energy available for excitation and relative kinetic energy.

The solution of eqn. 2.22 proceeds in three stages

i. The evaluation of \( E_T \) for various nuclear divisions and various deformations of the two fragments.

ii. An adequate evaluation of the level densities.

iii. A reasonable estimate of the division of the total energy \( E_T \) between excitation and relative translation of the two fragments.
The total energy release for fission into two fragments \((A_1, Z_1)\) and \((A_2, Z_2)\) is given by

\[
F = M^* (A, Z) - M (A_1 Z_1) - M (A_2 Z_2)
\]

where \(M^*\) is the mass of the excited compound nucleus undergoing fission

and \(M(A_1, Z_1)\) are the masses of the primary fission fragments in their ground states.

The total energy \(F\) at the scission point is shared between the coulomb energy of the two deformed nuclei in contact \(C(=C_1+C_2)\), the deformation energy of the two fragments \(D(=D_1+D_2)\) and the sum of the internal excitation energy of the two fragments and their relative kinetic energies \(E_T\). Pong calculated the total coulomb and deformation energy of the two fragments from the liquid drop model assuming \(P_3\) deformations only

\[
C(\alpha_{31}, \alpha_{32}) = \frac{Z_1 Z_2 e^2}{[r_{01}(1+0.9314 \alpha_{31})+r_{02}(1+0.9314 \alpha_{32})]}
\]

\[
D(\alpha_{31}, \alpha_{32}) = \sum (0.7143 \alpha_{31}^2 E_{si}^0 - 0.2041 \alpha_{31}^2 E_{ci}^0)
\]

where \(r_{0i}\) is the undistorted radius of fragment \(i\)

\(E_{si}^0\) is the undeformed surface energy of fragment \(i\)

\(E_{ci}^0\) is the undeformed electrostatic energy of fragment \(i\)

We have

\[
E_T = F - C(\alpha_{31}, \alpha_{32}) - D(\alpha_{31}, \alpha_{32})
\]

From the argument that the fission mode probability is proportional to the density of quantum states, the probability is highest for largest \(E_T\) and therefore, for a particular mass division, the most probable coulomb and deformation energy is obtained by minimising the sum.
C(a_{31}, a_{32}) + D(a_{31}, a_{32}).

ii. The energy level density formula assumed by Fong for spin zero nuclei were of the form

\[ \omega(E) = c \exp \left\{ \frac{1}{2} \right\} \]

The parameters a and c are functions of the mass number A and were evaluated by Fong. He assumed a smooth variation and set \( a = 0.050A \) and \( c = 0.38 \exp (-0.005A) \). Even-odd, shell and deformation effects were ignored. He assumed that these effects were already suitably contained within the mass formula. Thus, the density of excitation states of the two nucleon system may be written

\[ \Omega_0(E) = \int_0^E c_1 \exp \left[ 2(a_1 E_1)^2 \right] c_2 \exp \left[ 2(a_2 |E-E_1|)^2 \right] dE_1 \]

Eqn. 2.28 represents the density of states for spin zero nuclei. To include excitation states for higher angular momentum, Fong used the formula given by Bethe (1937).

\[ \Omega_j(E) = (2j+1) \exp \left[ \frac{1}{2} \right] \Omega_0(E) \]

where

\[ g = \frac{2}{5} (MR^2/\hbar^2) \sim A^{5/3} \]

\[ T = \left( \frac{E}{a} \right)^{1/2} \]

The total density of excitation states can be shown to be

\[ \Omega(E) \sim c_1 c_2 \left( \frac{A_1^{5/3} A_2^{5/3}}{A_1^{5/2} + A_2^{5/3}} \right)^{3/2} \left( \frac{(a_1 a_2)^2}{(a_1 + a_2)^2} \right) E^{3/2} \]
The density of momentum states of the two nucleon system is given by

\[
\omega(E_T - E) = \frac{4\pi V}{h^3} \left[ 2\mu^3(E_T - E) \right]^{1/2}
\]

where \( \mu \) is the reduced mass.

Eqn. 2.22 can be rewritten as

\[
Q(E_T) = \int_0^{E_T} \frac{c_1 c_2}{c_1^5/3 + c_2^5/3} \left( \frac{A_1^{5/3} A_2^{5/3}}{A_1^{5/3} + A_2^{5/3}} \right)^{3/2} \frac{1}{(a_1 + a_2)^2} E^{3/2} \times \exp \left\{ 2 \left[ (a_1 + a_2) E \right]^{1/2} \right\} \left[ 2\mu^3(E_T - E) \right]^{1/2} dE
\]

iii. The most probable partition of the total energy \( E_T \) can be determined by maximising the integrand of eqn. 2.22. Fong obtained for the most probable value \( k_o \) of the translational energy

\[
k_o = \frac{1}{2} \left( \frac{E_T}{a_1 + a_2} \right)^{1/2} \left( 1 - \frac{7}{4} \frac{1}{(a_1 + a_2) E_T^{1/2}} \right) + \ldots
\]

From eqn. 2.33 it can be seen that \( k_o \) is only a very small proportion of \( E_T \) and Fong in his treatment replaces the variable energy term \( (E_T - E) \) in the level density for the momentum states by the energy \( k_o \).

Fong has integrated eqn. 2.32 and obtains for the probability of fission proceeding through the channel \( (a_{31}, a_{32}, A_1, A_2, Z_1, Z_2) \)
The mass distribution and charge distribution can be obtained from eqn. 2.33 by summing over the appropriate parameters.

Fong obtained excellent agreement with experimental mass yield data existing for thermal neutron fission of U235 at that time. However, in the same year, Perring and Story (1955) using the same input data as Fong obtained a four humped mass distribution for Pu239. Perring and Story concluded that the calculated mass distribution was a particularly sensitive function of the input data. Furthermore they were somewhat critical of the corrections Fong applied to the semi empirical mass formula. Subsequent experimental data on the mass yield curve showed fine structure which was not reproduced in the predictions of the Fong theory. The predicted kinetic energy distributions showed a peak at symmetric fission in variance with later experimental data.

The appropriate level density formulae were re-examined by Newton (1956a), and Cameron (1958a). They showed that shell effects play a very important role in the functional form of the level density formula. Newton (1956) showed that, if the correct level densities
were used in Fong's formula, then the experimental mass distributions were not produced. One criticism that has been made of Fong's derivation from the statistical theory viewpoint was the extensive use made of the liquid drop model, particularly in determining the coulomb and deformation energy of the touching fragments. Newton (1956) has handled the problem of determining the effects of coulomb distortion in a different way. His approach uses the formalism of the nuclear reaction theory of Wigner and Eisenbud (1947). In the Wigner-Eisenbud theory, configuration space is divided into internal and external regions. The internal region is characterised by the condition that all the particles are relatively close together and interact with each other via coulomb and nuclear forces. In the external region, the system may appear as a large number of pairs of separated components which interact with each other via their coulomb fields. Each different representation is termed a channel. The probability of decay of the compound system via the fission channel \( s \) may be written

\[
\Gamma_s = \frac{2k_s R_s \gamma_s^2}{F_s^2 + G_s^2}
\]

where \( k_s \) is the relative wave number of the fragment pair at infinity, \( R_s \) is the smallest radius at which separation from internal conditions occurs, \( \gamma_s^2 \) is the reduced width of the fission mode, \( F_s \) and \( G_s \) are the regular and irregular external wave functions.
\((F_s^2 + G_s^2)^{-1}\) is often called the barrier penetrability. The problem with this approach lies in the definition of the radius \(R\). Newton adopted the simple procedure of treating \(R\) as a parameter to be determined. The probability of fission proceeding via the channel \((Z_1 Z_2 A_1 A_2 E_1 k)\) where the wave number of the two fragments lies between \(k\) and \(k+dk\) and the excitation of fragment 1 lies between \(E_1\) and \(E_1+dE_2\) is given by

\[
\Gamma(Z_1 Z_2 A_1 A_2 E_1 k) dE_1 dk = \frac{2kY^2 R}{F^2 + G^2} \omega_1(E_1) \omega_2(E_2) dE_1 k^2 dk
\]

Newton assumed that the reduced width is largely independent of the fission mode. It is possible then to write for the yield of the \((A_1 Z_1) (A_2 Z_2)\) configuration

\[
Y(Z_1 A_1 Z_2 A_2) = \text{const} \int_0^{E_T} dE_K \int_0^{E_T - E_K} dE_1 \frac{R \mu^2 E_K}{1 + \frac{R \mu^2 E_K}{E_T - E_K - E_1}} \omega_1(E_1) \omega_2(E_T - E_K - E_1)
\]

where \(E_K = \frac{k^2}{2 \mu}\)

Newton was not able to reproduce the experimental data using his formula. He concluded

1. There is a significant variation of the reduced width with fission mode.
2. There is a significant variation of the reduced width with energy.

Wilets (1964) has pointed out that the similarity of the results produced by both Fong and Newton should be expected as the level density formulae dominate the fission probabilities.
Cameron (1958) was led to the same conclusion as Newton, i.e. that the reduced widths are apparently functions of the fission mode, of the excitation energy and possibly other factors. He suggested this inherent difficulty might be overcome as follows.

The values of the reduced widths are again assumed to be constant and the effects that their variation would have introduced are included by allowing $R$ the radius of the boundary between the internal and external regions of configuration space to become a free parameter dependent on the mass ratio. Furthermore the reduced width dependence on excitation energy can also be included in a dependence of $R$ on the excitation energy. Cameron wrote the radius in the form

$$ R = R_0 + a \left( E_T - E_K \right)^{3/2} $$

The value of the parameter $a$ (and the value of the power index) was obtained by fitting the half width of the kinetic energy distributions for the thermal neutron fission of U235 and the spontaneous fission of Cf252. A value of $a = 0.0055 \text{ fermi MeV}^{-3/2}$ was obtained. The non-energy dependent part $R_0$, of the scission radius was used as an adjustable parameter to fit the mass yield curve for thermal neutron fission of U235. The required variation of $R_0$ with mass division as derived by Cameron is shown in fig. 2.11. The curve for $R_0$ has a variation similar to the mass yield curve. To obtain a large dip at symmetric division in the mass yield curve it has been necessary to reduce the separation of the fragment centres and therefore to
Figure 2.11 Plot of term $R_0$ in the separation radius of the fission fragments. From Cameron (1958)
increase the total coulomb energy. Increasing the coulomb energy decreases the excitation energy, and hence the number of quantum states. Thus there is a smaller symmetric fission yield. Cameron then used his model to predict the total kinetic energy as a function of the mass ratio. A comparison is shown in Fig. 2.12 with experimental data. The agreement is not particularly good.

An important discussion of the applicability of the Statistical Model to the fission process appears in the paper by Ericson (1960). His derivation of the fission mode probabilities is similar to that of Newton (1956) but warrants consideration. From the principle of detailed balance the transition probability from a state a to a state b, \( w_{ab} \), is related to the transition probability from state b to state a, \( w_{ba} \).

\[
\rho_a w_{ab} = \rho_b w_{ba}^* 
\]

where \( \rho_a \) and \( \rho_b \) are the densities of states a and b and the star on \( w_{ba} \) indicates the time reversed transition i.e. the transition in which all velocities and orbital angular momenta have changed sign. Applying this principle to the fission process we can write for the probability of decay per unit time of a compound nucleus with spin I and a level density \( \rho_c(I) \) into two excited nuclei of spins \( j_1 \) and \( j_2 \), excitation energies \( E_1^* \) and \( E_2^* \), and level densities \( \rho_1(E_1^*, j_1) \) and \( \rho_2(E_2^*, j_2) \)

\[
P_{12}(I;E,n)d\ Omega dE = \frac{1}{\rho_c(I)} \frac{p^2 dp d\Omega}{h^3} v_{12} \int \delta(n, \ell) T_{\ell}^{12}(E) d^3 \ell \ X
\]
Figure 2.12 The calculated average total fragment kinetic energy for different mass divisions compared with data from chapter 7. From Cameron (1958).

Figure 2.13 The deformation parameter $D_2$ as a function of mass number. The data used in Fong (1956) is also shown. From Fong (1963).
\[
X \int \int \int \int \delta^3 (\ell + j_1 + j_2 - I) \rho_1 (E_1^*, j_1) \rho_2 (E_2^*, j_2) X
\]
\[
X \delta (E^* - E_1^* - E_2^*) \ dE_1^* dE_2^* d^3 j_1 d^3 j_2
\]

where \( p \) is the relative momentum of the two fragments

\( v_{12} \) is the relative velocity

\( \delta (n, \ell) \) is the Dirac delta function

\( n \) is the direction of decay

\( \ell \) is the orbital angular momentum of the system

\( T_{12} \) is the coulomb penetrability of the two fragments

\( \delta^3 (\ell + j_1 + j_2 - I) \) is the ordinary three dimensional Dirac delta function.

The level densities can be written in terms of the level densities for zero angular momentum employing the normal formula for distributions of nuclear spin, i.e.

\[
\rho_1 (E_1^*, j_1) = \rho_0 (E_1^*) \exp \left( -\frac{j_1^2}{2\sigma^2} \right)
\]

where \( \sigma^2 = \frac{\mathcal{J} T}{\hbar^2} \)

\( \mathcal{J} \) is the moment of inertia of the fragment

\( T \) the temperature.

Introducing the variable \( j = j_1 + j_2 \) Ericson integrates 2.40 to the form
\[
P_{12}(\ell;E,n)dE\,d\Omega = \frac{dp\,d\Omega}{\mathcal{C}(I)4\pi^2\hbar} \int_0^\infty \int_0^\infty \left(\frac{2\pi\sigma_1^2\sigma_2^2}{\sigma_1^2 + \sigma_2^2}\right)^{3/2} X \\
\times \int_0^{E_1^*} \int_0^{E_2^*} \delta(E_1^* - E) \delta(E_2^* - E) \, dE_1^* \, dE_2^* X \\
\times \int \int T_{\ell}^{12}(E)\delta(n,\ell) d^3\Omega d^3(\ell - I) \exp \left( \frac{-j^2}{2(\sigma_1^2 + \sigma_2^2)} \right) d^3j \tag{2.42}
\]

The angular momentum barrier for fission is very small and it is the importance of the \(\sigma_1^2\) and \(\sigma_2^2\) factors rather than the penetrabilities which restrict the angular momentum. Therefore to a first approximation it is reasonable to replace the penetrabilities \(T^{12}_{\ell}\) by their value at \(\ell = 0\). The last part of the equation can now be written

\[
\int \int T_{\ell}^{12}(E)\delta(n,\ell) d^3\Omega d^3(\ell - I) \times \exp \left( \frac{-j^2}{2(\sigma_1^2 + \sigma_2^2)} \right) d^3\ell
\tag{2.43}
\]

This clearly demonstrates that the direction of emission \(n\) is preferably perpendicular to \(\ell\), the spin of the compound nucleus.

It is possible to derive an expression for the angular distribution of the fission fragments from equation 2.43 for those cases in which the anisotropy is most marked i.e. for near threshold fission of a zero or near zero ground state spin nucleus. Here the spin of the compound nucleus is due entirely to the spin of the incident projectile and we can average over all directions perpendicular to the beam direction i.e. over the azimuthal angle \(\varphi\). The part of equation 2.43 containing the angular correlation becomes accordingly
The predicted fragment angular distribution has the exact form as that derived by Halpern and Strutinsky (1958) from the Bohr (1956) collective model. This is a most important result as it indicates that the anisotropy of the fission fragment angular distribution can be accounted for within the framework of the Statistical Model. The fragment angular distributions do not therefore allow a choice to be made between the Adiabatic Model and the Statistical Model.

The relative probability of various mass divisions has been calculated from Ericson's formula by Erba et al (1964). They assumed the level densities to be given by

$$\rho(E^*) = E^{2} \exp \left( 2 \sqrt{\alpha E^*} \right)$$

where the $\alpha$ values were those obtained in a previous publication, Erba et al (1963). The coulomb penetrability was assumed to be empirically represented by the formula

$$T(E) = \left[ 1 + \exp \left( \frac{\alpha (B-E)}{BE} \right) \right]^{-1}$$

where $B = e^2 Z_1 Z_2 / r_o A^{1/3}$

$$\alpha = 180$$
Erba et al reported excellent agreement between the predicted mass distributions for the thermal neutron fission of U235 and the experimental data. The authors also claim equally good agreement for the case of Pu239 (unpublished).

Fong (1963) has reviewed his early work and endeavoured to improve the method. In his original derivation the fragment deformations considered were assumed to be of the $P_3$ type i.e. the scission radius of a particular fragment was given by

$$ R = R_0 \left[ 1 + \alpha_3 P_3 \left( \cos \theta \right) \right] $$

and the deformation energy (see equation 2.25) by $\frac{1}{2} D_3 \alpha_3^2$. Apart from the difficulties with the mass distributions, the fragment kinetic energies predicted on this basis had a maximum at symmetric fission in disagreement with experiment. Fong attributed the disagreement to the non-inclusion of shell effects on nuclear deformability. He suggests that this is best introduced by considering deformations of the $P_2$ type where the deformation energy is given by $\frac{1}{2} D_2 \alpha_2^2$. The values of $D_2$ for primary fission fragments were obtained by extrapolation from data points for stable nuclei fig. 2.13. The kinetic energy distributions are now in agreement with experiment. The severe variation in the new parameter $D_2$ introduced by Fong is not unlike that of the variable radius of the Cameron formalism.

Stavinski (1964) and Abdelmalek and Stavinski (1964) have investigated the Statistical Model using Fong's formalism and level density formula similar to those of Erba (1963). As in the work of
Erba et al (1964) it is the values of the constants in the energy level formula rather than the excitation energy which dominate the shape of the predicted mass curve.

The statistical model has had undoubted success in predicting many aspects of the fission process. However, it suffers from two major defects. Firstly, the predictions are incredibly sensitive to the input data and although in principle this sensitivity does not affect the veracity of the model any agreement between experiment and theory has to be completely unambiguous to be conclusive. This situation has not been reached. Secondly, although the original formulation was largely devoid of adjustable parameters improvements in the experimental data have necessitated the introduction of additional parameters to maintain agreement with experiment. Finally, there is the question of the basic assumption of the model namely statistical equilibrium either before or at scission. The likelihood or otherwise of equilibrium is related to the relative period of particle motion to that for collective motion. If this ratio is large the statistical approximation is more appropriate but if the ratio is small equilibrium will not be achieved. At high excitation energies the strong-coupling, statistical approximation may be valid, but at low energies the situation may be different.
2.3 References

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3. RESEARCH PROGRAM

Attempts to obtain a complete theoretical description of fission have failed. Although the early stages of the process appear to be reasonably well understood our lack of knowledge of the complex reorganisation of the nucleus from the saddle point to scission does not allow an unambiguous choice to be made between the weak coupling assumption (Adiabatic Model) or the strong coupling assumption (Statistical Model). Additional data on this late stage in the process are required. What type of information can be obtained? Since it is impossible to study this process in real time one must resort to a comparison of the saddle point conditions with the systematics at scission and endeavour to determine any correlations.

In this research program the distribution of energy at scission has been studied in detail and an attempt has been made to determine any influence of saddle point conditions in the energy balance. The total energy of the fissioning system at scission is made up of the coulomb repulsion of the fission fragments which gives rise to their kinetic energies and the deformation and excitation energies of the individual fragments which are released by neutron evaporation and gamma ray emission. The following experiments were performed:

3.1 Measurements of the average number of prompt neutrons emitted per fission, $\bar{v}_p$, for thermal neutron fission of $^{233}$U, $^{235}$U, $^{239}$Pu and $^{241}$Pu. (Chapter IV)

3.2 Measurements of $\bar{v}_p$ for the spontaneous fission of $^{240}$Pu and $^{242}$Pu. (Chapter V).
3.3 The energy dependence of $\bar{\nu}_p$ for U233 and U235 for neutron fission in the range 0-2 MeV. (Chapter VI).

3.4 Measurements of the variation of the average total kinetic energy of the fission fragments for neutron fission of U235. (Chapter VI).

3.5 Prompt neutron emission from individual fission fragments in thermal neutron fission of U235. (Chapter VII).
CHAPTER IV

4. PROMPT NUBAR FOR THERMAL NEUTRON FISSION

4.1 Introduction

Accurate measurements of \( \bar{\nu}_p \), the average number of prompt neutrons emitted per fission, are of prime importance in the design of nuclear reactor systems. The requirements of present design teams are for accuracies better than 0.5% particularly for the relative yields from different reactor fuels. These numbers have further value as general fission data since they provide a measure of the average excitation and deformation energy of the two fragments at scission. In the present investigation, measurements have been made of the yields of prompt neutrons in the thermal neutron fission of \( \text{U}^{233} \), \( \text{U}^{235} \), \( \text{Pu}^{239} \) and \( \text{Pu}^{241} \), relative to the yield from the spontaneous fission of \( \text{Cf}^{252} \). Data have also been obtained of the probability of emission of one, two, three, etc. neutrons per fission. These probability distributions can be fitted approximately with gaussians and it is interesting to compare the width of the neutron distribution with the width of the excitation distribution obtained under a variety of assumptions from nuclear data and experimental kinetic energy distributions. Some information can thereby be obtained about the correlation of the excitation energy of one fragment with that of the other. The competition of gamma ray emission with neutron emission as a de-excitation mode may be estimated as well.

In this investigation and all subsequent measurements of \( \bar{\nu}_p \) and associated parameters, the large liquid scintillator method has been used and is discussed in some detail in the following section.
4.2 The Liquid Scintillator Method

The large liquid scintillator technique was originally devised by Reines and Cowan (1955) to observe neutrinos and was subsequently applied to nubar measurements by Diven et al. (1956). Recent developments of the technique are due to Hopkins and Diven (1963), Mather et al. (1964) and Asplund-Nilsson et al. (1963). In this technique the neutron detector consists of a large liquid scintillator which is loaded with a high neutron capture cross section material such as gadolinium or cadmium. A fission counter containing the appropriate target is placed at the centre of a tube which runs axially through the scintillator tank and allows entry and exit of a neutron beam. Neutrons produced by fission in this counter enter the scintillator, are moderated there and, after a mean lifetime generally of the order of 10 \( \mu \)secs, are captured by the gadolinium or cadmium. The capture gamma rays so produced cause scintillations which may be observed by photomultiplier tubes mounted on the outside of the scintillator tank. By this method a multiplicity of neutrons produced in any fission event may be counted individually. Excellent discrimination against background radiation can be obtained by gating the output of the photomultiplier tubes with the fission pulse and only counting scintillation pulses for several neutron lifetimes. The neutron detection efficiency of such liquid scintillators may be calibrated using an \((n,p)\) scattering technique devised by Hopkins and Diven (1963) and Asplund-Nilsson et al. (1963). More often the detection efficiency is obtained by comparing the neutron count rate per fission for the spontaneous fission of Cf252 with an assumed value of \( \bar{\nu}/p \) for this process. At the present time, a discrepancy
exists between different methods of calibration of the standard $\bar{\nu}_p$ for Cf252 (see G. Hanna et al. (1969) for a discussion) but this does not affect the relative measurements made throughout this thesis. The important features of the various elements of the experimental set up used for the thermal $\bar{\nu}_p$ measurements are described below. Full details of the experimental system are reported in Boldeman and Dalton (1967).

4.3 The Fission Counters

The fission counters were high speed ionisation chambers, with a parallel plate spacing of 3 mm, across which a positive voltage of 450 volts was applied (figure 4.1). The counting gas, methane, was purified using the technique described by Cunningham and Kitt (1964). The fissile foils were prepared by electroplating the particular isotope onto nickel discs (0.006 inches thick). The weights of the targets and their isotopic concentrations are listed in Tables 4.1 and 4.2.

Table 4.1

<table>
<thead>
<tr>
<th>Isotope</th>
<th>No. of Chambers</th>
<th>Weight of Material (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U233</td>
<td>1</td>
<td>2.1</td>
</tr>
<tr>
<td>U235</td>
<td>2</td>
<td>8.1</td>
</tr>
<tr>
<td>Pu239</td>
<td>1</td>
<td>0.7</td>
</tr>
<tr>
<td>Pu241</td>
<td>1</td>
<td>2.8</td>
</tr>
</tbody>
</table>
Fig. 4.1. Fission Counter.
Table 4.2
Isotopic Analysis of Fissile Materials

<table>
<thead>
<tr>
<th>Isotope Poil</th>
<th>U233</th>
<th>U234</th>
<th>U235</th>
<th>U236</th>
<th>U238</th>
<th>Pu239</th>
<th>Pu240</th>
<th>Pu241</th>
<th>Pu242</th>
</tr>
</thead>
<tbody>
<tr>
<td>U233</td>
<td>99.27</td>
<td>0.07</td>
<td>0.04</td>
<td>0.07</td>
<td>0.53</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>U235</td>
<td>1.28</td>
<td>92.72</td>
<td>0.254</td>
<td>5.75</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu239</td>
<td>99.83</td>
<td>0.17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu241</td>
<td>1.44</td>
<td>5.69</td>
<td>91.78</td>
<td>1.09</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The important characteristics of this particular design of the fission counters were as follows:

(a) A high detection efficiency (98 per cent for Cf252, 94.5 per cent for U235).

(b) A high discrimination against amplifier noise.

The freedom from amplifier noise was achieved by operating the fission counters in coincidence with a scintillator signal resulting from the detection of prompt fission $\gamma$-rays and neutron induced proton recoils. The fast response time of the fission counters and the associated electronics made possible the use of a coincidence time resolution of 21 nsecs.

(c) Complete freedom from the effects of alpha pile-up.

Typical pulses from the fission counters had pulse durations of approximately 25 nsecs. This feature combined with the close electrode spacing of the ionisation chamber, the use of methane as the counting gas and the requirement of a coincident scintillator pulse, reduced the alpha pile-up probability to negligible proportions.
The performance of the fission counters may be observed in figures 2, 3(a) and 3(b) in Boldeman and Dalton (1967).

4.4 The Liquid Scintillator Tank

The liquid scintillator tank (figure 4.2) was 76 cm in diameter and held 240 litres of NE323, a trimethyl-benzene-based scintillator containing a loading of 0.5 per cent by weight gadolinium. Scintillations resulting from neutron capture within the tank were viewed by twelve 5 inch EMI 9618A photomultiplier tubes. To minimise spurious events produced by noise or after pulsing within any one photomultiplier tube, the photomultiplier tubes were connected in three coincident banks of four with a coincidence resolving time of 25 nsec. An axial tube 3 inches in diameter allowed the passage of a neutron beam. The appropriate fission counter was centered in this tube. The time distribution of neutron capture after fission of this particular scintillator tank is shown in figure 4.3.

4.5 Electronics

The logic of the electronics is shown in figure 4.4. A genuine fission event established by a coincidence between a fission counter pulse and a scintillator pulse, was used to initiate a 40 μsec counting gate. Neutrons from the fission event were detected with the time characteristic of figure 4.3 and recorded within the temporary store. The specific time of 40 μsec was chosen as 99 per cent of detected neutrons occurred within this interval and extension of the gate time became uneconomical because of the relative increase in the average background count. Following each 40 μsec fission neutron counting gate, a 100 μsec waiting period was introduced after which a second
Figure 4.2 - Liquid Scintillator Tank
Fig. 4.3. Time distribution of...
Neutron Capture.
Fig. 4:4. Block Diagram of Electronics.
A 40μsec counting gate allowed background to be recorded. Separate channels of the temporary store were available for this purpose. At the end of the entire counting cycle of 180 μsec duration, provided a second fission event had not occurred therein, the contents of the temporary store were transferred to the multiple event counter. A second fission event caused all information in the temporary store to be erased. The multiple event counter had a total of 17 channels; 11 corresponded to occasions on which 0 to 10 events were recorded during the first counting gate and 6 for occasions when 0 to 5 background events occurred during the second gate. The number of channels used was more than adequate to store accurately all multiple events during either counting gate.

4.6 Treatment of Experimental Data

The data recorded in the first eleven channels of the multiple event counter had to be corrected for three effects to obtain the average number of neutrons emitted per fission event:

(a) dead time of the recording equipment
(b) background counts
(c) neutron detection efficiency of the scintillator

The first two effects were removed simultaneously (Boldeman and Dalton 1967).

Pulses occurring during the neutron counting gate were subject to pulse overlap of three types

(i) neutron-neutron overlap
(ii) neutron-background overlap
(iii) background-background overlap
while pulses occurring during the background counting gate were subject to background-background overlap only.

If $k_{nn}$ is defined as the probability that two neutron pulses, occurring during the neutron counting gate, overlap and appear as one pulse, then

$$k_{nn} = 2T \int_{0}^{T} f^2(t) \, dt = 0.00675 \quad (4.1)$$

Here $f(t)$ is the normalised time distribution of neutron capture (figure 4.3), $T$ is the measured dead time of 73 nsec, and $T$ is the gate length of 40 µsec. A considerable effort was expended in determining accurately the dead time of the counting system and the various methods used are discussed in Boldeman and Dalton (1967).

The probability that a neutron and a background pulse, occurring during the neutron counting gate, overlap and appear as one pulse is $k_{nB}$ where

$$k_{nB} = 2T \int_{0}^{T} \frac{f(t)}{T} \, dt = 0.00365 \quad (4.2)$$

Similarly, the probability of overlap of two background pulses in either gate is given by

$$k_{BB} = \frac{2T}{T} = 0.00365 \quad (4.3)$$

$F'$ is defined as the probability per fission of recording $\ell$ pulses during the neutron counting gate and $B_x$ the probability of recording $x$ background pulses during the background counting gate. Then, if $D_x$ and $B_x$ are the real probabilities of occurrence of $x$ neutron pulses during the neutron counting gate and $x$ background pulses during either
gate respectively (in other words, the probabilities with the effects of dead time removed), the following equations may be written

\[ F'_g = \sum_{x=0}^{\ell} D_x B_{\ell-x} \left[ 1 - x C_2 k_{nn} - x (\ell-x) k_{nB} - (\ell-x) C_2 k_{BB} \right] + \]

\[ + \sum_{x=0}^{\ell+1} D_x B_{\ell+1-x} \left[ x C_2 k_{nn} + x (\ell+1-x) k_{nB} + (\ell+1-x) C_2 k_{BB} \right] \]  

(4.4)

\[ B'_x = B_x (1 - x C_2 k_{BB}) + B_x+1 C_2 k_{BB} \]  

(4.5)

These equations only correct for the probability of a single overlap per gate. The probabilities of either triple overlap or two overlaps per gate have been ignored in view of their extremely small magnitude. The 17 equations resulting from the above expressions have been solved on an IBM 360/50 computer to obtain the probabilities \( D_x \). For any particular measurement, \( \bar{\nu}_p \) is given by

\[ \bar{\nu}_p = \epsilon^{-1} \sum_{x} xD_x \]  

(4.6)

where \( \epsilon \) is the neutron detection efficiency of the scintillator. The neutron detection efficiency of the scintillator was measured by determining the average neutron count rate per spontaneous fission of Cf252 and comparing it with the value of \( \bar{\nu}_p \) that has been assumed throughout the course of this thesis: \( \bar{\nu}_p (\text{Cf}252) = 3.782 \).

4.7 The Neutron Source

A one inch diameter, collimated beam of thermal neutrons from the 10 kW reactor, MOATA, (Marks 1962) was passed through the axial hole of the tank; collimation was achieved with lead, graphite and borated
paraffin (figure 4.5). The scintillator was surrounded by 3 ft.
of heavy concrete to minimise background. For the neutron spectrum
emerging from the collimator the ratio of thermal neutron fission to
fission induced by neutron above 1 keV was estimated to be greater
than $10^4$ so no correction for the fast neutron component of the beam
was necessary.

4.8 Corrections and Experimental Accuracy

For measurements of parameters such as $\bar{\nu}$, the most significant
aspect of the data is the experimental accuracy achieved. An
exhaustive investigation has been made of the various sources of
experimental error in measurements of this type. The effects which
have been considered are:

1. Statistical accuracy
2. Drifts in counter efficiency
3. Fission by fast neutrons
4. Impurities
5. Inaccuracy of fission counter location
6. False gates
7. Fission fragment detection efficiency
8. Anisotropy of fission fragment detection
9. Fission spectra differences
10. Dead time correction
11. Additional fission occurring during the gate
12. Delayed gamma rays
13. Electronic errors
14. Variations in background
Fig. 4-5. Neutron Collimator and Shielding.
A complete discussion of these factors is contained in Boldeman and Dalton (1967) and only the significant effects will be mentioned here. Table 4.3, taken from this reference, lists the various correction and estimates of all sources of error. It will be observed that the magnitude of the delayed gamma ray contribution in Table 4.3 differs slightly from that in the original reference. The revised value has been obtained from recent and more accurate data from Sund and Walton (1968).

4.8.1 Statistical Accuracy

The statistical accuracy of the mean number of events per gate whether they be neutron or backgrounds, for a particular measurement, is given by

$$\left[ \sum_{i=0}^{i_{\max}} i^2 g_i - \left( \sum_{i=0}^{i_{\max}} i g_i \right)^2 \left( \sum_{i=0}^{i_{\max}} g_i \right)^{-1} \right] \frac{i}{2} \left[ \sum_{i=0}^{i_{\max}} g_i \right]^{-1} \quad (4.7)$$

where $g_i$ is the number of gates for which $i$ events were detected. The accuracy of a particular measurement of $\bar{v}_p$ was obtained by combining the statistical accuracy of the Cf252 calibration with the statistical accuracies of the mean number of events per gate and the mean number of background events per gate. A large number of measurements of $\bar{v}_p$ was made for each isotope. The distribution of the $\bar{v}_p$ values within any particular set was consistent with the statistical accuracy of the individual measurements assessed as above.
### Table 4.3 - Accuracy of Results

<table>
<thead>
<tr>
<th></th>
<th>U233 % correction to expt. result</th>
<th>U235 % correction to expt. result</th>
<th>Pu239 % correction to expt. result</th>
<th>Pu241 % correction to expt. result</th>
<th>% error from effect listed</th>
<th>% error from effect listed</th>
<th>% error from effect listed</th>
<th>% error from effect listed</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Statistical accuracy</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.168</td>
<td>0.103</td>
<td>0.155</td>
<td>0.108</td>
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<td>2. Counter drifts</td>
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<td>negligible</td>
<td>negligible</td>
<td>negligible</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. Fission by fast neutron</td>
<td>0.0001</td>
<td>0.0001</td>
<td>0.0001</td>
<td>0.0001</td>
<td></td>
<td></td>
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<tr>
<td>4. Impurities</td>
<td>0.000</td>
<td>0.0013</td>
<td>0.000</td>
<td>0.004</td>
<td>0.0013</td>
<td>0.004</td>
<td>0.224</td>
<td>0.043</td>
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<tr>
<td>5. Inaccuracy of counterlocation</td>
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<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6. False gates</td>
<td>0.015</td>
<td>0.005</td>
<td>0.010</td>
<td>0.005</td>
<td>0.02</td>
<td>0.01</td>
<td>0.000</td>
<td>0.01</td>
</tr>
<tr>
<td>7. Preferentialisation</td>
<td>0.02</td>
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<td>0.05</td>
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<td>0.0</td>
<td>0.01</td>
<td>0.02</td>
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<tr>
<td>8. Anisotropy</td>
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</tr>
<tr>
<td>9. Fission spectra differences</td>
<td>-0.51</td>
<td>0.26</td>
<td>-0.55</td>
<td>0.28</td>
<td>-0.34</td>
<td>0.18</td>
<td>-0.32</td>
<td>0.17</td>
</tr>
<tr>
<td>10. Dead time correction</td>
<td>-0.20</td>
<td>0.04</td>
<td>-0.30</td>
<td>0.06</td>
<td>-0.05</td>
<td>0.01</td>
<td>-0.10</td>
<td>0.02</td>
</tr>
<tr>
<td>11. Double fission inhibit</td>
<td>0.0000</td>
<td>0.0003</td>
<td>0.0000</td>
<td>0.0003</td>
<td>0.0000</td>
<td>0.0003</td>
<td>0.0000</td>
<td>0.0003</td>
</tr>
<tr>
<td>12. Delayed gamma rays</td>
<td>0.000</td>
<td>0.10</td>
<td>0.000</td>
<td>0.10</td>
<td>-0.18</td>
<td>0.10</td>
<td>-0.18</td>
<td>0.10</td>
</tr>
<tr>
<td>13. Electronic errors</td>
<td>0.000</td>
<td>0.03</td>
<td>0.000</td>
<td>0.02</td>
<td>0.0000</td>
<td>0.03</td>
<td>0.0000</td>
<td>0.02</td>
</tr>
<tr>
<td>14. Varying background</td>
<td>negligible</td>
<td>negligible</td>
<td>negligible</td>
<td>negligible</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>-0.675</td>
<td>±0.033</td>
<td>-0.790</td>
<td>±0.325</td>
<td>-0.546</td>
<td>±0.058</td>
<td>-0.334</td>
<td>±0.232</td>
</tr>
</tbody>
</table>
4.8.2 Fission Spectra Differences

Corrections were applied to the $v_p$ values to account for differences between the fission neutron spectrum of each isotope and that of the standard (Cf252) since the response of the detector was energy dependent. The variation in the neutron detection efficiency has been computed using Monte Carlo methods by AWRE (Mather, Moat and Fieldhouse) and independently B. McGregor (1962). Both calculations gave identical results and the response of the detector is tabulated for several energies in Table 4.4.

Table 4.4

<table>
<thead>
<tr>
<th>Neutron Energy (MeV)</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>95.5</td>
</tr>
<tr>
<td>0.5</td>
<td>94.9</td>
</tr>
<tr>
<td>1.5</td>
<td>94.8</td>
</tr>
<tr>
<td>3.0</td>
<td>93.1</td>
</tr>
<tr>
<td>5.0</td>
<td>83.4</td>
</tr>
<tr>
<td>7.0</td>
<td>77.1</td>
</tr>
<tr>
<td>10.0</td>
<td>71.1</td>
</tr>
<tr>
<td>14.0</td>
<td>61.1</td>
</tr>
</tbody>
</table>
Numerous experiments have shown that the fission neutron energy spectra may be accurately described by a Maxwellian distribution $\sqrt{E} \exp \left( -\frac{E}{T} \right)$ in which $T$ is a parameter describing the average neutron energy in the laboratory system, $\tilde{E} = \frac{3T}{2}$. The variation in the response of the scintillator was determined for a range of values of $\tilde{E}$. The corrections to the $\tilde{\nu}_p$ values were obtained from this curve using estimates of $\tilde{E}$ obtained from Terrell's (1962) empirical relationship

$$\tilde{E} = 0.74 + 0.653 (\tilde{\nu}_p + 1)^{\frac{1}{2}}$$

and the measured $\tilde{\nu}_p$ values. In view of the uncertainties in the response curve and present difficulties with fission neutron spectra data, errors of ± 50 per cent were attached to this correction.

4.8.3 Dead time correction

Dead time corrections were generally of the order of one per cent. However, for relative measurements of this type errors tend to compensate and the actual error in the correction is almost negligible.

4.9 Thermal $\tilde{\nu}_p$ Results

The measured values of $\tilde{\nu}_p$ for the thermal neutron fission of U233, U235, Pu239 and Pu241 are listed in Table 4.5.
Table 4.5
Thermal $\bar{\nu}_p$ Results Relative to $\bar{\nu}_p = 3.782$ for Cf252

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\bar{\nu}_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>U233</td>
<td>2.491 ± 0.008</td>
</tr>
<tr>
<td>U235</td>
<td>2.415 ± 0.008</td>
</tr>
<tr>
<td>Pu239</td>
<td>2.897 ± 0.008</td>
</tr>
<tr>
<td>Pu241</td>
<td>2.940 ± 0.007</td>
</tr>
</tbody>
</table>

It will be noted that the values listed here differ slightly from those in Boldeman and Dalton (1967). The revision results from the improved delayed gamma ray data from Sund and Walton (1968). The $\bar{\nu}_p$ data has been compared with previous determinations, Boldeman and Dalton (1967) and the agreement is satisfactory. (See also Fillmore 1968, and Hanna et al. 1969).

In addition to the above data the probabilities of emission of $\nu$ neutrons per fission event, $P_\nu$, were calculated from the experimental $D_x$ probabilities in accordance with formulae given by Diven et al. (1956) for the values of $\nu$ in the range 0 to 10 as follows:

$$P_\nu = \sum_{x=\nu}^{x=10} \frac{x!}{\nu!(x-\nu)!} \left(1 - \frac{1}{\bar{\nu}}\right)^{x-\nu} \left(\frac{1}{\bar{\nu}}\right)^{\nu} D_x$$  \hspace{1cm} (4.9)

For each distribution the second moment about the origin $<\nu^2>_{av}$, the second moment about $\bar{\nu}_p$, var, and a shape dependent parameter, R, were calculated as follows:
\[ \langle \nu^2 \rangle_{av} = \sum_{\nu=0}^{\nu=10} \nu^2 p_{\nu} \]  
(4.10)

\[ \text{var} = \sum_{\nu=0}^{10} p_{\nu} (\nu - \bar{\nu}_p)^2 \]  
(4.11)

\[ R = \frac{\langle \nu^2 \rangle_{av} - \bar{\nu}_p}{(\bar{\nu}_p)^2} \]  
(4.12)

The relevant parameters are listed in Table 4.6 together with their assessed accuracy. The accuracies were obtained from experimental reproducibility. This data will be discussed with similar data for spontaneous fission in the following chapter.
Table 4.6

Neutron Emission Parameters

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>U233</th>
<th>U235</th>
<th>Pu239</th>
<th>Pu241</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt;\nu^2&gt;_\text{av}$</td>
<td>$7.416 \pm 0.034$</td>
<td>$7.073 \pm 0.032$</td>
<td>$9.838 \pm 0.040$</td>
<td>$10.063 \pm 0.037$</td>
</tr>
<tr>
<td>var</td>
<td>$1.099 \pm 0.004$</td>
<td>$1.112 \pm 0.004$</td>
<td>$1.185 \pm 0.005$</td>
<td>$1.173 \pm 0.004$</td>
</tr>
<tr>
<td>$R$</td>
<td>$0.7932 \pm 0.0013$</td>
<td>$0.7979 \pm 0.0013$</td>
<td>$0.8221 \pm 0.0016$</td>
<td>$0.8190 \pm 0.0010$</td>
</tr>
<tr>
<td>$P_0$</td>
<td>$0.0259 \pm 0.0010$</td>
<td>$0.0313 \pm 0.0060$</td>
<td>$0.0094 \pm 0.0010$</td>
<td>$0.0097 \pm 0.0010$</td>
</tr>
<tr>
<td>$P_1$</td>
<td>$0.1526 \pm 0.0020$</td>
<td>$0.1729 \pm 0.0016$</td>
<td>$0.0990 \pm 0.0027$</td>
<td>$0.0877 \pm 0.0025$</td>
</tr>
<tr>
<td>$P_2$</td>
<td>$0.3289 \pm 0.0034$</td>
<td>$0.3336 \pm 0.0029$</td>
<td>$0.2696 \pm 0.0034$</td>
<td>$0.2636 \pm 0.0030$</td>
</tr>
<tr>
<td>$P_3$</td>
<td>$0.3282 \pm 0.0035$</td>
<td>$0.3078 \pm 0.0029$</td>
<td>$0.3297 \pm 0.0035$</td>
<td>$0.3343 \pm 0.0032$</td>
</tr>
<tr>
<td>$P_4$</td>
<td>$0.1320 \pm 0.0017$</td>
<td>$0.1232 \pm 0.0016$</td>
<td>$0.1982 \pm 0.0030$</td>
<td>$0.2099 \pm 0.0035$</td>
</tr>
<tr>
<td>$P_5$</td>
<td>$0.0252 \pm 0.0020$</td>
<td>$0.0275 \pm 0.0020$</td>
<td>$0.0924 \pm 0.0040$</td>
<td>$0.0811 \pm 0.0040$</td>
</tr>
<tr>
<td>$P_6$</td>
<td>$0.0045 \pm 0.0020$</td>
<td>$0.0038 \pm 0.0015$</td>
<td>$0.0119 \pm 0.0020$</td>
<td>$0.0112 \pm 0.0020$</td>
</tr>
</tbody>
</table>
4.10 **References**


5. PROMPT NUBAR FOR SPONTANEOUS FISSION

5.1 Introduction

Measurements of $\bar{\nu}_p$ for spontaneous fission have obvious relevance to reactor physics. However, in the context of this thesis, they are important for the data they provide concerning fission at extremely low excitation. For example, spontaneous fission of Pu240 may be regarded as equivalent to neutron fission of Pu239 at a compound excitation of $-6.3$ MeV (the neutron binding energy) relative to thermal neutron fission. It should be interesting then to compare neutron emission data from spontaneous fission with that reported in the previous chapter for thermal neutron fission.

5.2 $\bar{\nu}_p$ and Associated Data

Measurements of $\bar{\nu}_p$ for the spontaneous fission of Pu240 and Pu242 relative to $\bar{\nu}_p$ for the spontaneous fission of Cf252 have been made using the liquid scintillator method. Complete experimental details are given in Boldeman (1968), a copy of which is attached. The isotopic analyses of the fissile foils and their weights are listed in Table 5.1.
Table 5.1

Isotopic Analysis of Fissile Foils

<table>
<thead>
<tr>
<th></th>
<th>Pu 240</th>
<th>Pu 242</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wt. of material</td>
<td>800 mg</td>
<td>600 mg</td>
</tr>
<tr>
<td>Per Cent of weight Pu 239</td>
<td>24.46%</td>
<td>0.017%</td>
</tr>
<tr>
<td>Pu 240</td>
<td>74.14%</td>
<td>0.039%</td>
</tr>
<tr>
<td>Pu 241</td>
<td>1.29%</td>
<td>0.064%</td>
</tr>
<tr>
<td>Pu 242</td>
<td>0.10%</td>
<td>99.88%</td>
</tr>
<tr>
<td>Am 241</td>
<td>0.18%</td>
<td></td>
</tr>
</tbody>
</table>

The $\bar{\nu}_p$ data are listed in Table 5.2 and neutron emission parameters (defined as in Chapter IV) are tabulated for Pu240, Pu242 and Cf252 in Table 5.3.

Table 5.2

$\bar{\nu}_p$ Values Relative to Cf 252 $\bar{\nu}_p = 3.782$

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\bar{\nu}_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu 240</td>
<td>2.167 ± 0.009</td>
</tr>
<tr>
<td>Pu 242</td>
<td>2.156 ± 0.009</td>
</tr>
</tbody>
</table>
Table 5.3

Neutron Emission Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Pu 240</th>
<th>Pu 242</th>
<th>Cf 252</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle \nu^2 \rangle$</td>
<td>6.023 $\pm$ 0.035</td>
<td>5.981 $\pm$ 0.036</td>
<td>15.925 $\pm$ 0.007</td>
</tr>
<tr>
<td>$\text{var}$</td>
<td>1.150 $\pm$ 0.004</td>
<td>1.153 $\pm$ 0.004</td>
<td>1.268 $\pm$ 0.002</td>
</tr>
<tr>
<td>$R$</td>
<td>0.820 $\pm$ 0.002</td>
<td>0.822 $\pm$ 0.002</td>
<td>0.8479 $\pm$ 0.0005</td>
</tr>
<tr>
<td>$P_0$</td>
<td>0.0639 $\pm$ 0.010</td>
<td>0.0648 $\pm$ 0.011</td>
<td>0.00197 $\pm$ 0.00008</td>
</tr>
<tr>
<td>$P_1$</td>
<td>0.2284 $\pm$ 0.0021</td>
<td>0.2296 $\pm$ 0.0021</td>
<td>0.02447 $\pm$ 0.00025</td>
</tr>
<tr>
<td>$P_2$</td>
<td>0.3299 $\pm$ 0.0022</td>
<td>0.3362 $\pm$ 0.0024</td>
<td>0.1229 $\pm$ 0.0005</td>
</tr>
<tr>
<td>$P_3$</td>
<td>0.2536 $\pm$ 0.0028</td>
<td>0.2470 $\pm$ 0.0027</td>
<td>0.2707 $\pm$ 0.0008</td>
</tr>
<tr>
<td>$P_4$</td>
<td>0.1047 $\pm$ 0.0031</td>
<td>0.1019 $\pm$ 0.0032</td>
<td>0.3058 $\pm$ 0.0010</td>
</tr>
<tr>
<td>$P_5$</td>
<td>0.0166 $\pm$ 0.0015</td>
<td>0.0163 $\pm$ 0.0015</td>
<td>0.1884 $\pm$ 0.0007</td>
</tr>
<tr>
<td>$P_6$</td>
<td>0.0030 $\pm$ 0.0005</td>
<td>0.0030 $\pm$ 0.0006</td>
<td>0.0677 $\pm$ 0.0006</td>
</tr>
<tr>
<td>$P_7$</td>
<td></td>
<td></td>
<td>0.0160 $\pm$ 0.0003</td>
</tr>
<tr>
<td>$P_8$</td>
<td></td>
<td></td>
<td>0.0021 $\pm$ 0.0002</td>
</tr>
</tbody>
</table>

The $\bar{\nu}_p$ values obtained have been compared with other determinations in Boldeman (1968) and the agreement is satisfactory. The neutron emission parameters are in excellent agreement with those from Soleilhac et al (1966), (1969).

5.3 Correlation of $P_\nu$ data

It should be possible to calculate the probability $P_\nu$ of emission of any integral number of prompt neutrons $\nu$ from the distribution of
the excitation energy provided accurate nuclear data is available and the correct assumptions regarding the competition of gamma ray emission and correlations between the light and heavy fragments are made. Such calculations have been performed by Leachman (1956) who obtained good agreement with experimental data existing at that time. Briefly his procedure is as follows. The total energy release, i.e. the sum of the kinetic and excitation energy, is obtained from the mass balance equation. This sum is evaluated for three typical mass divisions using extrapolated atomic masses and the procedure of Coryell (1953). Assuming the excitation distributions for the light and heavy fragment are the same and independent, he then obtains these distributions from the measured distribution of the total kinetic energy. With the further assumption that gamma ray emission does not compete with neutron emission, the $P_\gamma$ probabilities were computed using evaporation theory. The residual energy, namely that remaining after all the neutrons that are energetically possible are emitted, appears as gamma rays. The computed total gamma ray energy was in serious disagreement with experiment, i.e. a computed value of 3.8 MeV versus the measured value of $7.2 \pm 0.8$ MeV for U235; see Maier-Leibniz et al (1965).

Terrell (1957) has endeavoured to correlate the various sets of experimental data by means of simpler calculations based on a minimum of parameters. He assumes

1. that the emission of any neutron from any fission fragment reduces its excitation by a value of $\Delta E$ which is nearly
equal to the average value $E_o = \langle \Delta E \rangle_{av}$.

2. That the total excitation energy of the two primary fragments from binary fission has a gaussian distribution with r.m.s. deviation $\sigma E_o$ from the average $\bar{E}$.

On the basis of these postulates, Terrell shows that the $P_v$ probabilities are given approximately, in cumulative form, by the gaussian distribution:

$$\sum_{n=0}^{v} P_n = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{(v - \bar{v} + \frac{1}{2} + b)\sigma} \exp \left(-\frac{t^2}{2}\right) dt$$

where $t$ is defined as $(E - \bar{E})/\sigma E_o$ and $b$ is a small adjustment ($\approx 10^{-2}$).

Of course it is tacitly assumed that gamma ray emission does not compete. Terrell has also considered the effects of various assumed correlations between the excitation energies of the light and heavy fragments. The results of this investigation suggest that eqn 5.1 should be independent of the form of the correlation. Rather than attempt to predict the $P_v$ probabilities, Terrell's procedure was now to fit gaussian distributions to the measured $P_v$ distributions to obtain $\sigma$. Terrell's analysis showed that the data did fit a gaussian distribution rather well. Fig. 5.1 is reproduced from Terrell and shows the quality of the fit. A straight line in this figure of course represents a gaussian distribution. If the present data is plotted in this fashion it will be observed (Fig. 5.2) that the fit is not as good. The experimental non-cumulative neutron emission probabilities from Terrell are plotted in Fig. 5.3. The continuous curve is that for a gaussian with $\sigma = 1.08$. 
Figure 5.1 Experimental cumulative neutron emission probabilities. The straight line represents a "Gaussian" distribution.
Fig. 5.2. Cumulative Emission Probabilities, Present Data. Thermal Fission Data are O. Spontaneous Fission Data are △
Figure 5.3 Experimental noncumulative neutron emission probabilities. The continuous curves are for a "Gaussian" distribution.
The present data have been plotted in a similar manner in Fig. 5.4 where fine differences will be observed between the different data sets, e.g. the distributions for spontaneous fission appear to be slightly broader than for thermal neutron fission.

The present $P^v_\nu$ probabilities have been fitted with gaussian distributions (zero probabilities were ignored and the distribution was allowed to extend to negative numbers). The calculated widths of the discrete distributions are listed in Table 5.4. Although the analytical fits to the data were poor when measured in terms of the experimental accuracy, this should not be taken too seriously. The poor quality of the fit only implies that the neutron distribution is not an exact gaussian distribution. In fact the neutron distribution is sufficiently close to a gaussian distribution that regarding it as such does not introduce any error in the subsequent analysis. A better measure of the likelihood of gaussian representation of the neutron emission probabilities can be obtained (in addition to Fig. 5.2) from a comparison of the fitted widths with the parameter specified as 'var'. The two numbers should be identical for each isotope after making suitable allowance for the fact that the measured distribution is terminated at zero neutrons whereas a fitted gaussian does in principle extend to negative numbers. For those isotopes (e.g. Cf252) where $v$ is high and the zero emission probability is quite small the agreement is very good. For Pu240 and Pu242 good agreement cannot be expected in the simple analysis above. In subsequent discussion the fitted widths have been selected as the best representation of the data.
Fig. 5.4. Neutron Emission Probabilities. The peaks of the distribution are inversely related to the widths. Note the peaks for U233, U235 > Pu239, Pu241 > Cf 252.
Table 5.4
Gaussian Fits to $p'v$ Probabilities

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\bar{v}_p$</th>
<th>$\sigma$ (fitted)</th>
<th>$\text{var} = \sum_p (v-\bar{v})^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Thermal</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>U233</td>
<td>2.491 ± 0.008</td>
<td>1.094</td>
<td>1.099</td>
</tr>
<tr>
<td>U235</td>
<td>2.415 ± 0.008</td>
<td>1.135</td>
<td>1.122</td>
</tr>
<tr>
<td>Pu239</td>
<td>2.897 ± 0.008</td>
<td>1.213</td>
<td>1.185</td>
</tr>
<tr>
<td>Pu241</td>
<td>2.940 ± 0.007</td>
<td>1.176</td>
<td>1.173</td>
</tr>
<tr>
<td><strong>Spontaneous</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu240</td>
<td>2.167 ± 0.009</td>
<td>1.243</td>
<td>1.150</td>
</tr>
<tr>
<td>Pu242</td>
<td>2.156 ± 0.009</td>
<td>1.225</td>
<td>1.153</td>
</tr>
<tr>
<td>Cf252*</td>
<td>3.782</td>
<td>1.255</td>
<td>1.268</td>
</tr>
</tbody>
</table>

* Standard Value assumed for normalisations.

Two simple observations may be made from the data in Table 5.4:

1. For thermal neutron fission, the widths are slightly related to $\bar{v}_p$ - there is a slight increase in $\sigma$ with increasing $\bar{v}_p$.

2. The widths of the fitted gaussians for spontaneous fission are similar to each other but in all cases are greater than for thermal fission.

The second of these observations may be of some significance when it is remembered that spontaneous fission of Pu240 and Pu242 correspond to neutron fission of Pu239 and Pu241 at compound excitation approximately 6.3 MeV below the neutron binding energy. This observation is treated in conjunction with additional data in Chapter 6.
Subsequent development of the above analysis by Terrell (1959) gave rise to a total gamma ray energy yield of 4.9 MeV, again considerably smaller than the experimental value.

Gordon and Aras (1965) have considered the energy balance in fission and their treatment yields a figure for the width of the neutron probability distribution. Briefly, their procedure is as follows. First they calculate the total energy release for a particular mass and charge split from the mass equation 5.2 using Seeger's (1961) mass formula.

\[ E_T = \Delta M (U^{236*}) - (\Delta M_L + \Delta M_H) \]  

They assume the average total kinetic energy \( \bar{E}_K \) is independent of the charge split \((Z_L/Z_H)\) for a particular mass division \((A_H/A_L)\). Then the excitation energy is given simply by

\[ E_x = E_T - E_K (A_H/A_L) \]

where the kinetic energy data may be taken from instrumental studies. Eqn 5.3 requires the shape of the excitation distributions to be the same as those for the kinetic energy distributions, i.e. for the assumed gaussians

\[ \sigma_{E_x} = \sigma_{E_K} \]

If the excitation energies of the light and heavy fragment are uncorrelated then

\[ \sigma_{E_{xL}}^2 + \sigma_{E_{xH}}^2 = \sigma_{E_x}^2 \]
They further assume

\[ \frac{\sigma_{E_{xL}}}{\sigma_{E_{xH}}} = \frac{E_{xL}}{E_{xH}} \]

\( \frac{E_{xL}}{E_{xH}} \) was a free parameter in the calculations which was allowed to vary until the correct ratio was obtained for \( \frac{\nu_L}{\nu_H} \).

De-excitation by emission of neutrons and gamma rays of a particular primary fragment with a selected excitation energy was determined using the Monte Carlo method described by Dostrovsky et al (1959). In their treatment the level densities of the residual nuclei as a function of excitation energy, \( E \), were assumed to be given by

\[ w(E) = \text{const exp}\left[ 2\sqrt{a(E - \delta)} \right] \]

in which \( \delta \) is the 'characteristic level' or pairing-energy correction to account for reduced level density when the number of protons and/or neutrons is even. This treatment assumed that a neutron is emitted whenever possible except that no neutron emission may leave the residual nucleus with an excitation energy less than \( \delta \) if \( \delta \) is non-zero. The parameter \( \delta \) has the effect of correcting for the effects of high angular momenta of the primary fragments which tend to favour gamma emission rather than neutron emission.

To obtain the required information, Gordon and Aras then averaged over the excitation energy distribution for a primary fragment
and followed with averaging over the primary fragment yields. They thereby obtained excellent agreement with experimental data on several important aspects.

1. The correct dependence of $\bar{v}_A$ upon mass number $A$ was obtained.
2. The calculated neutron energy spectrum agreed well with experimental determinations.
3. The average total gamma ray energy release was in good agreement with experiment i.e. a calculated value of 7.66 MeV versus the experimental figure of $7.2 \pm 0.8$ MeV.

Item 3 was the most important improvement over the previous analyses by Leachman (1956) and Terrell (1957). Despite this improvement, Gordon and Aras noted two important discrepancies. The average number of prompt neutrons emitted per fission for U235 was estimated to be 2.59, slightly higher than the experimental value of 2.415 (Chapter IV). But of far greater significance, the width of the neutron distribution was found to be $\sigma = 1.39$ which is in serious disagreement with the experimental value of 1.135. To resolve this important discrepancy between the calculated and experimental width of the neutron distribution Gordon and Aras have considered the effects of assuming either positive or negative correlation between the excitation energies of the light and heavy fragments. Neither positive nor negative correlation improved the overall fit to the experimental data. They concluded that the most obvious method of resolving the discrepancy was to relax the implied assumption of 5.3, namely that the average total kinetic energy is independent of the
charge division for a particular mass division. In this way some of the experimentally observed dispersion in total kinetic energy can be accounted for, without increasing the dispersion in the total excitation energy. However the recent experimental data from Glendenin et al (1969) confirms assumption 5.3 i.e. they find the total kinetic energy to be independent of the charge division. Consequently the discrepancy in the width of the neutron emission distribution remains unresolved.

A more sophisticated explanation of the large total gamma ray release in fission fragment de-excitation has been derived by Thomas and Grover (1967). They suggest that the discrepancy between the theoretical values and the experimental value is an effect due to the high angular momentum of the fission fragments. For nuclei with excitation energies somewhat in excess of the neutron binding energy, neutron emission is not necessarily inevitable but gamma ray emission may be a competing process if the neutrons must carry away a large amount of orbital momentum.

The quantitative calculations of Thomas and Grover which include such angular momentum considerations are based on the work of Grover (1967a, 1967b, 1967c). Grover has made shell model calculations of the yrast levels for a large number of nuclei with a given angular momentum. The yrast level of a given nucleus is that level with the least energy for a particular angular momentum. Grover and Gilat (1967a) have shown that for a nucleus of a given angular momentum, gamma ray emission competes favourably with neutron emission if the excitation energy is less than the sum of the neutron
binding energy and the yrast energy. In another paper, Grover and Gilat (1967b) provide a procedure to follow the de-excitation of a nucleus with specified excitation and total angular momentum. Emission of neutrons, dipole and quadrupole gamma rays, protons and alpha particles were considered. Thomas and Grover have applied these observations and the calculative procedure referred to above to the de-excitation of the fission fragments. They assume

1. The energy and angular momentum distribution of the fragments are uncorrelated.

2. The angular momentum distributions are given by equation 5.8

\[ N(J) \propto (2J+1) \exp \left[ -J(J+1)/2b^2 \right] \]  

where \( N(J) \) is the probability of forming a fragment with a particular angular momentum \( J \). This form (eqn 5.8) was used by Vandenbosch and Warhanek (1964) and by Sarantites et al (1965) in their analyses of isomer yields from fission. The parameter \( b \) gives the best fit to the isomer ratio results if it is placed equal to 6.

3. Perfect positive correlation exists between the excitation energy of light and heavy fragments. The excitation energy distributions and their mean were obtained from nuclear data in a fashion similar to that described in other analyses by, for example, Gordon and Aras (1965).

Thomas and Grover estimate the total gamma ray release to be 7.1 MeV in good agreement with experiment and also predict the correct average photon energy of 0.9 MeV. They also obtained reasonable values for \( \bar{\nu}_p \) and average neutron energy.
Fig. 5.5 is reproduced from Thomas and Grover and shows the calculated mean total gamma release from Sr96 and Xe140 excited to various excitation energies $E^*$. Assuming values of $E_L = 13.0 \text{ MeV}$ and $\sigma_L = 5.2 \text{ MeV}$ for the light fragment and $E_H = 10.1 \text{ MeV}$ and $\sigma_H = 4.0 \text{ MeV}$ for the heavy fragment we have calculated the variances of the gamma ray emission from the light and heavy fragments. The values obtained in this very simple manner are listed in Table 5.5. Of course it is assumed that there is no distribution about the mean gamma ray emission for a particular excitation energy.

Table 5.5

Dispersion of Total Gamma Ray Release for the Light and Heavy Fragments

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\sigma_{E_{\gamma L}}$</th>
<th>$\sigma_{E_{\gamma H}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sr 96</td>
<td>0.67 MeV</td>
<td></td>
</tr>
<tr>
<td>Xe 140</td>
<td></td>
<td>0.60 MeV</td>
</tr>
</tbody>
</table>

If it is assumed that data for these isotopes are representative for all light and heavy fragments and if it is further assumed that the excitation energies of the light and heavy fragments are perfectly correlated then the total gamma ray dispersion is given by 5.9 and is equal to 1.3 MeV.

$$\sigma_{E_{\gamma T}}^2 = \sigma_{E_{\gamma L}}^2 + \sigma_{E_{\gamma H}}^2 + 2\sigma_{E_{\gamma L}} \sigma_{E_{\gamma H}}$$

5.9
FIGURE 5-5. CALCULATED MEAN TOTAL ENERGY CARRIED AWAY BY $\gamma$ RAYS IN THE DE-EXCITATION OF Sr$^{96}$ & Xe$^{140}$ EXCITED TO ENERGY $E^*$ from THOMAS & GROVER (1967)
The finite width for the gamma ray dispersion may be a contributing factor in the discrepancy between the neutron width calculated by Gordon and Aras of 1.39 and the experimental width of 1.135 for U235. For example if one assumed perfect correlation between neutron emission and gamma ray emission then the calculated neutron width is reduced to 1.20 which is certainly closer to the experimental value. Furthermore it is possible that some effect due to gamma ray emission might cause the neutron width for spontaneous fission to be significantly larger than for thermal neutron fission.

The details of the arguments expressed above should not be taken too seriously, however they do suggest the possibility that gamma ray emission may have an influence on the systematics of neutron emission. In Chapter 6, this influence is more positively observed and the arguments above have really a supporting role for the later data.
5.4 References


REFERENCES (CONTD)


6. THE ENERGY DEPENDENCE OF $\bar{\nu}_p$ AND $\bar{E}_K$

§ 6.1 Introduction

The precise dependence of $\bar{\nu}_p$ and $\bar{E}_K$ on the energy of the incident neutrons producing fission has been the object of considerable experimental and theoretical research. One motivation for this research has of course been for reactor physics applications but by far the more important aspect has been the information that such data provide concerning the division of compound excitation between fragment kinetic energy and fragment excitation. In particular the question of the strength of the coupling of the compound excitation to the nucleonic degrees of freedom at scission is investigated.

Earliest speculation on this matter derives from Fowler (Leachman, 1956) who postulated that the fragment kinetic energy was independent of the compound excitation for a particular compound nucleus. This view was borne out by measurements from Okolovitch et al. (1962) who found the average total kinetic energy to be the same for thermal neutron and 5 Mev neutron fission of U235. In addition, early measurements of the variation of $\bar{\nu}_p$ for the important fissile nuclei showed $\frac{d\bar{\nu}_p}{dE_n}$ to be approximately equal to 0.13 MeV$^{-1}$. (Leachman 1958).

This slope was that expected on the basis of all additional compound excitation appearing as fragment excitation and contributing entirely to increased neutron emission.
However, when more precise measurements were made of the $\tilde{v}_p(E_n)$ dependence a different picture emerged. Table 6.1 summarises the U235 data from Meadows and Whalen (1962), Hopkins and Diven (1963) and Mather et al. (1964).

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Linear Fits to Data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mather et al. (1964)</td>
<td>$\tilde{v}_p(E_n) = (2.418 \pm 0.008) + (0.109 \pm 0.006) E_n$</td>
</tr>
<tr>
<td></td>
<td>$\tilde{v}_p(E_n) = (2.200 \pm 0.023) + (0.181 \pm 0.005) E_n$</td>
</tr>
<tr>
<td>Hopkins and Diven (1963)</td>
<td>$\frac{d\tilde{v}_p}{dE_n} = 0.085 \text{ MeV}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$\frac{d\tilde{v}_p}{dE_n} = 0.16 \text{ MeV}^{-1}$</td>
</tr>
<tr>
<td>Meadows and Whalen (1962)</td>
<td>$\tilde{v}_p(E_n) = 2.414 + (0.097 \pm 0.008) E_n$</td>
</tr>
</tbody>
</table>

For $E_n < 1.5$ to 3 MeV, all three groups observed a slope significantly less than that expected from Fowler's hypothesis. Above this energy the observed slopes were larger than that expected. A similar dependence was observed for U233, whereas for Pu239 a single slope, similar in magnitude to the larger of the two slopes in Table 6.1, was observed. In no case were any concrete explanations given to account
for the change in slope.

The situation became more confusing with the appearance of the \( \bar{v}_p (E_n) \) data from Blyumkina et al. (1964) for U235. Between 0-1 MeV neutron energy they observed an average slope similar to that in Table 6.1, but in addition reported a marked deviation from the linear dependence between 200-700 keV neutron energy. In particular, they claimed evidence of a significant peak in their \( \bar{v}_p \) data at approximately 400 keV neutron energy. Supporting measurements of the variation of the average total kinetic energy with compound excitation showed complementary behaviour, although the quantitative agreement between the two measurements was quite poor. Blyumkina et al. (1964) interpreted the structure as an effect emanating from the discrete nature of the low lying fission channels, A. Eohr (1956). This conclusion was relevant to fission theory, as it suggested that the saddle point conditions were still influential at the scission stage.

The \( \bar{v}_p \) measurements of Meadows and Whalen (1967) for U235 supported the general principle of non-linearity in the \( \bar{v}_p (E_n) \) dependence but were in severe quantitative disagreement with the Blyumkina data. In fact, they found evidence of two maxima and two minima in the energy region 200-700 keV. Measurements (above 0.4 MeV) by Prokhorova et al. (1966) suggested fine structure in the U235 \( \bar{v}_p (E_n) \) dependence of a different kind, namely a somewhat stepped curve.

Recent measurements of the dependence of the average total kinetic energy provide data just as confusing. The data from Bolshov (1968) tend to support the original Blyumkina data without
being in conclusive agreement. However, the very accurate measurements of Dyachenko (1968) are in complete agreement with Fowler's hypothesis.

The object of the research reported here has been to examine the \( V_p (E_n) \) dependence with high accuracy to resolve the discrepancies in the existing data. Measurements will also be reported of the dependence of \( \bar{E}_K \) on \( E_n \).

6.2 Energy Considerations

In this preliminary discussion the general features of the energy balance will be considered. Scission neutrons have been ignored, but will be treated in a later section. We will be concerned with the specific case of U235. Other nuclei can be treated in a similar way.

If \( E_T \) is the total energy released in the fission process then

\[
E_T = E_o + B_o + E_n = \bar{E}_K + \bar{E}_\nu + \bar{E}_\gamma
\]

where \( E_o \) is the total energy released in a hypothetical spontaneously fissioning U236 nucleus, \( B_o \) is the binding energy of the incident neutron and \( E_n \) is the incident neutron energy. \( \bar{E}_K \) is the average total kinetic energy of the fission fragments (averaged over the mass distribution), \( \bar{E}_\nu \) is the average energy expended in neutron evaporation and \( \bar{E}_\gamma \) is the average total gamma ray emission.

The average number of neutrons emitted is given by

\[
\bar{V}_p = \frac{\bar{E}_\nu}{\bar{B}_n + \bar{E}}
\]
Here $\bar{B}_n$ is the average binding energy of the emitted neutrons and $\bar{\epsilon}$ their average centre of mass kinetic energy. It follows that

$$
\frac{d\bar{\gamma}_p}{d\bar{E}_\gamma} = \frac{1}{(\bar{B}_n + \bar{\epsilon})} \left[ 1 + \frac{\bar{\epsilon}}{(\bar{B}_n + \bar{\epsilon})^2} \left( \frac{d\bar{B}_n}{d\bar{\gamma}_p} + \frac{d\bar{\epsilon}}{d\bar{\gamma}_p} \right) \right]^{-1}
$$

Equation 6.3

The quantities on the right hand side of equation 6.3 have been listed in Meadows and Whalen (1967). $\bar{B}_n$ and $d\bar{B}_n/d\bar{\gamma}_p$ may be obtained by making suitable averages over the mass yield distribution (Milton and Fraser 1962) of quantities calculated by an empirical mass formula (Cameron 1957; Milton 1962) and are

$$
\bar{B}_n = 5.0 \text{ MeV}
$$

$$
\frac{d\bar{B}_n}{d\bar{\gamma}_p} = 0.12 \text{ MeV}
$$

The other quantities may be obtained from experimental information listed in Terrell (1965) and (1962). $\frac{d\bar{\epsilon}}{d\bar{\gamma}_p}$ may be obtained from the semi-empirical relationship given by Terrell (1965)

$$
\bar{\epsilon} = 0.65 (\bar{\gamma}_p + 1) \frac{1}{2}
$$

Equation 6.4

The values used are

$$
\bar{\epsilon}_{\gamma} = 15.0 \text{ MeV}
$$

$$
\bar{\epsilon} = 1.21 \text{ MeV}
$$

$$
\frac{d\bar{\epsilon}}{d\bar{\gamma}_p} = 0.18 \text{ MeV}
$$
If we now assume Fowler's hypothesis, namely that all additional compound excitation appears as fragment excitation, and further assume that the mass division and the average total gamma ray energy are constant, then \( \frac{dE}{n} \) becomes \( \frac{d\bar{E}}{n} \). Thus

\[
\frac{d\bar{E}}{dE} = 0.14 \text{ MeV}^{-1}
\]

This slope disagrees with the average slope of the recent U235 data below about 3 Mev (see Table 6.1). Furthermore, no fine structure can be predicted on the basis of these simple minded arguments.

Let us consider what effects may arise from the discrete nature of the low lying fission channels. In the original A. Bohr (1956) theory, a fissioning nucleus with compound excitation near the fission threshold is cold with respect to internal excitation as it passes the saddle point. All available energy is bound up in potential energy of deformation and the spectrum of transition states should resemble the collective states of the heavy deformed nuclei near their ground states. These states are characterised by the quantum number \( K \) being the projection of the total spin \( I \) of the compound nucleus on the symmetric axis. For an even-even nucleus such as the compound nucleus U236, the lowest state is expected to have \( K=0 \) and correspond to an entirely paired configuration. The band with \( K=0 \) consists of rotational levels with energies given by
\[ E_{I,K} = E_K + \frac{\hbar^2}{2J} \, I \,(I+1) \]  

where \( J \) is the moment of inertia. For the highly deformed transition state nuclei, \( \frac{\hbar^2}{2J} \) is equal to approximately 2 keV, versus 7 keV for ground state nuclei. If the nucleus does not possess reflection symmetry about its centre, the ground state splits into an inversion doublet, the higher band of which possesses spin states of negative parity \( 1^-, 3^-, 5^- \) etc. and the lower band positive parity states \( 0^+, 2^+, 4^+ \) etc. The separation of the two bands is \( \hbar \omega \) where \( \omega \) is the tunnelling frequency of the nucleus between its mirror shapes. The spectrum of transition states is also expected to contain bands with \( K=1 \) (bending mode) and \( K=2 \) (gamma band). At higher excitation, sufficient energy is available to split the entirely paired configuration and single particle states become available (see Fig. 2.7).

For the U236 compound nucleus, the two \( K=0 \) bands have been identified with the two lowest thresholds observed in \((d,pf)\) studies on U235 by Northrop et al. (1963). The positive parity band is located 600 keV below the neutron binding energy and the negative parity band 200 keV above the neutron binding energy. The \( K=1 \) and \( K=2 \) bands have not been observed in angular distribution studies, however their presence has been postulated by Bolshov et al. (1968) and Strutinsky (1965) to explain their particular interpretation of the \( \frac{\nu_n}{\nu_p} (E_n) \) data. The lowest states associated with single particle excitations were located 2.3 to 2.9 MeV above the fission threshold or 1.7 to 2.3 MeV above the neutron binding energy by Britt et al. (1963).
This anomalously large value of the nuclear pairing gap, $2\Delta_0$, has been the subject of some controversy. For the heavy deformed nuclei the pairing gap is generally of the order of 1 MeV. Griffin (1965) has discussed this matter in some detail. Other comments on this subject include papers by Stephen and Syzmanski (1968) and Gustafson et al. (1966). The known transition state energy level scheme is shown in fig. 6.1

Let us consider the distribution of energy at the saddle point. The variable energy at the saddle point consists of a discrete collective term corresponding to a particular transition state and a continuous term which takes up the excess energy by which the compound excitation exceeds the transition state energy. The most likely form for the excess energy is the relative kinetic energy of the two components of the saddle point nucleus as it passes the saddle point. The strength of the coupling of the saddle point collective energy to the excitation energy of the fragments is a matter of profound significance to the theory of the fission process. Can measurements of the energy dependence of $\nu_p$ or $E_K$ determine the validity of either the weak or strong coupling assumptions?

Since the ground state of the U235 target nucleus is $^7/2^-$, s-wave fission at low excitation will proceed via the negative parity band and p-wave fission via the positive parity band. Optical calculations by Moldauer (1961), Auerbach and Perey (1962) indicate that p-wave fission is the dominant mode of compound nucleus formation from 0.1 MeV to 3 MeV. Thus as the incident neutron energy is increased
Fig. 6.1. Collective States for Compound Nucleus U236.
from thermal to 0.1 MeV the dominant fission channels cease to be the negative parity group and become the positive parity group. As a consequence, there is a sizeable reallocation of the total energy in the saddle point system - in particular, a decrease in the average collective energy and a corresponding increase in the excess energy. It is expected that the excess energy appears entirely as fragment excitation. Therefore, preferential appearance of the saddle point collective energy as a particular de-excitation mode should be readily observable in the energy dependence of $\tilde{v}_p$ or $E_K$. For example, if the coupling is weak then a peak is expected in the $\tilde{v}_p (E_n)$ dependence for U235 in the changeover from s-wave to p-wave fission. The data of Blyumkina et al. (1964) and Meadows and Whalen (1967) have been interpreted as evidence of weak coupling. Alternatively, strong coupling should lead to linear $\tilde{v}_p (E_n)$ behaviour and constancy for $E_K$.

However the interpretation of the $\tilde{v}_p (E_n)$ dependence is not as simple as it would appear from the above discussion. As a consequence of the improvement of our knowledge of the nature of nuclear shells, Strutinsky (1969, 1968, 1967) has shown that the fission barrier is characterised not by a single maximum in the potential energy curve as in the original LDM but by at least two humps in the curve separated by a pronounced potential well (fig. 6.2). (see Chapter II). With each potential barrier one can expect collective states of the A. Bohr type. However the theory has not been developed to the state where the characteristics of these two barriers are well known. At
Figure 6.2. Double Humped Fission Barrier From Strutinsky (1969).
this stage it is not even clear which of the two barriers is higher for the general body of fissile nuclei. There is some evidence to suggest that for $A<235$ the second barrier is higher while for $A>239$ the first barrier is higher (Strutinsky 1969). Bach et al. (1969) have analysed the resonances observed in their $(d, pf)$ fission cross section studies of $^{233}U$, $^{235}U$, $^{239}Pu$ and $^{241}Pu$ assuming the first potential barrier to be slightly higher than the second ($E_A = 6.0$ MeV, $E_B = 5.8$ MeV). Britt et al. (1969) have assumed $E_A = 5.95$ MeV and $E_B = 5.25$ MeV in their analysis of the observed resonances in the $^{240}Pu(p,p'f)$ reaction cross section. The relative heights of the two potential barriers is a crucial factor in determining the sensitivity of the $\tilde{\nu}_p (E_n)$ data to channels effects should they exist. It is obvious that the channel structure of the second potential barrier will determine any structure in $\tilde{\nu}_p (E_n)$ caused by weak coupling of the collective energy to the nucleonic degrees of freedom. This fact is independent of which barrier is higher. However the transition state energy level scheme depicted in Fig. 6.1 will be a feature of the higher potential barrier. Thus with varying neutron energy one selects preferentially different channels at the higher potential barrier, and should this not be the second potential barrier, interpretation of any observed fine structure becomes exceedingly difficult, if not impossible, at the present time.

6.3 The $\tilde{\nu}_p (E_n)$ Dependence for $^{235}U$ (Experimental Details)

Precise measurements have been made of the energy dependence of $\tilde{\nu}_p$ for neutron fission from $E_n = 0-2$ MeV. The experimental method
was similar to that described in Chapter 4. The principal difference relates to the source of neutrons and the shielding arrangements. The U235 fission counter was that described in Tables 4.1 and 4.2.

Neutrons of appropriate energy were obtained using analysed proton beams from a 3 MeV Van de Graaff accelerator. The reactions used were $^7\text{Li}(p,n)^7\text{Be}$ and $^3\text{T}(p,n)^3\text{He}$. The first reaction was used for studies between 0-1 MeV neutron energy and the second for neutron energies above 1 MeV. The proton beam (50-150 µA) was focussed through a 2 mm aperture onto the target assembly. Focussing was achieved in the usual way with a quadrupole magnet and a moving magnet system. To prevent burn-off of the target material, the target was cooled using annular water cooling and was also wobbled to distribute the heat. Details of the target design may be observed in Fig. 6.3. The neutron beam was collimated to a 1 inch diameter at the centre of the scintillator - a distance of 1.6 metres from the lithium or tritium target. The collimator materials consisted of successive thicknesses of cast iron and borated polythene. The collimator hole tapered to a diameter of 2mm at the target. The details of the radiation shield surrounding the scintillator tank may be seen in Fig. 6.4. The incident neutron energy resolution was set by the Li target thickness and in the case of the tritium targets by the evaporated Ti thickness. In both cases the energy resolution was determined experimentally using the threshold technique described by Marion and Fowler (1960).

The raw data were corrected for dead time losses as described in Chapter 4. In addition, corrections were applied for fission neutron
Fig. 6.3  WOBBLING TARGET SYSTEM.
Fig. 6-4. Neutron Collimator and Shield for Scintillator Tank.
spectra differences between U235 for different incident neutron energies and the calibration material Cf·252. The correction varied from $-0.55 \pm 0.22$ per cent for the 110 keV point to $0.46 \pm 0.22$ per cent at the 1.9 MeV point. Corrections were necessary for the impurities in the U235 target (see Table 4.2) and, for the higher energy measurements with the Li target, a correction was also made for the second neutron group from the $\text{Li}^7(p,\alpha \gamma)$ reaction using the data from Marion and Fowler (1960) and the $\bar{\nu}_p(E_n)$ data from this work. For the original collimator assembly, the fission rate from neutrons degraded in energy was approximately $11\pm3$ per cent of the total fission rate and a correction was applied for these neutrons assuming they represented a thermal fission background. Although this system was satisfactory for the lower energy determinations, it was inadequate for those at higher energies where the correction was considerably larger and the error in the correction became significant. Consequently the collimator system was improved considerably (final system as in fig. 6.4) and the thermal fission background rate was reduced to $2.0\pm0.5$ per cent. The background fission rate was determined by pulsing the accelerator and employing time of flight methods. Fig. 6.5 shows a typical time of flight spectrum for 1.9 MeV neutrons from a tritium target.

For the spontaneous fission of Cf252, the fission fragments are emitted isotropically in the laboratory system whereas in the fast neutron fission of U235 the fragment angular distribution has a small peak in the direction of the neutron beam. Since the angular distribution of post scission neutrons is strongly correlated with
Fig. 6-5. Neutron Time of Flight Spectrum (Beam Pulse 25 nsec wide, $E_n = 1.90 \pm 0.05$ MeV)
fragment direction, the angular distribution of fission neutrons is slightly different in the two cases of spontaneous fission of Cf252 and fast neutron fission of U235. Because of the axial tube through the centre of the tank, the scintillator detection efficiency is dependent to some extent on the average angular distribution of fission neutrons. The systematic error introduced in relative measurements by such considerations has been calculated by Mather et al (1964) for a large liquid scintillator tank identical with the tank employed here, using fragment anisotropy data of Simons and Henkel (1960) and the neutron angular distribution data of Ramanna and Rama Rao (1958). These calculations showed that for 7.5 MeV neutron induced fission, the systematic error introduced in the U235 measurement is 0.2 per cent relative to the Cf 252 measurement. For measurements below 7 MeV the error was shown to be negligible. On this basis we have made no systematic correction for fragment anisotropy in this work and have regarded the error in such an approach as negligible.

6.4 U235 Results

The final results after correction and including all sources of error are listed in Table 6.2 and displayed in Fig. 6.6. All results are relative to Cf252 for which $\bar{\nu}_p$ is assumed to be 3.782 without error.
Fig. 6. $\bar{V}_p$ Versus Incident Neutron Energy ($E_n$) - This work.
The relative accuracy of each point with respect to the others is slightly better than that stated as all errors include a contribution from the error in the fission spectra differences correction. A straight line fit to the data points indicates that they are adequately
represented by the straight line.

\[(2.412 \pm 0.005) + (0.114 \pm 0.008) E_n\] 6.7

where \(E_n\) is in MeV.

It is significant that all the measured points are statistically consistent with the linear fit and there is no evidence whatsoever in the data points of any deviation which could be interpreted as fine structure.

6.5 Comparison with Previous U235 Measurements

All previous measurements of reasonable accuracy in the energy range 0-2 MeV are listed in Table 6.7. All measurements are normalised to \(\bar{\nu}_p\) (Cf 252) = 3.782 except those cases where the thermal \(\bar{\nu}_p\) value for U235 has been the reference. For the latter the reference normalisation has been made to the evaluated thermal value from Fillmore (1968) i.e. \(\bar{\nu}_p\) U235 (thermal) = 2.418.

[Note: As mentioned in Chapter 4, there still exists a discrepancy of approximately 2 per cent between various absolute calibration methods for Cf252. A recent analysis of existing data has been carried out by Hanna et al. (1969) who recommend a preliminary value of \(\bar{\nu}_T\) (Cf252) of \(3.765 \pm 0.012\) i.e. \(\bar{\nu}_p = 3.756\). The final results from this evaluation were not available at the time of preparation of this thesis and we have continued to use the value \(\bar{\nu}_p\) (Cf252) = 3.782. This value has been most frequently used over recent years and our choice of it has been to minimise any confusion.]
Table 6.7

Previous $\tilde{y}$ Values for U235

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>$\tilde{\gamma}_p$</th>
<th>Experiment</th>
<th>Energy (keV)</th>
<th>$\tilde{\gamma}_p$</th>
<th>Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>1360</td>
<td>2.551±0.014</td>
<td>Soleilhac et al (1969)</td>
<td>1020</td>
<td>2.534±0.027</td>
<td></td>
</tr>
<tr>
<td>1870</td>
<td>2.631±0.014</td>
<td></td>
<td>1230</td>
<td>2.551±0.037</td>
<td></td>
</tr>
<tr>
<td>39</td>
<td>2.422±0.017</td>
<td>Meadows &amp; Whalen (1967)</td>
<td>1440</td>
<td>2.555±0.037</td>
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<tr>
<td>46</td>
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<tr>
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<td>2.610±0.032</td>
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<tr>
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<tr>
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<tr>
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<tr>
<td>550</td>
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<tr>
<td>640</td>
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<td>2.455±0.038</td>
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</table>

Note: The values for $\tilde{\gamma}_p$ are given with uncertainties in the form of a mean + standard deviation.
<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>$\bar{\nu} / \bar{\nu}$</th>
<th>Experiment</th>
<th>Energy (keV)</th>
<th>$\bar{\nu} / \bar{\nu}$</th>
<th>Experiment</th>
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<td>470</td>
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<td>990</td>
<td>2.507±0.029</td>
<td></td>
<td>815</td>
<td>2.476±0.026</td>
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</tr>
<tr>
<td>Thermal</td>
<td>2.404±0.012</td>
<td>Mather et al. (1964)</td>
<td>1080</td>
<td>2.536±0.026</td>
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</tr>
<tr>
<td>40</td>
<td>2.415±0.042</td>
<td></td>
<td>30</td>
<td>2.439±0.026</td>
<td>Meadows &amp; Whalen (1962)</td>
</tr>
<tr>
<td>230</td>
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<td></td>
<td>200</td>
<td>2.454±0.016</td>
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<tr>
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<td>620</td>
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<td>1110</td>
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<tr>
<td>700</td>
<td>2.449±0.016</td>
<td></td>
<td>1580</td>
<td>2.599±0.020</td>
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<tr>
<td>840</td>
<td>2.521±0.021</td>
<td></td>
<td>1760</td>
<td>2.594±0.021</td>
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</tr>
<tr>
<td>930</td>
<td>2.491±0.020</td>
<td></td>
<td>210</td>
<td>2.440±0.015</td>
<td>Butler et al. (1961)</td>
</tr>
<tr>
<td>1170</td>
<td>2.548±0.021</td>
<td></td>
<td>620</td>
<td>2.482±0.022</td>
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<tr>
<td>1470</td>
<td>2.575±0.020</td>
<td></td>
<td>1120</td>
<td>2.515±0.019</td>
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<tr>
<td>1940</td>
<td>2.648±0.021</td>
<td></td>
<td>1580</td>
<td>2.582±0.020</td>
<td></td>
</tr>
<tr>
<td>Thermal</td>
<td>2.432±0.020</td>
<td>Hopkins &amp; Diven (1963)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The data of Mather et al. (1964) have been adjusted by approximately -0.33 per cent to conform with an improved fission neutron spectra difference correction (Boldeman and Dalton, 1967). All data are plotted in Fig. 6.7.

### 6.6 Comments on Consistency

First observation of the plotted \( \bar{v}_p \) data in Fig. 6.7 suggests considerable discrepancies between the various sets. However some clarification is possible. The present results are consistent with those from Soleilhac (1969), Hopkins and Diven (1963), Mather et al. (1964), Meadows and Whalen (1962) and Butler et al. (1961). A least squares fit to the present data and the five sets above shows that the combined data are well represented by the straight line

\[
(2.415 \pm 0.004) + (0.110 \pm 0.006 E_n)
\]

The only point significantly different from the linear fit is the 230 keV value from Mather et al. (1964), and in this case the divergence is less than two standard deviations. In view of the conformity with the linear fit of the other sets with values in this energy region, no significance is attached to the deviation of this 230 keV point.

Blyumkina et al. (1964). Blyumkina et al. (1964) interpreted their data as evidence of fine structure in the vicinity of 200-600 keV neutron energy. They claimed that further evidence in support of this view was the correlated structure observed in their measurements of the average total kinetic energy of the fission fragments with
FIGURE 6.7 $\bar{\nu}_p$ VERSUS INCIDENT NEUTRON ENERGY ($E_n$) — THIS WORK + OTHER MEASUREMENTS
incident neutron energy. Although the qualitative trend of their two sets of measurements is similar the quantitative agreement is particularly poor. We have ignored the Blyumkina et al. (1964) $E_K$ data during this evaluation and have considered their $\bar{\nu}_p$ data purely on its own merits. The kinetic energy data will be reviewed in a subsequent section. Preliminary perusal of the Blyumkina data suggests that a straight line might be a reasonable fit. We have made a straight line fit to the Blyumkina et al. data and find the straight line (6.9) an excellent representation of their data.

$$\bar{\nu}_p = (2.439 \pm 0.018) + (0.056 \pm 0.033) E_n$$ \hspace{1cm} 6.9

Although the slope is somewhat different from that in 6.8 it still agrees within two standard deviations. More significantly, a comparison of each point from Blyumkina with the linear fit (6.8) indicates that only at 390 keV does the difference exceed one standard deviation, and then only marginally. A satisfactory conclusion from these observations is that the Blyumkina et al. data is in good statistical agreement with the present results. We find no basis within Blyumkina's $\bar{\nu}_p$ data for her claim of fine structure.

Prokhorova et al. (1966). Prokhorova et al. (1966) consider their data to be consistent with a stepped dependence of $\bar{\nu}_p$ on compound excitation. However, the statistical accuracy of their data points is such that a linear dependence is still the most acceptable fit and furthermore, each of their data points is statistically consistent with the linear dependence (6.8).
Meadows and Whalen (1967). The data from Meadows and Whalen (1967) are in complete statistical disagreement with the present set of values in the interesting region 200-700 keV. They find two maxima which are statistically significant at 325 keV and 425 keV, and two minima at 358 keV and 548 keV. We have abandoned the attempt to reconcile their data with the present results. It should be mentioned however, that their data are far more incompatible with the present set of values than with any other set.

Colvin and Sowerby (1965). The data points from Colvin and Sowerby (1965) lie mainly outside the region where fine structure in the $\langle v_p | E_n \rangle$ dependence was previously reported. Their values from 500 keV to 600 keV are consistently higher than the present data, and are, in fact, higher than the majority of the data, apart from Meadows and Whalen (1967).

**Consistency Summary:** A straight line fit has been made to all the data points excluding Meadows and Whalen (1967). The linear fit so obtained

$$\begin{align*}
2.416 + 0.004 + 0.107 + 0.004 E_n
\end{align*}$$

is found to be an excellent representation of the $\langle v_p | E_n \rangle$ data. The inclusion of the Colvin and Sowerby (1965) data does not significantly affect the quality of the fit.

The quality of a linear fit to the majority of the data between 0-2 MeV cannot be taken alone as adequate refutation of the previously proposed fine structure in the $\langle v_p | E_n \rangle$ dependence, as a significant
and consistent discrepancy at a particular point would be minimised by the weight of the data at all other points. It is significant, however, that the present set of values shows no fine structure whatsoever, nor, in general, do the majority of previous determinations. Where data points in one set deviate appreciably from the straight line, there is no significant correspondence in other sets.

6.7 Variation of the Average Total Kinetic Energy

The $\bar{E}_p$ data show no fine structure and therefore in accordance with equation 6.1 are in conflict with some of the average total kinetic energy data which do show significant structure. Consequently, the energy dependence of $E_K$ has been re-examined.

Coincident fission fragments were recorded by two solid state surface barrier detectors placed at 0.3 cm distance on either side of the fissioning source. The detectors were made of n-type silicon wafers of reactivity 7000 ohm-cm, had an active area of 2.5 cm$^2$ and were operated at 90 V reverse bias. The source consisted of 15 $\mu$gm cm$^{-2}$ of 93 per cent enriched U235 electro-sprayed on gold and resin coated VYNS plastic film. The resin had the property of improving the uniformity of the deposit. Thicknesses of gold, resin and VYNS layers were 17, 10 and 18 $\mu$gm cm$^{-2}$ respectively. Care was taken to prepare a uniform source whose fission fragment energy spectrum had a good peak to valley ratio ($\approx 7:1$) with a fast falling tail on the low energy side.

The source and detectors were mounted in a vacuum chamber in a plane perpendicular to the incident neutron beam as shown schematically
in Fig. 6.8. The maximum angle of divergence of the neutrons reaching the source was 25°. Neutrons of specific energy were obtained using analysed proton beams from a 3 Mev Van de Graaff accelerator incident on a 30 keV thick spray-cooled lithium target. Currents of the order of 60 µA were used. The variation in the average total kinetic energy was measured by comparing the mean total kinetic energy at a specific neutron energy with that for thermal neutron fission. A fast neutron measurement lasted an hour and was preceded and followed by a thermal calibration. Thermal neutrons were obtained using a paraffin block to moderate the fast neutrons. The counting rates obtained were typically of the order of 10 min⁻¹ for the fast neutron runs and 600 min⁻¹ in the thermal runs.

A block diagram of the electronics is shown in Fig. 6.9. The pulses from the detectors were taken through time pick off units, suitably amplified and then fed to the inputs of a dual analog to digital converter which formed part of a PDP-7 on line computer. The time pick off units were set to reject natural alpha activity from the source. A fast coincidence (2T = 100 nsecs) was required to gate the linear inputs of the dual A.D.C.'s. Of the six regions available in the PDP-7 computer (each of 1024 channels) only two were employed. The PDP-7 was programmed to store the individual pulse height analysed distributions of the two detector channels in the first region (i.e. from 0 to 511 and from 512 to 1023) and the summed distribution in the second region. The gains of the two channels were approximately matched and increased to the maximum possible extent when the channel width corresponded to about 0.2 MeV.
Fig 6.8. DETECTOR SYSTEM FOR TOTAL
FRAGMENT DETECTORS

FISSIONING SOURCE

KINETIC ENERGY MEASUREMENTS.
Fig. 6.9. LOGIC OF ELECTRONICS FOR FISSION FRAGMENT KINETIC ENERGY EXPERIMENT.
The sum distribution, aside from a few minor corrections, was proportional to the total kinetic energy distribution. Sufficient counts were collected in each thermal run to define the average of this distribution to better than one channel accuracy. Similarly sufficient counts were taken for each fast neutron energy such that the average of the sum distribution was defined to an accuracy of better than 0.1 per cent. All the events i.e. pairs of numbers for each run (fast as well as thermal) were punched out on paper tape which was later read onto magnetic tape for analysis on an IBM 360/50 computer.

6.8 Analysis of $\bar{E}_K$ Data

In order to determine $\Delta \bar{E}_K (E_n)$, the difference between the average total fragment kinetic energy for fission by neutrons of energy $E_n$ and thermal neutron fission, the raw data were analysed as follows

1. Gaussian fits were made to the light and heavy fragment pulse height distributions of the thermal runs before and after each fast run. The average channel width was then determined by using the known difference of average kinetic energies of the light and heavy fragments in thermal neutron fission of U235. (Milton and Fraser 1962).

2. The difference between the average of the sum pulse height distribution obtained in the fast run and the mean of the averages obtained for the sum distribution of the thermal runs preceding and following the fast run, was converted
into energy units by using the above calibration.

3. The data for each fast run were analysed in this way and the weighted average of $\Delta \tilde{E}_K$ for all fast runs at a particular energy is shown in the second column of Table 6.8.

The corrections to $\Delta \tilde{E}_K$ considered were:

1. A correction for energy loss in the source foil and the gold layer on the detector surface which depended on the anisotropy of fragment angular distribution. This correction was estimated by performing a Monte Carlo calculation to determine the average thickness seen by fragments having an anisotropic angular distribution with respect to the incident neutron beam (Simmons and Henkel, 1960). This correction is shown in column 3 of Table 6.8.

2. A correction for the variation in the prompt neutron emission with compound excitation. This correction accounts for the momentum effects produced by the variation in the number of neutrons emitted. The correction is quite small and is virtually insensitive to fine structure in the $\tilde{\gamma}_p (E_n)$ curve. (column 4 of Table 6.8).

3. The correction for the centre of mass motion of the fragment pairs was calculated and found to be negligible. The increase in the energy of one fragment due to the momentum brought in by the incident neutron is approximately equal to the decrease in energy of the other fragment.
4. The fraction of fission events due to neutrons inelastically scattered from the surroundings was estimated by operating the accelerator in a nanosecond pulsing mode and observing the time distribution of the fission events after each neutron burst. The scattered contribution was less than 2 per cent of the total and so the results are not affected in any significant way.

The final results are shown in the last column of table 6.8 together with the errors which include the statistical errors and the errors due to the finite channel width.

<table>
<thead>
<tr>
<th>$E_n$ (keV)</th>
<th>$\Delta E_K$ (MeV)</th>
<th>Anisotropy Correction</th>
<th>Neutron Emission Correction</th>
<th>$\Delta E_K$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
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<td>-0.105</td>
<td>-0.016</td>
<td>+0.017</td>
<td>-0.104±0.214</td>
</tr>
<tr>
<td>300±17</td>
<td>+0.252</td>
<td>-0.022</td>
<td>+0.026</td>
<td>+0.256±0.196</td>
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<tr>
<td>350±15</td>
<td>+0.041</td>
<td>-0.025</td>
<td>+0.030</td>
<td>+0.046±0.180</td>
</tr>
<tr>
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<td>+0.192</td>
<td>-0.028</td>
<td>+0.034</td>
<td>+0.193±0.189</td>
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<tr>
<td>450±15</td>
<td>-0.058</td>
<td>-0.031</td>
<td>+0.038</td>
<td>-0.051±0.195</td>
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<tr>
<td>500±15</td>
<td>-0.135</td>
<td>-0.034</td>
<td>+0.043</td>
<td>-0.123±0.211</td>
</tr>
<tr>
<td>600±15</td>
<td>+0.116</td>
<td>-0.040</td>
<td>+0.051</td>
<td>+0.126±0.202</td>
</tr>
<tr>
<td>700±15</td>
<td>-0.066</td>
<td>-0.045</td>
<td>+0.060</td>
<td>-0.051±0.242</td>
</tr>
<tr>
<td>900±15</td>
<td>-0.053</td>
<td>-0.056</td>
<td>+0.077</td>
<td>-0.032±0.250</td>
</tr>
</tbody>
</table>

6.9 Discussion of $E_K$ Data

The present results have been plotted in Fig. 6.10. Also shown there are the previous results from Blyumkina et al. (1964), Bolshov et al. (1968) and Dyachenko (1968).
Fig. 6.10. $\Delta E_k$ versus incident neutron energy $E_n$: Comparison of data by different authors for U235.
The agreement between the present results and those from Dyachenko et al. (1968) who used a similar method is excellent. There does appear to be some discrepancy between our data and Blyumkina et al. (1964) for the energy region 200-400 keV. However the disagreement is more with the interpretation placed on the Blyumkina data rather than with the data itself. In fact the probability that the Blyumkina data belongs to the linear average of the present data and that of Dyachenko et al. is approximately 1 in 100. The data from Bolshov et al. (1968) lie mainly outside the interesting region and are not in conclusive agreement with any group.

It will be observed that the present data are in good agreement with Fowler's hypothesis namely, constant average total kinetic energy with compound excitation, and do not show any evidence whatsoever of fine structure. In this respect the $\bar{E}_K$ data strongly support the $\bar{v}_p (E_n)$ data reported earlier in this chapter which also show no evidence of fine structure. However the constancy of the average total kinetic energy does introduce a problem with the slope of the $\bar{v}_p (E_n)$ dependence. The discrepancy between the measured slope of 0.107 MeV$^{-1}$ and that of 0.14 MeV$^{-1}$ calculated from conservation of energy requires an explanation.

6.10 Implications of Lack of Fine Structure in $\bar{v}_p (E_n)$ and $E_K (E_n)$

In terms of the original A. Bohr theory the lack of fine structure in $\bar{v}_p$ and $E_K$ has significant relevance to fission theory.
The data imply that the collective energy is strongly coupled to the nucleonic degrees of freedom at scission. The excess energy discussed previously also contributes entirely to the excitation energy of the fragments. Of course, we have at this stage overlooked the problem with energy balance i.e. the difference between the measured slope of \( \frac{\nu}{p} (E_n) \) of 0.107 MeV\(^{-1}\) and the calculated one of 0.14 MeV\(^{-1}\). This will be the subject of section 6.11.

The adiabatic model of the fission process is characterised by the assumption that the single particle motion follows the collective motion adiabatically. In other words the collective motion is slow compared to the single particle motion. Thus in the absence of viscosity coupling between the collective degrees and the nucleonic degrees must be weak. Alternatively the statistical model implies strong coupling. The present results in terms of the A. Bohr theory therefore favour the Statistical model or at least an adiabatic picture with high viscosity.

This conclusion can no longer be drawn from the linearity of the \( \tilde{E}_K \) and \( \tilde{\nu}_p \) data in view of the recent Shell Model developments brought about by Strutinsky. The crucial feature of the Strutinsky doublehumped potential barrier to fission that is required for an interpretation of the \( \tilde{\nu}_p \) or \( \tilde{E}_K \) data is which of the two humps is higher. If the second potential barrier is higher (barrier B in fig. 6.2) then the situation is effectively the same as described above and the present data are consistent only with strong coupling. Alternatively if the first potential barrier A is higher then no
conclusion on the strength of the coupling can be drawn from the linear data. It is possible, for example, that the nucleus spends a sufficiently long period in the potential well between the humps such that it forgets the properties that it had when passing over the first barrier. Furthermore, even if these properties are indirectly preserved, only a small difference is required in the relative heights of the two barriers such that, for a particular incident channel at the first barrier, a large number of channels is available at the second barrier. Averaging occurs and any structure effects that could be expected from weak coupling are smeared out. In this case, weak coupling is not excluded by the linear $\tilde{\nu}_p$ and $E_K$ data. Unfortunately, it is not known for the compound nucleus U236 which barrier is higher.

For the compound nucleus U234, however, there is reasonable evidence to suggest that the second potential barrier is higher, Strutinsky (1969). An investigation of the energy dependence of $\tilde{\nu}_p$ for neutron fission of U233 may therefore resolve the question of weak or strong coupling. Before consideration is given to this matter, it is necessary to attempt an explanation of the apparent lack of energy balance for U235.

6.11 The Energy Balance

In section 6.2 it was shown that with simple assumptions the slope of the $\tilde{\nu}_p (E_n)$ dependence for U235 should be approximately 0.14 MeV$^{-1}$. The measured slope in the region 0-2 MeV has been shown
to be 0.107 MeV\(^{-1}\). Furthermore the apparent loss of total energy does not appear in the kinetic energy of the fission fragments which has been found to be constant between 0 and 1 MeV. Obviously, one of the assumptions made in section 6.2 in the derivation of the slope of 0.14 MeV\(^{-1}\) must be relaxed. The assumptions were:

1. Mass distribution unchanged in the region 0-2 MeV
2. Scission neutrons could be ignored
3. Constant average gamma ray energy.

Data for other fissile nuclei will assist in the consideration of these assumptions and these measurements are now described.

6.12 The Energy Dependence of \(\bar{\nu}_p\) for U\(_{233}\) and Pu\(_{239}\)

Measurements have been made of the energy dependence of \(\bar{\nu}_p\) for U\(_{233}\) and Pu\(_{239}\) using the techniques described previously in this thesis. Table 6.9 gives details of the U\(_{233}\) and Pu\(_{239}\) fission counters.

<table>
<thead>
<tr>
<th>Counter</th>
<th>No. of Chambers</th>
<th>Wt. of Material</th>
<th>U233</th>
<th>U234</th>
<th>U235</th>
<th>U236</th>
<th>U238</th>
<th>Pu239</th>
<th>Pu240</th>
<th>Pu241</th>
</tr>
</thead>
<tbody>
<tr>
<td>U233</td>
<td>2</td>
<td>8 mgms</td>
<td>99.27</td>
<td>0.07</td>
<td>0.04</td>
<td>0.07</td>
<td>0.53</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu239</td>
<td>2</td>
<td>6.7 mgms</td>
<td></td>
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<td></td>
<td></td>
<td>96.9</td>
<td>3.0</td>
<td>0.1</td>
</tr>
</tbody>
</table>

The usual corrections were made to the data and, in the case of Pu239, a correction was applied to account for the spontaneous fission rate of
Pu240. The correction was estimated from the data in Chapter V. The final results relative to \( \tilde{\nu}_p \) (Cf252) = 3.782 are listed in Table 6.10. The U233 \( \tilde{\nu}_p \) data are plotted in Fig. 6.11.

### Table 6.10

\( \tilde{\nu}_p \) Data for U233 and Pu239

<table>
<thead>
<tr>
<th>Neutron Energy (keV)</th>
<th>( \tilde{\nu}_p )</th>
<th>Neutron Energy (keV)</th>
<th>( \tilde{\nu}_p )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal</td>
<td>2.491( \pm )0.008</td>
<td>Thermal</td>
<td>2.897( \pm )0.008</td>
</tr>
<tr>
<td>300( \pm )25</td>
<td>2.502( \pm )0.014</td>
<td>200( \pm )65</td>
<td>2.919( \pm )0.019</td>
</tr>
<tr>
<td>485( \pm )31</td>
<td>2.508( \pm )0.010</td>
<td>350( \pm )52</td>
<td>2.926( \pm )0.032</td>
</tr>
<tr>
<td>600( \pm )32</td>
<td>2.546( \pm )0.012</td>
<td>550( \pm )35</td>
<td>2.946( \pm )0.015</td>
</tr>
<tr>
<td>700( \pm )25</td>
<td>2.546( \pm )0.011</td>
<td>700( \pm )35</td>
<td>2.963( \pm )0.015</td>
</tr>
<tr>
<td>917( \pm )33</td>
<td>2.564( \pm )0.012</td>
<td>900( \pm )45</td>
<td>2.980( \pm )0.019</td>
</tr>
<tr>
<td>1500( \pm )50</td>
<td>2.645( \pm )0.019</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1870( \pm )50</td>
<td>2.685( \pm )0.022</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The U233 data in fig. 6.11 have been fitted by the two straight lines

\[
\tilde{\nu}_p = (2.491\pm0.008) + (0.035\pm0.026)E_n \text{ for } E_n < 0.40 \text{ MeV} \quad 6.11
\]

and

\[
\tilde{\nu}_p = (2.458\pm0.013) + (0.123\pm0.014)E_n \text{ for } E_n > 0.40 \text{ MeV} \quad 6.12
\]
Fig. 6.11. Data for U233. Present Results.
which are considered the best representation of the data (see evaluation in section 6.13).

6.13 Evaluation of \( \bar{\nu}_p \) data for U233, U235 and Pu239 from 0-5.0 MeV

In this section, all existing data for each of U233, U235 and Pu239 between 0-5.0 MeV neutron energy have been considered to obtain the best fit to the energy dependence. We have limited the data to that below 5.0 MeV because of the observation of several changes of slope above that energy by Soleilhac et al (1969). Furthermore, changes in the mass yield should begin to have some observable effect upon the \( \bar{\nu}_p (E_n) \) dependence above this energy.

6.13.1 U233 Data

Previous \( \bar{\nu}_p \) data for U233 between 0 and 5 MeV are tabulated in Table 6.11. As before, all data have been normalised directly or indirectly to the assumed value of \( \bar{\nu}_p = 3.782 \) for the spontaneous fission of Cf252. The data from Mather et al. (1965) have been adjusted by approximately -0.30 per cent to conform with an improved fission neutron spectra difference correction (see Section 6.4). The agreement between the present data and previous measurements is satisfactory.

A preliminary inspection of our data in fig. 6.11 suggests that a change in slope probably occurs between 300 and 500 keV neutron energy. This was borne out by linear fits to our data and the combined data. The linear fits were poor - the principal
Table 6.11
Previous $\tilde{\nu}_p$ Data for U233

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Neutron Energy (MeV)</th>
<th>$\tilde{\nu}_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mather et al. (1965)</td>
<td>0.0</td>
<td>2.525±0.031</td>
</tr>
<tr>
<td></td>
<td>0.96±0.21</td>
<td>2.524±0.036</td>
</tr>
<tr>
<td></td>
<td>1.98±0.15</td>
<td>2.631±0.033</td>
</tr>
<tr>
<td></td>
<td>3.00±0.12</td>
<td>2.846±0.038</td>
</tr>
<tr>
<td></td>
<td>4.00±0.09</td>
<td>2.914±0.043</td>
</tr>
<tr>
<td>Hopkins &amp; Diven (1963)</td>
<td>0.0</td>
<td>2.480±0.026</td>
</tr>
<tr>
<td></td>
<td>0.28±0.09</td>
<td>2.496±0.033</td>
</tr>
<tr>
<td></td>
<td>0.44±0.08</td>
<td>2.509±0.033</td>
</tr>
<tr>
<td></td>
<td>0.98±0.05</td>
<td>2.560±0.035</td>
</tr>
<tr>
<td></td>
<td>1.08±0.05</td>
<td>2.517±0.030</td>
</tr>
<tr>
<td></td>
<td>3.93±0.29</td>
<td>2.992±0.040</td>
</tr>
<tr>
<td>Graves (1963)</td>
<td>4.0</td>
<td>2.99±0.12</td>
</tr>
<tr>
<td>Smirenkin et al. (1959)</td>
<td>4.0±0.03</td>
<td>2.99±0.10</td>
</tr>
<tr>
<td>Colvin &amp; Sowerby (1965)</td>
<td>0.0</td>
<td>2.477±0.022</td>
</tr>
<tr>
<td></td>
<td>0.58</td>
<td>2.46±0.05</td>
</tr>
<tr>
<td></td>
<td>0.95</td>
<td>2.56±0.09</td>
</tr>
<tr>
<td></td>
<td>1.48</td>
<td>2.51±0.09</td>
</tr>
<tr>
<td></td>
<td>2.12</td>
<td>2.58±0.05</td>
</tr>
<tr>
<td></td>
<td>2.58</td>
<td>2.81±0.06</td>
</tr>
<tr>
<td>Diven et al. (1956)</td>
<td>0.08</td>
<td>2.527±0.062</td>
</tr>
</tbody>
</table>
difficulty being the relatively high value of the thermal point. In an endeavour to determine the most likely energy at which the \( \bar{\nu}_p(E_n) \) slope changes, the fitting program was varied to exclude, firstly the thermal data, and subsequently all data below 300, 400 and 500 keV. The outcome of this analysis was inconclusive. Apart from the obvious improvement in the fits to the combined data and, in particular, to our data when thermal values are excluded, it was not possible to determine unambiguously where the change in slope occurred. This is not surprising in view of the extremely small magnitude of the effect under investigation. The only conclusion that can be drawn is that the slope probably changes between 300 and 500 keV. In the absence of further information we have placed the change in slope at approximately 400 keV, and have fitted linear relationships above and below this energy. The linear fits to the present data are those shown in section 6.12. (eqns 6.11 and 6.12). The fitted lines to the combined data are

\[
\bar{\nu}_p = (2.491 \pm 0.007) + (0.035 \pm 0.050)E_n \quad E_n < 0.44 \text{ MeV} \quad 6.13
\]

\[
\bar{\nu}_p = (2.453 \pm 0.008) + (0.122 \pm 0.007)E_n \quad E_n > 0.44 \text{ MeV} \quad 6.14
\]

The combined data have been plotted in Fig. 6.12 which also shows the linear fits, 6.13 and 6.14. It is interesting to note that the recent evaluation of \( \bar{\nu}_p \) data for U233 by Fillmore (1968) also shows a change in slope - in this evaluation at 0.85 MeV. Here, the higher energy slope was heavily weighed by the 14 MeV data.
Fig. 6.12. $\bar{V}_p$ Data for U233 Straight Lines are the Linear Fits to the Data.
6.13.2 U235 Data

The $\bar{\nu}_p$ data for U235 below 2.0 MeV have already been evaluated in section 6.9. Above 2.0 MeV, we have taken the excellent data from Soleilhac et al (1969) as the most accurate indication of the $\bar{\nu}_p (E_n)$ dependence. Soleilhac et al. (1969) have fitted their $\bar{\nu}_p$ data between 1.36 and 5.06 MeV with the linear relationship

$$\bar{\nu}_p = 2.373 + 0.1293 E_n \text{ for } 1.36 \text{ MeV} \leq E_n \leq 5.06 \text{ MeV}$$

6.15

Our evaluated fit below 2.0 MeV is given by

$$\bar{\nu}_p = 2.416 + 0.107 E_n \text{ for } E_n < 2.0 \text{ MeV}$$

6.16

The change in slope of the $\bar{\nu}_p (E_n)$ dependence occurs at 1.95 MeV.

6.13.3 Pu239 Data

Previous $\bar{\nu}_p$ data for Pu239 are listed in Table 6.12. The data from Soleilhac et al (1969) have all been decreased by 0.005. This is the correction that has been applied to the present $\bar{\nu}_p$ data for Pu239 to account for the delayed gamma ray contribution.

The agreement between the present data and previous data is satisfactory. A linear fit to all the data points below 5.0 MeV is a reasonable representation of the data and there is little evidence within the data to suggest that a change in slope might occur. However the magnitude of the change, if it exists, will be quite small and difficult to observe with the accuracy of present techniques. Furthermore, there are two reasonable arguments to
Table 6.12

Previous $\bar{\nu}_p$ Data for Pu239

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Neutron Energy (MeV)</th>
<th>$\bar{\nu}_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soleilhac et al. (1969)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.36$\pm$0.165</td>
<td></td>
<td>3.055$\pm$0.013</td>
</tr>
<tr>
<td>1.87$\pm$0.150</td>
<td></td>
<td>3.147$\pm$0.013</td>
</tr>
<tr>
<td>2.45$\pm$0.125</td>
<td></td>
<td>3.217$\pm$0.013</td>
</tr>
<tr>
<td>2.98$\pm$0.105</td>
<td></td>
<td>3.299$\pm$0.012</td>
</tr>
<tr>
<td>3.50$\pm$0.100</td>
<td></td>
<td>3.367$\pm$0.013</td>
</tr>
<tr>
<td>4.03$\pm$0.090</td>
<td></td>
<td>3.462$\pm$0.011</td>
</tr>
<tr>
<td>5.06$\pm$0.070</td>
<td></td>
<td>3.623$\pm$0.011</td>
</tr>
<tr>
<td>Mather et al. (1965)</td>
<td>0.0</td>
<td>2.931$\pm$0.034</td>
</tr>
<tr>
<td>0.99$\pm$0.19</td>
<td></td>
<td>3.103$\pm$0.053</td>
</tr>
<tr>
<td>1.99$\pm$0.14</td>
<td></td>
<td>3.170$\pm$0.040</td>
</tr>
<tr>
<td>3.00$\pm$0.11</td>
<td></td>
<td>3.243$\pm$0.049</td>
</tr>
<tr>
<td>4.02$\pm$0.10</td>
<td></td>
<td>3.325$\pm$0.050</td>
</tr>
<tr>
<td>Diven et al. (1956)</td>
<td>0.08</td>
<td>2.979$\pm$0.079</td>
</tr>
<tr>
<td>Hopkins &amp; Diven (1963)</td>
<td>0.0</td>
<td>2.839$\pm$0.028</td>
</tr>
<tr>
<td>0.25$\pm$0.05</td>
<td></td>
<td>2.940$\pm$0.039</td>
</tr>
<tr>
<td>0.42$\pm$0.11</td>
<td></td>
<td>2.966$\pm$0.046</td>
</tr>
<tr>
<td>0.61$\pm$0.07</td>
<td></td>
<td>2.912$\pm$0.041</td>
</tr>
<tr>
<td>0.90$\pm$0.08</td>
<td></td>
<td>3.013$\pm$0.041</td>
</tr>
<tr>
<td>3.90$\pm$0.29</td>
<td></td>
<td>3.432$\pm$0.039</td>
</tr>
<tr>
<td>Bondarenko et al. (1958)</td>
<td>2.1</td>
<td>3.12$\pm$0.15</td>
</tr>
<tr>
<td>Graves (1963)</td>
<td>4.0</td>
<td>3.36$\pm$0.11</td>
</tr>
<tr>
<td>Bethe et al. (1955)</td>
<td>4.25</td>
<td>3.69$\pm$0.4</td>
</tr>
<tr>
<td>Smirenkin et al. (1959)</td>
<td>4.0$\pm$0.3</td>
<td>3.42$\pm$0.09</td>
</tr>
</tbody>
</table>
suggest that such a change does occur. Firstly, a change in slope occurs for both U233 and U235. Secondly, the fitted slopes to the Pu239 data never intersect the value for the spontaneous fission of Pu240. Okolovich and Smirenkin (1963) and Holmberg and Conde (1965) have discussed this problem. If it is assumed that a change in slope does occur for Pu239, then the two linear relationships,

\[ \bar{\gamma}_p = (2.890 \pm 0.005) + (0.115 \pm 0.016)E_n \text{ for } E_n < 1.19 \text{ MeV} \]  
6.17

\[ \bar{\gamma}_p = (2.847 \pm 0.023) + (0.151 \pm 0.006)E_n \text{ for } E_n > 1.19 \text{ MeV} \]  
6.18

are a very good representation of all the Pu239 data and the value for the spontaneous fission of Pu240 (Chapter V). The change in slope occurs at 1.19 MeV. The Pu239 data and the fitted lines are shown in fig. 6.13. It should be mentioned that the two lines 6.17 and 6.18 are a slightly better representation of the Pu239 data than a single line.

The U233, U235 and Pu239 data are summarised in Table 6.13.

<table>
<thead>
<tr>
<th></th>
<th>U233</th>
<th>U235</th>
<th>Pu239</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Lower Slope MeV^{-1}</strong></td>
<td>0.035</td>
<td>0.107</td>
<td>0.115</td>
</tr>
<tr>
<td><strong>Higher Slope MeV^{-1}</strong></td>
<td>0.122</td>
<td>0.129</td>
<td>0.151</td>
</tr>
<tr>
<td><strong>Position of Change of Slope</strong></td>
<td>0.44</td>
<td>1.95</td>
<td>1.19</td>
</tr>
<tr>
<td>(corresponding E_n value)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 6-13. Comparison of Pu239 $\bar{\nu}_p$ Data.
It will be observed from Table 6.13 that the data for all three nuclei are characterised by a change of slope at apparently random energies. However, it is more reasonable to determine the energies corresponding to the slope change with respect to the fission thresholds, rather than with respect to the neutron binding energy. Fission thresholds have been taken from the data of Northrop et al. (1963). We have ignored recent discussion of the experimental accuracy of such determinations (see Strutinsky 1969, and Britt et al. 1969). The recalculated energy points for the three nuclei are listed in Table 6.14. It is observed that they are all similar in magnitude and approximately equal to the pairing energy. (Griffin et al. 1963).

Table 6.14

Position of Slope Change with Respect to Fission Thresholds

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U233</td>
<td>1.95</td>
</tr>
<tr>
<td>U235</td>
<td>2.55</td>
</tr>
<tr>
<td>Pu239</td>
<td>2.80</td>
</tr>
</tbody>
</table>

We will now consider which of the three assumptions referred to in section 6.12 is best relaxed.

6.14 Unchanged Mass Distribution

It is well known that the mass distribution does change dramatically at large compound excitation. In particular, symmetric fission becomes
more probable with higher excitation. However the general view has been that the mass distribution does not change over the first few MeV of excitation. Nevertheless, there is the data of Cowan et al. (1965) who show significant variation in the peak to valley ratios for the eV resonances of U235. Other radiochemical data from Cunninghame et al. (1961) show considerable variation in the peak to valley ratios in the region of hundreds of keV. To be useful in any explanation of the $\bar{\nu}_p (E_n)$ dependence, the mass yield data required must be far more accurate and more comprehensive than that existing at the present time. Consequently, development of any explanation along these lines is well-nigh impossible. It should be noted however that considerable difficulties would be experienced in accounting for the change in slope at the pairing gap within the framework of an explanation of this type.

6.15 Scission Neutrons

Fraser (1965) has suggested that scission neutrons might have an energy slope adequate to explain the observed $\bar{\nu}_p (E_n)$ dependence. Measurements of the angular distribution of fission neutrons with respect to fragment direction by Milton and Fraser (1965) and Bowman et al. (1963) have shown that a small component of the neutron emission is uncorrelated with fragment direction. It is believed that these neutrons originate from the fissioning nucleus either at the moment of scission or fractionally later - before the fragments have reached their terminal velocities. It is most probable that
these neutrons are emitted from the neck between the two fragments although this is difficult to substantiate experimentally. The exact magnitude of the scission component is not known with great reliability but appears to be of the order of 15 per cent for thermal neutron fission of U235 i.e. approximately 0.36 neutrons per fission. To explain the observed \( \frac{\bar{\nu}}{p} (E_n) \) dependence in this manner would therefore require a scission neutron component slope of \(-0.18 \text{ MeV}^{-1}\). Energy balance could be achieved if scission neutrons have a smaller binding energy than neutrons evaporated in the ordinary way. If \( \bar{\nu} \) is defined as the total number of neutrons of each component, \( \nu_s \) the scission component and \( \nu_F \) the fragment component, then

\[
\frac{d\nu_s}{dE_n} = \frac{d\nu}{dE_n} - \frac{d\nu_F}{dE_n}
\]

Also \( \frac{d\nu_F}{dE_n} \) can be shown to be

\[
\frac{d\nu_F}{dE_n} = \frac{d\nu_{\text{exp}}}{dE_n} - \frac{d\nu_s}{dE_n} \left( \frac{\bar{B}_s + \bar{\xi}_s}{\bar{B}_n + \bar{\xi}_n} \right)
\]

where \( \frac{d\nu_{\text{exp}}}{dE_n} \) is the expected energy dependence (0.14 MeV\(^{-1}\)), \( \bar{B}_s \) is the average binding energy of the scission neutrons and \( \bar{\xi}_s \) their average centre of mass energy. \( \bar{B}_n \) and \( \bar{\xi}_n \) are similar values for fragment neutrons. This approximate derivation gives \( \bar{B}_s + \bar{\xi}_s = 5.3 \) MeV. If the centre of mass energy of the scission neutrons is the
same as that for fragment neutrons, then \( B_s = 4.1 \text{ MeV} \) (compare \( B_n = 5.0 \text{ MeV} \)). It is not unreasonable to expect the binding energy of neutrons in the neck between the fragments to be less than that for undeformed nuclei. The nucleus at scission is in a very stretched configuration and the nuclear forces will be somewhat relaxed. Furthermore, it is conceivable that the scission component might reduce with increasing excitation. The probability of emission of scission neutrons will be in some vague way related to the length of time the nucleus spends in the elongated scission configuration. In a simple picture of fission, it might be expected that the process speeds up with increased compound excitation.

The principal difficulty with an explanation of this type lies in accounting for the significance of pairing in the \( \tilde{\nu} (E_n) \) dependence. There are other problems, such as the severe negative gradient for \(^{233}\text{U}\) and the uncomfortably large scission component predicted for spontaneous fission.

6.16 Gamma Ray Competition

It was noted in Chapter V that gamma ray competition may account for the discrepancy between calculated widths of the neutron emission probabilities and experimental values. Certainly, Thomas and Grover (1967) have shown that gamma ray competition does account for the high value of total gamma ray release in thermal neutron fission.

Their calculations may be used to estimate the rate of change of the total gamma ray yield with compound excitation. Fig. 5.5 in
Chapter V shows the calculated gamma ray emission for Sr96 and Xe140 at various excitation energies. If, for thermal fission, we assume a mean excitation of 13 MeV and a width of 5.2 MeV for Sr96, and 10.1 MeV and 4.0 MeV respectively for Xe140, then the rate of change of gamma ray emission from each of these isotopes with excitation may be calculated provided the widths of the excitation distributions do not change with mean excitation. The calculated rates of change were \( \frac{dE_{\text{gamma}}}{dE^*} = 0.09 \) and \( \frac{dE_{\text{gamma}}}{dE^*} = 0.06 \). If these two fragments are representative for the light and heavy fragments then the rate of change of total gamma ray emission is approximately 0.08 MeV per MeV of total fragment excitation.

It is interesting to speculate on whether the variation of gamma ray emission is affected above the pairing gap. Gamma ray emission competes in the first place with neutron evaporation because of the high angular momentum of the fission fragments and the unavailability in daughter nuclei (produced by neutron emission) of high angular momentum states at low excitation. The yrast levels i.e. the energy level with least energy for a particular angular momentum have been calculated by Grover (1967). The most important consideration in these calculations of the yrast levels was the number of unpaired nucleons. For a particular excitation energy, the maximum angular momentum available is related to the number of unpaired nucleons. If one assumes as Norenberg (1969) has done, that the number of unpaired nucleons at scission increases abruptly above the pairing gap, then it is possible that the gamma ray
competition is decreased and may vanish altogether. In this case a change of slope in the $\tilde{\nu}_p(E_n)$ dependence would occur at the pairing gap and the magnitude of the effect would be such as to change a slope of $0.129 \text{ MeV}^{-1}$ above the pairing energy to approximately $0.118 \text{ MeV}^{-1}$ below the pairing energy. The latter value is not too different from that actually measured for U235.

It may be concluded therefore that a major contributing factor to the change in slopes of the $\tilde{\nu}_p(E_n)$ dependence for U235 and Pu239 at the pairing energy is gamma ray competition below this energy. This particular explanation is not sufficient for U233 where the magnitude of the change in slope is far larger ($0.035 \text{ MeV}^{-1}$ below the pairing energy versus $0.122 \text{ MeV}^{-1}$ above). Clearly in this case some further influence is involved.

6.17 Weak or Strong Coupling?

We return now to the question of the strength of the coupling of the collective energy at the saddle point to the nuclear degrees of freedom at scission. It was shown in section 6.10 that interpretation of the linear $\tilde{\nu}_p$ and $E_K$ data for U235 required additional information of the relative heights of the two Strutinsky barriers. It is now proposed that between mass numbers A=234 and A=236 the higher of the two barriers changes from the first barrier to the second. Then weak coupling would considerably affect the $\tilde{\nu}_p(E_n)$ dependence for U233 without influencing the dependences for U235 and Pu239. Behaviour of this kind has been observed. In the U233 $\tilde{\nu}_p(E_n)$ dependence,
weak coupling would be expected to produce a smooth effect rather than fine structure, because in this case, in the measurable energy range below the pairing energy (0 - \(\approx 440\) keV), the excitation energy is well above threshold and there are a considerable number of collective states available at the saddle point. Lack of exact information of the spectrum of available collective states does not permit an estimate of the expected slope below the pairing energy for U233. Above the pairing energy, the availability of single particle excitation states causes all additional compound excitation to appear at scission as fragment excitation.

If the explanation proposed above is true, then an observable effect should be noticed in the average total kinetic energy data for U233. We have not obtained any \(\bar{E}_K\) data for U233 because of the lack of a suitable target. Consequently we have had to rely on previous data. This is unfortunate because we have observed some differences between our \(\bar{E}_K\) data for U235 and that from previous experiments, notably Blyumkina et al. (1964). Fortunately, Dyachenko et al. (1968) whose data are in agreement with ours for U235 have obtained data for U233. Fig. 6.14 shows the \(\bar{E}_K\) data from Dyachenko et al. (1968), Bolshov et al. (1968), Kuzminov et al. (1967) and Blyumkina et al. (1964). Also shown in Fig. 6.14 are the equivalent \(\bar{E}_K\) data determined from the linear fits to the U233 \(\bar{v}_p(E_n)\) data using a slope of 0.122 MeV\(^{-1}\) for the variation in \(\bar{v}_p\) with total fragment excitation and including a correction for gamma ray competition below the pairing energy. The \(\bar{E}_K\) data are in good
Fig 6.14 VARIATION IN TOTAL KINETIC ENERGY FOR $^{233}\text{U}$:

$$\Delta E_k = E_n - E_k (\text{thermal})$$

Straight line has been calculated from $\bar{V}_p$ data fits.
agreement with the data calculated from the $\bar{\nu}_{p_n}(E)$ dependence and
tend to confirm the proposal of weak coupling.

It is possible that weak coupling has a minimal effect on the
$\bar{\nu}_{p_n}(E)$ dependences for U235 and Pu239 below the pairing energies.
No comment can be expressed on this possibility as the only
confirmatory data, the $\tilde{E}_K$ data for U235, have errors in excess of the
likely effect.

The influence of the relative heights of the Strutinsky barriers
has been observed in other fission phenomena - notably the magnitude
of the anisotropy in the fission fragment angular distributions.
Bjornholm and Strutinsky (1969) have noticed this effect and
fig. 6.15 is reproduced from that paper. It will be observed that
the magnitude of the anisotropy of the fragment angular distributions
changes quite markedly from the compound nucleus U235 to U239.
Figure 6.15 Anisotropies of the Fission Fragment Angular Distributions. (Compound nuclei shown) The magnitude of the anisotropy changes dramatically between $A = 235$ and $A = 239$. 
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7. **NEUTRON EMISSION FROM INDIVIDUAL FISSION FRAGMENTS**

7.1 **Introduction**

Previous chapters have described measurements of the systematics of fragment neutron emission averaged over the entire mass and charge distributions. The natural extension of this work is to obtain data such as the average neutron emission (and the distribution about that mean) from fragments of specific mass and charge for particular values of the total kinetic energy.

The data sought are of the form \((M_1, Z_1, \bar{\nu}_1, \sigma\nu_1, E_K)\). This data can be obtained experimentally by simultaneous measurement of the kinetic energies of both fragments, the charge of one fragment via its K X-ray energy and neutron emission data using a large liquid scintillator tank. Of course, high X-ray energy resolution is required for precise determination of the nuclear charge. Although some progress has been made with this experiment, reasonable statistical accuracy for all parameters has not yet been achieved.

The experiment was designed to investigate thermal neutron fission of U235 using a reactor neutron beam. The high background in the neutron detector has limited the value of the neutron distribution data, and unfortunately it has been necessary to terminate the experiment temporarily during modifications to the reactor system. During the reactor shutdown, measurements will be made for the spontaneous fission of Cf252. In the meantime, however, data have been obtained for the mean neutron emission versus fragment mass for specific values of the
total kinetic energy i.e. \((M_1, \tilde{v}_1, E_R)\) for the thermal fission of U235.

Measurements of the variation of neutron emission with fragment mass have been performed by Milton and Fraser (1965), Apalin et al. (1965) and Maslin et al. (1967). The experimental methods used were all based on the fact that at least 85 per cent of the neutrons are emitted from the fragments after they have reached their terminal velocities. The angular distribution in the laboratory system of neutron emission from a particular fragment is therefore strongly peaked in the fragment direction. Consequently, a neutron detector geometrically located in the fragment direction will detect preferentially neutron emission from that fragment. The neutron data obtained have been important in understanding the energy balance at scission. The variation of \(v\) with mass has revealed that this is more a factor of the properties of the fragments than of the mass ratios. Nevertheless the variation has been further evidence of the role shell effects play in determining the scission configuration. The data have been used - see for example Terrell (1965) - to obtain deformation parameters of the neutron rich fission fragment species of nuclei. Some discrepancies do exist between different experiments and the clarification of this situation has been one of the motivations for this research program. However the ultimate aim (not achieved in this experiment) is to determine, from a comparison of the neutron distributions of the light and heavy fragments, the correlation of the light and heavy fragment excitation energies.
7.2 Experimental System

A schematic representation of the experimental system is shown in Fig. 7.1. A highly collimated beam of thermal neutrons was obtained from the 10 M Watt reactor HIFAR. Special care was taken in the design of the collimator system as it was intended to perform the experiment without shielding of the X-ray and fragment detectors, and the divergence of the neutron and gamma ray beams was therefore the limiting factor in determining the geometry of the detector system. The neutrons originate in the graphite reflector of the reactor and spectrum measurements have shown the fast neutron and epithermal components to be very small. $[\phi_F$ (corrected for collimator geometry) of $3 \times 10^4$ n cm$^{-2}$ sec$^{-1}$, Boldeman et al. (1962)]. The thermal flux at the experiment was $1.5 \times 10^7$ n cm$^{-2}$ sec$^{-1}$ and the beam diameter there was 1.5 cms.

The two fission fragment detectors were typical surface barrier diodes made of n type silicon and operated at 90 V reverse bias. Both detectors were collimated to active areas of 3 cm$^2$. Detector 1 was located approximately 2.5 cms from the U235 target and consequently subtended an angle of $\pm 22^0$. Detector 2 was mounted on a linear motion feedthrough and its position in the vacuum system could be accurately varied externally. This detector was positioned 6.5 cms from the U235 target and defined the maximum divergence from the axis of the detector system of the selected fission fragments ($\pm 8^0$). The particular geometrical arrangement used prevents discrimination.
Fig. 7.1 Schematic representation of Experimental System.
against fragments emitting high numbers of neutrons. In principle it would have been preferable to have detector 1 define the fragment geometry, but the spatial requirements of such an arrangement reduce the geometrical efficiency of the scintillator tank.

The K X-ray detector was a lithium drifted silicon diode with an active area of \( \approx 0.5 \text{ cm}^2 \) and depletion depth of 3 mm. The detector was cooled to \(-190^\circ\text{C}\) and the associated FET on the detector preamplifier to \(-160^\circ\text{C}\). The liquid nitrogen reservoir was of the feed through type and some difficulties were initially experienced with acoustic pick up in the silicon detector. The active area of the X-ray detector was covered with 0.0015 inch thick Aluminium to prevent fission fragment radiation damage. The X-ray resolution was reasonably good for a detector of this active area and was found to be 780 eV for the 14 keV gamma ray from Co57 decay.

The U235 targets were prepared by electrospraying the U235 material onto gold plated VYNS films. The U235 target, gold layer and VYNS film thicknesses were respectively 30 \( \mu \text{gms cm}^{-2} \), 15 \( \mu \text{gms cm}^{-2} \) and 20 \( \mu \text{gms cm}^{-2} \). Initially the support ring for the VYNS film was approximately 8 cms diameter in order to minimise neutron and gamma ray scattering into the various detectors. However, it became necessary to reduce this diameter to 3 cms as the larger foils had a maximum lifetime of 2 days in the vacuum environment of the detector system. The U235 target was placed at 45\(^0\) to the axis of the fragment detectors and the neutron beam.
The U235 target contained approximately 0.2 μgms of Am241. A third surface barrier detector was operated in coincidence with the X-ray detector to record coincident α and X-ray emission from Am241 decay. This method was used for stabilisation of the X-ray detector system.

The entire detector system was operated under reasonably high vacuum of approximately $2 \times 10^{-8}$ mm Hg. The vacuum system had entry and exit windows of approximately 0.005 inch Aluminium for the neutron beam.

The neutron detector was a large liquid scintillator tank containing approximately 60 litres of NE 323. The tank was specially constructed for this particular experiment and its size was a compromise between neutron efficiency and background count rate in the reactor environment. Two 9618A photo multiplier tubes were mounted on the outside of the tank. The mean time for neutron detection after fission was approximately 8 μsecs. The $4\pi$ geometry neutron detection efficiency of the scintillator under optimum conditions was approximately 65 per cent. For use in the present experiment there was no axial tube through the tank.

7.3 Electronics

A block diagram of the electronics is shown in Fig. 7.2. In principle fast logic timing data and slow pulse height data were taken from each detector (apart from the scintillator tank). For the two fragment detectors and the α detector, fast timing was achieved with inductive pick offs. A timing signal was obtained from the X-ray detector using a fast current amplifier, Rush (1963).
FIG 7.2  BLOCK DIAGRAM OF ELECTRONICS.
The coincidence timing resolutions used were

\[ \tau = 12 \text{nsecs for the fragment coincidence} \]

\[ \tau = 100 \text{nsecs for the X-ray - double fragment coincidence} \]

\[ \tau = 50 \text{nsecs for the X-ray - } \alpha \text{ coincidence.} \]

The coincidence outputs were used to gate the 256 channel analogue to
digital converters on the pulse height lines. A multiple event
counter similar to that used for the \( \bar{\nu}_p \) measurements was also gated
300 nsecs after each coincidence of any type. The neutron and
background counting gates of the multiple event counter were 15 \( \mu \text{secs} \)
and the delay between the two gates was 60 \( \mu \text{secs} \). The multiple event
counter could store up to 15 counts in the foreground channel and
seven counts in the background channel. The gate length time of
15 \( \mu \text{secs} \) was chosen to minimise background with respect to the
fission neutron count rate. Under optimum conditions a gate time of
30 \( \mu \text{secs} \) would have been ideal. Because of the severe background
problem it was necessary in addition to operate the scintillator tank
at reduced efficiency. The approximate 4\% operating efficiency for
the entire experiment was 25 per cent. At this efficiency the data
on the distribution of neutron emission becomes very poor and the
analysis of the data was not extended to obtain this information.

All data were recorded event by event on magnetic tape using
an incremental tape recorder. Any particular record consisted of
five bytes of information. The first was a number (numbers 241-248
were reserved for this purpose) specifying the type of coincidence
and therefore the nature of the data recorded. The event encoder
generated this experiment number. (Triple coincidence data had
priority over all other types of data). The two successive bytes were the digitised outputs from the fragment detectors, the fourth byte contained the X-ray detector pulse height and the last byte contained the foreground and background data from the scintillator tank. It was more convenient to gate all data lines with each coincidence rather than to record only the appropriate data for a particular coincidence.

7.4 Analysis of Data

The triple coincidence rate \( (E_1, E_2, K \text{ X-ray}) \) was less than 1 in 50 secs and the accumulated data (continuous operation for 1 month) before the experiment was terminated by modifications to the reactor was of insufficient accuracy to be meaningful. A total of \( 2 \times 10^7 \) fission fragment coincidences have been recorded. However, the subsequent data are based on the analysis of \( 5 \times 10^6 \) fission events. The preliminary stages of the analysis of the data consisted in visual inspection of the fragment pulse height spectra to determine electronic drifts and inspection of the scintillator background data to determine constancy of its efficiency. For this purpose raw data were printed out in groups of 100,000 fission events. In practice it was found that pulse height drifts (determined by fragment kinetic energy peak positions) were less than 0.1 per cent per \( 5 \times 10^5 \) fission events and that variations in the scintillator background rate were less than 2 per cent per \( 5 \times 10^5 \) fission events. As a consequence, the data were analysed in groups of \( 5 \times 10^5 \) fission events. This turned out to be a convenient size in terms of computing time.
The raw kinetic energy data were used to obtain the pre-neutron emission masses and kinetic energy using the procedures of Schmitt et al. (1965) and Terrell (1962). The method was as follows. Firstly, a linear calibration of the pulse height scales for each detector was made using the fragment spectra peak positions and experimental data from Milton and Fraser (1962). The approximate pre-neutron emission masses were then obtained from the kinetic energy data using the relationships

\[
M_1 = \frac{236 E_2}{E_1 + E_2} \quad 7.1
\]

\[
M_2 = 236 - M_1 \quad 7.2
\]

Post neutron emission masses were obtained from \( M_1 \) and \( M_2 \) using the \((v, E_K)\) data from Maslin (1967). The detector energy scales were recalibrated using the procedure of Schmitt et al. (1965). They have shown the energy scale of surface barrier detectors for fission fragment detection is given by

\[
E = (a + a'M) x + b + b'M \quad 7.3
\]

where \( E \) is the fragment kinetic energy,

\( M \) is the fragment mass,

\( x \) is the pulse height,

and \( a, a', b, b' \) constants tabulated in Table 7.1.
Table 7.1

Calibration Constants

<table>
<thead>
<tr>
<th>a = $\frac{30.9734}{P_L - P_H}$</th>
<th>$a' = \frac{0.04596}{P_L - P_H}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$b = 87.8626 - aP_L$</td>
<td>$b' = 0.1345 - a'P_L$</td>
</tr>
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The values $P_L$ and $P_H$ are the observed pulse heights corresponding to the mid points between the 3/4 maximum points in the light and heavy mass groups. This calibration procedure has been discussed in further detail by Schmitt et al. (1966). The recalculated post-neutron emission kinetic energies were converted into pre-neutron emission energies using equation 7.4.

$$E_{\text{Pre}} = \frac{E_{\text{Post}}}{1 - \frac{\nu}{M}}$$  \hspace{1cm} 7.4

where the $\nu$ data as a function of mass and total kinetic energy were obtained as before from Maslin et al. (1967). The entire process was repeated until the pre-neutron emission masses before and after a particular iteration were the same to within 0.1 a.m.u. The output data for each particular fission event consisted of pre-neutron emission masses and total kinetic energy, plus neutron and background data.

To correct the neutron data for scintillator geometry and backscatter from the complementary fragment, the data were sorted into two matrices giving the number of events and the measured mean number of neutrons
detected for each value of the mass and total kinetic energy. The mean number of neutrons is obtained from the difference of the mean count in the foreground and background channels of the multiple event counter. The mass groups were 2 a.m.u. wide and the total kinetic energy groups 5 MeV wide. The neutron data were not corrected for dead time losses as this correction was less than 1 per cent. In determining the detector geometry and backscatter correction we have assumed

1. The excitation energies of the two fragments are correlated.

2. The detection efficiency of the scintillator is constant with neutron energy.

In principle, the correction procedure was as follows. Firstly, an approximate correction for detector geometry and backscatter was made assuming all neutrons are emitted from the moving fragments. The data obtained were normalised to $\bar{v}_p$ (thermal) $U_{235} = 2.415$ (Chapter IV). Thus an approximate value of the average scintillator efficiency and the variation of $\nu_T$ (total neutron emission from both fragments) with fragment mass was obtained. Assuming now that 15 per cent of the neutrons are emitted isotropically in the laboratory system (Milton and Fraser, 1965) the experimentally observed probabilities were adjusted to remove the scission neutron component contributions. The remaining
contributions (i.e. from neutrons correlated with the fragment
direction) were corrected for detector geometry and backscatter
and the variation of $\varepsilon \nu_F$ with fragment mass was obtained. Here $\varepsilon$
is the neutron detection efficiency of the scintillator and $\nu_F$
the mean number of neutrons emitted from the moving fragments. It
was assumed that, for a particular mass division, the scission
neutrons were emitted from each fragment in the same proportions
as those from the moving fragments. Thus the total neutron
emission from a particular fragment $v$, is given by

$$\varepsilon \nu = \frac{\varepsilon \nu_F}{0.85}$$  \hspace{1cm} 7.5

The data were normalised as before to $\nu_p$ (Thermal) $U235 = 2.415$. It
was unnecessary to repeat the process with the more accurate $\nu_T$
and scintillator efficiency data as subsequent corrections change the
final data by less than 1 per cent. The details of the detector
geometry and backscatter corrections are as follows. If $P_i(M_i,E_K)$
are the observed experimental probabilities of neutron detection, then

$$P_1(M_1,E_K) = \nu_1 \varepsilon P_{11}(M_1,E_K) + \nu_2 \varepsilon P_{22}(M_2,E_K)$$  \hspace{1cm} 7.6

and

$$P_2(M_2,E_K) = \nu_2 \varepsilon P_{21}(M_2,E_K) + \nu_1 \varepsilon P_{12}(M_1,E_K)$$  \hspace{1cm} 7.7

where $M_1$ and $M_2$ are the complementary masses,
$\nu_1$ and $\nu_2$ are the neutron emission probabilities from
the complementary fragments,
\[ \varepsilon \] is the liquid scintillator efficiency,

\[ P_{i1} \] is the probability of forward neutron emission
into the scintillator geometry of neutrons emitted
from fragment \( i \),

and \[ P_{i2} \] is the probability of backward emission into the
scintillator geometry of neutrons from fragment \( i \).

The \( P_{i1} \) and \( P_{i2} \) probabilities were calculated as follows. It was
assumed that the correlated neutrons were emitted isotropically in
the centre of mass of the fragment and that the centre of mass neutron
spectra were accurately represented by the usual evaporation spectra
of temperature \( T \) i.e.

\[ \phi(E) \propto \frac{E}{T^2} \exp\left(-\frac{E}{T}\right) \] 7.8

It was assumed that the temperature distribution for each fragment
could be adequately represented by the experimentally determined
mean centre of mass energy (i.e. \( T = \frac{2}{3} \bar{E} \)). The evaporation temperature
data were taken from Kluge and Lajtai (1968). The laboratory
probability distribution with respect to the fragment direction
becomes

\[ \rho_i(v_i, \theta) = \frac{a^2 v_i}{T_i^2} v_i^2 \sin \theta \exp\left(-\frac{av_i^2}{T_i}\right) \] 7.9
where $V_i$ is the laboratory velocity of the neutrons

$n_i$ is the neutron centre of mass velocity

$a = 0.5228$

$\theta$ is the neutron emission angle with respect to the fragment direction.

The laboratory velocity $V_i$ is given by

$$V_i^2 = v_i^2 - W_i^2 + 2v_i W_i \cos \theta$$  \hspace{1cm} 7.10

where $W_i$ is the laboratory velocity of fragment $i$.

The probabilities $P_{i1}$ and $P_{i2}$ are given by

$$P_{i1} = \int_0^\infty \int_0^\infty \rho_i(v_i, \theta) \, dv_i \, d\theta$$  \hspace{1cm} 7.11

$$P_{i2} = \int_0^\infty \int_{155.5^\circ}^{155.5^\circ} \rho_i(v_i, \theta) \, dv_i \, d\theta$$  \hspace{1cm} 7.12

where the scintillator subtends $\pm 24.5^\circ$. These expressions were integrated numerically and equations 7.6 and 7.7 were solved to obtain $\epsilon v_i(M_i, E_K)$. For each particular fragment mass group, the neutron emission data were averaged over the kinetic energy distribution to obtain $\epsilon v_i(M_i)$. The effect of the selected fragment distribution with respect to the axis of fragment detectors ($\pm 8^\circ$) on the geometry factors $P_{i1}$ and $P_{i2}$ was investigated and found to be relatively insignificant. ($<1$ per cent for $A = 80$ where geometrical corrections have their greatest effect).
7.5 Results

7.5.1 Mass distribution

The pre-neutron emission mass distribution, calculated from the raw kinetic energy data as described in section 7.4, is shown in Fig. 7.3. The yield curve shown is based on $5 \times 10^6$ fission events. The statistical accuracy of each data point is very high and the accuracy is limited only by the experimental method. The input $(v, E_K)$ data used in the correction procedure were taken from Maslin et al. (1967). In view of the similarity of the present $(v, E_K)$ data to that of Maslin et al., it was unnecessary to recalculate the mass distribution using our $(v, E_K)$ data set.

The measured mass distribution is in good agreement with those obtained using more accurate methods, e.g. radiochemical studies (Wahl 1965) and double velocity measurements using time of flight techniques (Milton and Fraser 1962). The fine structure observed in the mass distribution by Milton and Fraser (1962) is not strongly reproduced in the present data although both the light and heavy fragment distributions do have shoulders. The magnitude of the fine structure actually observed is acceptable in view of the poorer mass resolution in double energy studies and the need for reasonable count rate.

The ratio of the asymmetric peak yield to the symmetric yield was approximately 110:1, which was somewhat disappointing. This value compares unfavourably with those from radiochemical studies of 650:1. In the symmetric region therefore, only one in six events is genuine. Similar difficulties were experienced by Maslin et al. (1967) who
Fig. 7-3. Pre-Neutron Emission Mass Distributions (Based on $5 \times 10^6$ Fission Events).
obtained a ratio of 115:1. In our case, the raw fragment kinetic energy spectra are reasonably good and a better yield curve should be expected. The origin of the problem has not yet been traced.

7.5.2 Kinetic Energy Data

The distribution of the pre-neutron emission total kinetic energy is shown in Fig. 7.4. The mean total kinetic energy was found to be 170.8 MeV, which is in good agreement with the value of 171.9±1.4 MeV from Schmitt et al. (1965) on whose work the correction procedure is based. The mean total kinetic energy as a function of the heavy fragment mass is plotted in Fig. 7.5 together with similar data from Schmitt et al. (1966) and Maslin et al. (1967). Apart from the symmetric region the present data are in good agreement with previous measurements. The dip in the kinetic energy curve for symmetric fission is 30 MeV which is slightly larger than recent estimates. Apalin et al. (1965) obtained a value of 21 MeV. Alexander et al. (1963) from measurements of fission fragment ranges placed the dip between 18 and 27 MeV. As in the data from Maslin et al. (1967) who measured a symmetric dip of 33 MeV, the present measured decrease has been increased due to the anomalously large symmetric mass yield produced by degraded fission fragments.

7.5.3 Neutron Data

The measured variation of neutron emission with pre-neutron emission fragment mass, corrected as in section 7.4, is shown in Fig. 7.6. The data shown are based on the analysis of 5 x 10^6 fission events. The errors shown in Fig. 7.6 are purely statistical and are
Fig. 7.4 The Distribution of the Average Total Kinetic Energy.
Fig.7-5. Mean Total Fragment Kinetic Energy versus Heavy Fragment Mass.
Fig. 7.6. Neutron Emission versus Pre Neutron Emission Fragment Mass.
typically about 3 per cent at masses corresponding to the peaks in the mass distribution. As the statistical accuracy is poor and only one in six events is genuine, neutron data have not been plotted in the symmetric region. The curve in Fig. 7.6 shows the usual trends that have been observed in previous measurements. In particular, the neutron yield near the spherical closed shell nuclei \((N=50, Z=50)\) is very small and the yield from the easily deformed complementary fragments correspondingly high. The small peak in the neutron yield at \(A=97\) is probably genuine. Milton and Fraser (1965) have also observed a peak corresponding to \(A=96\). In addition they have observed smaller peaks at \(A=90\) and \(A=101\) which have not been reproduced in the present experiment, presumably because of the poorer mass resolution. The neutron yield from the light fragments to that from the heavy fragments \(\frac{\nu_L}{\nu_H}\) was found to be 1.18. The statistical accuracy of this value is very high, however the exact magnitude is quite sensitive to the assumptions made in the correction procedure (section 7.4).

The present data have been compared with that from Maslin et al. (1967) in Fig. 7.7. It will be observed that the agreement between the two sets of data is particularly good. This of course should be expected as the experimental methods were similar. The only minor differences that occur are the small peak at \(A=97\) and the larger decrease at \(A=129\) in the present set. These small differences may be due to the slightly better mass resolution in the present experiment.

The present data have been plotted with previous data from Maslin et al. (1967), Milton and Fraser (1965) and Apalin et al. (1965) in
Fig. 7.7. Comparison of present neutron emission data with Maslin et al (1967).
Fig. 7.8. As in Fig. 7.7, data for symmetric fission have not been plotted although the latter two experiments do have reasonably accurate data in this region. Fig. 7.8 is a comparison of experiments employing direct neutron counting methods. Data obtained from a comparison of initial and final mass yields (see e.g. Terrell, 1962) have not been included. Although the general features of all the data are similar, there are large discrepancies in the magnitude of the neutron emission. For the heavy fragments, the agreement is reasonably good although the large yield observed above $A>145$ by Apalin et al. (1965) has not been reproduced in any other data set. For the light fragments the agreement is very poor. Milton and Fraser (1965) and Apalin et al. (1965) both find the light fragment peak neutron emission to be significantly higher than either the present experiment or Maslin et al (1967). For neutron emission at masses corresponding to the peak in the light fragment mass yield, Milton and Fraser (1965) are significantly higher than the other three sets. Milton and Fraser (1965) have pointed out that backscatter corrections have not been made to their data and this correction has a significant effect on the neutron emission from the light fragment. For the very light fragments, $A<90$, Apalin et al. (1965) measure a significantly smaller yield than the other three experiments.

The discrepancies between the various experiments may in part be due to the effects of different mass resolution. However, they are more probably related to the various geometrical corrections required in each particular experiment. The geometrical correction factors
Fig. 7-8. Comparison of present data with previous direct neutron counting data.
derived in section 7.4 will be re-examined for the present experiment.

7.5.4 Neutron Emission versus Total Fragment Kinetic Energy

The measured variation of neutron emission with total fragment kinetic energy for various fragment masses is very similar to the previous data from Maslin et al. (1967). Fig. 7.9 shows a sample of the present data for two reasonably wide mass groups. The complete data set is not shown in view of the similarity with the previous data.
Fig. 7.9. Average Neutron Emission per Fragment versus Total Fragment Kinetic Energy for Two Mass Groups.
7.6 References


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CHAPTER VIII
FINAL COMMENTS

Following initial studies made to determine values of $\bar{v}_p$ for thermal fission of U233, U235, Pu239 and Pu241 to high precision, it became clear that high precision would only be achieved by a detailed study of all effects which, though small, could materially affect the results. For thermal and spontaneous fission, an accuracy has now been achieved which is sufficiently high to satisfy the requirements of reactor design teams provided the absolute value of the standard, $\bar{v}_p$ for the spontaneous fission of Cf252, is known without ambiguity. In view of the discrepancies which exist between various absolute calibrations of this standard, a re-examination of $\bar{v}_p$ for the spontaneous fission of Cf252 is essential at the present time.

The next step in this study was to measure $\bar{v}_p(E_n)$ with the precision developed for the thermal measurements to resolve discrepancies between results of various investigators, some of whom claim fine structure in the $\bar{v}_p$ variation with compound excitation. Since the discrepancies were greatest for the neutron fission of U235, the first measurements were the determination of the $\bar{v}_p(E_n)$ dependence for U235. These measurements showed no evidence of any fine structure within the experimental error. The $\bar{v}_p(E_n)$ data obtained for U235 was not, for the most part, statistically inconsistent with the results of the majority of previous investigations.
To confirm the lack of structure in $\bar{\nu}_p(E_n)$ for U235, measurements were made of the variation of the average total kinetic energy of the fission fragments $\langle \bar{E}_K \rangle$ with compound excitation. Because $\bar{E}_K(E_n)$ is correlated with $\bar{\nu}_p(E_n)$, it should reflect the corresponding structure in that parameter. No structure has been found in $\bar{E}_K(E_n)$ within experimental error, and this is consistent with the observed lack of structure in $\bar{\nu}_p(E_n)$.

It became apparent that an explanation of the nature of the $\bar{\nu}_p(E_n)$ dependence for U235 within the framework of existing fission theories required additional $\bar{\nu}_p$ data — in particular the $\bar{\nu}_p(E_n)$ variation for both U233 and Pu239. These dependences were obtained. From an evaluation of the present and existing data for U233, U235 and Pu239, it was observed that the $\bar{\nu}_p(E_n)$ dependence, though linear, is characterised by a change in slope at the pairing energy. For U235 and Pu239, the magnitude of the change in slope was quite small and considerably less than that for U233. An explanation for these facts has been presented in terms of the double-humped fission barrier with the adiabatic assumption of weak coupling of the collective saddle point energy to the nuclear degrees of freedom at scission. The change in character of the $\bar{\nu}_p(E_n)$ dependence between U233 on the one hand and U235 and Pu239 on the other is associated with the change in the relative heights of the two humps of the fission barrier. To improve the likelihood of the proposed explanation, further data are required. In particular, the $\bar{E}_K(E_n)$ dependence for U233 should be examined and an attempt made to measure $\bar{\nu}_p(E_n)$ below
the neutron binding energy.

Some effects have been observed in the systematics of neutron emission which may be attributed to the competition of gamma ray emission as a fragment de-excitation mode. This matter has not been studied in great detail in the literature and further investigation in the light of the comments made in this thesis is warranted.

Subsequent development of the research program has been concerned with measurements of neutron emission from fission fragments of specific mass and charge. There remains much which is unknown about the final stages of the fission process. A preliminary study of neutron emission versus fragment mass has shown that an adequate experimental method has been developed to contribute to the improvement of knowledge in a variety of aspects of fission physics. It is clear that the data which may be derived from these more complex experiments will be essential for the detailed explanation of the data presented earlier in this thesis.