International Atomic Energy Agency

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# INTERNATIONAL NUCLEAR DATA COMMITTEE

PROCEEDINGS OF THE TOPICAL CONFERENCE ON "NEUTRON INDUCED FISSION" HELD DURING THE FOURTH INDC MEETING

Bhabha Atomic Research Centre, Bombay Wednesday, July 14, 1971

Organised by International Nuclear Data Committee, International Atomic Energy Agency, Vienna

IAEA NUCLEAR DATA SECTION, KARNTNER RING 11, A-1010 VIENNA

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#### FOREWORD

The Fourth International Nuclear Data Committee meeting was held during July 12-16, 1971 at the Bhabha Atomic Research Centre, Trombay, Bombay. When the programme for this meeting was being planned it was felt that it would be worthwhile for every one concerned if a topical conference on a relevant topic could be arranged as a part of this meeting. It was decided after some correspondence that the topical conference should be on 'Neutron Induced Fission'. By arranging this conference it was intended to establish a strong interaction between the visiting scientists from various countries and the scientists at the Bhabha Atomic Research Centre and other laboratories in India by informing the visiting scientists about the work going on in India and by stimulating local scientists with information and ideas concerning the work done at other laboratories of the world. As the papers at the conference and the discussions indicate, the purpose of the topical conference was well served.

The Indian Nuclear Data Group would like to convey its thanks to all the speakers who participated at this conference and to all others who contributed to making the conference a success.

Aspinatia

A.S. Divatia Convenor Indian Nuclear Data Group

, SESSION 1

Chairman : W.G. Cross, AECL, Canada

### EXCITATION ENERGY DEPENDENCE OF SHELL EFFECTS IN NUCLEAR FISSION<sup>+</sup>

S.S. Kapoor Bhabha Atomic Research Centre Trombay, Bombay-85

In this talk, I shall like to present some of those aspects of nuclear fission studies which are related the nuclear shell effects and their dependence on excitation energies. The last few years have been some very interesting observations both in the theory and experiment. which have focussed our attention to several new features which appear in the fission process due to the presence of nuclear shell effects for both spherical and deformed nuclear configurations. For example, the theoretical studies 1, 2 of the stability of very heavy nuclei after incorporating the effects of shells have made some exciting predictions about the possibility of existence of an island of super-heavy nuclei and have also shown that the fission barriers, are, in general of a double humped type which is in fact responsible<sup>3</sup> for a number of interesting experimentally observed phenomena such as observations of fission isomers in a number of nuclei ranging from uranium to berkelium and gross structures in the subthreshold neutron fission resonances for  $Np^{237}$  and  $Pu^{240}$ . These developments have demonstrated that studies of the fundamental aspects of the fission process are important not only for obtaining information on nuclear structure of highly elongated nuclear shapes, but also for a proper interpretation and evaluation of nuclear data needed for reactor design. Most of these phenomena relate to the single particle effects in the ground and very slightly excited states of nuclei. However, an important question arises as to how the ground state nuclear shell corrections influence the observed phenomena in fission in the case of a "hot" nucleus having an excitation energy much above the fission barrier. We have carried out some investigations in this regard which I would like to present in this talk.

Before going into details of this, it will be appropriate to give a short introduction into the subject. On purely energy consideration, any nucleus with A > 120, if split into two parts can lead to a release of energy. Yet all of these nuclei do not disintegrate sponta-What keeps a nucleus as a whole and stable is the presence neously. of a fission barrier. In terms of the liquid drop model (LDM) this barrier arises as a result of a delicate balance of energy between the Coulomb disruptive forces and the nuclear attractive forces. When a nucleus is deformed, upto a certain deformation the increase in the nuclear surface energy is slightly more than the decrease in the Coulomb energy resulting in an increase in the total energy. But after this deformation is reached the total energy decreases with further deformation. This gives rise to a fission barrier with a corresponding saddle point in the deformation space The height of the barrier and

and the nuclear shape at barrier depends on the fissionability parameter  $X = E_{co}/2 E_{so}$  where  $E_{co}$  and  $E_{so}$  are the Coulomb and surface energies of the undeformed nucleus. Detailed LDM calculations have been carried out by Swaiatecki and his collaborators<sup>4</sup> to calculate  $E_B$  and shape of the saddle point versus parameter X.

The other important development<sup>5</sup> in fission came with the realisation that a deformed nuclear configuration passing over the fission barrier (or saddle point) can exhibit quasi-stationary states which are characterised by the total angular momentum I, its projection K along the symmetry axis, and the projection M of I on the beam direction and consequently the fragment angular distributions in fission induced by energetic projectiles depend on the quantum numbers of the fission channels open at the saddle point. At medium excitation energies where several K channels are open, a statistical approach<sup>6</sup> is taken and it has been shown that from the fragment angular distribution data it is possible to derive the value of  $K^2$ , where  $K^2_0$  is the mean square projections of the angular momentum on the symmetry axis and is related to the effective Moment of Inertia Jeff and nuclear temperature T by the relation  $K_0^2 = J_{eff}^T / \hbar^2$ . It has therefore been possible to infer<sup>7</sup> the effective moment of inertia for the saddle point configuration and thereby the shape of the transition state nucleus from the measurements of fragment angular distributions at medium excitation energies. We shall later see how the barrier shapes derived from angular distribution data compare with those calculated on the liquid drop model. It may be remarked here that in the framework of statistical approach. the transition state, by defination, corresponds to the deformation where the nuclear entropy is minimum.

I now come to a more recent development concerning shell effects in fission. The gross systematics of the ground state energies are very well described by LDM mass formula. However small but systematic deviations are observed from this average behaviour. The differences between the experimental masses and the liquid drop model masses attributed to the ground state shell corrections are found to be negative (-5 to - 10 MeV) at the closing of the shells and this accounts for the extra binding of the magic nuclei. Since the measured barriers in actinide region are only about 4-6 MeV, the shell corrections of the same order become important and it is to be expected that the measured barriers do not fit the barriers calculated on the LDM. These shell corrections to LDM energies are now theoretically understood on a combined microscopic-macroscopic approach first suggested by Swiatecki<sup>1</sup> and now further refined by Strutinsky<sup>2</sup>. In terms of the single particle picture, LDM can be considered as the one referring to a smoothly varying continuous distribution of nucleons in the various energy levels while the actual distribution has discreteness and discon-The difference between the total energies computed from the tinuities. two schemes is the shell correction which is correlated with the density of single particle states near the Fermi surface. Lower than average level density leads to negative shell correction and vice versa.

It is now known<sup>2</sup> that as a nucleus deforms the single particle density at the Fermi surface oscillates, about an average value, which results in similar oscillations in the shell correction energy  $\Delta$  as a function of deformation. In order to arrive at the total energy of the nucleus, the shell correction energies are added to the LDM energies in this two part approach. This synthesis of a smooth LDM energy surface with the oscillating shell correction-energy, then leads to a double-humped fission barrier for a number of nuclei in the actinide region, as shown in Fig. 1. The occurance of a secondary minimum in the potential energy of deformation has provided a natural explanation for a large number of experimentally observed phenomena such as observations of fission isomers and sub-barrier fission resonances in a number of nuclei in the actinide region,

The existence of the two fission barriers instead of only one, however, poses a new question, That is, in the statistical limit of high excitation energies, what is the shape of the transition state nucelus where the angular distributions are decided. Is it the first barrier, or second barrier or the LDM barrier shape? In order to get an answer to this question, one needs to find out the configuration of minimum nuclear entropy. If one uses the Fermi gas expression  $S = 2(aE_x)^{\frac{1}{2}}$  this point of minimum entropy coincides with the point of minimum excitation energy E, and the transition states would coincide with the nuclear shapes at the top of these two barriers. However, this expression is not valid ' for a nucleus having shell effects and should For a given single particle level sequence, the numeribe modified. cal calculations of nuclear entropy have been carried out by Ramamurthy, Kapoor and Kataria<sup>8</sup> and the results are shown in Fig. 2, where  $S^2$  versus E is plotted for the two cases of  $Pb^{208}$  and  $Pu^{242}$  (spherical The shell corrections to LDM energies for these two cases shape). are found to be - 9.2 MeV and + 14.5 MeV respectively, as obtained by the Struntinsky procedure. If the Fermi gas relation was applicable, a plot of  $S^2$  versus  $E_x$  should be a straight line. Clearly there are deviations observed from a stright line. However at high excitation energies, a relation of the form  $S^2 = 4a(E_x \pm \Delta E_x)$  is apparent. It was actually found that  $\Delta E_x$  is equal to the ground state shell correction. This, in other words, means that in the asymptotic limit of high auditation high excitation energies (> 30 MeV) a usual Fermi gas relation of the type S<sup>2</sup> = 4a E<sub>x</sub> can be used, provided the excitation energy is measured from a reference surface which coincides with the LDM energy This can be interpreted to mean that the shell effects are surface. not manifesting themselves in nuclear entropy for excitation energies exceeding 30-40 MeV. This work further suggests that the ground state shell corrections in different nuclei can be obtained in a direct way through a calculation of  $S^2$  versues  $E_x$  and this method of obtaining shell correction may have a more general applicability than the Strutinsky prescription.

Applying these ideas to a nucleus having a double humped barrier, it follows that although for low values of  $E_x$ , the points of minimum entropy would correspond to the shapes at the top of the barriers I and II, at medium excitation energies ( $E_x$  30-50 MeV) the point of minimum entropy would correspond to LDM barrier shapes.

In Fig. 3, we show that in the fragment anisotropy data, the evidence for this new effect does exist. The values of  $J_0/J_{eff}$  shown in the figure are evaluated from the fragment anisotropy data? in which the fissioning nuclei have excitation energies  $\gtrsim 30$  MeV for all cases except for nuclei lighter than Thorium. Also shown in the figure are the calculated  $J_0/J_{eff}$  for transition state shapes coinciding with barriers I, II and LDM barrier shape. It is clear from this figure that the fragment anisotropies at medium excitation energies are indeed characteristic of the LDM barrier shapes, and therefore the transition state shape does not coincide with barrier I or II but with the LDM barrier shape. It has been shown<sup>3</sup> that in near threshold fission, the fragment anisotropies are in fact characerized by the distribution of the K values of the open channels at the top of the barrier II only.

If the near threshold anisotropies are decided by barrier II and at medium excitation energies by LDM barrier, there should be a change of shape of the effective transition state as a function of energy. The evidence for this effect is also found<sup>7</sup> and shown in Fig. 4 where the observed variation of  $K_0^2$  vs  $E_x^s$  for the transition state nucleus  $Pu^{242}$  is shown. It is evident that different shapes at low and medium excitation energies of the experimental curve of  $K_0^2$ versus  $E_x$  arise as a result of the change of the shape of the transition state nucleus from that of barrier II to that of LDM barrier in the energy range of about 4 to 30 MeV.

We shall now discuss the effects of the excitation energy dependence of the shell effects on the determination of the LDM fission barrier heights from the analysis of the measured fission excitation functions. To bring out certain specific effects we consider, as an example, the fission excitation function for the reaction  $_{79}Au^{197} + _{2}He^{4} \rightarrow T1^{201*}$  fission. From these measurements of Burnett et al<sup>9</sup>  $f_{7}/f_{7}$  have been determined and the results are shown in Fig. 5. The main purpose of these kinds of measurements is to obtain information about the fission barrier height.

According to the standard transition state theory first developed by Bohr & Wheeler<sup>10</sup>,  $\int_{\mathcal{L}}$  and  $\int_{\mathcal{T}}$  are essentially proportional to the nuclear level densities in the transition state (saddle) shape and in the nucleus after neutron emission. Calculations of  $\int_{\mathcal{T}}$  essentially involves the level densities of the residual nucleus, which can be microscopically calculated with our method without any free parameters

starting from the single particle, level sequence for the ground state nucleus. There is therefore very little uncertainty about the calculated In. On the other hand, the s.p. level sequence corresponding to a highly deformed saddle configuration are not known to determine by a microscopic calculation, the saddle point level densities and therefore . If, however, one assumes that there are no shell effects at the saddle point for this nucleus one can use the standard expression  $S = 2(aE_v)^2$ for saddle point level density calculation. If this is done, the calcuated  $f_{f}/f_{h}$  do not fit the experimental values as shown in Fig. 5. We have therefore used the experimental  $f_{\beta}/f_{m}$  to calculate the level densities at the saddle point and evaluate shell correction at the saddle point. It is found that in order to fit the experimental values, the level densities  $f^*$  ( $E_x^s$ ) at the saddle point need to be replaced by f' ( $E_x^s + \Delta E_x$ ), where  $\Delta E_x$  is found to be energy dependent for low energies but becomes constant for higher energies. This result shows that the saddle point configuration has a positive shell correct-The magnitude of the shell correction ion as explained in Fig. 6. energy derived from this data, as shown in Fig. 7, is + 2.4 MeV. Myer and Swiatecki<sup>1</sup> have used the LDM barrier height for this nucleus as an input parameter in their mass formula to arrive at the Coulomb energy coefficient. If this positive shell correction of 2.4 MeV at the saddle point is taken into account, the actual liquid drop barrier height becomes 15.0 MeV instead of 17.4 MeV used by This may therefore necessiate a redtermination of the coeffithem. cients of the mass formula, and it is possible that the Coulomb energy radius anamoly pointed out by Myer and Swiatecki<sup>1</sup>, may be partly or wholly attributable to this.

I shall now come to a discussion of the excitation energy dependence of shell effects on the production of superheavy nuclei by heavy ion bombardment. Intensive efforts are now being made to produce these superheavy nuclei in the laboratory by means of heavy ion reactions.

Let us consider the typical case of the following reaction  

$$94^{Pu} + 20^{Ca} \xrightarrow{48} 114^{X^{296*}} 114^{X^{292}} + 4_0^{n^1}$$

Since the kinetic energy of 20 Ca<sup>48</sup> ion should be sufficient to penetrate the Coulomb barrier, the compound nucleus is always formed with a minimum of 30 - 40 MeV excitation energy, which has to be got rid of by means of neutron and gamma emission. As shown in Fig. 8, the fission and neutron emission compete at each stage. For an excited nucleus with  $E_x = 30 - 40$  MeV, we have shown that the shell effects do not manifest themselves on nuclear entropy. Therefore the calcuated  $\frac{1}{2}/\frac{1}{2}$  comes out to be very large-the same which would be expected if there was no fission barrier in the ground state. The calculated values of  $\frac{1}{2}/\frac{1}{2}$  and the total probability for the nucleus to end up in its ground state by a successive cascade of neutron emission for different initial excitation energies are shown in Fig. 9. This shows that even after the compound nucleus is formed with an excitation energy of about 40 MeV, only one nucleus out of about  $10^5$  is expected to survive fission and end up in the ground state forming a superheavy nucleus.

To summarize, this talk was intended to point out certain new effects which may arise due to the excitation energy dependence of the shell effects in the fissioning nuclei at the barrier and at the ground state deformation. The role of shells and of their excitation energy dependence in deciding the mass and charge distribution in fission is a subject of a separate paper in this conference and has not been included in this talk.

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Fig. 1. Double himped barrier in the deformation energy of a heavy nucleus resulting from the addition of the shell correction energy to the LDM deformation energies.



Fig.2. Plot of  $S^2$  versus  $E_x$ , for the cases of the doubly magic nucleus  $Pb^{208}$  and the nucleus  $Pu^{242}$  (spherical shape). The dashed curves in each case represent the asymptotic behaviour at high excitation energies.



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Fig. 3. Variation of  $J_0/J_{eff}$  with  $Z^2/A$ . The continuous curve gives the calculated variation for the LDM barrier shapes, and the patches for the shapes corresponding to barrier I and II.



Fig. 4. Variation of  $K_0^2$  with excitation energy of the transition state nucleus  $Pu^{242}$ . The experimental points are taken from the work of R. Vandenbosch, H. Warhanek and J. R. Huizenga, Phys. Rev. 124, 846 (1961). The calulated variations for the nuclear shapes corresponding to the LDM barrier, barrier I, and barrier II are shown by the different curves, which also take into account the shell and pairing effects on the effective moment of inertia for specified shapes of the nucleus.



Fig. 5. This figure shows the disagreement between the experimental values of  $\frac{1}{4}/\frac{1}{10}$ , and the values calculated with the assumption that the saddle point shape has no shell effects.



Fig. 6. In this figure it is shown that if the saddle point deformation has a positive energy shell correction, and the relation S = 2 (a  $E_x^S$ )  $\frac{1}{2}$  is used to calculate saddle point level densities, the value of  $E_x^S$  is equal to  $E_x - B_{\zeta} - \Delta$  (E) where  $\Delta$  (E) is energy dependent and becomes equal to the positive shell correction in the asymptotic limit of high excitation energy.

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Fig.7. The values of  $\Delta$  (E) required in order to obtain an agreement between the experimental and the calculated values of  $\frac{1}{5}/\frac{1}{5}$ are shown by the dashed curve. Note that the disagreement shown in Fig. 5 refers to the case where  $\Delta$  was assumed to be zero. The solid curve refers to the values of after taking into account the expected changes in the barrier height with increasing angular momentum.





Fig.8. Competition between fission and neutron emission at each stage during the deexcitation of the superheavy nucleus X<sup>296</sup>. 114



Fig.9. Calculated values of  $\int_{\mathcal{F}} / \int_{\mathcal{F}}$  and the total probability for the nucleus to end up in its ground state by a successive cascade of neutron emission for different excitation energies.

DISCUSSION

- M.K. Mehta : You have quite clearly shown that in estimating the production rates for the superheavy elements in heavy ion bombardment one has to remember the fact that one gets a very highly excited compound nucleus in which ground state shell effects are not felt as regards fission probability. How about the possibility of superheavy nuclei production by a reaction of the type U<sup>238</sup> + U<sup>238</sup>, where one of the fission fragments may be formed as a super heavy nucleus.
- S.S. Kapoor : There are several suggestions as to how one can produce these nuclei, and the one you mentioned is one of these. I would think that all these suggestions should be explored.
- V. S. Venkatavardhan: Do you think that the super heavy nuclei could be formed in ground state by successive neutron capture in the stellar interior with appreciable cross section.
- S.S. Kapoor : This will depend on the path in which the intermediate nuclei lie on the N-Z plot and the fission half lives of the intermediate nuclei. Calculations in these directions are still being carried out.

### REPORT ON WORK ON NUCLEAR SPECTROSCOPY OF HIGHLY DEFORMED Th-231

J.E. Lynn and G.D. James A.E.R.E., Harwell, U.K.

and

L.G. Earwaker University of Birmingham, Birmingham, U.K. \* (Reported by E.R. Rae, A.E.R.E., Harwell)

A measurement of the neutron-induced fission cross-section of Th-230 has been made in the neutron energy range 680 keV to 1.4 MeV with a neutron energy resolution of 5 keV. At selected energies near a prominent resonance at 715 keV, the angular distribution of fission products with respect to the neutron beam has been measured with neutron energy resolution of about 18 keV. The results obtained are interpreted in terms of a  $\beta$  -vibration in the secondary fission potential barrier minimum and enable a direct estimate of the effective moment of inertia of the Th-231 nucleus in its shape isomeric state to be obtained.

Did Superheavy Nuclei Exist in Our Solar System ?

Narendra Bhandari Tata Institute of Fundamental Research Bombay 5

#### ABSTRACT

The available evidence on the existence of superheavy elements is evaluated and the results are discussed in terms of their possible physical and chemical properties. There are two lines of evidences, both obtained from a study of primitive objects of our solar systems, the meteorites and lunar dust grains which indicate that superheavy elements were extant at the time these objects solidified. The rare gas evidence based on the presence of an anamolous component of fissiogenic xenon has been discussed in detail by Anders and Heymann (1969) and by Rao (1970). Recently Bhandari et al (1971) have observed fission fragment tracks characteristic of nuclei Z >110. These tracks are present in a number of meteorites as given in table 1. The estimated number of tracks due to  $Pu^{244}$  and element Z >110 relative to  $U^{238}$  as observed in certain enriched regions of the silicate crystals is also given in this table.

Based on these observations arguments are developed for possible chemical nature of elements responsible for the characteristic long (>15 micron) tracks. The fact that the meteorite Angra Dos Reis is highly depleted in such tracks but unusually enriched in U, Pu, Ba etc. lead us to conclude that the element() may not be a chemical homologue of these groups. However its exact chemical properties can not be ascertained from the available data. Near absence of the superheavy tracks in lunar rocks which are younger by about 1 billion years indicates that they must have a half life  $\leq 10^8$  years and must now be extinct.

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# Table I

# Relative abundances of fossil tracks in extraterrestrial samples (Bhandari et al 1971)

SAMPLES	GLASS		• <b></b> -	ABUN	DAN	ICE *
	-	$\hat{\rho}_{U}^{2}$	38	$P_{\rm Pu}^{24}$	4	<i>P</i> (Z > 110)
NORTON COUNTY	AUBRITE	1	:	60	:	22 - 330
MOORE COUNTY	EUCRITE	1	:	39	:	18
STEINBACH	STONY-IRO	N 1	:	х	:	0.4X
NAKHLA	NAKHLITE	1	:	x	:	1.3X
ANGRA DOS REIS	ANGRITE	1	:	19	:	0.002
LUNAR DUST	?	1	:	25	;	4 - 58
LUNAR ROCKS	BASALTIC	1	:	0	:	0
******			• • • • •			*

\* The values refer to certain enriched zones and not to "whole" samples.

DISCUSSION

R. H. Iyer : Is the range of 15-18 microns that you are referring to due to one fragment or both? If it is due to both fragments, have you looked into the possibility that 2 adjacent U atoms or Pu<sup>244</sup> atoms underwent fission and the tracks formed along side added on and appeared as a single track of 25-30 microns?. Bear in mind also that the number of events of long tracks that you see are very small.

Answer : In case of fission, both the fragments are recorded so the length measurements refer to total etchable range of both fragments. The possibility of two tracks falling in a line, next to each other is negligible since the total track density is itself small. Number of long fragments is relatively so large that it cannot be explained by such coincidences.

N. N. Ajitanand : Since the specific ionisation of fragment tracks is expected to be very high is it not possible to differentiate from cosmic-ray tracks by measuring the width and shape of the tracks?

Answer : Certainly it should be possible to estimate the charge by measuring the diameter of tracks but this would require measurement of the residual range also. There are some experimental difficulties in this method and the deductions have to be based on very small differences in diameters. For this reason the total length measurements are much superior.

- P. P. Chakraborty: Can you explain why the tracks crowd around the cleavages?
- Answer
  This was the first observation we made before taking up a systematic search for these "excess" tracks. We now understand that some elements, which cannot form a part of the crystal lattice due to their ionic radius or chemical properties, migrate to the boundaries of the crystal while the crystal is forming. Heavy elements like uranium fall in this category. This is very well established by looking at neutron induced fission tracks of U<sup>2.35</sup> which cluster in such grain boundaries, exsolution planes or faults which appear as cleavages on etching. Transuranic elements can be expected to behave likewise.

R.F. Taschek	:	Were you able to identify any ternary fission fragment tracks since superheavy element fission is supposed to have a large fraction of its disintegrations going this way?
Answer	:	Since we look at tracks across a broad (1-2 micron) cleavage, experimentally it is very difficult to establish which prongs belong to a single event. Extrapolations have to be made to find out if the prongs meet at some point within the cleavage. This is least doubtful in case of two prongs (binary fission) since they should lie in a straight line. Therefore so far we are very doubtful of ternary fission events although such tracks seem to exist. Also sometimes one of the three prongs may not be recorded in the sili- cate mineral, if its charge is smaller than the detector threshold. We are therefore not yet sure of the origin of some V tracks we have seen. Of course, if a mineral

could be easily identified.

exists which retains this fissioning element within the crystal structure, a ternary fission
SESSION 2

Chairman : S. Cierjacks, F.R. Germany

Mass Distributions in Fission and Some Related Phenomena

J. Felvinci, E. Melkonian, W.W. Havens, Jr. Columbia University

#### Introduction

This paper primarily discusses the experiments performed at Columbia University on the several types of mass distributions and their determination, the variation of the mass distribution with the mass of the fissioning nucleus, with its degree of excitation, and with its spin state and also brings in the results of experiments performed elsewhere to illuminate the same subject. Also discussed is a further elaboration of the mass distribution as a function of the kinetic energies of the fission fragments as well as a more detailed account of the emission of neutrons from the fragments. The possibility is raised that the observed mass distributions may be related more to the K quantum number of the transition state than to the spin of the fissioning nucleus.

There are three mass distributions to be considered: I. that just after scission but before any neutrons have been emitted\*, II. that after emission of prompt neutrons, which takes place in less than  $10^{-11}$  seconds, and III. that after emission of all neutrons, including delayed, the most delayed decaying with a half life of 55 seconds. Usually a distinction between II and III is not considered since the delayed neutron fraction is generally less than 1% and spread over many masses.

Radiochemical determinations of the mass distribution yield III (possibly mixed with II if the chemistry is done rapidly enough). Most of the data on the mass distribution falls into this category, so that most trends (e.g. with atomic weight and neutron energy) have been determined by radiochemical means.

\*In the case of  $Cf^{252}$ , and possibly for other fissioning nuclei, there is estimated to be ~10% of the neutrons emitted just before scission. How this affects the following considerations has not yet been determined. Since prompt neutrons are emitted in a time short compared with the time for the fragments to move over a short distance, the pre-neutron mass distribution I is difficult to determine and must be done directly on each fission event just after it occurs, rather than waiting (as in radiochemical determinations) after many fissions have occurred. The very first determinations of mass distribution by physical means were done by simultaneous measurement of the energies of both fragments, initially in gridded ionization chambers and subsequently by means of solid state detectors. If energy and momentum changes arising from the emission of neutrons and gamma rays are ignored, the double-energy data lead directly to a mass distribution through the relationship

$$M_1 = \frac{E_2 M}{E_1 + E_2}$$

where  $E_1$  and  $E_2$  refer to the energies of the two fragments, and M is the mass of the fissioning nucleus. However, consideration of the emission of neutrons leads to the conclusion that the resulting mass distribution corresponds strictly to neither the pre- nor to the post-neutron mass distribution. Application of the "universal" neutron yield versus mass relationship enables correction of these data to an approximate pre-neutron mass distribution. The reason the double-energy measurement does not give a clean I or II distribution is that the emission of a neutron almost always decreases the energy of the fragment because the mass is reduced, thus reducing the energy, even though the velocity averaged over many events remains unchanged.

A direct determination of the pre-neutron mass distribution can be made by measuring simultaneously the velocities of both fragments, instead of the energies, as above, and using the above formula with V's replacing E's. The emission of a neutron changes the velocity of a fragment in a symmetric manner; that is, the velocity is sometimes increased and sometimes decreased, but averaged over many events there is no net change in velocity. Thus a true unbiased pre-neutron mass distribution results, but "smeared" out by a resolution function associated with the neutron emission.

It is possible to determine the post-neutron mass distribution by physical means, but now both energy and velocity must be measured for one fragment for each event. The post neutron mass of one fragment is then obtained from the relationship  $E = MV^2/2$ . We have chosen to measure the energies of both fragments together with the velocity of one in the experiment to be described subsequently. By suitable treatment of the data to simulate a double velocity measurement, the pre-neutron mass distribution can also be deduced.

The motivation for doing this is that in a hypothetically perfect experiment, it becomes possible to deduce the average number of neutrons emitted corresponding to each mass and energy. The "universal" curve gives the number of neutrons emitted for each mass summed over all possible energies. For many masses, yields occur from the highest to the lowest values of the total kinetic energy corresponding respectively to scission occurring at a time when the nucleus is only slightly distorted to the time when the nucleus is stretched out with a substantial "neck." Thus, considerable additional information can be obtained by the simultaneous determination of both pre- and post-neutron mass distributions.

The "real world" measurements suffer from practical considerations such as (a) the solid state detectors, which are the best available for this purpose, have some dispersion in the relationship between pulse height and energy, so that a completely accurate determination of the fragment energies cannot be made; (b) there are limitations on the fragment flight time determinations imposed by existing equipment, noting that our time resolution was as 0.7 nanoseconds; and (c) attempts to get better timing by the use of longer flight paths are limited by intensity considerations. Nevertheless, we have carried out a set of measurements on the thermal neutron fission of U-235 and have obtained some interesting results. Because of resolution limitations, the differences between pre-neutron and post-neutron masses are not integral values corresponding to the emission of individual neutrons, but

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rather show continuous distributions such that even negative values occur as part of the distributions.

## The Fission Fragment Mass Distribution

On the basis of both energetics and a simple liquid drop model, the fissioning nucleus is expected to break up into approximately equal mass fragments. Almost all of the mass distributions observed are startlingly different from this, indicating a double-humped distribution with very little yield at half the original mass (i.e. symmetric fission).

The existence of this asymmetric fission is thought to be determined by a resistance to breaking up of two doubly-magic substructures in the fissioning nucleus. These are (1) Z=50, N=82 giving A=132, and (2) Z=28, N=50, giving A=78. It is seen in the mass distributions that most of the heavy fragment peak is confined to the region A=132 to A=M-78 and that most of the light fragment peak is similarly confined to the region A=78 and A=M-132, where M is the mass of the fissioning nucleus, and neutron emission is ignored.

In addition to this coarse structure, there is frequently observed to be some fine structure. Figs. (1) and (2) show the mass distribution in the case of spontaneous fission of Pu-240<sup>(1)</sup>. The results are somewhat unusual in that there is a rather large and broad peak near mass 134 and a broad shoulder between masses 140 and 146. However, this is only an exaggerated version of features found in other isotopes; e.g. U-235 shows an unusually high yield at mass 134 as well as some additional fine structures. Fig. (1) includes the mass distribution in the thermal-neutron-induced fission of Pu-239 for comparison, showing a considerable difference. Fig. (2) includes the radiochemical data of Laidler and Brown<sup>(2)</sup>.

#### Variation with Mass of the Fissioning Nucleus

The differences in mass distribution amongst the various fissioning nuclei have received considerable attention. (Since these effects are fairly large, the differences amongst the various mass distributions discussed above may be ignored here.) The general observation is that the average mass of the heavy mass peak is almost the same (138-139) for all of the fissioning nuclei for a large range of excitations as long as the double-humped distribution is the predominant feature. Thus for a sequence of isotopes, it is the average mass of the light peak which increases as the atomic weight is increased.

We have added another observation on the low mass end. Fig. (3) shows the single-fragment kinetic energy distribution for the resonance-neutroninduced fission of Th-229 and U-235 taken recently with the Columbia synchrocyclotron used as a pulsed neutron source (3). Using the U-235 data to estimate the pulse-height defect of the solid state detectors used, we find the ratio of the average energies of the light and of the heavy fragment peaks to be 1.59 for Th-229. This ratio is the same as the ratio of average masses of the two distributions. Assuming an average emission of 2.2 neutrons per fission, we find for the heavy mass peak an average mass of 139.7, consistent with the general findings.

Fig. (1) shows that the average mass of the heavy fragment in the case of the spontaneous fission of Pu-240 is again around 138.

# Mass Distribution as a Function of Fragment Kinetic Energy

The mass distribution in fission has another dimension, that of the kinetic energies of the fragments, or probably more significantly, the sum of kinetic energies of the two fragments for each event. Since the total kinetic energy for each event is related to the separation of the fragments at the instant of scission, the mass distributions as a function of total kinetic energy give information about the conditions of the various states of elongation of the fissioning nucleus. Fig. (4) shows the mass distribution in the case of thermal-neutron-induced fission of Pu-239 for 5 MeV slices of the total kinetic energy centered about the indicated values. At the high energy end, the two distributions are very narrow and centered about masses 105 and 133, with essentially no symmetric fission. This is the case of scission occurring very early in the elongation process so that the shell structure prevails. At the other extreme of low total kinetic

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energy, the nucleus has survived so that some breakup of the shell structure has occurred, increasing the yield of symmetric fission. However, the bulk of the yield is still confined to the mass ranges indicated above, and in particular there is very little yield below mass 78.

# Primary and Post-Neutron Mass Distributions and the Emission of Prompt Neutrons

As indicated above, we have made double-energy single-time-of-flight measurements of fission fragments in the thermal-neutron-induced fission of U-235<sup>(//)</sup> to obtain both pre-neutron and post-neutron mass distributions and details on the emission of prompt neutrons as a function of fragment mass and energy. The two resulting mass distributions are shown together in Fig. (5). The mass dispersion of the pre-neutron distribution is about 2 amu FWHM, determined mostly by the effects of neutron emission. The mass dispersion for post-neutron distribution is about 1.9 amu at the center of the light-fragment peak and 3.8 amu at the center of the heavy-fragment peak, both being FWHM, determined mostly by the intrinsic resolution of the solid state detectors.

Fig. (6) shows some representative mass distributions for 6 MeV intervals of total fragment kinetic energy at the high end of the peak, at the middle, and at the lower end of the total kinetic energy distribution. Examination of all of the data shows evidence for a slight preference for fission into mass pairs with heavy fragment masses of 134. 140, 146, and 153 mass units. The evidence here is that all of the structure in the mass distributions is already present at the instant of scission and not produced by neutron emission, although the latter may sharpen some of the structure. The opposite view has been expressed that the primary mass distribution is relatively featureless, and that the observed structure is the cumulative effect of the slow variation of neutron emission with mass  $\binom{13}{2}$ .

Fig. (7) shows the average neutron emission as a function of primary fragment mass as determined in this experiment, while Fig. (8) shows the comparison with other data, all determined by the direct counting of neutrons. The best agreement is with the data of Maslin, Rodgers, and Core, with the largest discrepancy occurring near the doubly magic nucleus of mass 132, where we observe almost no neutron emission, a result which seems to us to be reasonable. (14)

It is difficult to present the data on neutron emission for all combinations of mass and energy in a meaningful way, so only representative data are presented here. Fig. (9) shows the neutron emission for selected pairs of complementary primary fragments integrated over all energies. Examination of all of the data shows that the light fragment yields peak at the emission of one or of two neutrons. For most heavy fragments below mass 140, the most probable number of neutrons emitted is zero, while above mass 140, the neutron distributions usually peak at one neutron, with some peaking at two neutrons and some having two peaks at two and zero.

For the purpose of giving an indication of dependence on fragment kinetic energy, Fig. (10) shows the neutron number distributions for two selected mass numbers (96 and 140) for 5 MeV total kinetic energy intervals. As expected, the peaks shift to larger neutron numbers as the total kinetic energy decreases.

Fig. (11) includes the average neutron emission as a function of total kinetic energy for selected complimentary mass pairs.

## Effect of Energy on the Mass Distribution

When the energy of the incident neutron is increased, the extra excitation energy all goes into neutron emission, the average kinetic energies of the fragments will in fact slightly decrease. The position of the peaks in the mass distribution stays in the same place but the whole distribution broadens, with an increased symmetric fission as a consequence.

In general there seems to be a tendency to have more symmetric fission with increasing excitation energy and this trend can be observed at MeV

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energies. (One has to separate these effects from those of the onset of p and d wave fission.) On the other extreme the spontaneous fission has zero excitation energy (proceeds by barrier penetration) and thus has less symmetric fission, narrower mass distributions than thermal neutron fission.

The fact that the average number of emitted neutrons is smaller for spontaneous fission than for neutron induced fission does affirm the trend. The fewer neutrons emitted from the fragments, the more structure is expected as the features of the primary division are more noticable.

This has been also observed by Toraskar and Melkonian<sup>(1)</sup> in the spontaneous fission of Pu-240. The mass distribution is much narrower and also it shows more structure than the neutron induced fission of Pu-239. The symmetric fission yield is quite low and so is the very asymmetric yield (Fig. 1).

Radiochemical data on other spontaneously fissioning nuclei, U-238, Pu-240, Cm-242, also show the characteristics mentioned above and also indicate more structure in the mass distributions than in most thermal neutron induced fission.

## Variations of Mass Distribution with Spin and/or K Quantum Number

In the previous section it was shown that the mass distribution can vary as the available excitation energy is changed. It has also been observed that the mass distributions can differ for different neutron resonances. To understand this effect one should recall that in a fission process most of the excitation energy is tied up in deformation and thus the nucleus is "cold." <sup>(6)</sup> The effect of this is that the so-called transition states at the saddle point are few in number and are members of different low lying rotational and vibrational bands. The channel theory of fission assumes that the fission proceeds through these few open channels which lie between the deformed ground state of the compound nucleus and the fission threshold energy. Using this knowledge, Wheeler<sup>(7)</sup> predicted that the mass distribution could be different for the different spin states of the compound nucleus. U-235, after capturing an s-wave neutron, could have  $3^{-}$  or  $4^{-}$  spin and thus could fission through the  $3^{-}$  level of the K=0 band, the  $3^{-}$ ,  $4^{-}$  members of the K=1 band or the  $3^{-}$ ,  $4^{-}$  members of the K=2 band. A. Bohr pointed out that the  $3^{-}$  levels going through the K=0 band would have more asymmetrical fission because of the symmetry properties of the band. Experiments based on these ideas were performed and spins of resonances assigned using ratios of symmetric to asymmetric fission (8)(9). Other experiments determined spins directly through neutron scattering measurements or capture  $\gamma$ -ray multiplicities. There is considerable contradiction between these findings.

In Pu-240 the spin of the compound nucleus formed from Pu-239 by s-wave neutron capture is  $1^+$  or  $0^+$ . The ground state rotational band K= $0^+$ has a  $J=0^+$ , and a  $K=1^+$  band at around 1.2 MeV above ground state has a  $J=1^+$ spin state, both of which are definitely open channels (Fig. (12)). A higher lying second  $K=0^+$  band having  $J=0^+$  spin state is possibly partially open. Thus in Pu-240 the levels divide quite nicely into two groups -narrower levels having presumably  $J=1^+$  and wider levels  $J=0^+$ . Both from the excitation energy and symmetry arguments, the mass distributions should show more symmetric fission for the  $0^+$  levels and less for the  $1^+$  levels. This has been observed by Cowan<sup>(8)</sup>, who determined radiochemically the ratios of Mo-99 to Cd-115 corresponding to asymmetric and symmetric fission as a function of incident neutron energy. To test this hypothesis in more detail and also with physical means, an experiment was performed at Columbia by Toraskar and Melkonian<sup>(11)</sup>. In this measurement, the kinetic energies and mass distributions were obtained for the induced fission of Pu-239 by neutrons filtered through beryllium and through samarium. The beryllium filter enhances the negative level, and the samarium filter the 0.297 eV level. These two levels are known to have different spins, the negative energy level having spin  $0^+$ . the 0.297 eV level 1. The results in Fig. (13a) and (13b) show that the mass distribution induced by the beryllium filtered neutrons has indeed a higher symmetric fission yield as expected from the  $0^+$  spin. The ratio of the symmetric fission induced by the beryllium filtered neutrons to that of the samarium filtered neutrons is around 2. There are also smaller differences in the most probable fission yields between the two levels (Figs. (15a) and (15b)). The total kinetic energies were also measured, of course, and they show a shift of 0.7 MeV, with the beryllium filtered neutron-induced fission lower than that of the samarium filtered neutron-induced fission (Pig. (14)). This is in line with expectations from excitation energy considerations.

Studying these and other results, we feel that some of the inconsistencies in spin assignments are the result of the fact that the mass distributions are determined not so much by the spins of the transition states, but by the available excitation energy and thus they are correlated rather with the K values of the different rotational and vibrational bands (5)(12).

Is there any possiblility of obtaining selection rules which would tell us which bands are more likely to be responsible for fission and thus predict the magnitudes of mass distribution effects? Let us assume that when the compound nucleus is formed, K is still a good quantum number. This would mean that the most favored fissions would be those where K varies least, and the least favored those with the largest change in K. The U-236 compound nucleus has 3<sup>-</sup> and 4<sup>-</sup> spins and assume it also has a certain distribution of K values from 0 to 4. The rule suggested above would minimize fission for the K=0<sup>-</sup> band. The fact that measured differences in the ratios of symmetric to asymmetric fission in the U-235 resonances are very small would also argue against a transition through the K=0<sup>-</sup> band.

There is added information about the capture  $\gamma$ -rays in U-235 which supports this argument. It has been observed that the probability of El transition to the 2<sup>+</sup> and 4<sup>+</sup> members of the K=0<sup>+</sup> ground state band is very small. This was quite unexpected and an assumption that the compound nucleus level has a definite K value would help to explain this. K selection rules in  $\gamma$ -transition require  $\Delta K \leq L$  where L is the multipolarity of the  $\gamma$ -ray.  $\Delta K = 2,3,4$  is thus highly prohibited.

This is not the case in Pu-239 where some high energy transitions are observed, and members of  $\Delta K = 0^{-}$ , 1<sup>-</sup> bands could be excited.

Radiochemical experiments done by Regier<sup>(10)</sup> on U-233 also indicate that the mass distribution might be quite different for different resonances. A simple experiment to test this and its possible effects on cross section measurements was performed during 1970, using the neutron beam from the Columbia synchrocyclotron. Single fragment kinetic energies and neutron time-of-flights were recorded and subsequently analyzed by taking selected cuts in the pulse height spectrum and calculating the corresponding resonance areas (Fig. (16)). Significant differences were observed between selected resonances when the lowest energy fragments were compared to all fragments<sup>(3)</sup>. The lowest energies correspond mostly to the heaviest fragments and thus give us the effects of the wings of the mass distribution. Looking again at the K bands in U-234, we can see that the ground state band K=0<sup>+</sup> has a J=2<sup>+</sup> but no J=3<sup>+</sup> level; the K=2<sup>+</sup> band around 0.7 MeV and the K=1<sup>+</sup> band at around 1.4 MeV have both 2<sup>+</sup> and 3<sup>+</sup> levels.

We expect three families of mass distributions in U-233 which would correspond to fissions through the K=0<sup>+</sup>, 1<sup>+</sup> and 2<sup>+</sup> bands respectively. As mentioned earlier the energy differences between different K bands are generally larger than between the two spin states within the same band. If the assumption of a definite K value in the compound nucleus is valid, we expect only few fissions to proceed through the ground state band. The mass distribution for the K=1<sup>+</sup> band would have more symmetric fission than that for the K=2<sup>+</sup> band. The energy difference between these two bands in U-233 is smaller than the differences in Pu-239 and thus the expected variation in symmetric fission from resonance to resonance is also smaller in U-233 than in Pu-239. Both Regier's<sup>(10)</sup> radiochemical results and our experiment would confirm this assumption.

## Effects of the Mass Distribution on Fission Cross Section Measurements

The measurement of mass distributions is important not only in underst nding the physics of the fission process itself, but also in the more "practical" realm of cross section measurements. As has been pointed out previously, the fission yields of the different neutron resonances depend to a certain degree on the bias used in the fission fragment energy measurements. Certain techniques, e.g. ionization chamber measurements, can cut off the low energy fission fragments through biasing (as much as 10-30% of the fragments) and thus slightly distort the cross section measured at the different resonances. The results of Felvinci and Melkonian<sup>(3)</sup> would indicate this possibility in U-233. In Pu-239 this effect could be even more serious. Because the differences between the bands in U-235 are small, its cross section measurement would be affected least. Unfortunately, at higher energies, say above 10 keV, p-wave fission becomes important and thus increased effects of this on the mass distributions and consequently on  $\sigma_{\rm f}$  measurements cannot be ruled out.

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Fig. 1. Mass distribution for the spontaneous fission of  $Pu^{240}$  superimposed on the normalized mass distribution for the induced fission of  $Pu^{239}$  by beryllium-filtered neutrons.



Fig. 2. Mass distribution for the spontaneous fission of  $Pu^{240}$  superimposed on the radiochemical mass distributions.





Fig. 4. Mass distribution for the beryllium-filtered neutron induced fission of Pu-239, considering fixed 5 MeV intervals of the total kinetic energy distribution.



Fig. 5. Primary and post-neutron mass yields. Error bars indicate statistical error.



Fig. 6. Primary and post-neutron mass distributions at selected values of primary fragment total kinetic energy.



Fig. 7. Average neutron emission as a function of primary fragment mass. Error bars include only statistical error. The total neutron emission is the sum of the single-fragment values for complementary mass pairs. The pre-neutron mass distribution is shown for reference.



Fig. 8. Comparison of average neutron-emission results of the present experiment with those obtained in the direct neutron-counting experiments.



Fig. 9. Neutron number distributions from single fragments of complementary mass. Circles; light fragment. Crosses; heavy fragment.



Fig. 10. Neutron number distributions from fragments of mass 96 (circles) and 140 (crosses), at different values of fragment total kinetic energy. Error bars indicate statistical error.



Fig. 11. Results at fixed values of primary fragment mass. Left: total-kinetic energy distributions before neutron emission. Right: average neutron emission as a function of primary total kinetic energy for complementary fragment mass pairs.



 $F_{ig}$ . 12. Spectrum of a fissioning Pu<sup>240</sup> nucleus at saddle point compared with the low-lying spectrum of the stable Pu<sup>240</sup> nucleus. Reference: Proc. of the IAEA Symposium on Phys. & Chem. of Fission, 1965.



Fig. 13a. Mass distributions  $N(\mu)$  for the  $Pu^{239}$  fissions induced by the beryllium filtered neutrons.



Fig. 13b. Mass distributions  $N(\mu)$  for the  $Pu^{239}$  fissions induced by the samarium filtered neutrons.



Fig. 14. Measured total kinetic energy distributions for the  $Pu^{239}$  fissions induced by the beryllium and samarium filtered neutrons.



Fig. 15a. Mass distributions  $N(\mu)$  and the corresponding preneutron mass distributions N(m) for the  $Pu^{239}$  fissions induced by the beryllium filtered neutrons.



Fig. 15b. Mass distributions  $N(\mu)$  and the corresponding preneutron mass distributions N(m) for the  $Pu^{239}$  fissions induced by the samarium filtered neutrons.



Fig. 16.  $U^{233}$  single fragment kinetic energy distribution. Different energy groups are numbered 2 to 8.

DISCUSSION

- M.R. lyer : Has there been any measurement of neutron emission as a function of the charge of the fragment for the same mass? We are interested in the mass yield data for the spontaneous fission of Pu<sup>240</sup>.
- W.W.Havens : I do not know of any such measurements. Prof. Melkonian has the data on the spontaneous fission of Pu<sup>240</sup> stored on magnetic tape. Write to him and ask him for the specific data you would like and if he has it available, I'm sure he will be happy to send it to you.
- D. M. Nadkarni : 1. You showed evidence to suggest a dependence of mass distribution on the K quantum number of the saddle point nucleus, and also some influence of the shell structure of the fission fragments. Do you think there is any contradiction in these two findings from the point of view of finding where the mass distribution is decided during the fission process?
  - : 2. What were the values of the peak to valley ratio of the mass distributions for the two different incident neutron energy cases?
- : Our treatment of the shell structure of the nucleus W.W. Havens is not as detailed as your question implies. We invoked shell structure to explain the gross assymetry of the mass distribution in nuclear fission. Our opinion is that the K quantum number of the deformed nucleus just before the nucleus splits appears to determine the gross mass distribution. Probably the final picture will require a combination of collective model theory for the deformed nucleus and the shell structure of the fragments. The peak to valley ratios for Pu<sup>240</sup> are given in Fig. 13, a for Be filtered neutrons and in Fig. 13 b for Samarium filtered The ratios were about 4/1 for Be neutrons. filtered neutrons and 8/1 for Samarium filtered neutrons.

## FISSION ISOMERIC STATE IN U-236\*

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In a recent experiment at the Nevis cyclotron, we looked for evidence of an isomeric state of U-236 in fission induced by resonance energy neutrons in a U-235 target. The neutron energies involved ranged from a few hundredths of an eV to about 3 keV. Thus, the excitation energies of the compound nucleus in this experiment were restricted to much lower values than in previous work<sup>(1)</sup> on U-236 fission isomers, where the excitation energies ranged upto several MeV above the neutron binding energy.

In this experiment, we used a time-to-amplitude converter (TAC) to measure the time interval between the detection of a fission event and a gamma ray associated with the fission. The fission detectors were silicon surface barrier detectors, and the gamma rays were detected by means of a 3 by 3 inch NaI crystal. The energy of the fission-inducing neutron was also recorded.

We expected fission to be delayed with respect to the gamma ray in the case of a  $(\gamma, f)$  reaction in which the nucleus decays to a state below the fission barrier prior to fissioning by barrier penetration. This should produce an exponential fission decay, which we would see superimposed on a prompt peak in the TAC spectrum. With our time resolution of 3.4 nsec, the prompt gamma rays originating from the fragments would appear to take place simultaneously with fission, along with any  $(\gamma, f)$  reactions to states above the barrier.

The figure shows the TAC spectrum we obtained. We have made an effort to eliminate the possibility of an istrumental origin for the observed asymmetry to the right of the peak. We have investigated and ruled out the effects of walk due to pulse-height variation, pile-up due to high count rates in the gamma detector, gain drift in the phototube, instability in the phototube high voltage supply, and radio frequency pickup from the cyclotron.

The spectrum is consistent with a half-life of  $7\pm1$  nsec for the delayed fission. There is a suggestion of the presence of a longer half-life decay, but our statistics are not good enough to allow us to draw any conclusions about its lifetime. Previous work<sup>(1)</sup> on U-236 has resulted in half-life determinations ranging from 66 to 130 nsec. The ratio of delayed to prompt fission in our results appears to be quite large. Our preliminary lower limit is 2%

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difference between  $\gamma$ -rays and fission fragments from the neutron induced fission of U-235. The prompt peak is in center of the figure.

DISCUSSION

- M.K. Mehta : Is there any further work planned on the 7 ns. isomer in <sup>236</sup>U? The reason why I ask is that in our work at Seattle and in the work of the Copenhagen group this isomer was not seen. We saw the decay period of about 120 ns and did not see the 7 ns decay although the time resolution was good enough. Your delayed to prompt ratio of 2% also seems to be quite high.
- W.W. Havens : I think that Charles Bowman at Lawrence Livermore Laboratory expects to look for an isomeric state in U<sup>236</sup> with low energy neutrons. If he has not done all the experiments by the time our Cyclotron is reconstructed we will redo these experiments with better resolution and intensity. We were using much lower energy neutrons than the Copenhagen and Seattle groups and the 7 nanosec state may be confined to very low energy neutrons. Our total number of counts was not sufficient to say anything about the 46 130 nanosec state.
#### THEORY OF FRAGMENT MASS AND CHARGE DISTRIBUTION IN FISSION

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The asymmetric division of fragment mass in low energy fission of heavy nuclei is one of those longstanding problems in Nuclear Physics which inspite of many detailed experimental investigations have defined all attempts towards a physical understanding of the process in terms of a simple theory. The Liquid Drop Model of fission which is the historical starting point of all fission theories and which has been quite successful in explaining qualitatively many known features of fission such as the limit of stability of naturally occuring nuclei against spontaneous fission and the existence of a fission threshold even for nuclei having a large positive Q-value for fission, has failed conspicuously in explaining the fission fragment mass asymmetry. It is now known that this failure arises mainly as a result of neglecting the internal structure of the nucleus.

Considerable progress has been made in the last few years in our understanding of nuclear shell effects on the deformation energies of nuclei. On the assumption that the fission process is adiabatic with respect to the collective degrees of freedom such as elongation and rotation of the nucleus as a whole, compared with the individual nucleon motions, many calculations have been done for the deformation potential energy surfaces of nuclei. These calculations have had remarkable success in explaining many of the pre-saddle point properties of fissioning nuclei, in particular the existence of a secondary minimum in the potential energy surface, which has strikingly changed the traditional notion of the fission process. Extrapolation of these calculations into the region of very large deformations of fissioning nuclei has also brought out another important feature, that is the preformation of fragments during the last stages of the division process. It has been shown earlier(1) that considerable simplification can occur in our understanding of the division process of the fissioning nucleus if one recognizes the fact that during the last stages of the process, the fissioning nucleus can be well approximated by two nearly independent nuclei in close proximity; they interact through the exchange of a relatively small number of nucleons from inert cores of the nascent fragments. On the assumption that the fission process is adiabitic with respect to the collective degrees of freedom of elongation and rotation, the process can be treated as a stochastic process. It is shown here that with certain simplifying assumptions regarding the structure of single particle levels in the nascent fragments the observed distributions of fragment mass and charge in the fission of a wide range of nuclei as well as their dependence on the excitation energy of the fissioning nucleus can be explained quite satisfactorily.

The mathematical formulation used here is essentially the same as that described in Ref. (1) and is summarised below: If  $\omega_{NZ}$  is the probability that the configuration of the fissioning nucleus is that with N neutrons and Z protons on the heavy side and  $P_{NZ}$ , N'Z' is the probability that a configuration with N neutrons and Z protons in the heavy side goes over to one with N' neutrons and Z' protons on the same side then one has

$$\omega_{NZ}(t = t_o + \Delta t) = \begin{cases} \xi & \xi & \omega \\ Z' & N' & N'Z' \end{cases} (t = t_o) P_{N'Z', NZ} \qquad \dots (1)$$

If the nascent fragments are in equilibrium,

$$\omega_{NZ}(t = to + \Delta t) = \omega_{NZ} (t = to)$$

That is

$$\omega_{NZ} = \sum_{N'}^{\xi} \sum_{Z'}^{\xi} \omega_{N'Z'} P_{N'Z',NZ}$$

...(2)

By definition

$$\begin{array}{c} \xi & \xi \\ N & Z \\ \end{array} \begin{array}{c} NZ \\ \Sigma \\ N' \\ Z' \\ \end{array} \begin{array}{c} NZ \\ NZ, \\ N'Z' \\ \end{array} \begin{array}{c} 1 \\ 1 \\ 1 \\ 1 \end{array}$$

If the unit of time  $\triangle$  t is sufficiently small one can neglect multinucleon transfers; that is

$$P_{N'Z', NZ} = O \qquad N' \neq N, \quad N\underline{+}1$$
$$Z' \neq Z, \quad Z\underline{+}1$$

The transition probabilities  $P_{N'Z', NZ}$  are simply related to the nuclear transfer probabilities  $P_{L \rightarrow H}$  and  $P_{H \rightarrow L}$  where  $P_{L \rightarrow H}$  is the probability of a nucleon transfer from the light to the heavy fragment and  $P_{H \rightarrow L}$  is the probability of an inverse transfer. An estimate of the relative probability of nucleon transfer in a given direction can be made by using the expression

$$P_{L \rightarrow H} \sim \int \mathcal{J}_{L}(E) \, \mathcal{J}_{L}(E) \quad T_{L \rightarrow H}(E) \, \mathcal{J}_{H}(E) \left[ 1 - \mathcal{J}_{H}(E) \right] \, dE$$

$$P_{H \rightarrow L} = \int \mathcal{J}_{H}(E) \, \mathcal{J}_{H}(E) \quad T_{H \rightarrow L}(E) \, \mathcal{J}_{L}(E) \left[ 1 - \mathcal{J}_{L}(H) \right] \, dE \quad \dots (3)$$

 $\mathcal{F}_{L}(E)$  and  $\mathcal{F}_{H}(E)$  are the single particle energy level densities in the light and the heavy fragment respectively.  $\mathcal{F}_{L}(E)$  and  $\mathcal{F}_{H}(E)$  are the Fermi-Dirac distribution functions.

The probability of a nucleon transfer from one fragment to an identical energy level on the other may be assumed to be equal and independent of energy i.e.

 $T_{H} \rightarrow L = T_{L} \rightarrow H = Constant$ 

The absolute values of  $T_L \rightarrow H$  and  $T_H \rightarrow L$  depend on the actual potential energy profile in the region between the nascent fragments. However, the equilibrium probability distributions of fragment mass and charge are independent of the actual values of  $T_{L} \rightarrow H$  and  $T_{H} \rightarrow L$ A numerical evaluation of the nucleon transfer probabilities on the basis of Eq. (3) require only a knowledge of the nucleon single particle energy level densities  $g_L(E)$  and  $g_H(E)$  in the light and the heavy fragment and the corresponding Fermi-Dirac distribution functions  $f_L(E)$  and  $f_H(E)$ .

The distribution of single particle levels  $g_{L}(E)$  and  $g_{H}(E)$  in the light and the heavy nascent fragments respectively are model dependent and carry the nuclear information. If one neglects nuclear shell effects,  $g_{I}(E)$  and  $g_{H}(E)$  are simple functions of energy and correspond to an assembly of noninteracting fermions confined to a given volume. If, however, one has to take into account nuclear shell effects, the distributions are to be suitably modified. Also in the high energy limit, where the nucleons populate a large number of levels, nuclear shell effects are expected to have minimum influence on the nucleon transfer probabilities and therefore on the resulting distributions of fragment mass and charge. In order to illustrate the details of the present mechanism, we first present an analysis without including nuclear shell effects. (The Liquid Drop Model limit). For  $g_{I}(E)$  and  $g_{H}(E)$  we use the average single particle level densities obtained by Strutinsky-smearing(2) of the shell model single particle energy levels of Seeger and Perisho<sup>(3)</sup>. A value of  $0.7 \frac{1}{7} \omega$  was used for the smearing parameter and is sufficient to remove all shell effects, as was shown by Strutinsky<sup>(2)</sup>. For the case of protons, one has also to take into account the effect of the Coulomb self energy of the individual nascent fragments and also of the Coulomb interaction energy between the nascent fragments. Consequently, all the proton levels are raised by an amount  $(C_3Z/A^{1/3} + kZ')$  where Z and Z' are the atomic numbers of the fragment pairs under consideration. C<sub>3</sub> is the Coulomb energy coefficient of the semi-empirical nuclear mass formula, k is a measure of the interfragment distance. Anestimate of k can be made from the measured average kinetic energy of the fragment pair under consideration. The Fermi-Dirac distribution function f(E) is given by

$$f(E) = \frac{1}{1 + \exp(E - \mu) / T}$$

where  $\mu$  is the chemical potential and T is the thermo-dynamic temperature. T is determined from the condition that the integral of f(E)g(E) over all energies should be equal to the total number of nucleons (protons or neutrons) on either one of the nascent fragments. The assumption of statistical equilibrium between the fragment pairs implies equal temperature for any fragment pair. We also make the further assumption that one can represent the situation by an average temperature T for all fragment pairs, T will be a measure of the excitation energy of the fissioning nucleus near the scission point and therefore should also be simply related to the initial excitation energy of the compound nucleus. With these assumptions we first present the results of the calculations for the case of the high energy fission of the nucleus <sup>210</sup>Po. For nuclei in this region of mass numbers, the mass division<sup>84</sup> is known to be symmetric and the Liquid Drop Model description of the fission process has had reasonable success.<sup>(4)</sup> Because of the decisive role played the relative disposition of the chemical potential of the pair fragments in deciding the direction of prefrential nucleon transfers, we have shown in Fig.1 the proton and the neutron chemical potential of the pair fragments for a mass ratio R = 1.625, (M<sub>H</sub> = 130; M<sub>L</sub> = 80). It is seen that the point of equal chemical potential is very well defined for both protons and neutrons. Away from the point of equal chemical potential, their relative dispositions in the nascent fragment pairs are such that there is always a tendency for a preferential transfer of nucleons leading towards the point of equal chemical potential. Since this mass division is not the most probable mass division for the fissioning nucleus, the points of equal chemical potential for the protons and the neutrons are different and the most probable charge division for this mass division can only be obtained by solving the equilibrium equation (2). It can be however, be immediately seen that the most probable charge division coincides very nearly but not exactly with the prediction of the unchanged charge Density (UCD) hypothesis. The heavy fragment acquires a somewhat lesser charge density than the complementary light fragment. Fig. (2) shows the fragment mass distribution, obtained by solving Eq. (2). The calculation was carried out with a value of 1.2 MeV for the average nascent fragment temperature. Fig. (3) shows the corresponding plot of the most-probable charge versus the mass of the heavy fragment. As is customary, the deviation of the most probable charge from the prediction of the UCD hypothesis is plotted instead of the most probable charge itself in order to bringout the finer details, Fig.4 shows a plot of the percentage yield of various fragment charges for the same fragment mass. It is seen that distribution is very nearly a gaussion with a width of about 1.5 charge units. Similar calculations have also been carried out for the case of the fission of heavier nuclei. It was found that in all cases the mass distribution was predominantly symmetric as expected on the basis of the Liquid Drop Model calculations and should be associated with the high energy fission of these nuclei.

In low energy fission, the assumption of the complete absence of shell effects is not valid. Shell effects manifest themselves as a deviation of  $g_{I}(E)$  from a smooth dependence on energy. A precise knowledge of  $g_{L}(E)$  and  $g_{H}(E)$  requires the details of the shape of the fissioning nucleus near the scission point. In the absence of such a precise information we have carried out calculations on the basis of some simplifying assumptions to bring out the essential validity of the proposed mechanism of fragment mass and charge division. It is known that, in general, nuclei near closed shells possess extra rigidity against deformation whereas midshell nuclei are more soft towards deformation. Consequently, near the scission point, nascent fragments in the region of closed shell nuclei tend to retain their spherical shape. where as fragments in the region of midshell nuclei tend to get de-The consequence of such a deformation is twofold: Though formed. with increasing deformation there is always a formation of secondary shells, these secondary shells have in general a lesser strength compared to spherical nuclei. Also because of the increasing Liquid Drop Model deformation energy, there is an overall increase in the energy of the levels (neglecting shell effects, which might raise or lower any one particular level) with these qualitative remarks in kind, we introduce the following refinements in the assumed spectrum of single particle levels  $g_T(E)$  and  $g_H(E)$  in the light and the heavy nascent fragments. Fragments in the deformed region defined by

32	Л	Z	$\leq$	46
54	\$	Z	4	78
54	٤	N	\$	78
86	4	N	4	122

are deformed further near scission and compared to spherical nascent fragments may be assumed to have little or no shell effects. One can therefore use for these nascent fragments the shell independent single particle energy level densities. Also for these fragments, all the levels near the Fermi Level are raised by an amount C. The exact magnitude of C depends on the deformation of the fragment and is treated as a free parameter in the present calculations. For nascent fragments near closed shells, one should use a shell dependent level density distribution. This was obtained from the shell model level scheme of Seeger and Perisho by partially smearing the levels to take into account the small deformations that these fragments might also possess and the splitting of levels due to the interfragment interaction. A value of 0.4 hw for the smearing parameter was found to be most suitable. Calculations were carried out for the fissioning nuclei  $^{226}$ Ra and  $^{236}$ U for different nascent fragment temperatures. Fig. 5 shows the calculated fragment mass distribution in the fission of  $^{226}$ Ra for two nascent fragment temperatures, T = 1.0 and T = 1.1 MeV. The pronounced triple peaked structure is immediately seen. One can qualitatively understand this structure in the fragment mass

distribution as follows: For nascent fragment configurations in the region 113  $\leq$  M<sub>H</sub>  $\leq$  125, both the light and the heavy fragment are in the deformation region. Consequently, those fragments have been assumed to have little or no shell structure and the resulting distribution of fragment mass yield in this region is symmetric, as was shown to be the case when one neglects shell effects. However, the presence of a doubly closed shell in the region  $M_{\rm H} \sim 130$ , results in an asymmetric peak in this mass region. The mass yield curves, the reference shows a triple peaked structure, one centered around the symmetric division and the other around the asymmetric division. As the temperature is increased, the shell effects tend to disappear, resulting in a relatively smaller asymmetric peak. At very high excitation energies, shell effects completely disappear, and the mass distribution is completely symmetric. Fig.6 shows the calculated fragment mass distribution in the fission of <sup>236</sup>U for two nascent fragment temperatures, T = 0.5 and 0.75 MeV with C = 0.2 MeV. Also shown in figure, is experimental post neutron emission mass distribution taken from Ref. (5). It is seen that the calculated distribution agrees very well in shape and magnitude with the experimental distribution. An increase in the nascent fragment temperature tends to fill up the symmetric region, which is also consistent with the experimental results that there is an increase in the symmetric yield with increasing excitation energy of the compound nucleus. Fig. 7 shows a plot of the most probable charge versus the fragment mass. This shows that the general trend is well reproduced, though the calculations tend to underestimate the most probable charge consistently by about 0.2 charge units. The experimental points are from Ref. 6.

It is thus seen that most of the systematics in the fission fragment mass distributions are very well brought out by the proposed mechanism, with simple assumption regarding the distribution of single particle levels in the nascent fragments. The near absence of nuclear shell effects in all nascent fragments except those in the doubly closed shell region is consistent with the fact that most of the fragments are quite deformed near the scission point. It can also be said that secondary shells do not play any significant role in deciding the mass and charge distributions. The use of a more realistic set of single particle level distribution for the nascent fragments can also be expected to give a better quantitative fit to the experimental data.

It is of interest at this stage to ask the question "what is the relevance of the proposed model to the more fundamental approach taken by Nix<sup>4</sup> where one writes down the Hamiltonian and solves the equations of motion". The present model assumes statistical equilibrium in the asymmetry degree of freedom, and has neglected the effects of dynamical motion in the rest of the degrees of freedom on the final mass asymmetry. This can be interpreted to imply that the effective mass in the asymmetry degree of freedom is quite small compared to the effective masses in the other collective degrees of freedom. Though a theoretical justification for this assumption is yet to be made available, it has enabled one to take into account in a simple way the influence of single particle effects into mass division process.

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Fig. 1: Plot of the chemical potential versus the nucleon number for a given mass division (M<sub>H</sub>=130) in the fission of <sup>210</sup>Po. The temperature of the nascent fragments is taken as 1.2 MeV and no shell effect have been included.



Fig. 2: Calculated fragment mass distribution in the fission of 210 Po. Nascent fragment temperature T=1.2 MeV. No shell effects have been included.



Fig. 3: Calculated most probable fragment charge versus the fragment mass. As is customary, the deviation of the most probable charge from the UCD hypothesis value rather than the most probable charge itself is given in the y-axis. Nascent fragment temperature T=1.2 MeV. No shell effect have been included.



Fig. 4: Percentage yield of various fragment charges for a given fragment mass. (M<sub>H</sub>=130) nascent fragment temperature T=1.2 MeV. No shell effects have been included.





$$T = 1.0 \text{ MeV}$$
  
-----  $T = 1.1 \text{ MeV}$ 

Fragments in the doubly closed shell region are assumed to have shell dependent single particle energy level density distribution, and C = 0.2 MeV.



Fig. 6: Calculated fragment mass distribution in the fission of 236U for two different nascent fragment temperatures

$$T = 0.5 \text{ MeV}$$

Fragments in the doubly closed shell region have been assumed to have a shell dependent single particle energy level density distribution and C=0.2 MeV. The points give the experimentally obtained post neutron emission mass dis ribution in thermal neutron induced fission of  $^{235}$ U. (Ref. 5).



Fig.7: Calculated most probable fragment charge versus the fragment mass in fission of 236U for the nascent fragment temperature T=0.5 MeV. The points give the experimental results obtained into thermal neutron induced fission of  $^{235}$ U (Ref. 6)

#### DISCUSSION

M.R. Iyer : Did you notice any odd/even effects in the form of fine structures in the calculated mass yield curve? Have you estimated the width of charge distribution as a function of mass? Don't you expect magic numbers to influence the width of the charge distributions?

V.S.Ramamurthy: I have not specifically looked for this effect though our calculation gives this information. I can only say at the moment that no drastic odd/even effects are exhibited.

> The width of the charge distribution was found to be about 1.8 charge units (full width at half maximum) in reasonable agreement with Wahl's value. Of course our value refers to the prompt fragments while Wahl's value refers to post-neutron emission distribution.

The influence of magic numbers on the width was also observed. But the change in the width was not drastic as would have normally been expected.

## DISTRIBUTION OF THE TOTAL FISSION WIDTH

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#### ABSTRACT

Some recent developments in the distribution of the total fission width are presented. General expressions for the correlation coefficients of the resonance parameters which take into account the effects due to unitarity are given. A two-resonance two-channel model is used to derive an expression for the distribution of the total width.

#### I. INTRODUCTION

The reactions which go through the formation of a compound nucleus have been studied for a fairly long time. Compared to the direct reactions they occur much more slowly. What one usually observes in these reactions are a bunch of resonances which are fitted using a Briet-Wigner amplitude. The resonance form of the scattering matrix is derived either using the R-matrix theory<sup>1</sup> of Wigner and Eisenbud or the unified theory of nuclear reactions developed by Feshbach<sup>2</sup>. So far as the fitting of the data is concerned both these theories give the same parameteric form of the scattering matrix.

Till very recently it was assumed that the resonances are non interfering type and therefore the resonance parameters of the scattering matrix are the same as the ones of the R-matrix theory. Even within this approximation, it was known that the distribution of fission widths cannot be handled in a simple way since the fissioning process may give rise to correlations among them. Lately the statistical theory has also been applied to the heavy ion reactions<sup>3</sup>, where the ratio of the average width to the average spacing is definitely not small. For such situations the theoretical expressions have to be rederived without any restriction on the above ratio.

The purpose of this paper is to present some of these general expressions without going into their derivations and reexamine the problem of correlations and mean-square deviations of the total widths for this general situation.

#### IL RELATIONS BETWEEN S MATRIX AND R MATRIX

The unitary scattering matrix S based on R matrix theory or Feshbach's unified theory can always be written in the form

$$S = V \left[ I - i \sum_{\lambda, \mu=1}^{N} (\overline{X}_{\lambda} * \overline{X}_{\mu}) A_{\lambda \mu} \right] V$$
<sup>(1)</sup>

where V is unitary and symmetric,  $X_{\lambda}$  is a real vector in channel space. It is the R-matrix width amplitude. N is the number of resonances and  $A_{\lambda\mu}$  are the matrix elements of the level matrix, which are given by

$$(\overline{A}')_{\lambda\mu} = (\overline{E} - \overline{E}_{\lambda}) \delta_{\lambda\mu} + \frac{i}{2} (\overline{X}_{\lambda}, X_{\mu})$$
<sup>(2)</sup>

where  $E_{\lambda}$  are the real eigenvalues of the compound nucleus Hamiltonian and  $(\overline{X}_{\lambda}, \overline{X}_{\lambda})$  denotes the scalar product of the vectors  $\overline{X}_{\lambda}$  and  $\overline{X}_{\lambda}$  in the channel space.

The pole resonance form of the scattering matrix S is given by

$$S = V \left[ 1 - i \sum_{\lambda=1}^{N} \frac{G_{\lambda} \times G_{\lambda}}{E - Z_{\lambda}} \right] V \qquad (3)$$

where  $Z_{\lambda}$  are complex poles,  $Z_{\lambda} = \xi_{\lambda} - \frac{i}{2} \prod_{\lambda}$  and  $\mathcal{G}_{\lambda}$  is now the complex width amplitude. Comparing expressions (1) and (3) we can express  $\mathcal{G}_{\lambda}$ ,  $Z_{\lambda}$  in terms of R-matrix parameters  $\mathcal{X}_{\lambda}$ ,  $\mathcal{E}_{\lambda}$ . The view point which one takes is that the properties of the R-matrix parameters are fixed while those of S-matrix can change but are completely determined by the relations which are provided by the identity in E.

For non-interfering resonances  $A^{-1}$  is diagnonal and we can immediately see from relations (1), (2) and (3) that S has the same parameters as R. When the resonances start interfering the parameters  $\mathcal{E}_{\mu}$ ,  $\mathcal{I}_{\mu}$  become functions of both  $\mathbf{E}_{\mu}$ ,  $\mathbf{X}_{\mu c}$ . We give some of these relations here. They are:

$$\sum_{\substack{M=1\\M=1}}^{N} \mathcal{E}_{\mu} = \sum_{\substack{M=1\\M=1}}^{N} \mathcal{E}_{\mu}, \qquad ,$$

$$\sum_{\substack{M=1\\M=1}}^{N} I_{\mu} = \sum_{\substack{M=1\\M=1}}^{N} \sum_{\substack{m=1\\K=1}}^{m} \chi_{\mu c}, \qquad ,$$

$$\sum_{\substack{M=1\\M=1\\K=1}}^{N} \left( \mathcal{E}_{\mu} \mathcal{E}_{\lambda} - \frac{1}{4} \Gamma_{\mu} \Gamma_{\lambda} \right) = \sum_{\substack{M=1\\M=1\\K=1}}^{N} \mathcal{E}_{\mu} \mathcal{E}_{\lambda} - \frac{1}{4} \mathcal{E}_{\lambda} \Gamma_{\mu} \right) = \sum_{\substack{M=1\\K=1}}^{N} \mathcal{E}_{\mu c} \left( \sum_{\substack{M=1\\X\neq\mu}}^{2} \mathcal{E}_{\lambda} \right), \qquad ,$$

$$\sum_{\substack{M=1\\K\neq\mu}}^{N} \mathcal{E}_{\lambda} \mathcal{E}_{\lambda} \right), \qquad ,$$

$$(4)$$

In the next section we present various correlation coefficients, which can be worked out using relations (4).

#### III. CORRELATION COEFFICIENTS

The correlation coefficient  $(\mathbf{T}_{\mu},\mathbf{T}_{\lambda})$  between two total widths  $\mathbf{\Gamma}_{\mu},\mathbf{T}_{\lambda}$  of the S-matrix is given by

$$(\mathbf{\Gamma}_{\mathcal{M}},\mathbf{\Gamma}_{\mathcal{A}}^{\prime})^{-1} = -(\mathcal{N}-1)^{-1}$$
(5)

This is the same as the one which is obtained for the case of non interfering resonaces. Thus unitarity constraint does not enhance the correlation between total widths. The same is not true for the correlation coefficient between  $\xi_{\mathcal{M}}$ ,  $\xi_{\mathcal{A}}$ . It is now given by

$$C_{\mathcal{E}_{\mathcal{M}},\mathcal{E}_{\mathcal{A}}} = -(N+I)^{-1} - \frac{1}{2} \left[ (N+I)(N-I) \right] < \frac{\Gamma_{\mathcal{M}}}{\sum \lambda} - \langle \Gamma_{\mathcal{M}}^{2} \rangle + \frac{1}{2} (N+I)^{-1} \frac{\langle \Gamma_{\mathcal{M}}^{2} \rangle}{\langle \mathcal{E}_{\mathcal{M}}^{2} \rangle} + \frac{1}{2} N \left[ (N+I)(N-I) \right] < \frac{\Gamma_{\mathcal{M}}}{\sum \lambda} + \frac{1}{2} \langle X_{\mathcal{M}_{\mathcal{C}}}^{2} \rangle < \frac{\kappa_{\mathcal{M}_{\mathcal{C}}}}{\langle \mathcal{E}_{\mathcal{M}}^{2} \rangle} It is easy to see from expression (6) that the correlation$$

coefficient  $C_{\xi_{\mu},\xi_{\lambda}}$  becomes  $-(N + 1)^{-1}$  for the non-interfering (6) resonances, as it should be.

We next give general expressions for the transmission coefficient and the average corss section<sup>4</sup>.

#### IV. TRANSMISSION COEFFICIENT AND AVERAGE CROSS SECTION

To analyse the heavy ion cross sections one needs expressions which are valid for all values of the ratio of the average width to the average spacing. The transmission coefficient  $T_c$  is given by

$$T_{c} = 1 - \exp\left(-2\pi \langle \Gamma_{vc} \rangle / D\right)$$
(7)

For small values of  $\pi < T_{\mu c} > / b$  it reduces to its usual form

$$T_{c} = \frac{2\pi \langle \Gamma_{\mu} \rangle}{D}$$

The average cross section  $\langle \epsilon_{cc'} \rangle$  is given by

$$\langle \mathcal{C}_{cc'} \rangle = \frac{\pi}{h_c^2} \left[ 1 - \exp\left(\frac{\left[l_n\left(1 - \overline{l_c}\right)\right] \left[l_n\left(1 - \overline{l_c'}\right)\right]}{\left[l_n\left(1 - \overline{l_c}\right) + l_n\left(1 - \overline{l_c'}\right)\right]}\right) \right]$$
(8)

which should be compared with the Hauser-Feshbach expression<sup>5</sup>

$$\left\langle \frac{HF}{6_{cc'}} \right\rangle = \frac{T}{k_c^2} \frac{\overline{T_c} \overline{T_c'}}{\overline{T_c} + \overline{T_c'}}$$
(9)

which is derived for small values of T ,  $T_{c'}$  . In expression (8), (9)  $k_c$  denotes the wave number in channel c .

ω

If one does not take the effects of nnitarity into account, then one can derive<sup>6</sup> an expression for the characteristic function of the total fission width. In this section we would like to give the joint distribution of  $\boldsymbol{\epsilon} = |\boldsymbol{\epsilon}_1 - \boldsymbol{\epsilon}_2|, \boldsymbol{\Gamma}_1, \boldsymbol{\Gamma}_2$  for the case of two channels and two resonances which takes fully into account the unitarity constraint. This distribution is given by

$$P(\epsilon, \Gamma_{1}, \Gamma_{2}) = (8 < s^{2})^{-1} 8[(\Gamma_{1} + \Gamma_{2}) - 2(D_{c_{1}} + D_{c_{2}})]$$

$$[(\Gamma_{1}, \Gamma_{2} - 4 D_{c_{1}} D_{c_{2}})[\epsilon^{2} + (D_{c_{1}} - D_{c_{2}})^{2}]]^{-\frac{1}{2}}[4 \epsilon^{2} + (\Gamma_{1} - \Gamma_{2})^{2}]$$

$$exp[(-\frac{1}{4}, \frac{\pi}{\langle s \rangle^{2}})(\epsilon^{2} + \Gamma_{1}, \Gamma_{2} - 4 D_{c_{1}} D_{c_{1}})] (10)$$

where  $Dc_1$ ,  $Dc_2$  are the eigenvalues of the correlation matrix, the diagonal elements of which give the variances of  $\lambda_{\lambda c}$  and the off-diagonal element is related to the correlation of  $\lambda_{\lambda c}$ , and  $\lambda_{\lambda c$ 

The distribution of the total width 
$$\Gamma'$$
 is obtained by integrating  
over  $\mathcal{L}$  and one of the widths in expression (10). It turns out to be  
$$P(\Gamma) = (\mathcal{M}_{3} \pi^{-1})(e \rtimes \mathcal{M}_{1}) \left[ \Gamma'(z - \Gamma) - \mathcal{M}_{2} \right]^{-1/2} \left\{ \mathcal{M}_{1} \mathcal{M}_{3}^{-1} \left[ \mathcal{K}_{1}(\mathcal{M}_{1}) + \mathcal{K}_{6}(\mathcal{M}_{1}) \right] - \mathcal{M}_{2} \right]^{-1/2} \left\{ \mathcal{M}_{1} \mathcal{M}_{3}^{-1} \left[ \mathcal{K}_{1}(\mathcal{M}_{1}) + \mathcal{K}_{6}(\mathcal{M}_{1}) \right]^{-1/2} \right\}$$
  
here  $\mathcal{M}_{1} = \left( \left[ \mathcal{T} / \mathcal{K}_{2} (\mathcal{M}_{1}) \right]^{-1/2} \left( \left[ \mathcal{L}_{2} - \Gamma \right]^{-1/2} \right]^{-1/2} \left( \left[ \mathcal{L}_{2} - \Gamma \right]^{-1/2} - \mathcal{M}_{2} \right]^{-1/2} \right)^{-1/2}$ 
  
 $\mathcal{M}_{3} = \left( \left[ \left[ \mathcal{T} / \mathcal{K}_{2} (\mathcal{K}_{2})^{2} \right]^{-1/2} \right]^{-1/2} \left( \left[ \mathcal{L}_{2} - \Gamma \right]^{-1/2} \right)^{-1/2} \right)^{-1/2}$ 

This should be compared with the total dimensionless width of the R-matrix which is given by

$$P(y) = \pi^{-1} [y(2-y) - \mu_2]^{-\frac{1}{2}} (12)$$

We find from expressions (11) and (12) that the effect of unitarity is to broaden the width distribution.

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DISCUSSION

- J.J. Schmidt : To my knowledge the non-diagonal elements of  $OL^{-1}$ die out with increasing number of channels, because they are essentially products of reduced channel widths which can be assumed uncorrelated and, because positive and negative signs of these widths have equal probability of occurrence (at best, one cannot a priori decide about the sign; random sign approximation); Where did this random sign approximation enter into your calculations?
- N. Ullah : No random sign approximation is used in the derivation of our expressions. If one uses this approximation, one has to examine higher terms in the expansion of  $A_{\lambda_{i}^{i}\ell}$  (see for example, A. M. Lane and R.G. Thomas, Revs. Mod. Phys. <u>30</u>, 257 (1958)) before throwing them away.

# SESSION 3

Chairman : N. Cindro, IRBZ, Yugoslavia

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## PROMPT RADIATIONS EMITTED IN LOW ENERGY FISSION

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In this talk I shall present results of some recent work carried out at Trombay on prompt radiations emitted in fission and also briefly review some of the earlier work done here. There are two main reasons why the study of prompt radiations emitted in fission forms such an important part of fission physics. One reason is that because these radiations are emitted at different stages of the post-saddle part of the fission process they are perhaps the only probes available to learn about the post-saddle characteristics of the The different stages of emission of these radiations fission process. are shown schematically in Fig. 1. As the nucleus passes past the saddle stage it quickly breaks up into two fragments at scission in about  $10^{-20}$  sec. During the actual breaking process or near about it, on rare occasions, a very high energy particle, called the long range charged particle (LRCP), is emitted. The two nascent fragments produced get accelerated due to their mutual Coulomb repulsion to very large kinetic energies ( $\sim$  170 MeV) and each of these fragments has an excitation energy of about 14 MeV. A large fraction (  $\sim 70\%$ ) of this excitation energy is emitted in the form of 2 to 3 neutrons in about  $10^{-18}$  to  $10^{-14}$  sec and the remaining energy is carried away by a cascade emission of about 8 gamma rays in a time around  $10^{-10}$  sec. About 6% of these gamma transitions, mostly low energy transitions, are interhally converted in the K shell with the consequent emission of conversion electrons and characteristic K X-rays. After this stage the fragments produced being neutron-rich and unstable, undergo 2 to 3 beta decays and end as fission products. Thus, a study of these prompt radiations can provide information about these post-saddle The second reason is that the fission fragments stages of fission. are highly neutron rich and cover a large region of the nuclear periodic table. (Fig. 2). Hence by examining the spectra of radiations emitted from fission fragments of specific mass and charge very useful spectroscopic information on nuclei far from the line of  $\beta$  -stability could be obtained.

The earlier investigations dealt with the emission of prompt neutrons and gamma rays emitted in thermal neutron fission of  $U^{235}$ and were carried out using the reactor Apsara. The energy distribution of neutrons, emitted along the direction of motion of the light and heavy fission fragments, as well as the angular distribution of neutrons of different energy with respect to the light fission fragment direction were measured<sup>(1)</sup>. The two main results of this investigation were that (a) about 10% of neutrons are not emitted from moving fragments and (b) the energy spectrum of neutrons from each fragment is a linear superposition of various evaporation spectra corresponding to a linear distribution of temperatures upto a certain maximum temperature  $T_m(Fig. 3)$ . The angular distribution of gamma-rays with respect to the direction of selected light-fragments was measured<sup>(2)</sup> for two gamma-ray energy groups (Fig. 4) and an anisotropy of about 10-15% was observed suggesting<sup>(3)</sup> that fragments have a significant angular momenta correlated with their direction of motion.

During the last several years extensive investigations have been carried out on K X-ray emission in thermal neutron fission of  $U^{235}$  and spontaneous fission of  $Cf^{252}$  and in most of these studies, high resolution Lithium-drifted silicon detector systems were used. The yields of K X-rays from  $U^{236}$  fission fragments of specified masses were determined<sup>(4)</sup> in which the fragment masses were identified by recording the kinetic energies of both the fragments and in Fig.5 these results are compared with the previously measured<sup>(5)</sup> K X-ray yields from  $Cf^{252}$ fission fragments. The yield of K X-rays from each fragment depends on the average probability of creation of a vacancy in the K electronic shell which in turn depends on the number of transitions and the energy and multipolarity of each of these transitions. From Fig. 5 it is seen that  $U^{236}$  fission fragments extend the data for fragment masses less than 100 and that elsewhere, although the gross features of X-ray yields from  $Cf^{252}$  and  $U^{236}$  fission fragments are similar, for masses greater than 144 the abrupt increase in K X-ray yield observed in the former case was not observed in the latter case. These results were interpreted on the basis that the X-ray yield depends not only on the characteristics of low-lying states but also on the initial spin of the These studies were extended<sup>(6)</sup> by determining the K X-ray fragments. yields emitted in the time range of up to 1/4 sec by fragments of specified nuclear charges in thermal neutron fission of  $U^{235}$  (Fig. 6). The observed increase in X-ray yield as one moves away from the closed shell region of N=50, Z=50 and N=82 have been qualitatively correlated with the expected properties of the low-lying states in these neutron-rich nuclei.

In the study of K X-ray emission it is important to know the average X-ray emission times and their dependence on the nuclear This is because the times of emission of K X-rays following species. fission are determined by the life times of the nuclear transitions being internally converted, since, once a vacancy is created in the K-shell, the atomic transitions giving rise to K X-rays take place very quickly ( ~  $10^{-16}$  sec). In a recent investigation we have determined<sup>(7)</sup> the average K X-ray emission times from  $U^{236}$  fission fragments by detecting the X-rays in the two cases of the emitting fragment moving towards and away from the X-ray detector in the time ranges of 110  $\mu$  sec and 1 $\mu$  sec after fission. (Fig. 7). It was observed that for fragments in the region of N = 50 shell the X-ray emission times are of the order of 0.1 nsec and the yields of K X-rays are very low suggesting the presence of widely spaced levels which give rise to faster decay and low internal conversion. For a few fragment charges 43Tc, 52Te and 58Ce comparatively larger emission times were observed. A predominantly delayed component of K X-ray was observed in the case of Tellerium which is consistent with the isomeric gamma-transition observed recently by John et  $al^{(8)}$ . For the remaining fragment nuclei the average emission times were found to be around 1 nsec which shows the absence of any intense isomeric transitions of long half life in these nuclei.

Another important aspect of X-ray emission in fission is the multiplicity of K X-ray emission from fragments of specified atomic number. Although for most of the fragment nuclei the yield of K Xrays per fragment is appreciably less than one, it is possible that in certain cases more than one X-ray is emitted in a cascade. One can define a X-ray emission distribution function  $f_{\tau}(n)$  which represents the fraction of events in which n X-rays are emitted in a cascade from fragments of charge Z. The average yield of K x-rays per fragment is then equal to the first moment of  $f_{v}(n)$ . In a recent investigation<sup>(7)</sup> we have determined both the first and second moment of  $f_r(n)$  for Cf<sup>252</sup> fission fragments and this was accomplished using two cooled Si(Li) detectors to measure the energies of coincident K x-rays emitted from fission fragments. Fig.8 shows the normal K x-ray yield per fragment as well as the yield of additional K X-rays per fragment when one K X-ray is known to have been already emitted from  $Cf^{252}$  fragments of specified atomic numbers. The observation that for almost all fragment atomic numbers the probability of an additional X-ray emission is significantly large suggests that X-ray emission is, in general, a cascade process. In the case of some fragment charges the yield of additional x-rays is greater than unity indicating a fairly large probability of emission of more than two K X-rays in these cases. The same feature is evident from the second moment of  $f_{z}(n)$  shown in Fig. 9. This further implies that a large fraction of fission events do not lead to x-ray emission.

As was mentioned earlier, the emission of LRCP is perhaps the first of the series of radiations emanating from the fission process. It is known that rarely (once in about 500 fissions) a LRCP, usually an alphaparticle, is emitted. On the basis of the strong 90<sup>0</sup>-peaked angular correlation of the LRCP and fission fragments, the former were assumed<sup>(9)</sup> to be emitted in a region between the nascent fragments before the latter acquire any velocity. However the exact mechanism of emission of these particles is not known and in view of the stage of fission at which they are believed to originate a detailed study of this radiation should provide invaluable information about the last stage of fission process. At Trombay we have investigated several aspects of the emission of LRCP in fission. One of these is the dependence of the yield of LRCP on the initial excitation energy of the fissioning nucleus. Fig. 10 shows the yield of LRCP per  $10^3$ fissions observed (10, 11) in the fission of  $U^{235}$  induced by thermal, 2-. 3- and 4 MeV neutrons. These measurements were made using the 5.5 MeV Van de Graaff accelerator. It was observed that LRCP emission probability is not sensitively dependent on the initial excitation

energy of the fissioning nucleus. If the probability of LRCP emission depends on the energy available for LRCP emission and on the configuration of the fissioning nucleus during the very last stages, as is assumed in some models, the data suggests that the energy available for LRCP emission as well as the shape of scissioning nucleus are insensitive to the initial excitation energy of the compound nucleus. In addition, the energy spectrum of LRCP emitted in 3 MeV neutron fission was found (11)to be similar to that emitted in thermal neutron fission and the average energy of LRCP in the two cases were equal within (+.4 MeV) thereby indicating a rather weak dependence of the average energy of LRCP on the initial excitation energy of the fissioning nucleus (Fig. 11). Measurements have also been made of angular anisotropy of both the LRCP(11) and fission fragments(12) with respect to the incident 3 MeV neutron direction and it was observed that the former is forward-peaked (with an anisotropy  $\sim$  32%) whereas the latter is 90°-peaked (with an anisotropy  $\sim 10\%$ ). These observations were found to be consistent with the predictions of the evaporation  $model^{(13)}$  of LRCP emission in fission.

At Trombay some studies of emission of prompt radiations in LRCP-accompanied spontaneous fission of  $Cf^{252}$  have been made. The yield and energy spectrum of K X-rays emitted by  $Cf^{252}$  fission fragments in coincidence with LRCP have been measured<sup>(14)</sup> and compared with that in the normal binary fission (Fig. 12). The observation of 14-25% increase in the X-ray yield in the former case compared to the latter is consistent with the assumption that these particles originate from the fissioning nucleus as a whole and not at the expense of nucleons from either of the fragment groups alone. In a measurement<sup>(15)</sup> of yield of gamma-rays from normal binary fission and LRCP-accompanied fission of  $Cf^{252}$  it was observed that about 16% more gamma-rays are emitted in the former case compared to that in the latter case suggesting a slight change in the gamma-de-excitation process of fragments produced in the two cases.

Recently we have measured (16) the energy spectrum of LRCP emitted at four average angles  $(90^{\circ}, 46^{\circ}, 27^{\circ} \text{ and } 11^{\circ})$  with respect to the fission fragment direction in thermal neutron fission of  $U^{235}$  and some of these spectra are shown in Fig. 12. From an analysis of these data the angular coreelation of LRCP and fission fragments and its dependence on the energy of LRCP were determined. In Fig. 14 is shown the width  $\sigma$  ( $\Theta_{\alpha}$  f) of the angular correlation versus the energy of LRCP. It was found that the angular correlation (peaked at  $90^{\circ}$ ) has nearly the same width for all energies from 11 to 20 MeV and then it starts broadening until at very high LRCP energies it becomes almost isotropic or even a distribution having a minimum at 90°. Similar results were observed in an earlier investigation (17) on LRCP The last observation suggests the need for considering the emission. dynamic effects due to the possible motion of nascent fission fragment on LRCP emission. It was observed that for the overall LRCP energy spectrum, many more LRCP are emitted near forward angles  $(\overline{\Theta}_{44} = 11^{\circ})$ 

than expected on the basis of a gaussian distribution which fits the data for the angular region  $(30 - 90^{\circ})$ . This suggests that even at forward angles there exists an appreciable probability of LRCP emission which therefore seem to be emitted from regions of the fissioning nucleus other than the neck region connecting the nascent fragments.

Although the bulk of gamma radiation is emitted in a time of the order of  $10^{-10}$  sec, a number of isomeric gamma transitions having half lives ranging from  $10^{-8}$  to  $10^{-4}$  sec have been reported. These isomeric transitions are of interest from the point of view of the spectroscopic information on these neutron-rich high-spin fission fragments. Recently investigations on isomeric transitions from Cf<sup>252</sup> fission fragments have been started at Trombay. Fig. 15 shows the spectrum of delayed gamma rays emitted between 300 and 800 nsec after fission from  $Cf^{252}$  fission fragments which was measured<sup>(18)</sup> with a 30 c.c. Ge(Li) detector and the assignment of fragment mass number to the various peaks has been made by comparison with the recent data of John et  $al^{(8)}$ . Fig. 16 shows the spectrum of prompt gamma rays recorded in coincidence with these delayed gamma rays where the assignment of most probable fragment masses giving rise the various prompt gamma-ray peaks have been made by comparison with the data of Watson et  $al^{(19)}$ . It may be noted that although the delayed gamma rays are emitted from fragment masses in the regions 96-99, 108-110 and 133-137, in the coincident gamma spectrum only the lines from the 108-110 mass group and their complementary fragments are observed.

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Fig. 1: Radiations emitted at the different stages of Fission







Fig. 3: Neutron emission spectra from light and heavy fragments in  $U^{235}(n, f)$ .



Fig. 4: Angular distribution of gamma rays in  $U^{235}(n, f)$ 

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Fig. 5: Number of K x-rays per fragment vs  $M_f$  in  $U^{235}(n, f)$ 



Fig. 6: Number of K X-rays per fragment vs Z in  $U^{235}(n, f)$


-101-







-103-



-104-



Fig.11: Energy Spectra of LRCP in thermal and 3 MeV neutron fission of U<sup>235</sup>.









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DELAYED GAMMA RAYS FROM CET SPONTANCOUS FISSION

Fig. 15: Energy spectrum of gamma rays emitted between 300 to 800 nsec after fission in  $C\ell^{352}$  fission fragments.



# PROMPT GAMMA RAYS IN COINCIDENCE WITH DELAYED GAMMA RAYS COMING 300-800 nano secs. AFTER FISSION

Fig. 16: Prompt gamma rays emitted in coincidence with those emitted in 300-800 usec in Cl<sup>252</sup> spontaneous fission DISCUSSION

- P. Ribon : For which purpose did you measure the prompt gamma rays in coincidence with the delayed gamma rays emitted in isomeric transition of fragments?
- D. M. Nadkarni : We measured the spectrum of prompt Y -rays emitted in coincidence with the delayed (300 n sec. -800 n sec) Y -rays from Cf<sup>252</sup> fission fragments with a view to look for Y -ray transitions which populate the level which is responsible for the isomeric transition. It is possible, in principle, to build up a level sequence for those fragments which emit delayed Y -rays. It is found that a relatively small number of fragment nuclei, concentrated in a few regions of A, emit delayed gamma rays (300 nsec-800 nsec). At the moment, these measurements are in progress.
- **R.F. Taschek** : What were the properties of the 10% of the neutrons which were not associated with moving fragments and how was it established that they were not correlated with moving fragments?
- D. M. Nadkarni : In answering your second question, it was found that the experimentally observed angular distribution of neutrons, with respect to selected region of light fragments, shows a lesser correlation than that computed on the assumption of isotropic evaporation from the moving fragments. The extent of deviation from that calculated was particularly large for high energy neutrons. This discrepancy could be removed if it is assumed that a small fraction ( $\wedge 10\%$ ) of these neutrons are not emitted from the moving fragments. As to your first question, these 10% neutrons were found to be emitted with an evaporation like spectrum having an average temperature of about 1.6 Mev. It has been suggested these are emitted just before or at scission before the fragments have acquired their full velocity by a mechanism similar to that proposed by Fuller or, alternatively, by a evaporation process during saddle to scission descent.

J.	Rowlands	:	Have you measured the energy spectrum of the 10% of neutrons emitted before scission?
D	. M. Nadkarni	2	Yes. The energy spectrum of these 10% of neutrons emitted before the fragments acquire their full velocity were obtained indirectly. This was done by finding the fraction of neutrons that must be deducted from the experimental angular distribution in order to get an agreement between the observed angular distribution and that compu- ted on the assumption of isotropic emission from fully accelerated fission fragments. This fraction was determined for neutrons of different energies and thus the spectrum of neutrons emitted before scission was determined.

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## RECENT STUDIES ON PROMPT NEUTRON AND GAMMA RAYS FROM FISSION

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#### 1. INTRODUCTION

The paper gives a review of some recent experiments on prompt neutrons and gamma-rays from fission. Special attention is paid to  $\overline{V}$ -values i.e. the average number of prompt neutrons emitted in fission and the energy spectra of the fission neutrons. Furthermore, a summary is given of a measurement by ALBINSSON (1971a) of prompt gamma-rays from fission.

Studies of prompt neutrons and gamma-rays give besides of nuclear data for reactor physics and shielding calculations also fundamental information about the fission process e.g. about energetics in fission and about type of excitation and decay modes of the neutron-rich highly excited fission fragments. However, a better understanding of the fission process is needed to interpret many of the results experimentally observed in the measurements of fission neutrons and gamma-rays.

## 2. ENERGY DEPENDENCE OF $\overline{\nu}$

The dependence of  $\vec{\nu}$  on the energy of the incident neutron has recently been measured with good accuracy for the neutron induced fission of <sup>235</sup>U in the whole energy region from thermal to 15 MeV (Ref. see figure 1). Therefore, it has been used here to illustrate general trends and problems in the measurements of  $\vec{\nu}$  in the different energy regions and for different fissioning nuclei.

From 2 MeV upto about 14 MeV the  $\vec{v}$  energy dependence is with good approximation a straight line with a slope corresponding to 0.14 n/MeV. This slope is expected if all added energy goes into the production of neutrons.

However, around 6 and 14 MeV first and second chance fission start to be energetically possible, i.e. the nucleus can undergo fission after separation of one or two neutrons. The expected steps in  $\overline{y}$  are about 0.1-0.2 and are also spread out over several MeV. The experimental results by SOLEILHAC et al. (1969) do also indicate a weak step at about 6 MeV (figure 1).

If one compares  $\overline{\nu}$  -values for different isotopes of uranium in the energy region above 2 MeV, the  $\overline{\nu}$ -values all fall within 3% of the same line (figure 2). For other elements the  $\overline{\nu}$ -values are quite different. Thus the Z-value seems to be the critical parameter.

Several experiments have indicated a structure in the energy dependence of  $\overline{\nu}$  for 235U below 2 MeV (figure 1). Steps at 400 and 1000 keV of about 1-3% in  $\overline{\nu}$  have been reported by SOLEILHAC et al. (1970), SAVIN et al. (1970) and NESTEROV et al. (1970), while BOLDEMAN et al. (1970) observed a linear energy dependence with a slope of about 0.11 n/MeV. Earlier measurements (see e.g. FILLMORE (1968)), also give a different slope (about 0.10 n/MeV) of the linear fit below 2 MeV than above this energy. A better theoretical understanding of the fission process is certainly needed to solve the problems with the structure in this energy region.

For even-even isotopes like  $^{232}$ Th,  $^{236}$ ,  $^{238}$ U accurate measurements have been made of  $\overline{\nu}$  close to the fission threshold. In this region the fission cross sections show a structure depending on subthreshold fission. A linear energy dependence of  $\overline{\nu}$  is observed except for  $^{232}$ Th where  $\overline{\nu}$  increases close to the threshold (figure 3).

In the fission resonance region recent measurements have been made on  $\overline{\nu}$  of <sup>235</sup>U for different resonances by WEINSTEIN et al. (1969), and RYABOV et al. (1970) (figure 1). Small changes in  $\overline{\nu}$  (1-2%) were observed going from one resonance to another. However for the induced fission of <sup>239</sup>Pu changes of 3-5% in  $\overline{\nu}$  for different resonances were reported by the same groups. WEINSTEIN et al. observed the same  $\overline{\nu}$ -values for earlier assigned 0<sup>+</sup>resonances and these  $\overline{\nu}$ -values were about 3% higher than the corresponding values for the 1<sup>+</sup>-resonances. Recently, the measurements on <sup>239</sup>Pu were repeated by WESTON et al. (1971). Contrary to the earlier measurements this group could not see any differences within  $\frac{1}{2}$ -1% in  $\overline{\nu}$  for different resonances.

The  $\overline{\nu}$ -value for the spontaneous fission of <sup>236</sup>U has recently been measured by CONDE and HOLMBERG (1971). In the energy dependence diagram (figure 1) this value corresponds to a point at -6.4 MeV, which is equal to the neutron binding energy in <sup>236</sup>U. Compared with the thermal value the change in  $\overline{\nu}$  is  $\Delta \overline{\nu} = 0.51$  and thus if the change is expressed in neutrons per MeV  $\Delta \overline{\nu}/\Delta E = 0.08$  n/MeV. This value of  $\Delta \overline{\nu}/\Delta E$  is low compared to the corresponding value observed above thermal neutron energy. A similar result was also obtained for <sup>240</sup>Pu and <sup>242</sup>Pu where comparisons can be made with neutron induced fission of <sup>239</sup>Pu and <sup>241</sup>Pu (CONDE et al. (1968)).

#### 3. FISSION NEUTRON SPECTRA

The experimental fission neutron spectra are very well fitted to distributions of the Maxwellian type  $N(E) \sim \sqrt{E} e^{-E/T}$ . This general type of spectrum is expected if one assumes that the neutrons are emitted from moving fragments.

Another type of spectrum was calculated by WATT (1952) assuming a Maxwellian type of spectrum in the centre-of-mass system which results in the following type of spectrum in the laboratory system

$$N(E) \sim e^{-E/T} \sinh \frac{2}{T} \sqrt{EE}_{f}$$
,

where  $E_f$  is the average kinetic energy per nucleon of the fragments. As pointed out by TERRELL (1965) the Watt spectrum does not fit quite well if the energy  $E_f$  is given the actual average value, about 0.75 MeV. This would be an indication that the centre-of-mass spectrum is broader than a Maxwellian distribution and should be better represented by the sum of two Maxwellian distributions. This in turn would give the laboratory spectrum as the sum of perhaps four Watt distributions and the result of which would be close to a Maxwellian distribution.

At higher neutron energies, around 6 and 14 MeV, it is energetically possible, that the fission occurs after the evaporation of one and two neutrons, respectively. The neutron energy spectra measured for incident neutron energies above 6 MeV will therefore have a low energy contribution caused by inelastically scattered neutrons (HANNA and CLARKE (1961)).

The existence of so called scission neutrons was suggested by BOWMAN et al. (1962) in order to explain their experimental results of fission neutron angular distribution from  $^{252}$ Cf. They suggested that the scission neutrons are emitted isotropically from the fissioning nucleus at the moment of scission. BOWMAN et al. estimated the number of scission neutrons for  $^{252}$ Cf to be approximately 0.4 neutrons/fission. MILTON and FRASER (1965) estimated that the number of scission neutrons in the thermal fission of  $^{233}$ U and  $^{235}$ U might be as many as 30%. In the fission neutron energy spectra no clear evidence for the existence of scission neutrons have been reported.

TERRELL (1965) also derived from evaporation theory a semiempirical relation between the average energy of fission neutrons,  $\tilde{E}$ , and the average number of neutrons emitted per fission  $\tilde{V}$ 

 $\overline{E} = 0.74 + 0.65 (\overline{\nu} + 1)^{\frac{1}{2}} (MeV)$ 

mostly very heavy. All these problems have probably hindered a faster progress of the knowledge of prompt fission gamma radiation from the thermal-neutron induced fission of  $^{235}$ U, and so far most studies have concerned gamma-rays from fragments formed in the spontaneous fission of  $^{252}$ Cf.

Measurements of the prompt gamma radiation have been made recently with Ge(Li) detectors on  $^{252}$ Cf fission (CHEIFETZ et al. (1970), WILHELMY et al. (1970)), and also on  $^{235}$ U fission (HORSCH et al. (1969)). The experimental technique has been improved considerably during the last five to seven years, for instance by the introduction of Si(Li) detectors, with which K X-ray energy spectra from the fragments can be recorded in a simple way. These X-rays are formed through conversion of the prompt gammarays, and studies of them can therefore yield complementary data to the knowledge of the prompt gamma radiation. Recently experiments have also been performed with X-rays and gamma-rays in coincidence (RUEGSEGGER et al. (1970)), and so it is now possible to determine a few of the lower gamma-ray cascades in some mass regions.

The main difference between the experiment performed by Albinsson and most others reported so far is the use of a collimator to select different time intervals after the fission event. The study follows in basic principle the ideas outlined by JOHANSSON (1964). With the collimator (see figure 4) it is possible to study the time distribution of the gamma radiation, such as decay curves of the integral radiation from all fragments or from certain fragments, and also to record gamma-ray energy spectra from certain fragments during different time intervals after the fission event. This means that gamma radiation of different half-lives as a function of fragment mass can be studied (figure 4). Another interesting parameter in these investigations is the total fragment kinetic energy. This measurement was reported separately (ALBINSSON (1971b)). The data acquisition system was a two-parameter analyzer, so that there is no possibility to add more parameters to the two whose interrelations were studied: gamma-ray energy and fragment mass.

It is sometimes possible to use more than one existing model in nuclear physics for the interpretation of fission data. One often used model is the collective model, the reason being of course that the fission process is really a collective, many-particle process. Some extra problems arise, however, as the fragments, just after their formation, are very neutron-rich, and it may be difficult to compare results of fission gamma-ray studies with those of other nuclear reactions. Of great interest for purposes of comparison are the data from prompt neutron emission studies, and quite a lot of the discussion from these studies can be adopted for the interpretation of prompt fission gamma radiation. Unfortunately the situation surrounding prompt neutron emission is far from clear, and The experimental values for the thermal fissions of  $^{235}U$  and  $^{239}Pu$ and for the spontaneous fission of  $^{252}Cf$  seem to indicate a more rapid change in  $\overline{E}$  with  $\overline{V}$  than given by the above relation (SMITH (1970), JEKI (1971)).

There exists through the years a lot of measurements of fission neutron spectra using different experimental techniques (see e.g. CONDE and DURING (1965)). A typical feature for many of the measurements is that one claims a very good accuracy compared to the spread in the Maxwellian temperatures obtained from different experiments. This might be due to systematic errors which have not been correctly accounted for (JEKI et al. (1971)). The difficulties might be, just to point at some of them, to convert a continuous time-of-flight spectrum to an energy spectrum corrected for elastic and inelastic neutrons, delayed gamma-rays etc. or they might be to convert a continuous pulse-height spectrum to an energy spectrum, taking into account the response function, efficiency, multiple scattering etc.

Especially the fission neutron spectrum for the thermal fission of  $^{235}$ U has been deduced recently from measured spectralaverage cross sections utilizing activation detectors (GRUNDL (1968), FABRY (1967)). The results from these measurements give about 15% higher E-values than earlier microscopic measurements and are not strictly described by Maxwellian distributions (MCELROY (1969), SMITH (1970)). The reason for the difference between the macroscopic and microscopic data is not quite well understood but there might be problems associated with the calculation procedure and the differential cross sections used in the first type of measurement.

## 4. PROMPT FISSION GAMMA-RAYS

A summary is given of a measurement by ALBINSSON (1971a) of prompt gamma-rays from the thermal fission of  $^{235}$ U made at the reactor R2, Studsvik.

It is of interest to study prompt gamma radiation from fission fragments for two reasons. Firstly, a knowledge of this radiation should be of value for any detailed theory of the fission process, and secondly, it can provide information for designing shielding around a reactor.

The gamma-ray energy spectra are very complicated owing to the many nuclei (fragments) which emit this radiation. Furthermore, these nuclei can start emitting their radiation from states in a rather wide energy range depending on the way in which the fragments were formed. Studies of gamma rays from fragments formed in a fission process induced by neutrons from a reactor often involve experimental difficulties, because the background at a reactor is The gamma radiation studied by Albinsson is the part which is characterized as prompt. Somewhat arbitrarily the radiation is usually divided into two parts, namely a prompt part whose components have half-lives shorter than 1 ns, and a delayed part with longer half-lives. This division is further justified by the fact that the experimental techniques for study of the two parts differ and that the properties of the radiation in the two cases show a distinct difference (JOHANSSON (1964)).

In the experiment by Albinsson a special technique was adopted, namely that of using time-of-flight discrimination between the prompt neutrons and the fission gamma radiation. This was done by placing the gamma detector about 70 cm from the fission foil. This technique has not been used extensively so far, probably because of the small solid angles involved and, as a consequence, the low counting rates in the gamma detector.

## CONCLUSIONS

Studies have been performed of the gamma radiation from fission fragments in slow-neutron induced fission of  $^{235}U$ . Gammaray energy spectra were recorded as function of mass and time after fission (figure 5, 6, 7). The main conclusions from the investigation may be summarized as follows:

1. The gamma-ray energy spectra vary in shape with time after fission.

2. The gamma-ray energy spectra vary in shape with mass within each time interval, the variation being stronger the later the time interval studied.

3. When a time interval is selected in which half-lives of the radiation of 50 ps and more are enhanced, there is strongly massdependent yield of 1200 keV photons, which might come late in a cascade of gammas.

4. There is a very strong dependence on mass of the yield of photons of a few hundred keV in the time interval in which half-lives of 50 ps and more are enhanced.

5. The relationship between the average gamma-ray energies and the associated half-lives gives a strong indication that the bulk of prompt photons from fission fragments are of the quadrupole type. 6. Yield curves of the photons as function of mass can be interpreted on the basis of a model including the property of the varying resistance to deformation of the nuclei, depending on whether they contain nucleons of magic numbers or not.

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Fig. 1.  $\sqrt[n]{v}$  energy dependence for the neutron induced fission of  $^{235}$ U. In the upper left figure  $\sqrt[n]{v}$  is given for different resonances in the eV-range while the upper right figures give the energy dependence in the keV-range from four different experiments.





Fig. 3. Number of prompt neutrons emitted per fission, v, for neutron induced fission of <sup>232</sup>Th as a function of incident neutron energy. The open circles are from Condé and Holmberg (1965), the filled circles from Prokhorova and Smirenkin (1966), the open triangle from Condé and Starfelt (1961), the filled triangles are data of Mather et al. (1965), the open square is from Smith et al. (1959), and the filled squares are data of Kuzminov (1959).

The upper curves represent the energy dependence of the anisotropy of fission fragments and the total fission cross section of  $^{232}$ Th.









Fig. 5. Gamma-ray energy spectra from a number of different messes associated with half-lives of 50 ps and more.



Fig. 6. Gamma-ray energy spectra of the radiation with a halflife of about 20 ps for a number of different masses.

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Fig. 7. Gamma-ray energy spectra of the radiation with halflives shorter than 10 ps for some different masses.

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- A.H.W. Aten : 1. Are not the maxima and minima in Solheilac's curve for  $\tilde{y}$  of  $^{235}$ U+n. suspect, because practically all of them consist of a single point?
  - 2. If  $\overline{y}$  for  $^{235}$ U+n. is represented by a <u>curved</u> line is it not possible to draw such a line in such a way that all  $\overline{y}$  values lie on it, and also the  $\overline{y}$  value for spontaneous fission of  $^{236}$ U at an energy value of -6.4 Mev?
- H. Conde': 1. A least square fit to the experimental data gives a curve with two steps at approximately 0.4 and 1.1 MeV.
  - 2. Of course a curved line can be drawn through all the points and be used for data needs, but so far the fission theory only predicts a straight line dependence all the way down to zero excitation energy.
- S. Cierjacks : Have you carried out a statistical significance test? Looking on your data it seems to me that the data with these uncertainties do not show a significant deviation from a straight line.
- H. Conde<sup>i</sup> : I referred to the data by Soleilhac et al. and they have carried out least square fits to their data which show a nonlinear dependence.
- J.J. Schmidt : (Comments) : The statistically best available measurement of  $\overline{\mathcal{V}}$  (E<sub>n</sub>) on U<sup>235</sup> in the same energy range in which Soleilhac et al. measured, is due to Boldeman, from Lucas Heights, Australia. It does not show the deviation from a linear energy dependence as observed by Soleilhac. As a consequence, weighted least squares fitting of all available  $\overline{\mathcal{V}}(E_n)$  data for U<sup>235</sup> as carried out by the Nuclear Data Section of the IAEA in the last weeks does not yield any significant structure in  $\overline{\mathcal{V}}(E_n)$ (U<sup>235</sup>) and gives very nearly a straight line.

# RADIATIONS EMITTED IN FISSION

Pierre RIBON

## SUMMARY -

We briefly examine the properties of the various particles which are emitted during or after the fission process. Their emission can occur at three different stages :

1- at the scission time. They are mainly  $\alpha$  particles and other light particles; the results for these emissions are numerous and coherent, and allow to obtain informations on the scission configuration. On the contrary, the neutron emission is not well enough known.

2- by the fission fragments. These ones desexcited themselves mainly by neutron emission; nevertheless, if they have hight spin values ( $I \sim 15$ ), the  $\gamma$ -ray emission is expected to compete strongly with the neutron emission. But some very recent results give lower spin values ( $I \sim 7$ ).

3- by the fission products. The delayed neutron precursors are now quite well known, and the interest is turning more and more upon their energy spectra. The calculations which have been done to fit the previously known spectra does not describe the structure of the most recent results. The radiations emitted during or after the fission processus can be classed in 3 categories according their time of emission :

1- the scission particles, which are emitted at the time of the scission event, i.e. in less than  $10^{-18}$  s.

2- the radiations emitted by the fission fragments, which are highly deformed and excited nucleus; the characteristic time of this emission varies from a few  $10^{-14}$  s to about  $10^{-11}$  s.

3- the radiation emitted by the fission products, which are radioactive nucleus lying above the  $\beta$  stability line.

It has been treated of these radiations at the two conferences on "Physics and Chemistry of fission" [1, 2]. I will try to give a brief idea of the status of their knowledge, with emphasis on information obtained since the Vienna Conference [2].

## I - RADIATIONS EMITTED WHEN THE SCISSION PROCESS OCCURS.

I-1. The light charged particles are the best known of these radiations. The table I, a recent compilation by ASGHAR [3], gives the main results obtained up to now for the relative intensity, for the most probable energy and for the width at half maximum of the energy distribution. The general consensus that these charged particles are emitted at the time of scission and near the scission point is based on their angular distribution (which is markly peaked in a direction perpendicular to the fragments direction), on the variation of their angular distribution with the fragment mass ratio and, according to Halpern [4], on the non agreement of a statistical evaporation interpretation with the experimental yields.

It is generaly accepted that the properties of the fission fragments in the binary fission without the emission of light charged particle and of these fragments when this binary fission is accompanied by charged particles are the same. This interpretation is based on the similitude of the mass and energy distribution of the fission fragments and of the emitted neutrons, on the equality of the total kinetic energy, etc... In his review [5] Feather conclude that " the  $\alpha$  particles accompanied ternary mode develops out of the binary mode only when the total deformation excitation energy of the nascent binary fragments is, on average, some 10 MeV greater than the mean value for binary modes generally ".

The properties of these kinds of fission events are the best known when the light particles are  $\alpha$ . The properties of the other light charged particles are not so well known; nevertheless, Feather concluded that " with some slight reservation in respect to protons, it appears that the release process is of the same character for all charged particles ".

This reserve has been confirmed by results from NARDI et al [6] who show that the value of the correlation between the kinetic energy of the two fragments and the kinetic energy of the light particle may be 0 in the case of protons, while it is worth about - 0.4 for tritons or alphas.

They constitute a usefull tool to study the configuration of the 2 fission fragments at the scission point. Many trajectories computation have been performed [7 to 12]. But many quantities are involved in these calculations, such as:

- the initial position of the light particle
- its initial energy and angular velocity distributions
- the initial energy of the fragments and their initial distance
- the mass ratio R.

According the different authors, some of these parameters are fixed by some hypothesis, the others being adjusted to fit a set of experimental data - which is not always the same. As a result, it appears a great dispersion of the parameters: the initial energy of the  $\alpha$  particles varies from 0.5 MeV (FONG, 1970 - [12]) to 3 MeV (BONEH, 1967 - [7]) and the initial distance of the fragments lies between 20 fm [12] and 26 fm [7, 11].

But the definition of the initial time is ambiguous, and may explain a part of these discrepancies; for instance, the results are as if the initial time for FONG was earlier that for BONEH. Furthermore, KROGULSKI and BLOCKI [11] could describe the same data by the same methods and with the same hypothesis except for the scission configuration: they used successively separation distances of the fragments equal to 26 fm (elongated configuration) and to 20 fm (compact configuration).

I-2. <u>Neutron emission</u>: To our knowledge there are no new results since that Feather considered, in 1969, that " the most convincing evidence for the existence of these " central " neutrons comes from the pre 1965 investigation of BOWMANN and al [13] with  $^{252}$ Cf ".

Different experiments have concluded more or less directly to the existence of such a component, rather important for  $^{252}$ Cf ( $\sim 0.4$  central neutron per fission),  $^{236}$ U<sup>\*</sup>[14, 15],  $^{240}$ Pu<sup>\*</sup>[15],  $^{234}$ U<sup>\*</sup>[16],  $^{239}$ Np<sup>\*</sup>[17] (17 MeV excitation -  $\sim 0.28 \pm 0.26$  central neutron per fission after subs-tracting the prefission neutron evaporation), while such a central component was not found for  $^{232}$ Th<sup>\*</sup>[18].

Though theoretical calculations have shown that it may exist with an important yield [19], this kind of neutron emission, which would be of the same origin that the charged light particles, is not proved with enough confidence, and some more experiments, very difficult indeed, are necessary. In 1969 Feather concluded that " there would be added confidence, perhaps, if under the same experimental conditions the description of prompt neutrons from one mode of fission were to be found not to need the central component for its full description, whilst the distribution relative to another mode required such a component ". But this suppose that this central component does not always exist, if it does.

II - RADIATIONS EMITTED BY THE FISSION FRAGMENTS.

They are essentially neutrons and  $\gamma$ -rays.

II-1. <u>Neutrons</u>. We shall not consider the  $\bar{\nu}$  value, but will give some attention to the number of neutrons emitted by the fragments as a function of their mass number. The most recent results are those obtained by BOLDEMAN et al [20] on <sup>236</sup> U<sup>\*</sup> (fig. 1). They agree very well with those of MASLIN et al [21]

and show the same general trend as was previously obtained by APALIN et al [22] and by MILTON and FRASER [23] - but they don't obtained evidence for the structure which was claimed by these last authors.  $\ddagger$ 

They obtained a very straight linear variation of  $\vec{\nu}_{total}$  versus the total fragment kinetic energy with a slope  $\frac{dE}{d\vec{\nu}} = -0.167 \text{ MeV/n}$ , and deduced, from the MYERS-SWIATECKI mass formula [24], a value from the variation of excitation energy of fission fragments with  $\vec{\nu} : \frac{dE^*}{d\vec{\nu}} = 9.5 \text{ MeV/neutron}$ ; this result confirms the value previously obtained by NIFENECKER et al [25].

These values are greater than the  $\sim 6.6 \text{ MeV/n}$  value for emission of a neutron; this difference is interpreted as due to the  $\gamma$ -rays emission.

II-2. The y-rays emission may be studied from two points of view :

a- The first one concerns the identification of the  $\gamma$ -rays and of their origin. For the thermal neutron induced fission of <sup>235</sup>U, HORSCH [26] resolves 54 gamma-ray lines and, for about 35 of them, identifies the emitting nucleus; 7 of these identifications agrees with previous results obtained for <sup>252</sup>Cf by BOWMAN, WATSON et al [27].

b- The other point of view concerns the study of the angular distribution of these  $\gamma$ -rays which results of the alignment of the spins of the fission fragments. These spins can be deduced from the experimental angular distribution if one does some assumptions, such as their complete alignment.

At the 1969 Vienna Symposium, both ARMBRUSTER et al [28] and JEKI et al [29] concluded that the average spin value was of the order of 15; this value is much greater than the previously quoted ones: 7,8 or 10 according various physicist [31]. From this one may deduce that there is an important competition between  $\gamma$ -rays and neutron emissions.

But in a recent paper [32] ARMBRUSTER et al give their final results from which they concluded that:

المائة أراحا فالك اللها بيان الاثر بريان بعدة اللها فتحا فحة ليتي بريد بجار ووي بويه بعد المر سية بويه بجه فله الكل

\* Some very recent results from Columbia University have been presented by Professor W. W. Havens at this topical conference.

- the average decay time is of the order of 0.5 to 1  $10^{-11}$ s, the shortest life time being  $10^{-12}$ s
- the average energy of the  $\gamma$ -rays is of the order of 0.8 MeV
- dipole electric transition are not possible, and desexcitation occurs only by quadrupole transition E2 -

- that the statistical model for desexcitation, which would give  $\overline{I} \sim 16$ , does not predict several of the experimental results, which, on

the contrary, would be explained if one suppose a desexcitation by a collective E2 cascade

and they concluded that  $\overline{I}$  could not be greater than 8 or 9 and is probably worth 6 to 8 (varying from 5 to 10 according the fission product ratio).

It has to be noticed that this was the value given by VAL'SKII et al [33] in 1969 in interpreting the energy spectra and the angular anisotropy of the  $\gamma$ -rays accompanying the fission of <sup>252</sup>Cf from simple considerations. ×

**III - DELAYED RADIATIONS -**

They are the different radiations emitted by radioactive nuclei :  $\beta$ -rays,  $\gamma$ -rays,... We shall just say a few words about the  $\gamma$ -rays émission. A special emphasis is given to the delayed neutrons.

For a long time the descriptions of these 2 kinds of radiation were not detailed but gave some empirical law such as the Way Wigner law for decrease of  $\gamma$ -ray emission intensity ( $I \sim t^{-1.2}$ ). All these informations were reviewed in 1965 by KEEPIN [34]. Since that time efforts have been devoted to the study of the particular radiations outgoing from specific emitting nuclei.

III. 1. - Delayed  $\gamma$ -rays. The most completed work on individual delayed  $\gamma$ -rays is probably the one by GUY for  $^{252}$ Cf [35]. He resolved 144 lines with energies from 90 to 2860 keV, with periods from 5 ns to 3  $\mu$ s, and identified the emitting nucleus for most of them. The energies of these  $\gamma$ -rays are less than 400 keV, except for a few of them; among others, a  $\gamma$ -ray with energy of 1180 keV, probably emitted by  $^{134}$ Te, was reported at the Vienna Conference [36] and DIETRICH [37] pointed out that it could be interpreted as a possible transition between oblate and prolate states.

More recently, a paper by AJITANAND [38] confirms a part of GUY's results.

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**±** The same result is obtained by Nifenecker (private communication).
III. 2. Delayed neutron.

I - Precursors identification. In a recent report TOMLINSON [39] reviews the data concerning the delayed neutron emitters. He identifies 38 of them (21 for the light group of fission fragments, 17 for the heavy one) and sets 10 other fission fragments as possible emitters. There are still some discrepancies between the calculated yields and the experimental values, but the knowledge of these ones have very much progress in the recent years.

The fact that there exists at least 38 delayed neutron emitters does not mean that, in reactor kinetic studies, one has to take in account 38 (or more) periods because many of these radionuclei have the same precursors, or contribute only for a very few to the total delayed neutron yields; but the old 6 groups classification has to be improved. The second group  $(\tau = 22^{"})$  for instance is mainly composed by 88 Br  $(\tau = 15.9^{"})$  and by  $^{137}I (\tau = 24.4^{"})$ .

One of the remaining problems was the existence of a long period for delayed neutrons ( $\tau > 55$ "); some evidence for the existence of such a period was previously reported [40]. But at the 1969 Vienna Conference, TOMLINSON and HURDINS [41] reported a careful study from which they concluded that there are no true delayed neutrons with a period greater than 55", but that there exist some "pseudo delayed neutrons" with periods of 3.1, 17 and 111 minutes, which are due to prompt neutron occuring from the photofission of <sup>238</sup>U and <sup>235</sup>U, due to high energy gamma-rays emitted by some fission products. The yield of these pseudo delayed neutrons depends very much on the total amount of material used; it's worth w6. 10<sup>-8</sup> n/fission for the 3 mn period with 3.5 kg of natural U.

2 - Energy spectrum. The interest is turning more and more on the energy spectrum of the delayed neutrons. In fact, the theories predicting this spectrum also predicte the emission probabilities. The most sophisticated study is probably the one by GAUVIN and de TOURREIL [42] which allows a satisfying description of the old experimental data [43] (fig. 2). But according the recent results from CUTLER and SHALEV [44], the energy spectrum of the delayed neutrons shows much more structures than were previously obtained by BATCHELOR (fig. 2). Similar results have been obtained in Sweden  $\begin{bmatrix} 45 \end{bmatrix}$  and in USA  $\begin{bmatrix} 46 \end{bmatrix}$ 

It has to be emphasized that the knowledge of this energy spectrum is not only useful for reactor application but that it gives information on the excited level of the neutron emitting nucleus. In their calculations GAUVIN and de TOURREIL treated these levels by a statistical method; this procedure is sufficient to explain the absence of detailed structures in the theoritical curves.

## CONCLUSION -

1 - There is a good knowledge of the light particle emission at the scission stage but it seems that we need something else to allow us to obtain valuable information on the scission process.

2 - There are great improvements on the Y and n emission, but the interpretation of the Y-rays distribution seems to be uncertain, and it follows a misunderstanding of n and Y-ray emission competition.

3 - The knowledge of the delayed neutron precursors is nearly solved; now it is necessary to study experimentally their energy spectrum, and to elaborate practical theories which would allow us to describe these spectra and the emission probabilities.

## ACKNOWLEDGEMENTS -

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Baa	Relative intensity (extrapolated)				E pezk (MeV)				FWHM (MeV)			
ticle	233 U+nth	235 U+nth	239 Pu+nth	<sup>252</sup> Cf	233 U+nth	235 Uinth	239 Pu+nth	252 <sub>Cf</sub>	233. Uinth	235 U+ntl:	239 Pu+nth	252 <sub>C£</sub>
ЧН		1.15-0.15	1.9 <sup>±</sup> 0.1	1.75 -0.30		£6±23	8.40-0.15	7.8-0.8		6.9-0.5	7.2-0.3	6.5-1.6
2 <sup>2</sup> H	a41 ± a02	0.5 -0.1	a5 <sup>+</sup> a1	Q68 - Q03	84-0,2	7.9-03	8.2 -0.3	8.0-2.5	63±03	7 + 1	7.Z+0.5	7.2+1.0
<sup>3</sup> н	460 - 020	6.2 +0.5	68 <sup>±</sup> 03	846 -028	84-02	\$ 6-0.3	820-015	& 0 <sup>+</sup> 0.3	6.5-0.3	67-06	7.6-0.4	6.2-0.6
<sup>3</sup> He	0											
<sup>4</sup> He	100	100.	100	100	163±01	157-03	160 <sup>±</sup> a1	160-02	9.7-0.Z	9.8-0.4	10.6-0.4	10.2-0.4
<sup>6</sup> He	1.37 -0.07	1.1 -0.2	1.9 -0.2	263 - 218	11.5=02	12.9-05	11.8 <sup>±</sup> 0.4	120-05	9.5-0.3	87-07	10.6-0.6	8 - 2
<sup>8</sup> Не	0.036-0.004		008-002	0.090-0.012	9.7 <sup>±</sup> 0.3		12	10.2+1.0	6.9-0.5		9	s <sup>+</sup> ż
<sup>6</sup> Li	0											
7 <sub>Li</sub>	a.037 <sup>+</sup> a.002				15.8-0.3				121-04			
<sup>8</sup> Li	0.01 <b>8-</b> 0.002				14.4-0.5				10.6-0.8			
9 <sub>Li</sub>	<b>a.036</b> ±a.005				12.0-1.0				11.0+1.5			
Li	0.091-0.009	a12-a002		0.132-0.001								
<sup>7</sup> Be	O											
9 <sub>Be</sub>	a.037 <sup>±</sup> a.008											
<sup>10</sup> Be	0.43 +0.03				17.0-0.4				157-09			
LI Be	0											
Be	0.467±0.038	0.37 <sup>±</sup> 0.04		0.201±0.002								

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Fig. 1 - FROM BOLDEMAN ET AL  $\begin{bmatrix} 20 \end{bmatrix}$  COMPARISON OF NEUTRON EMISSION DATA (FOR DIRECT NEUTRON COUNTING DATA)



Fig. 2 - Energy spectra of the delayed neutrons for the  $^{235}$ U induced fission; the Y-axis graduations are arbitrary. The theoritical curve represents the spectra of the 2<sup>nd</sup> groups of delayed neutrons; the experimental curves, which correspond mainly to this group, show the resolution improvement.

DISCUSSION

J.J. Schmidt: (Comments): On the basis of R. Keepin's wellknown results reactor physicists are accustomed to using six delayed neutron groups. According to the recent Argonne results of Tomlinson et al. this seems to be much too coarse a picture, the number of detected precursors being as high as 38.

# FISSION BARRIER HEIGHTS, ENERGY RELEASE, AND FRAGMENT ENERGY PARTITION PROCESSES FROM THE VIEWPOINTS OF THE NUCLEAR STRUCTURAL MODELS

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# 1. INTRODUCTION

In this talk, I wish to make a brief summary of the theoretical work done in the Saha Institute on various aspects of the nuclear fission phenomena. I must mention the efforts of Ramanna and his associates<sup>1-4</sup> to develop a fission model. The consequences of their postulate that fission is a combined result of nucleonic Brownian motion and the magic shells (50 and 82 nucleo configurations) were studied<sup>1</sup> by applying the theory of random flights (Markov process<sup>2</sup>). The free Fermi gas model was used<sup>2,3</sup> to understand the fragment mass distributions as a function of fissioning mass and excitation energy, and also charge distributions<sup>4</sup>. Another wse<sup>5</sup> of the Fermi gas model has been made to study the vanishing of the shell structure effects with excitation.

#### 2. MICROSCOPIC MODELS

There have been recent serious attempts, after Strutinsky's major breakthrough work<sup>6</sup>, to attempt microscopic descriptions of the fission phenomena. Most notable among these are the work of Nilsson and his associates<sup>7</sup> and the work of Mosel and Greiner<sup>8</sup>.

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For the first time, as a combined result of these works, a systematic study of the fission barrier heights  $E_B$  and the energy release  $E_R$  has been possible, and the stability of superheavy nuclei against spontaneous fission has been examined.

In Calcutta (SINP), a series of studies since 1966 was undertaken to understand the energy transformation and energy partition processes in fission<sup>9-12</sup>. The work relies heavily in developing a suitable interaction-modified form of a model of a nuclear Fermi gas (called the renormalised Fermi gas model - the RGM) and connecting this up with the potential energy surface concept (the PES concept). The model is shown schematically in Fig. 1. The PES energy expansions used in our work were as these of  $M_{\mathbf{0}}$  sel and Greiner<sup>8</sup>. It has been possible to write out an energy balance equation<sup>9</sup> containing the intrinsic energies and the Coulomb interaction terms of the fragments expanded as functions of respective deformations. We were thus able to solve for the fission barrier height  $E_{\rm R}$  from the condition of energy minimisation at the saddle point and the respective critical deformations at the saddle and the scission points. The energy release  $E_R$  was solved directly from the energy balance equation itself. The partition of available energy into the kinetic and excitation energies of the fragments (T<sub>p</sub> and UF) was directly obtained from the terms used in the combined RGM-PES expansions.

3. OUR RESULTS

The consequênces of the simple elementary approach outlined above may be compared with many relevant experiments. The

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microscopically detailed predicted energy partitions are shown in Figs. 2(a - d) for  $^{233}$ U+n,  $^{235}$ U+n,  $^{239}$ Pu+H and  $^{252}$ Cf; where comparison has been made with the experimental data on kinetic and excitation energies. Extention of the work<sup>12</sup> to another ten nuclei, in the mass range  $226 \leq A \leq 256$ , predicts the energy partitions and the systematic behaviour of the fission barrier heights  $E_B$ . This predicted trend of  $E_B$  is shown in Fig. 3, and is compared with the liquid drop predictions and the experimental measurements. Since our procedure does not depend, at <u>any</u> stage, on the use of a mass formula, it is instructive to compare the systematics of energy release<sup>12</sup>. This is shown in Fig. 4. It is observed that while the fit and the general trends agree around mass 235 - 242 region, some systematic differences exist in the low and heavy mass fissioning regions.

Other detailed applications of the model have been in the study of the prompt gamma-decay processes<sup>10</sup> in  $^{235}$ U+n and  $^{252}$ Cf (Fig. 5) and the partition of energy in an excited fissioning nucleus<sup>11</sup>, e.g., in  $^{233}$ U+n,  $^{230}$ Th+et, and  $^{226}$ Ra+p, at different projectile energies (Fig. 6). Fairly satisfactory agreement of the predictions has been obtained in fill cases with the experimental data.

#### 4. CONCLUSION

It appears that the studies in fission phenomena has reached a stage of maturity in the sense that on the one hand, excellent fission models are being considered and developed, and on the other, sufficiently detailed microscopic nuclear structure information are -148-

being plugged in such models to understand the trends and systematics of this complex many-body transformation process.

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Fig. 1. (a) A schematic diagram of the basis of the RGM. The model defines an effective ground state  $\epsilon$  developed from the nuclear interaction energy corrections +f,  $-\Delta^2/G$ ,  $-\Delta$  and -S on the free Fermi surface  $\epsilon_0$  (Ref.<sup>9</sup>).



Fig. 1 (b) A schematic diagram connecting the RGM with the deformation dependent PES (Ref.<sup>9</sup>) suitable to apply to the fission studies.



Fig. 2(a): The energy partitions in the prompt fragments for fission in <sup>234</sup>U. For detailed discussions of the energy terms and the symbols used, see Ref. 9.



Fig. 2(b): The energy partitions in the prompt fragments for fiscion in <sup>236</sup>U. For detailed discussions of the energy icrus and the symbols used, see Ref. 9.



Fig. 2(c): The energy partitions in the prompt fragments for fission in 240 Ju. For detailed discussions of the energy terms and the symbols used, see Ref. 9.





Fig. 2(d): The energy partitions in the prompt fragments for fission in 252Cf. For detailed discussions of the energy terms and the symbols used, see Ref. 9.



Fig. 3. The fission barrier heights  $E_B$  as a function of the fissioning mass in the range 226  $\leq A \leq 256$ . The LDM predicted values (for two types of calculations with the fissionability parameter x= 48 and one calculation with x = 60) and the experimental observations (Hyde and Ignatiuk-Smeremkin) are compared with our calculations<sup>12</sup>.



Fig. 4. The mean fission energy release  $B_R$  plotted as a function of the fissioning nuclear mass. Our model-predicted values (solid curve) are compared with the predictions of the Wing-Fong type of mass formula (Ref.<sup>12</sup>).



Fig. 5. Calculated prompt gamma-ray yields<sup>10</sup> from prompt fragments for fiesion of (a) <sup>236</sup>U and (b) <sup>282</sup>Cf shown as triangles are compared with the experimental data (black dots with error bars).



Fig. 6(s): Number of prompt neutrons emitted from fission of 227Ac as a function of prompt fragment mass. Numbers at different excitations of the fissioning nuclei are compared with different sets of experiments (nee Ref. 11).



Fig. 6(b): Number of prompt neutrons emitted from fission of <sup>234</sup>U as a function of prompt fragment mass. Numbers at different excitations of the fissioning nuclei are compared with different sets of experiments (see Ref. 11).

DISCUSSION

- S. Mukherji : How do you justify equating the quadrupole interaction energy to the Y-decay energy?
- A. Chatterjee : I took terms of the type discussed in Wilets' Book and assumed that the quadrupole interaction term gives the **Y**-energies.

#### CONCLUDING REMARKS

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This topical conference on fission has certainly been one of the highlights of the INDC meeting, where it was made clear by various speakers that nuclear fission is still continuing to be as exciting as it was when it was just discovered and that the investigations of the physics of the fission process are important not only for the understanding of the nuclear structure of highly deformed nuclear configurations, but also for a reliable systematisation of the nuclear data needed for reactor design.

Nuclear fission indeed represents the changing fashions in physics. Many years ago when I had just started working in this field, one had to think in terms of the liquid drop model alone to explain everything about fission. This was so because there was not much choice. Over the years we have seen a rapid progress in our theoretical understanding, and today we see that most of the observed features in fission are infact the consequences of the single particle and other quantum effects. One of the landmarks on the theoretical side was the introduction of the transition state by A. Bohr in 1955. At the first IAEA Nuclear fission symposium held at Salzberg in 1965, the transition state was the subject of lot of discussions with regards to detailed theoretical fits to the various angular distributions and other experimental data. In the recent years, however, most of this earlier work has been overshadowed by the experimental and theoretical consequences of the discovery of a double-humped barrier by V. M. Strutinsky, as was also reflected by the proceedings of this conference. Double-humped barrier is just one consequence of the present theoretical understanding of the nuclear shell effects in spherical and deformed nuclear configurations. The other consequence, which I believe has been even more exciting, is the prediction of the possible existence of an island of stability of the superheavy nuclei around Z = 114 and N = 184. Dr. Bhandari's investigations of the tracks in the moon rocks seemed to fit the idea that these superheavy nuclei might have been present at some stage. However Dr. Kapoor's talk made us feel that it is going to be very difficult to produce these nuclei by heavy ion reactions for at least one important reason that the shell effects quickly disappear with excitation energy.

I do not know how many of you are convinced about the existence of these nuclei which is just one of the many problems in fission which have eluded a solution so far. Another such problem is the mass distribution in fission and Prof. Haven's talk showed the importance of the K-quantum number in this connection. In 1963 we had also carried out some work in Trombay to investigate the effects of K-quantum number on mass distribution by studying a possible correlation between anisotropy and asymmetry in the 4 MeV neutron induced fission of  $U^{235}$ . We did observe a correlation which. I believe, is consistent with Prof. Haven's observations. However, there have been one or two other experiments which have not shown the same results. I feel that this is an important problem and more experiments need to be carried out to determine how far the mass distributions are affected by the transition states. Dr. Ramamurthy's paper on the stochastic nature of the fission process finds further support from the concept of a double-humped barrier. For, hitherto, it was very much doubted whether the time from saddle to scission was enough for stochastic equilibrium but now with the presence of one or more humps in the potential surface it is possible for the fissioning nucleus to get stuck in the valley making the progress to scission rather slow.

The detailed and accurate results of measurements of the various aspects of the radiations emitted in fission are naturally of great interest to the delegates of the INDC meeting because of the role of such data in various nuclear energy programmes. I can see an urgent need of more and more accurate measurements on the average numbers and spectra of neutrons as a function of neutron energy in fast fission, for the design of fast breeders. There is also a lot of interesting physics in the studies of these radiations namely neutrons, gamma rays, conversion electrons and Kx-rays, and the occasionaly emitted alpha particles since several questions concerning physics of the fragment deexcitation process still do not seem to be satisfactorily resolved. I noted with interest that the duestions asked at the time of the Salzburg conference concerning prescission neutrons are still being asked. A distinguished scientist once said about nuclear spectroscopy: "Give me a hundred scientists and a hundred years and I can produce any number of problems". If this is true of an old subject like nuclear spectroscopy then how much more is this so for the relatively young field of nuclear fission?

Before closing this meeting, it is my pleasant duty to thank the chairman and the speakers of the various sessions of this very interesting and fruitful conference on the Neutron induced fission, which was organised as part of the INDC meeting held here. I particularly thank the IAEA sponsors for having decided to hold the INDC meeting at Trombay. I have no doubt that by holding such meetings at places like Trombay a large number of members working in this field are able to benefit. It is for this reason that in all my dealings with the IAEA, I have been urging them to give us the chance of hosting more such meetings and conferences.