PROCEEDINGS OF THE «SEMINAR ON FISSION»

Castle of Pont d'Oye (Habay-la-Neuve)

22-23 May 1986

C. WAGEMANS (Editor)
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BLG 586
SEMINAR ON FISSION
22-23 MAY 1986

PROGRAMME

THURSDAY 22 MAY 1986

9h00 : Welcome address C. Wagemans

SESSION I. INTRODUCTORY SESSION
Chairman : M. Nève de Mévergnies

9h05 : Fission revisited A. Deruytter
9h20 : How fission was discovered P. Van Assche

Break 10h00 - 10h30

10h30 : Photofission E. Jacobs
11h05 : Neutron induced fission below and at the barrier H. Weigmann
11h40 : Application of some fission properties to neutron dosimetry P. D'hondt

Lunch 12h00 - 14h00

SESSION II. LIGHT CHARGED PARTICLE AND NEUTRON EMISSION IN FISSION
Chairman : E. Jacobs

14h00 : Review of theoretical approaches to the emission of α-particles during nuclear fission N. Cârjan
14h45 : Ternary fission experiments J. Theobald

Break 15h30 - 16h00

16h00 : Neutron induced ternary fission C. Wagemans
16h25 : Emission of light charged particles in the photofission of actinides M. Verboven
16h50 : Fragmentation and neutron emission for 252Cf(s.f.) C. Budtz-Jorgensen

18h30 : Reception

20h00 : Dinner
FRIDAY 23 MAY

SESSION III. FISSION FRAGMENT PROPERTIES
Chairman : A. Deruytter

9h00 : The scission point model: possibilities and shortcomings J. Moreau
9h45 : Cold fragmentation: experiments and models F. Gonnenwein

Break 10h30 - 11h00

11h00 : Fission fragment energy and mass distributions in the spontaneous fission of the Pu-isotopes P. Schillebeeckx
11h35 : Energy, mass and charge characteristics of the $^{241}$Pu(nth,f) fragments studied with Cosi Fan Tute P. Schillebeeckx
11h35 : Fission fragment properties of $^{235}$U(n,f) in the neutron energy range from thermal to 1 MeV F. Hambsch

Lunch 12h00 - 13h30

13h30 : $^{235}$U(n,f) fragment mass, kinetic energy and angular distributions for incident neutron energies between thermal and 6 MeV H.-H. Knitter
14h05 : Photon induced fission of $^{232}$Th with 12 and 20 MeV bremsstrahlung M. Piessens

Break 14h30 - 14h50

SESSION IV. HEAVY ION INDUCED FISSION
Chairman : Y. El Masri

14h50 : Heavy ion induced fission at $E_i \leq 10$ MeV/u F. Hanappe
15h35 : Heavy ion induced fission above 10 MeV/u B. Tamanis
16h20 : Dynamical and statistical theory of deep inelastic, fast fission and fusion processes C. Le Clercq-Willain
16h45 : Binary fission of $^{44}$Ti P. Cohilis

17h10 Closing of the Seminar. C. Wagemans
WELCOME ADDRESS

Ladies and Gentlemen,

I am very glad that I may welcome you in this ancient castle of Pont d'Oye. I am convinced that this remote corner of Belgium with its magnificent nature will be an ideal frame for an interesting conference and for fruitful discussions. Also the presence and the active collaboration of our foreign guest speakers will be of great benefit to this meeting.

As you can see in the programme, fission is still an actively studied research field in Belgium, in which about thirty colleagues working in five different institutions are involved. So it was certainly worthwhile to organize a "Belgian" topical fission meeting, especially since the IAEA conference series devoted to the fission process has been finished and our French colleagues only talk about the souvenirs of the "Journées d'Etudes sur la Fission" however without organizing new ones. We hope that his conference will give you the opportunity to demonstrate once again the nice work which has already been realized and the importance of continuing it. Then the proceedings of this conference will become a very useful document.

Finally I want to express the thanks of the organizing committee towards the National Fund for Scientific Research and the the SCK/CEN Mol for their financial support, which made the organization of this conference possible.

Cyriel Wagemans
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P. del Marmol
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G. Grégoire
F.J. Hambsch
F. Hanappe
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SESSION 1

INTRODUCTORY SESSION

Chairman: M. Nève de Mévergnies
CEN/SCK, Mol
The theme of this seminar is 'fission revisited'. This violent collective phenomenon has now been studied for many years. Of course not only the great complexity of the process triggered that many studies, but it was also soon realised that the process was important for the liberation of nuclear energy. The accumulation of experimental data was very useful for the description of many aspects of this manifold reaction but did not lead for a long time to a coherent and thorough understanding of the process. First fission models were of a phenomenological nature (liquid drop model) and did not stem from basic nuclear theory. Only recently the progress in the knowledge of nuclear structure and nuclear reactions resulted in a more fundamental understanding.

Different phases can be identified in the fission process passing from (1) the initial state formation to (2) transition from the initial state over the saddle point to the scission in two or more primary fragments followed by (3) the formation of fission products by prompt processes and concluded (4) by the de-excitation of the fission products by delayed processes.

(1) The initial state formation via neutrons is most commonly used and the most important for practical applications. However, also photofission and recently even electrofission were studied. Light charged particles such as p, d, a are used and recently heavy ion induced reactions allow the study of the fission channel with a very large set of initial conditions. In neutron induced fission the initial states are the initial states of the compound nucleus with an excitation energy given by

\[ E^* = S_n(A^*,Z) + E_n \frac{A}{A+1} \]

where \( S_n \) (\( A^*,Z \)) is the neutron separation energy in the compound nucleus (\( A^*Z \)) and \( E_n \) the incident neutron energy. The spin \( J \) of the compound state is then determined by the coupling of the spin \( I \) of the target with the angular momentum \( \ell \) and spin \( s \) of the incoming neutron. The parity is determined by the parity of the target nucleus and the orbital angular momentum \( \ell \). In photofission the entrance channel is governed by the electromagnetic interaction with the target nucleus, leading to small cross-sections but only selective parity states are excited. For instance for \( 0^+ \) target nuclei (e.g. \( ^{238}_{\text{U}} \)) only \( 1^− \) states will be formed under E1- bombardment. Studies of the influence of the excitation energy and the spin and parity of initial states on fission characteristics, also in connection with the channel theory of fission, are still an important component of present research. In some cases also spontaneous fission can be used as a reference system for comparison.

In heavy ion collisions fission phenomena are seen in elastic scattering (Coulomb fission), inelastic and transfer reactions (fission after transfer), deep inelastic collisions (sequential
fission) and in fusion reactions (conventional and fast fission). This 'fast fission' is a new output channel encountered when the critical angular momentum value for fusion or capture reactions is larger than the angular momentum corresponding to a vanished fission barrier under centrifugal force effects. It exhibits an intermediary lifetime between deep inelastic collisions and conventional fission.

It should be remembered that fission is used in heavy ion reactions as a tool to obtain information on the entrance channel in heavy nuclei fusion studies. When excited nuclei are built, above 10 MeV/A bombarding energy, the fissioning nuclei are so hot that evaporation occurs during the fission process and the standard (static) statistical model cannot be used. A persisting problem here is that, although theoretically predicted, fission barriers do not vanish with temperature.

(2) The motion of the fissioning system from the formation of the initial state over the saddle point to scission is governed both by the statics and the dynamics of the process. This phase plays an essential rôle for the determination of the fission properties and in particular for the so important fission probability.

The statics of the process is determined by the knowledge of the total energy (potential energy) of the system (supposed to be at rest) as a function of deformation. To describe the statics of the fissioning system, it is therefore essential to define the shape of this system by means of a set of deformation parameters \( \{s\} \). Usually it is assumed that the nucleus has a sharp surface, defined by \( \{s\} \) in which the nuclear matter is supposed to be incompressible. Very often the potential energy is plotted as a function of only one deformation coordinate, mainly the elongation, the other deformation parameters being set equal to zero or given the values they take on the fission path when the first deformation parameter has the indicated value. This is the familiar one-dimensional fission barrier representation. In Fig. 1 the variation of the potential energy of \(^{240}\text{Pu}\) as a function of deformation along the fission path is shown\(^1\). The energy is expressed in MeV with the origin corresponding to the sum of the individual mass-energies of the nucleons in \(^{240}\text{Pu}\). The solid line illustrates the detailed energy variations, included the shell effects and shows the accuracy needed in the calculations for obtaining reliable fission barriers which are indeed very small (only about 6 MeV high for \(^{240}\text{Pu}\)) compared to the overall range of energy variations met during the fission process. Knowledge of the fission barrier, and possibly of the multidimensional potential energy surface is essential for the prediction of fission properties, especially the fission probability. The calculation of \( V \ (\{s\}) \) particularly at large deformation is difficult and is still a challenge for theoreticians. Basic calculations from first principles (nucleon-
nucleon interaction) are possible but do not give accurate results as yet (initially too high barriers). Phenomenological procedures are more precise but still need to be justified from a fundamental point of view. The hybrid model which combines the macroscopic and microscopic aspects of the nucleus as introduced by Strutinsky\textsuperscript{2)} allows the calculation of the potential energy \( V(s) \) as composed of two parts: the macroscopic energy \( V_M(s) \) which is the more important one and which can be derived from the liquid drop model; and a shell-energy correction \( A E_{sh}(s) \) which takes into account the shell structure of the nucleus at deformation \( s \). The deformation potential (minimum total energy of the nucleus as a function of deformation) has two minima which correspond to states of the nucleus with normal and extended deformation. This double-humped structure has important consequences for sub-barrier fission in particular (intermediate structure in the fission cross-section). It is further to be remarked that in contrast to the Pu- and heavier isotopes, the fission cross-section of most of the light actinides shows pronounced peaks in the threshold region\textsuperscript{3)} (e.g. \( ^{230}\text{Th} \) on Fig. 2).

The strong narrow peak A is interpreted as a vibration in a shallow minimum of the deformation potential. The present belief is that the shallow minimum is the third rather than the second one. This hypothesis of a third minimum needs further substantiation.

The dynamics of the fission process also plays an important rôle in the fission mechanism from two points of view: inertia and viscosity. In studying the motion of the fissioning system towards scission the effective mass of the system has to be specified. This mass appears e.g. in the expression of the kinetic energy of the nucleus at deformation \( s \). The mass tensor exhibits strong variations with deformation \( s \) as an effect of the variation of shell structure with deformation. The viscosity effects are due to the coupling of the collective degrees of freedom to the intrinsic ones.

This coupling induces excitations in the nucleus and therefore governs the sharing of the available energy between kinetic energy in the fission mode and excitation energy. Viscosity can play an important rôle in the rapid descent from saddle point to scission because the Rayleigh dissipation force contains first derivatives of the deformation parameters. The energy balance at scission can be written as follows:

\[ E^* (A^*, Z) + V(s_o) = V_{sc} + E_{sc}^* + E_K^sc \]

where \( V(s_o) \) and \( V_{sc} \) are the potential energies at ground state deformation and at scission respectively and \( E_{sc}^* \) and \( E_K^sc \) are the excitation and kinetic energies of the fissioning system at scission.

Fig. 2. The fission cross section of \( ^{230}\text{Th} \) in the threshold region.
Viscosity of nuclear matter is at present very poorly understood and its importance depends on many parameters including excitation energy. This makes it difficult to extrapolate viscosity properties observed at one energy to another energy (e.g. spontaneous fission and thermal neutron induced fission). Despite its complexity viscosity of nuclear matter is at present actively studied not only in fission but even more so in heavy-ion physics.

(3) The scission point, where the fragments start their own life is crucial because from then on detection of the fragments leading to angular, mass- and energy distributions of heavy fragments, light charged particles and neutrons becomes possible. Any theory of fission has to be able to explain these manifold distributions. The most successful model of nuclear fission presently is that by Wilkins, Steinberg and Chasman\(^4\), where agreement is reached with several striking trends in the fission of actinides. It assumes that collective and intrinsic degrees of freedom are separately in thermal equilibrium at the scission point such that \(T_{\text{coll}}\) may be different from \(T_{\text{intr}}\). This metastability is in contradiction to the foundations of fission theory, i.e. that the gross properties of fission can be understood by the changing preponderance of surface against Coulomb energy. The scission point remains a very unstable configuration, also when shell effects are included. This objection against the foundation of fission theory would not be important, unless measured properties prove the insufficiency of the model. One such property is the prodigious width of the mass distributions created by fission, not reproduced by the Wilkins model. Another property is the dependence on \(A\) of the multiplicity of evaporated neutrons that persists at high excitation energies. The sawtooth in low energy fission was explained as being due to shell effects in the nascent fragments. However, since shell effects quickly die out with rising nuclear temperature, the scission point model should deny the existence of the sawtooth at high excitation energies. However, the sawtooth has a remarkable persistence with increasing excitation and even seems to be present in deep-inelastic heavy-ion reactions with excitation energies higher than 100 MeV.

Recently a model was developed by Brosa and Grossman\(^5\)\(^6\) who propose that nuclear scission happens as a consequence of hydrodynamic instability triggered by random surface vibrations. Thus the scissioning complex ruptures at random positions. It is based on the instability against surface vibrations of long liquid jets when the wavelength exceeds the circumference (Rayleigh). Measured total kinetic energies as well as neutron emission data support the relevance of the proposed instability model. The model is illustrated in Fig. 3 and the persistence of the sawtooth curve with excitation energy is illustrated in Fig. 4 for \(^{238}\text{U}\) photofission\(^7\).

It is evident that the properties of fission fragments (angular distributions, mass distributions, \(\langle J \rangle\), odd-even effects, etc.) have to be studied in all possible detail, as a function of excitation energy of the compound system, and as a function of the excitation energy of the fragments. Mass-separators, such as \textit{LOHENGRIN}\(^{11}\) and \textit{COSI FAN TUTTE}\(^{12}\) at ILL, Grenoble, play here an important rôle for fission of the common fissile isotopes. Also the twin gridded ionization chamber as developed by Knitter and Budtz-Jørgensen\(^{11}\) is an excellent tool. The strong influence of shell effects in the fragments is becoming better recognized. Cold fragmentation, although a rather exotic limiting case of the fission process, deserves attention as it yields information on the most compact scission configurations, where the fragments had
no chance to acquire prescission kinetic energy. Also the light charged particle associated fission deserves further study, in particular because of its relevance to the scission point configurations and parameters, and the explanation of polar emission is required.

![Fig. 4. Neutron multiplicity $\bar{v}(A)$ over mass number $A$ for the photofission of $^{238}\text{U}$. Full lines calculated with model of Brosa and Grossman. Experimental data from Defrenne et al.][1]

![Fig. 3. Mass yield $Y$ and neutron multiplicity $\bar{v}(A)$ as functions of the fragment's mass number $A$ for the spontaneous fission of $^{252}\text{Cf}$. Full lines are calculated from model. Experimental data are from Bowman, Whetstone and Terrel.][2]

(4) The third phase from scission to the formation of the fission products in their ground state is dominated by the Coulomb repulsion of the two fragments and by prompt de-excitation by neutron- and gamma-ray emission. Recent measurements by Budtz-Jørgensen and Knitter[14] on $^{252}\text{Cf}$, in particular the neutron energy dependence of the $N(90^\circ)/N(0^\circ)$ intensity ratio, is in full accord with the assumption that all neutrons are emitted from the fully accelerated fragments and the existence of a hard ($T = 2 - 2.5$ MeV) scission neutron component which Märtens[15] concluded from the Bowman[16] angular distributions must be denied. They also observed a striking dip at $A = 130$ in the dependence of the average neutron energy on $A$, previously not observed by Bowman. See Figures 5 and 6.
These data illustrate again the importance of detailed information on low probability events. It is interesting to note that, although fragments formed at scission are neutron rich, this is not the reason for the non-emission of charged particles. It is the Coulomb-barrier which prevents their emission. So the situation at scission must be very different as charged particles are emitted at that instant, and only few neutrons!

The last phase, i.e. the de-excitation of the fission products by delayed processes, mainly by β-decay is much slower as it is due to a weak interaction. In a few cases delayed neutrons are emitted which are so important in the neutron balance in the starting up phase of fission reactors. This part of the fission process has its relevance to the study of nuclides formed far away from the stability line, but is less pertinent to our two-days discussion on fission.

This introduction was fragmentary and in particular the more application oriented aspects were not emphasized sufficiently, but this information finds its way easily to other types of conferences. I hope to have illustrated that fission has not revealed all its secrets and that this violent collective phenomenon deserves our further attention.
References


14) C. Budtz-Jørgensen and H.H. Knitter, this meeting.


Enrico Fermi was the first to use neutrons as a research tool. This idea went against the that-time conviction that you could not expect too much from these meager neutron intensities. Didn't Joliot and Curie use indeed very intense beams of alpha particles in 1934 to induce and observe the first artificial radioactivity in nuclei? The intensities of the beams, made available from cyclotrons and from the accelerator of Cockcroft and Walton exceeded by far anything that could be given by neutron sources.

Still Fermi and collaborators made a systematic survey of neutron-induced radioactivity for all the elements; already in 1934 they reached the element uranium. In contradiction to the decay curves of lighter elements - where one or at most two half-lives were observed - the behaviour of the decay of irradiated uranium was "rather complex", as Fermi wrote himself. After some chemistry, including coprecipitations with cerium and lanthanum ..., Fermi realized that the produced activity did not come from any known element with atomic number between 83 and 92. Hence the conclusion: this activity originates from a new element with atomic number larger than 92.

A prudent remark by Ida Noddack-Tacke, who together with her husband Walter Tacke and with Otto Berg discovered the element rhenium in 1925, could have shown the way to Enrico Fermi and to the whole radiochemical community. A few months after Fermi's publication she wrote:

"Es wäre denkbar, dass bei der Beschissung schwerer Kerne mit Neutronen diese Kerne in mehrere grössere Bruchteile zerfallen, die zwar Isotope bekannter Elemente, aber nicht Nachbarn der bestrahlten Elemente sind."

She therefore advised to exclude first the presence of light elements. Using an artificial mixture of different elements she made clear that many of them still passed through the separation methods used by Fermi. In spite of this notice and in spite of the frequent contacts of Ida Noddack with scientists as Otto Hahn, it was only at the end of 1938 that light elements as alkaline earth metals have been observed in the products from uranium, irradiated with neutrons.

How was this discovery realized? Here we can refer to the competition between the laboratory of Frédéric Joliot and his wife Irène Curie, and the one of Otto Hahn and Lise Meitner. Indeed, Irène Curie and her collaborator Paul Savitch had observed a radio-element with a half life of 3.5 hours; they carefully called the element
The previous years each publication on the product-elements of uranium had been deluged with names as EkaOs, EkaRe and other Eka's. Some quotation from this work:

"Nos expériences nous ont montré par la suite que $\text{Ra}_3^{3,5}h$ n'est pas un isotope du thorium, mais présente des propriétés semblables à celles des terres rares" 

"Mélangé avec du lanthane, on peut le séparer presque complètement du thorium" 

"Dans l'ensemble, les propriétés de $\text{Ra}_3^{3,5}h$ sont celles du lanthane, dont il semble jusqu'ici qu'on ne puisse le séparer que par fractionnement." 

Finally there was the unambiguous determination by Otto Hahn and Fritz Strassmann that during the irradiation of uranium with neutrons in fact elements as barium, lanthanum and cerium are being formed. Hahn has formulated this result in following sibylline terms:

"As Chemiker müssten wir aus den kurz dargelegten Versuchen das oben gebrachte Schema eigentlich umbenennen und statt Ra, Ac, Th die Symbole Ba, La, Ce einsetzen. Als der Physik in gewisser Weise nahestehende 'Kernchemiker' können wir uns zu diesem, allen bisherigen Erfahrungen der Kernphysik widersprechenden, Sprung noch nicht einschließen. Es könnten doch noch vielleicht eine Reihe seltsamer Zufälle unsere Ergebnisse vorgetäuscht haben." 

Table 1: Decay schemes, as observed by Hahn and Strassmann. To the right of these data we give the actual assignment of isotopes and half-lives.

<table>
<thead>
<tr>
<th>Element</th>
<th>Half-life</th>
<th>Element</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaI/143Ba</td>
<td>&lt; 1 Min/20s</td>
<td>AcI/143La</td>
<td>&lt; 30 Min/14,3 min</td>
</tr>
<tr>
<td>RaII/142Ba</td>
<td>14 ± 2 Min/10,7 min</td>
<td>AcII/142La</td>
<td>&lt; 2.5 Std/92,5 min</td>
</tr>
<tr>
<td>RaIII/139Ba</td>
<td>86 ± 6 Min/82,7 min</td>
<td>AcIII/139La</td>
<td>- mehrere Tage?/stable</td>
</tr>
<tr>
<td>RaIV/138Ba</td>
<td>250 ± 30 Std/12,8 d</td>
<td>AcIV/138La</td>
<td>&lt; 40 Std/40,2 h</td>
</tr>
</tbody>
</table>

a. Ref. 4
b. "Als Chemiker müssten wir eigentlich sagen, bei den neuen Körpern handelt es sich nicht um Radium, sondern um Barium" the authors wrote. The same applies for Ac/La.

Yet this carefully formulated discovery indicated for many specialists that the previously undiscussible stability of the atomic nucleus was no more so sure. Lise Meitner, together with her nephew Otto Frisch, could give a physical explanation to the break-up of the nucleus. Bringing together the liquid-drop model and an electrostatic repulsion was sufficient to explain a reduced stability of the shape of uranium nuclei, and to lead to the final fission into two parts.

Why didn't brilliant scientist like F. Joliot, E. Fermi and others even take into
consideration the break-up of a nucleus? According to O. Frisch⁷, first the analogy between a nucleus and a liquid drop had to be made, and this model was not available at the first suggestion of I. Noddack. Apparently the hard chemical fact of the presence of light elements was needed to break up this "liquid drop" into parts. It gives to think that everyone, involved with the difficult discovery of the fission exerted himself to demonstrate the uselessness of Ida Noddack's remarks. O. Frisch's arguments are not that convincing:

"Ida Noddack, a German chemist, quite rightly pointed out that they might be lighter elements; but her comments (published in a journal not much read by chemists...and hardly at all by physicists) were regarded as mere pedantry. She did not indicate how such light elements could be formed; her paper had probably no effect whatever on later work."

F. Strassmann also just gives a partial explanation:

"Im Jahre 1934 hatte die Chemikerin Ida Noddack zwar schon die Hypothese vertreten, dass ein Kern auch in mehrere Bruchstücke zerfallen könne, sie stützte ihre Meinung aber nicht durch Bezugnahme auf kernphysikalische Modellvorstellungen. Ihre Ansichten konnten darum weder von Fermi noch von anderen Kernphysikern ernst genommen werden. Denn das Uran von 23⁷ H.Z. war β-Strahler und ging dadurch in ein Element 93 über. Fermi hat es zwar nicht identifiziert, aber er hat es erzeugt. Da es aus Uran 238 entstand, das im Uran zu 99,3 % vorhanden ist, hatte er mit seiner Behauptung also recht. Für die restlichen 0,7 % (Uran 235) wurde die Berechtigung zur Erwartung eine Spaltung modellmässig erst durch Lise Meitner und ihren Neffen Otto Robert Frisch gegeben. Mit ihrer Hypothese hatte I. Noddack also nur einen Zufallstreffer erzielt."

It is right that the 23 min half-life (of uranium 239) corresponds to the beta decay, leading to the element with Z = 93, unknown at that time. Yet Fermi had very clearly mentioned a 13 (thirteen) min as well as many other half-lifes, which did not belong to the element uranium.

Even O. Hahn refers to Noddack's suggestion in a footnote of his Nobel lecture⁹, a reference that may report perfectly the mentality of "damals":

"Von noch anderer Seite (Ida Noddack) wurde sogar der Einwand gemacht, man müsse zunächst einmal alle Elemente des Periodischen Systems ausschliessen, bevor man die Behauptung aufstellen könnte, ein Element 93 zu haben. Dieser Einwand wurde damals als allen physikalischen Vorstellungen über Kernphysik widersprechend nicht ernstlich diskutiert."

A final false note, related to the discovery of nuclear fission. In 1945 the Nobel Prize for Chemistry of 1944 (reported that year) was awarded to Otto Hahn for the discovery of fission. This surely was a disappointment for Strassmann and Meitner, both closely involved in this discovery. Fritz Strassmann was the co-author, who after the results of Curie and Savitch did everything to prove unquestionably the presence of for instance the element barium¹⁰. For Lise Meitner this should have been even a greater disappointment. Since 1912 she had been working as an Austrian
citizen together with Otto Hahn in the Kaiser-Wilhelm Institut in Berlin-Dahlem, where she specially took care of the nuclear physics basis of the measurements, further developing the existing equipment. After the "Anschluss" in March 1938 she lost the last protection (a foreign passport) against the increasing persecution of Jews. With the help of Otto Hahn and the Dutch physicists Peter Debye and Dirk Coster she could escape to the Netherlands in July 1938 (ref. 11). This happened exactly at the decisive phase in the discovery of nuclear fission. Indeed, on July 12, 1938 the Journal de Physique received the publication of Curie and Savitsch³, and Hahn and Strassmann sent their work on the discovery of light elements in neutron-irradiated uranium to the editor on December 22, 1938.

With satisfaction we can premise that the scientific community unseparably involved the names of Meitner and Strassmann to the discovery of fission. Thus, in Berlin a new institute was called the "Hahn-Meitner Institut für Kernforschung"; in 1966 Hahn, Meitner and Strassmann received the Enrico Fermi award. Even at the Nobel seminar "Von den natürlichen Umwandlungen des Urans zu seiner künstlichen Zerspaltung" Otto Hahn refers five times to F. Strassmann - also five times to O. Frisch - and ten times to his long-time collaborator Lise Meitner. People who can wait until November 1995 (the statute of the Nobel Foundation includes a 50 year's wait) can examine all the exact facts of this case in the documents of the Nobel Committee.

Attempts to secrecy

Everyone was impressed by the enormous energy released by fission, millions of times more than in the case of the strongest chemical reactions. Imagine that in the fission process more than one neutron might be released and you are confronted with the possibility of a "chain reaction" and of - up to then unknown - powerful weapons. This prospect, at a moment that Europe was threatened by dictatorial regimes, made some scientists propose a secrecy of all results in connection with fission. All these attempts were almost complete failures. Only Fermi, who at first had entered a protest against these proposals of secrecy, changed his mind. Looking for good "moderators", namely materials that do slow down the fast neutrons from fission without absorbing them too much, he discovered together with H.L. Anderson of the Columbia University that pure graphite had a very small cross section for neutron absorption. Under the influence of Leo Szilard, Fermi eventually agreed to keep secret this information, turning out to be very essential for the operation of nuclear chain reactors.

EPILOGUE: How the first discovery of a fission product was ignored

In this Epilogue we give arguments for the statement that Walter Noddack, Ida Noddack-Tacke and Otto Berg should be credited for the discovery of the element with Z = 43. We also explain that, due to lack of knowledge of the phenomenon "fission", this discovery was unduly ignored.

How has it been possible that the advice from I. Noddack about a better search for
lighter elements in neutron-irradiated uranium was so completely neglected? One of
the reasons seemed to be that Ida Noddack-Tacke had lost all credibility with a
previous publication, together with Walter Noddack and Otto Berg\textsuperscript{13} viz. the discovery
of the elements \( Z = 75 \) (rhenium) and \( Z = 43 \) (masurium).

The discovery of the element \( Z = 43 \) could not be confirmed later on, such that -
except of course for the well-confirmed existence of rhenium - the work of Noddack,
Tacke and Berg was fully ignored, and that one had to wait for the observation of the
element \( Z = 43 \) by Perrier and Segrè.\textsuperscript{13} It was artificially producted in E.O.
Lawrence's cyclotron by bombardment of molybdenum for some months by a deuterium
beam. The new element was later called "technetium" because it was the first element
that seemingly did not exist in nature and had to be produced by technical means. At
the same time the authors of ref. 13 were heavily criticized for their "mistake" in
the earlier "discovery" of the element 43, a mistake that apparently removed all
their credibility.

Before having a closer look at the results from Noddack, Tacke and Berg in ref. 13,
we must realize that the element \( Z = 43 \) is only encountered in nature as a product of the
spontaneous fission of \(^{239}\text{U}\), phenomena unknown in these years. With the actual data

\[
T_{\alpha}^{(99\text{Tc)}} = \text{2.1} \cdot 10^5 \text{ y}
\]

\[
fission \text{ yield } ^{99}\text{Tc} = 6.1 \%
\]

\[
T_{\alpha}^{(239\text{U sf})} = 6 \cdot 10^{15} \text{ y}
\]

we obtain following estimate for the concentration of the \( Z = 43 \) element relative to
uranium, when we assume that both elements are in equilibrium in a uranium-containing
ore:

\[
\frac{m (Z = 43)}{m (U)} = \frac{0.06 \cdot 2.1 \cdot 10^5}{2 \cdot 6 \cdot 10^{15}} = 1.1 \cdot 10^{-12}
\]

Following observations and comments on the results of ref. 13 are important:

1) in "Platinerz", an ore that contains elements from Cr to Cu, Ru to Ag and Os to
Au, but not a single trace of uranium, only the element \( Z = 75 \) is observed without
any indication for the presence of the element \( Z = 43 \);

2) in "Columbite" an ore with - according to the authors - percentage quantities of
many transitional elements, among them uranium, the authors observed in most cases
the element 43 and only rarely also the element 75;

3) the authors estimate that the combination of their chemical separation and the
applied analysis of roentgen-ray spectra gives them in general a detection limit
of at least \( 10^{-9} \);

4) the observed energies and intensities of the roentgen rays indicate \underline{without any}
doubt the presence of the element with \( Z = 43 \), see annex;

5) from their data the strongest \( Z = 43 \) roentgen ray (K\(\alpha_1 \)) still seems to be much
weaker than weak transitions for the element 75; therefore we estimate the element
Quantities to be at least a factor of 100 smaller than those for the element 75; this brings the detection limit for the element 43 below $10^{-11}$.

Assuming that the investigated ore contained 5% of uranium, we get an estimated concentration of $6 \times 10^{-14}$ for the element 43; one needs only to increase the crude estimates for the detection of the element 75 ($10^{-10}$ instead of $10^{-9}$) and the intensity difference 43/75 in the roentgen spectra ($10^{-3}$ instead of $10^{-2}$) to come very close to the expected concentration of element 43 in the investigated ores; viz. $10^{-13}$ to $6 \times 10^{-14}$.

If both numbers differed by many orders of magnitude, one might be forced to question the evidence of the first four observations. In our opinion the opposite is true: by simply adding to the detection limits of ref. 13 our estimated intensity difference for the element 43, relative to the element 75, no major argument against a well-commented experimental result can be raised.

We therefore may state that Ida Noddack-Tacke came close to "nuclear fission" twice in her career, first in 1925 by touching a fission product, and second in 1934 by suggesting literally how "fission" could have been discovered; twice her work and the work of her collaborators was ignored.

COROLLARY

One may raise the question about the name of the element 43, a question to be decided upon by qualified instances.

Some arguments are in favour of the original name MASURIUM (Ma) given after Masurien, a region south-east in the previous Ostpreussen, where probably the Noddacks originated from:

1) Introducing again the name "masurium" would correct the tragedy the Noddacks had to live with;
2) Giving the element 43 the technologically ambitious name "technetium" ignores the fact that this element has always been present in nature by metric-ton quantities.
3) Perrier and Segrè did not question the results of ref. 13; they even took profit of the chemical expertise as published by the Noddacks.

ANNEX: The data of the roentgen ray analyses

As these data contain both the strongest evidence for the identification of the element 43 and the indication for its detection limit, we prefer to summarise these data, as given in table 4 of ref. 13.
For wavelength measurements the linewidths were reduced to 0.001 Å, yielding an energy resolution of about $1.6 \times 10^{-3}$. The authors claim correctly that no other known intense line can explain the observed $Z = 43$ roentgen rays; only a second order diffraction of Nd $K\alpha_2$ might coincide with the first order of $43 K\alpha_1$, but except for the strange appearing of Nd after the applied chemistry one should also observe then the more intense Nd $K\alpha_1$ and this was not the case.

The experimental intensities for $Z = 75$ are taken from a drawing that intends to reproduce the registered spectra. No similar drawing is given for the element 43 but the authors claim that the 43 lines appeared in the expected intensity ratio, viz mostly only the strongest $K\alpha_1$ and in a few cases also the $K\alpha_2$ and $K\beta_1$. The two- to four-times larger difference between experimental and calculated wavelength values gives us another hint that these lines are much weaker than the weakest $Z = 75$ lines.

<table>
<thead>
<tr>
<th>Designation</th>
<th>$Z = 43$</th>
<th>$Z = 75$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K\alpha_1$</td>
<td>0.672</td>
<td>1.4299</td>
</tr>
<tr>
<td>$K\alpha_2$</td>
<td>0.675</td>
<td>1.4407</td>
</tr>
<tr>
<td>$K\beta_1$</td>
<td>0.601</td>
<td>1.235</td>
</tr>
<tr>
<td>$L\alpha_1$</td>
<td>1.4306</td>
<td>1.2048</td>
</tr>
<tr>
<td>$L\alpha_2$</td>
<td>1.4406</td>
<td></td>
</tr>
<tr>
<td>$L\beta_1$</td>
<td>1.2355</td>
<td></td>
</tr>
<tr>
<td>$L\beta_2$</td>
<td>1.2041</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Δ (exp-th) mA</th>
<th>-1.4</th>
<th>-2.9</th>
<th>+1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative intensity b)</td>
<td>100</td>
<td>50</td>
<td>20</td>
</tr>
<tr>
<td>Experimental</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

a) The authors refer to calculations by M. Siegbahn; in the meantime change in calibration factors increased the wavelengths by about 2.1 promille. Once this correction factor applied, these calculated values correspond to the actual ones to within 0.000,2 Å (J.A. Bearden, NYO-10586, Oak Ridge (1964)).

b) These intensities are estimated, taken partially from A.E. Sandström in Encyclopedia of Physics XXX, Berlin (1957) p. 78-245.
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Abstract

This contribution is mainly dealing with two aspects of photofission studies. In a first part the photon absorption and the probability that this photon absorption is followed by fission of the compound nucleus is discussed. A general overview of the behaviour of the photofission cross section for photon energies between 10 and 300 MeV is given. More detailed information about the photofission cross section and the fission probability in the barrier and the giant resonance region is given. A second part of this contribution treats the characteristics and the distributions of the fragments emitted in photofission. A few examples of results obtained with 12-70 MeV bremsstrahlung are presented. The interest of future experiments with 6-12 MeV bremsstrahlung is shown.

I. INTRODUCTION

In studying photofission one can emphasize the investigation of the photon absorption and the probability that this photon absorption is followed by fission of the excited nucleus. Or on the other hand one can be interested in the study of the characteristics of the fragments emitted in photofission. In this contribution we will consider essentially the fission of heavy (actinide) nuclei that are excited with photons with an energy below ~ 20 MeV, that means that we will concentrate our attention to the region from the barrier up to the giant resonance. For such a situation the formed compound nucleus will decay by the emission of 1 or 2 neutrons or it will undergo fission, directly or after it was first emitting a neutron.

The experiments that we performed in our laboratory were in the first place concerned with the study of the characteristics of the fission fragments produced in the photofission of actinides with bremsstrahlung with endpoint energy between 12 and 30 MeV. Experiments were started at our new linac to extend these studies for bremsstrahlung with endpoint energies below 12 MeV. Before treating our own work on the fragment characteristics we will first discuss photofission cross section measurements.

II. PHOTON ABSORPTION AND FISSION PROBABILITY

A. General Aspects

An interesting general schematic overview of the behaviour of the photofission cross section was given already 30 years ago by Katz et al. These authors studied the photofission of $^{238}$U using bremsstrahlung with endpoint energy between 8 and 24 MeV. The yield of a few mass chains was determined by radiochemical methods and the photon difference method was used to deduce photofission cross sections for different fragment masses. Combining their results with previously
reported results, they constructed a 3-dimensional $\sigma(A,h\nu)$ surface, giving the photofission cross section as a function of the fission product mass and the energy of the photon inducing the fission. This surface was constructed for $10\text{MeV}<h\nu<300\text{MeV}$. It is shown in fig.1. Cuts parallel to the $(\sigma_{Yf},h\nu)$ plane give cross sections for the production of a fission product with given mass. Cuts parallel to the $(\sigma_{Yf},A)$ plane give mass distributions for fission induced with photons with a given energy. In fig.2 examples of photofission cross sections for symmetric mass (mass 115) and asymmetric mass (mass 139) are given. The cross sections show the well known giant resonance shape, but the maxima for symmetric fission (at $\approx 18$ MeV) and asymmetric fission (at $\approx 14$ MeV) are not coinciding, and the high energy tail, above $h\nu=25$ MeV, behaves quite different. For symmetric fission the cross section increases from a minimum value of $0.2$ mb at $h\nu=25$ MeV to $0.7$ mb at $h\nu=300$ MeV, which is practically the same as the peak value in the giant resonance region. On the other hand the cross section decreases with increasing photon energy for asymmetric fission, so that for high-energy photon induced fission the mass distribution becomes symmetric (with a FWHM of $\approx 20$ mass units).

B. Barrier to giant resonance region for $^{235,236,238}\text{U}$ and $^{232}\text{Th}(\gamma,F)$

The older cross section measurements were done with bremsstrahlung, having a continuous energy spectrum. The more recent experiments are performed with "monoenergetic" photons. These can be obtained using the tagged photon method or using photons produced by the annihilation in flight of monoenergetic positrons.

This can be illustrated e.g. with the recent work of Caldwell et al.\textsuperscript{2}). These authors studied the $(\gamma,n)$, $(\gamma,2n)$ and $(\gamma,F)$ processes from the $(\gamma,n)$ threshold or from the fission barrier up to 18 MeV for $^{235,236,238}\text{U}$ and $^{232}\text{Th}$. They used photons produced by the annihilation
in flight of positrons. The photon energy resolution was typically \( \sim 250 \) keV at 10 MeV up to \( \sim 325 \) keV at 18 MeV. This photon beam impinges on the U or Th sample, that is located at the centre of a high-efficiency 4\( \pi \) neutron detector. The neutron detector consists of a 61-cm cube of paraffin in which are embedded 48 BF3 proportional counters, arranged in an array of 4 coaxial rings, each containing 12 counters, cylindrically symmetric about the beam line. For a nuclear reaction the number of counts in each ring is measured, which provides the probability to determine not only the average number of neutrons \( \langle \bar{n} \rangle \) emitted in that reaction, but also the average neutron energy. This latter is obtained from the ratio of counts in the different rings. In the experiment, one-line the number of events in which one, two, three,... up to 8 neutrons were detected is recorded, together with the ring distribution for each category of event. From the data the number of \( (\gamma, n) \), \( (\gamma, 2n) \) and \( (\gamma, F) \) events is deduced and together with the average number of neutrons also the average neutron energy for a given photon energy.

In fig. 3 we show their \( \sigma(\gamma/\text{tot}) \), \( \sigma(\gamma/n) \), \( \sigma(\gamma/2n) \) and \( \sigma(\gamma/F) \) results for the photofission of \( ^{232}\text{Th} \) and \( ^{232}\text{U} \). The total photonuclear cross sections \( \sigma(\gamma/\text{tot}) = \sigma[(\gamma/n)+ (\gamma/2n)+(\gamma/F)] \) are given in part a of the figure. They have for the four investigated nuclei about the same peak value and the same integrated value up to 18 MeV, except for \( ^{236}\text{U} \) having values about 10% lower. They have also practically the same shape, being clearly split like for other statically deformed nuclei. The \( (\gamma/n) \), \( (\gamma/2n) \) and \( (\gamma/F) \) cross sections are given in the parts b, c and d of the figure. As the total photon-absorption cross sections are roughly the same, which is also expected from sum-rule considerations, increasing fissionability of the compound nucleus should correspond to increasing \( (\gamma/F) \) cross sections and as a consequence to decreasing \( (\gamma/n) \) and \( (\gamma/2n) \) cross sections. Thus the lowest \( \sigma(\gamma/n) \) and \( \sigma(\gamma/2n) \) is expected for \( ^{235}\text{U} \) and the highest for \( ^{232}\text{Th} \), in agreement with the experimental results. We can also remark that the measurement of only one of the \( (\gamma/n) \), \( (\gamma/2n) \) or \( (\gamma/F) \) cross sections would not give the correct view of the shape of the giant resonance. So would e.g. the \( \sigma(\gamma/n) \) even suggest that the considered nuclei are even not statically deformed.

Typical is also the sharp rise of the \( (\gamma/F) \) cross section just above the second chance fission barrier, \( B_p(\gamma, \text{nf}) \), reflecting the enhanced fission probability by the opening of this new fission channel.

Just below the \( (\gamma/n) \) threshold relatively large peaks are observed in the \( \sigma(\gamma/F) \) for \( ^{232}\text{Th} \) and \( ^{238}\text{U} \). These can be explained by the competition between fission and neutron channels. This is illustrated in fig. 4, where the fission probability \( P_f = \sigma(\gamma,F)/\sigma(\gamma/\text{tot}) \) is given versus the photon energy. Following Gavron et al.\(^3\) the heights of the fission barriers are related closely to the shapes of these \( P_f \) curves. The height of the inner barrier is related to the shape near "threshold", and that of the outer barrier to the asymptotic value. The shape of the curve for \( ^{232}\text{Th} \) indicates that a sizable component of the fission proceeds via an axially symmetric inner barrier, while for the other systems the inner barrier has to be essentially axially asymmetric.
Fig. 3. Photonuclear cross sections for $^{238}$U and $^{232}$Th: (a) $\sigma(\gamma,\text{tot})$, (b) $\sigma(\gamma,n)$, (c) $\sigma(\gamma,2n)$, (d) $\sigma(\gamma,F)$

The experimental set-up (ring-ratio technique) allowed Caldwell et al. to decompose $\sigma(\gamma,F)$ into its two components: $\sigma(\gamma,f)$ and $\sigma(\gamma,nf)$. In fig. 5 the ratio $\sigma(\gamma,f)/[\sigma(\gamma,f) + \sigma(\gamma,nf)]$ is shown for $^{236}$U and $^{238}$U. It appears that for both systems the first and second-chance photofission cross sections are approximately equal a few MeV above $B_F(\gamma,nf)$.

Below the $(\gamma,nf)$ barrier the $\Gamma_n/\Gamma_F$ ratio (giving the neutron emission to fission probability ratio for a certain level) is obtained directly from $\sigma(\gamma,n)/\sigma(\gamma,F)$. At higher energies one must separate $\sigma(\gamma,F)$ into $\sigma(\gamma,f)$ and $\sigma(\gamma,nf)$ in order to obtain $\Gamma_n/\Gamma_F$, which is then given by $\frac{\Gamma_n}{\Gamma_F} = \frac{\sigma(\gamma,n) + \sigma(\gamma,2n) + \sigma(\gamma,nf)}{\sigma(\gamma,F)}$. 


Fig. 4. Fission probability, $\sigma(\gamma,f)/\sigma(\gamma,\text{tot})$, for $^{235,236,238}\text{U}$ and $^{232}\text{Th}$.

The $\Gamma_n/\Gamma_f$ result for $^{236}\text{U}$ is shown in fig. 6. $\Gamma_n/\Gamma_f$ reaches an asymptotic value above 9 to 10 MeV. As a consequence its value determined at $\sim$ 11 MeV should be characteristic for the studied system.

C. Photofission cross section of $^{240,242,244}\text{Pu}$

We studied in our laboratory $\sigma(\gamma,f)$ for $^{240,242,244}\text{Pu}$ up to 30 MeV. The resulting $(\gamma,f)$ cross sections are given in fig. 7. They are deduced from fission product yield measurements for fission induced with bremsstrahlung with endpoint energy between 10 and 30 MeV, using the photon difference technique. The fission product yields were measured in our experiments, using four solid state heavy ion detectors placed in front of the $^{240}\text{Pu}$, $^{242}\text{Pu}$, $^{244}\text{Pu}$ target. Between 5 and 7.5 MeV the cross section data of Rabotnov et al. were used. In the remaining energy region, from 8.0 to 9.5 MeV, the cross section was determined by interpolation between the data of Rabotnov et al. and ours. The photofission cross section shows the giant resonance structure, again without exhibiting the splitting that can be expected for the total photon absorption cross section of statically deformed nuclei. The position of the maximum at $\sim$ 14 MeV and the width of the distribution (FWHM $\sim$ 7 MeV) is practically the same for the three cases. The maximum photofission cross section value however decreases.

Fig. 5. First chance to total photofission cross section ratio for $^{236}\text{U}$ and $^{238}\text{U}$. 
Fig.6. Neutron to fission branching ratio, $\Gamma_n/\Gamma_f$, for $^{236}$U.

Photon Energy (MeV)

<table>
<thead>
<tr>
<th>Photon Energy (MeV)</th>
<th>$\Gamma_n/\Gamma_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>1</td>
</tr>
<tr>
<td>10</td>
<td>2</td>
</tr>
<tr>
<td>14</td>
<td>3</td>
</tr>
</tbody>
</table>

Fig.7. Photofission cross section for (a) $^{240}$Pu, (b) $^{242}$Pu and (c) $^{244}$Pu.

from 340 mb for $^{240}$Pu over 290 mb for $^{242}$Pu to 250 mb for $^{244}$Pu. Above the giant resonance region, between 20 and 30 MeV, the cross section is low (≈ 20 to 30 mb) and constant within the error bars.

Assuming, as observed by Caldwell et al., that the total photon absorption cross section is practically independent of the $Z$ and $A$ of the actinide under study, and that the $\Gamma_n/\Gamma_f$ ratio is constant above 9 MeV, and by adopting from the same authors that $\Gamma_n/\Gamma_f = 1.4$ for $^{235}$U, we can deduce from our results $\Gamma_n/\Gamma_f = 0.63 \pm 0.15$ for $^{240}$Pu, $1.02 \pm 0.17$ for $^{242}$Pu and $1.14 \pm 0.19$ for $^{244}$Pu. Our $\Gamma_n/\Gamma_f$ results, when plotted as a function of $B_f-B_n$ ($B_f$ being the fission barrier and $B_n$ the neutron binding energy), show the same exponential dependence on $B_f-B_n$ as the results of Caldwell et al. and those deduced from 2-MeV neutron induced fission cross sections (see eg. ref.9) for even-even fissioning systems.

This is illustrated in fig.8, where also the results for even-odd fissioning systems are included. It was shown by Vandenbosch and Huizenga that in a simple model with a constant temperature nuclear level density expression this exponential dependence of $\Gamma_n/\Gamma_f$ on $B_f-B_n$ can be expected. These authors show also that the displacement between even-even and even-odd systems is equal to $\Delta f + \Delta_n$, with $\Delta f$ the neutron pairing gap for a system at the saddle point and $\Delta_n$ the neutron pairing gap for a system at equilibrium deformation. From fig.8 we can deduce $\Delta f + \Delta_n = 1.2$ MeV. This value is quite reasonable as $\Delta_n = 0.6$ MeV as deduced from even-odd fluctuations in the ground state mass behaviour in this mass region.

III. FISSION FRAGMENT CHARACTERISTICS AND DISTRIBUTIONS

In the foregoing we dealt essentially with the photon absorption and the probability that this absorption is followed by fission of the excited system. Our experimental photofission work was concentrated in the first place on the investigation of the characteristics of the fragments and their distributions. Although experiments were performed with bremsstrahlung with endpoint energy between 12 and 70 MeV
special attention was payed to the giant resonance region. With our new linac experiments are started with bremsstrahlung with endpoint energy between 6 and 11 MeV, which will extend our experimental results down to the vicinity of the fission barrier.

Basically we are using two different experimental techniques: energy correlation measurements and $\gamma$-spectrometric methods. In the energy correlation measurements the energy of two fragments is measured using surface barrier detectors. Energy and provisional mass distributions are deduced from these measurements. $\gamma$-spectrometric methods permit the determination of independent and cumulative yields, from which postneutron mass distributions, charge distributions, isomeric ratios and initial fragment spins are deduced. By combining provisional and postneutron mass distributions neutron emission curves and preneutron mass distributions can be obtained. Details about our experimental techniques can be found in ref.5,10,11 and in some references given there in.

I will treat her only a few typical examples and show that it is interesting to study them at lower bremsstrahlung endpoint energy.

A. Fine structure and odd-even proton effects

1. Photofission of $^{238}\text{U}$

In the postneutron mass distribution for $^{238}\text{U}(\gamma,F)$ we observed an increased yield around mass 134 (see fig.9). In a more detailed investigation an increased production of the $Z=52, N=82$ $^{134}\text{Te}$ could be demonstrated. Wilkins et al. showed that in their model the production of $^{134}\text{Te}$ will be favoured over that of $^{132}\text{Sn}$ due to the tendency to conserve in the fragments the $N/Z$ ratio of the compound nucleus. So that the increased yield of masses around 134 should, at least for a large part, be attributed to a $N=82$ closed shell effect.

Odd-even proton effects were not observed.
in our experiments. Assuming that the probability that an existing proton pair at
the saddle point will survive the transition from saddle point to scission point is
22% for $^{238}\text{U}(\gamma,\text{f})$—this is the same value as for $^{235}\text{U}(\text{n}_{\text{th}},\text{f})$—ref. 14—we can esti-
mate proton odd-even effects of $\sim 5\%$ and $\sim 3\%$ for the photofission of $^{238}\text{U}$ with re-
spectively 12- and 15-MeV bremsstrahlung. This cannot be distinguished within the ex-
perimental accuracy.

Making the same assumptions as above, it is expected that by decreasing the
bremsstrahlung endpoint energy down to 8 MeV or lower the proton odd-even effects
will increase to 10 to 20%, which would be observable within the experimental accu-
racry. At the same time, shell effects—being more important at lower excitation ener-
gy of the compound nucleus, an increase of the fine structure around mass 134
can be expected.

2. Fission of $^{240,242,244}\text{Pu}$

The provisional mass distributions for the spontaneous fission of $^{240}\text{Pu}$,
$^{242}\text{Pu}$ and $^{244}\text{Pu}$ show a sharp peak in the vicinity of mass 134-135 and a shoulder in
the mass region 143-144 (see ref.4-6). This sharp peak around mass 134-135 can again
be explained by the preferential formation of a $N=82$ shell stabilized fission frag-
ment with low deformation. Especially for $^{242}\text{Pu}$ s.f. this effect is very well expressed,
showing a very high peak yield of $\sim 9\%$ around mass 135. These structures are redu-
ced drastically for $^{239}\text{Pu}$ and $^{241}\text{Pu}(\text{n}_{\text{th}},\text{f})$ and they have practically disappeared
for the photofission of $^{240}\text{Pu}$, $^{242}\text{Pu}$ and $^{244}\text{Pu}$. In fig.10 the results for the fission of
$^{242}\text{Pu}$ are given as an example.

Thermal neutron capture in $^{239,241}\text{Pu}$ results in an excitation energy of
$\sim 6.5$ MeV in the formed $^{240,242}\text{Pu}$ compound nuclei. In the photofission of $^{240,242,244}\text{Pu}$
with bremsstrahlung with an endpoint energy of 7 to 8 MeV, the average excita-
tion energy of the compound nucleus will be comparable to the excitation energy
of the compound nuclei $^{240,242}\text{Pu}$ formed in $^{239,241}\text{Pu}(\text{n}_{\text{th}},\text{f})$. Thus performing photo-
fission experiments with 7 to 8 MeV bremsstrahlung will enable us to compare directly,
for the same excitation energy, the fission of the compound systems $^{240}\text{Pu}$ and $^{242}\text{Pu}$,
once formed predominantly in a $J^\pi=0^+$ or $1^+(^{240}\text{Pu})$ and $2^+ or 3^+(^{242}\text{Pu})$ state by thermal neutron capture in $^{239}\text{Pu}$
or $^{241}\text{Pu}$.

B. Kinetic energy behaviour for $^{232}\text{Th}$ fission

David et al. studied the total kinetic energy (TKE) of the fragments for
the fission of $^{232}\text{Th}$ via the reaction $^{232}\text{Th}(^4\text{He},^4\text{He}'\text{f})$ at $E(\text{He})=120$ MeV. Triple
coincidence experiments between the $^4\text{He}'$ and the two fission fragments were performed.
These authors found (see fig.11) that below $\sim 8$ MeV the average TKE increases with
increasing excitation energy ($E_x$) of the compound nucleus, with the slope $\frac{d\text{TKE}}{dE_x}=1.5$.
Between 8 and 12 MeV the TKE remains constant, or decreases slightly. Such an abnor-
mal behaviour of the TKE for the fission of $^{232}\text{Th}$ at excitation energies just above
the barrier has been observed already earlier. The statistical accuracy of the measure-
ments of David et al.\(^{15}\) didn’t allow an unambiguous conclusion concerning the dependence of the slope \(\frac{d\text{TKE}}{dE_x}\) on the fragment mass. Within the experimental accuracy, this slope would be independent of the fragment mass as well for \(E_x < 8\) MeV as for \(E_x > 8\) MeV. The same authors studied also the fragment TKE for the fissioning systems \(^{236}\text{U}(^4\text{He},^4\text{He}')\), \(^{235}\text{U}(d,pf)\) and \(^{238}\text{U}(^4\text{He},^4\text{He}')\). For these cases they found that the slope \(\frac{d\text{TKE}}{dE_x}\) is 0 over the entire excitation energy region studied (up to be second chance fission barrier).

An excitation energy of \(\approx 8\) MeV for \(^{232}\text{Th}\) corresponds practically to \(B_f + \Delta(B_f\text{ the fission barrier, } \Delta\text{ the pairing gap})\). The increase of TKE in the region of \(B_f, B_f + \Delta\) for the fission of \(^{232}\text{Th}\), and the non observation of it for heavier systems was correlated by David et al.\(^{15}\) to low damping in the lighter actinides, and a rapid increase of the defect of damping with \(Z/A\). One can remark that also proton odd-even defects decreases when moving from thorium through uranium to plutonium and californium\(^{16}\).

This behaviour of TKE with changing \(E_x\) was unexpected. It was not observed in other fissioning systems. However it can be correlated to the \(\overline{V}(E_x)\) behaviour observed by Calwell et al.\(^{17}\) in the study of the photofission of \(^{235,236,238}\text{U}\) and \(^{232}\text{Th}\). The \(\overline{V}(E_x)\) results of these authors are given in fig. 12. As expected, for \(^{235,236,238}\text{U}\) the number of emitted neutrons increases with increasing photon energy over the whole \(E_x\) interval. This is also the case for \(^{232}\text{Th}\) for photon energies above 8.2 MeV. However for photon energies between 6 and 8.2 MeV \(\overline{V}\) decreases for increasing excitation energy of the compound nucleus \(^{232}\text{Th}\).

We want to study the fission of \(^{232}\text{Th}\) with bremsstrahlung with endpoint energy between 6 and 11 MeV. Combining energy correlation measurements and \(\gamma\)-spectrometric results will yield information of the behaviour of TKE, TKE (\(m^*\)), \(\overline{V}\) and \(\overline{V}(m^*)\) with changing \(E_x\).
IV. CONCLUSIONS

Photofission reactions make use of the simplicity and directness of electromagnetic interactions as a powerful tool with which to explore the process of nuclear fission. It enables the study of systems that cannot be reached, or only with a lot of experimental difficulties, in other nuclear reactions. E.g. a complete study (post- and preneutron results) of $^{232}\text{Th}$, $^{238}\text{U}$,...becomes possible. In addition, as photon absorption is predominantly El absorption, fission of even-even systems, like $^{232}\text{Th}$, $^{238}\text{U}$, $^{240}\text{Pu}$,...can be studied from states with a well defined $J^\pi$ value. This spin value(1) is much lower than the average spin of the system formed with charged particle induced reactions. However, the use of bremsstrahlung, having a continuous energy spectrum, or the difficulties and loss of intensity associated with the production of monoenergetic photons restrict the possibilities of obtaining accurate, detailed and systemative data in photofission experiments.

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Neutron induced fission below and at the fission barrier.

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Abstract

After a brief reminder of the properties of intermediate structure in sub-barrier fission cross sections, the neutron induced fission of $^{242}$Pu is discussed as an example of a nucleus for which the cross section from the low resonance region up to and through the fission threshold can be understood with a common fission barrier model. Finally, the evidence for a third minimum in the deformation potential of light actinides is shortly reviewed.

1. INTRODUCTION: reminder of sub-barrier fission.

One of the most dramatic consequences of Strutinsky's (1) suggestion in 1965 that the fission barrier of most actinides should have a double-humped shape, was the occurrence of a pronounced narrow intermediate structure in sub-barrier fission cross sections. The situation is recalled with the aid of fig.1: The deformation potential (minimum total energy of the nucleus as a function of deformation) has two minima which correspond to states of the nucleus with normal and extended deformation. The excited levels indicated in the figure for these two values of deformation, which are called class I and class II states, respectively, are meant to include all possible excitations, not only vibrations. The class I states above neutron separation energy are excited as compound nuclear resonances when a neutron interacts with an actinide target nucleus, because their deformation is close to the one of the target ground state. The class II states at similar excitation act as doorways to the fission channel: Only if a resonance is close in energy to a class II state, the nucleus may undergo a transition from the first to the second well, and once this has occurred, the probability for it to undergo fission is large.

For energies several hundred keV below the top of the intermediate barrier, the coupling between class I and class II states is sufficiently weak to be described by perturbation theory: The class II state mixes with only a few neighbouring class I resonances, and its fission width, determined mainly by the penetrability of the outer barrier, is spread over the resulting mixed states. Often the coupling is so weak, that one of them still carries a major fraction (70 to 80%) of the original class II state and thus the same relative amount of its fission width. Again because of the weak coupling, this quasi-class II state has a very small neutron width,
usually much smaller than its fission width. Since the fission yield from this resonance is then practically independent of its fission width, it will be very difficult to measure the fission width. Quite often, the quasi-class II state will even have escaped detection due to its very small neutron width. This difficulty to measure the largest part of the fission strength of a class II state presents the major problem in an attempt to determine the fission barrier parameters from intermediate structure data.

There are only a few cases of intermediate structure resonances where one is completely sure to have detected the quasi-class II state and thus the bulk of the fission strength. One example is the lowest class II state in $^{238}$U+n, where accurate radiative capture data (2,3) have shown that the resonance at 721 eV neutron energy has a much smaller capture width than the neighbouring resonances and thus is very likely to be predominantly class II. Another instructive example is the structure at 1405 eV in $^{240}$Pu, because at the same time it provides a textbook example for the mixing of two nearly degenerate levels. The data (fig.2) (4) show two strong resonances which comprise 99% of the total fission width within this class II.
cluster. The fission widths of the two strong resonances are inversely proportional to their neutron widths. Therefore, they are interpreted as resulting from the mixing of the class II state providing the fission width with essentially only one (almost degenerate) class I level which provides the neutron width. The constructive interference in the region between the two resonances is an inherent feature of this situation.

We conclude this lengthy introduction with the remark that intermediate structure in fission provides the only example of doorway states where this term can be taken literally as an almost "geographical" doorway in configuration space.

2. THE NEUTRON INDUCED FISSION CROSS SECTION OF $^{242}$Pu

Fig. 3 shows the measured (5) fission cross section of $^{242}$Pu in some selected energy intervals, from a few eV to a few MeV. At the lowest energies one observes resolved class II clusters, i.e. the individual peaks are class I fine structure resonances. In the keV region this fine structure is no longer resolved, and the observed peaks represent individual class II states. At a few hundred keV also the class II states are no longer resolved, but we still observe structure due to the statistical fluctuations of their parameters. In the threshold region, finally, also this structure is averaged out.

We want to describe the fission cross section in all four regions with one common set of barrier parameters. In the threshold region the fission cross section is reproduced with the aid of a statistical model calculation. Apart from the fission barrier parameters, the calculated cross section depends on neutron transmission coefficients and on the density of transition states above the lowest barrier. The transmission coefficients for the elastic neutron channels are obtained from strength functions given in the literature (for details see ref.5), for the inelastic neutron channels the same strength functions are used in the lack of better knowledge. Since the applied statistical model routines (FISINGA-CAMEL, ref.6) require individual transition states rather than merely a state density, a specific sequence is found which fits the cross section best. However, the quality of the fit is not very sensitive to the detailed sequence which therefore has no particular meaning, but it is only sensitive to the total number of states within the first 400 keV above the lowest barrier which has been chosen in accord with the transition state density as proposed by Lynn (7). The final fit to the fission cross section in the threshold region is shown in fig.4 where also the barrier parameters used for this fit are indicated.

The height and curvature of the higher (inner) barrier are quite well determined from the position of the threshold and the slope of the cross section at threshold. In contrast, the parameters of the lower (outer) barrier are very uncertain, because
Fig. 3: The fission cross section of $^{242}\text{Pu}$ in selected energy intervals.
they depend strongly on the choice the other parameters needed, some of which like the transmission coefficients for the inelastic neutron channels, are badly known. A better estimate of the parameters of the lower barrier is obtained below from the resonance data at lower energies.

![Statistical model fit to the fission cross section of 242Pu in the threshold region.](image)

The class II state on which most information is available is the one at 762 eV, which is shown in fig. 3a. But even here we are faced with the problem that the fission width of the quasi-class II state at 762 eV is not experimentally known because the neutron width of this resonance is much smaller than its fission width which therefore cannot be determined from the fission yield. Nevertheless, it is possible in this case to obtain a reasonable estimate of the class II fission width primarily from the fission widths of the other resonances within this cluster together with an estimate of the coupling strength from the neutron width of the quasi-class II state. The resulting class II fission width of about 0.5 eV requires an outer barrier height of about 5.5 MeV, thus clearly higher than the uncertain estimate from the analysis of the threshold region, but in agreement with the statistical analysis in the intermediate region discussed below.

Finally we want to analyse the statistical properties both, of the class II resonances observed up to 70 keV neutron energy, and of the structure observed in the measured cross section up to 500 keV. For this purpose, a Monte Carlo routine is used which generates sequences of class I and class II levels with spin $J \leq 5/2$ from the relevant distribution laws. First, a sequence of class II levels with average spacing $D(II) = 1000$ eV (for spin 1/2 and correspondingly smaller for higher spin) is generated from a Wigner distribution. Their fission and coupling widths for each fission channel are sampled from Porter-Thomas distributions with expectation values calculated from the relevant fission barrier parameters. The sampled sequence of class I states with average spacing $D(I) = 17.5$ eV are given...
neutron widths also sampled from Porter-Thomas distributions with energy dependent expectation values corresponding to given neutron strength functions. The same sequence of transition states and neutron strength functions are assumed as in the statistical model calculations in the threshold region. The coupling between class I and class II states is then introduced in the usual way, again allowing for Porter-Thomas fluctuations of the individual fine structure fission widths. From the sequence of resonances constructed this way, the contribution of each class II level to the fission cross section is calculated, and the result compared to the experimental observation.

In the energy range below 70 keV, the quantities compared between the statistical calculation and the experiment are (i) the number of class II structures with a fission area above the energy-dependent experimental detection limit and (ii) their average fission area. Several simulations have been performed with different fission barrier parameters, and for each parameter set the simulation has been repeated a sufficient number of times to obtain meaningful averages and variances of these quantities. A comparison of the simulation which yielded the best result, with the experimental data is shown in table 1.

Table 1: Monte Carlo Simulation of sub-barrier fission resonances.

<table>
<thead>
<tr>
<th>Neutron energies [keV]</th>
<th>Number of levels</th>
<th>Average Area [b eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1-20</td>
<td>20-70</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Simulation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>32 ± 4.5</td>
<td>61 ± 4.4</td>
<td>4.4 ± 1.0</td>
</tr>
<tr>
<td>4.4 ± 1.0</td>
<td>6.2 ± 0.8</td>
<td></td>
</tr>
<tr>
<td>Experiment</td>
<td>29</td>
<td>67</td>
</tr>
<tr>
<td>3.6</td>
<td>8.3</td>
<td></td>
</tr>
</tbody>
</table>

The same parameter set yields the simulated fission cross section in the 200 to 500 keV range which is compared to the experimental cross section in fig.5. The similarity of the two cross sections of fig.5 with respect to magnitude and period of the oscillations is obvious. In order to get a more quantitative account of this similarity, the following quantities are calculated for both, the experimental and the simulated cross section:

(i) the relative variance

\[ v = \frac{1}{n-1} \sum (1 - o_i/o_s)^2 \]

where the sum is over the n data points representing the cross section o in a given energy interval, and o_s is a smooth cross section of the form \( o_s = \exp(aE - b) \) where a and b are adjusted such as to minimize v.

(ii) the number of runs r for o being continuously above (h) and below (l) o_s.
The quantities \( v, h, l, r \) and its expectation value \( E(r) \) for an uncorrelated statistical data set, are compared in table 2. The fact that \( r \) is smaller than its statistical expectation by about a factor of two tells that non-statistical structure is present in both data sets which, of course, does not come as a surprise. The interesting point is the good agreement between the \( r \)-values of the simulation and the experimental data, demonstrating that the fluctuations in the latter are well reproduced with the intermediate structure parameters used in the simulation.

![Graphs showing measured and simulated fission cross sections](image)

**Fig. 5:** measured (a) and simulated (b) fission cross section of \(^{242}\text{Pu}\) between 200 and 500 keV.

**Table 2:** Comparison of structures in measured and simulated fission cross section between 200 and 500 keV neutron energy.

<table>
<thead>
<tr>
<th>( E_n ) (keV)</th>
<th>Simulation</th>
<th></th>
<th></th>
<th></th>
<th>Experiment</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>( v )</td>
<td>( h )</td>
<td>( l )</td>
<td>( E(r) )</td>
<td>( r )</td>
<td>( v )</td>
<td>( h )</td>
<td>( l )</td>
<td>( E(r) )</td>
</tr>
<tr>
<td>200-300</td>
<td>0.073</td>
<td>62</td>
<td>120</td>
<td>83±6</td>
<td>45</td>
<td>0.047</td>
<td>93</td>
<td>89</td>
</tr>
<tr>
<td>300-400</td>
<td>0.016</td>
<td>54</td>
<td>54</td>
<td>55±5</td>
<td>28</td>
<td>0.015</td>
<td>46</td>
<td>62</td>
</tr>
<tr>
<td>400-500</td>
<td>0.014</td>
<td>33</td>
<td>41</td>
<td>38±4</td>
<td>20</td>
<td>0.016</td>
<td>32</td>
<td>42</td>
</tr>
</tbody>
</table>

Thus, apart from the inaccurate estimate of the height of the lower barrier from the statistical model fit in the threshold region, the measured data from a few eV neutron energy up to above 1 MeV can be understood with one common set of parameters for the double-humped fission barrier and the associated class II states.

3. **THE THIRD MINIMUM IN THE DEFORMATION POTENTIAL OF LIGHT ACTINIDES**

In contrast to the Pu- and heavier isotopes, the fission cross section of most of the light actinides shows pronounced peaks in the threshold region. One of the best studied examples is the case of \(^{230}\text{Th}\) shown in fig.6. The strong narrow peak is interpreted as a vibration (possibly the ground state vibration) in a shallow minimum of the deformation potential; the minimum must be shallow in order to explain the obvious lack of damping of the vibration into other degrees of freedom which in the Pu-isotopes produces the rather smooth cross section shape. The present
belief is that this shallow minimum is the third rather than the second one because of two reasons:

1) Calculations of potential energy surfaces by Moller and Nix (8) showed a deep rather than a shallow second minimum and a first barrier much lower than the neutron binding energy, but they also showed the development of a shallow third minimum on top of the second barrier when mass asymmetric deformations were allowed for in the calculations.

2) High resolution fission cross section measurements performed at Geel some years ago (9) revealed a fine structure which has two features supporting the third minimum hypothesis: As is shown in fig.7, the most convincing fit of the fine structure components requires two rotational bands with $K = \frac{1}{2}$ (necessary to explain the generally forward peaked angular distribution) and opposite parity, as is expected for a reflection asymmetric potential well. Secondly, the moment of inertia of the nucleus extracted from these rotational bands is about three times the moment of inertia in the ground state, whereas the moment of inertia in the shape isomeric state of the second well is only twice the ground state value (10).

What can be done to further substantiate the third minimum hypothesis? The most obvious thing would be to verify the angular momentum assignments of the individual members of the proposed rotational bands of fig.7 by measurements of the fission fragment angular distribution. Some angular distribution data do exist (11-13), but with the exception of those of ref.13 their neutron energy resolution is insufficient to resolve the rotational band components; the data of Veeser and Muir (13) which have played an important role in selecting the particular rotational band of fig.7 among several possible choices, have been taken under two angles (100 and 125 degrees), only. Thus the definite angular distribution measurement with a resolution at least comparable to the one of fig.7, is still missing. Although considerable progress has been made in the development of a suitable detector (14),

Fig. 6: The fission cross section of $^{230}$Th in the threshold region.
Another possibility has been pursued by Blons et al. (15,16): If the compound nucleus is excited by a (d,p) reaction, states with higher angular momenta are populated much more strongly than in neutron capture. Thus the high spin members of the rotational bands should pop up in 230Th(d,pf) as compared to 230Th(n,f). Their results are shown in fig. 8: Not only are the already assigned rotational band components 5/2+ and 7/2− increased according to expectation, but also the higher members of the rotational bands not seen in the (n,f) reaction, especially the 9/2+, 11/2− and 13/2+ levels, are found at essentially the predetermined locations. Inclusion of the exact locations of these levels into the over-all fit changes the rotational band parameters only very slightly. This success must be looked at as a strong support of the description of the 230Th data by two rotational bands with opposite parities and thus of the third minimum hypothesis.

Fig. 7: Fit of the 230Th fission cross section around 715 keV by two K=1/2 rotational bands with opposite parity.

Fig. 8: Comparison of 230Th(d,pf)16 with 230Th(n,f) in the region of the K=1/2 rotational bands this is still a very difficult measurement which is on the limit of feasibility with present day neutron sources.
How about neighbouring nuclei? An interesting case has since long been the $^{231}\text{Pa}(n,f)$ reaction, because it has very strong isolated resonances at low neutron energies where the experimental resolution is still very good. The data of Plattard et al.\(^{(17)}\) are shown in fig. 9. The tall resonance at 157 keV is assigned $(K,J^m) = (3,3^+)$ on the basis of its peak cross section and of angular distribution data by Sicre \(^{(18)}\). The associated $4^+$ rotational state is unobservable because it requires $I=3$ neutrons which at this low energy means too small a cross section. The first two members $(I=2)$ of the expected $K=3^+$ band may be associated with the narrow peaks at 173 and 189 keV. Thus the data do not provide direct evidence for rotational bands with opposite parities, but they are in accord with this assumption.

A basically more simple situation would be encountered in an even-even compound nucleus where one could expect isolated $K=0$ bands as the lowest rotational bands. Unfortunately, with neutron capture in an even-odd target one generally ends up above fission threshold and thus cannot study sub-barrier states. Blons et al.\(^{(16)}\) have investigated $^{229}\text{Th}(d,pf)$. The data which are not yet analysed, are shown in fig. 10. The observed fine structure is surprisingly weak. A possible explanation might be that the two barriers which enclose the third minimum have a rather different transparency. Nevertheless, we may expect with interest a detailed analysis of these data.

Fig. 9: The measured \(^{17}\) near-barrier fission cross section of $^{231}\text{Pa}$

Fig. 10: The fission probability of $^{230}\text{Th}$ as obtained by Blons et al.\(^{(16)}\) from $^{229}\text{Th}(d,pf)$
References.

APPLICATION OF SOME FISSION PROPERTIES TO NEUTRON DOSIMETRY

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Abstract

Two applications are treated:

1. Neutron flux determination by distinct fission fragment absolute radiometric measurements on fission foils.
2. Neutron flux determination by detection of the delayed neutrons emitted by a fissile sample.

For both applications the measuring technique, the calibration procedure, the application domain, the advantages of the method as well as the restrictions in absolute neutron flux determination are considered. As the detection technique for both applications is of a totally different nature, they are presented in separate sections.

1. NEUTRON FLUX DETERMINATION BY DISTINCT FISSION FRAGMENT ABSOLUTE RADIOMETRIC MEASUREMENTS ON FISSION FOILS

The specific absolute activity at the end of irradiation \( A(i) \), for a fission product \( i \), is related to the absolute fission rate \( \sigma_f \phi \), hence the absolute neutron flux \( \phi \) by:

\[
A(i) = \frac{N_0}{A} \cdot \alpha \cdot \sigma_f \cdot \phi \cdot Y_i \cdot f(\lambda_i T)
\]

with

- \( N_0 \): number of Avogadro
- \( A \): atomic mass
- \( \alpha \): isotopic abundancy
- \( \sigma_f \): fission cross-section
- \( \phi \): neutron flux
- \( Y_i \): fission chain yield
- \( f(\lambda_i T) \): activation function.

The activation function \( f(\lambda_i T) \) reduces for a square irradiation profile to \( (1 - e^{-\lambda_i T}) \), with \( T \) the irradiation time and \( \lambda_i \) the decay constant for the measured fission product.

When measuring a gamma-ray belonging to a fission product in radioactive equilibrium with its parent nucleus relation (1) has to be multiplied by
\[
\left( \frac{\lambda_d}{\lambda_d - \lambda_p} \right)
\]

with \( \lambda_d \): decay constant of the daughter nucleus,

\( \lambda_p \): decay constant of the parent nucleus.

The specific absolute activity at the end of irradiation \( A(i) \) is related to the number of counts \( N \) in a corresponding gamma-peak in the measured gamma-spectrum of the fission foil by:

\[
A(i) = \frac{N}{t_m} \cdot \frac{1}{\epsilon} \cdot \frac{1}{I_\gamma} \cdot \frac{\lambda_1 t_m}{1 - e^{-\lambda_1 t_m}} \cdot e^{-\lambda_1 t_d}
\]

(2)

with \( t_m \): measuring time

\( t_d \): time between the end of irradiation and starting time of the measurements

\( \epsilon \): absolute efficiency of the detector at the gamma-ray energy

\( I_\gamma \): gamma-ray emission probability of the measured gamma-line

\( \lambda_1 \): decay constant of the fission product measured.

Further corrections are applied to relation (2) for:

i) geometrical differences of the used fission foils compared to the absolute calibration sources

ii) self absorption in the fission foil

iii) fission fragment escape from the fission foil

iv) corrections for competing reactions

v) correction for flux perturbation caused by the fission foil.

The main advantage of the use of fission foils for neutron dosimetry lies in the large variety of fission fragments with half-lives ranging from very short to very long (for example \( ^{137}\text{Cs} \); \( T_{1/2} = 30.0 \text{ year} \)), which means that a fission foil can be used as a perfect integrator of the neutron environment for short as well as for long irradiation periods. The disadvantage however when performing direct radiometric measurements on distinct fission fragments lies in the uncertainty on the obtained absolute fission rate \( q_f \) which is mainly governed by:

* the uncertainty on the fission yields : 2 % up to more than 8 %.

Others might more or less contribute to the overall uncertainty on the absolute fission rate such as:

* measuring statistics

* decay constant of the fission product measured

* gamma-ray emission probability of the measured gamma-line

* uncertainties related to correction applied (see above).
Since all mentioned contributions are uncorrelated, propagation in quadrature can be applied to yield the overall uncertainty on the absolute fission rate, which is typically of the order of 3% to 4% if only fission products with well-known yields are taken into account. However, better accuracy can be obtained using the following calibration procedure, based on a back-to-back irradiation of a fission foil with a similar deposit in a double, parallel-plate absolute fission chamber. This procedure allows to link directly the radiometric measurements to the absolute fission chamber measurements. The accuracy is now contained in some "K-factors" linking directly the measured fission product activities to the absolute fission chamber measurements. With this calibration procedure an accuracy of typically 1.5% to 2.0% can be reached.

As a by-product of this calibration procedure one can look at those "K-factors" in terms of fission chain yields and see how well they agree with fundamental information stemming from many independent approaches or/and well-documented evaluations. This approach has been applied during a measuring campaign at the French Masurca fast critical facility in Cadarache and results are given in table I.

Table 1: $^{235}U$, Mass chain yields for fast reactor spectrum; comparison between this work, ILRR and other compilations

<table>
<thead>
<tr>
<th>Mass chain</th>
<th>This work</th>
<th>ILRR 1)</th>
<th>Crouch 2) (a)</th>
<th>Preliminary ENDF/B-VI 3) (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{95}Zr$</td>
<td>0.0661 (+ 3.2%)</td>
<td>0.0645 (+ 2.2%)</td>
<td>0.0643 (+ 1.8%)</td>
<td>0.0641 (+ 0.5%)</td>
</tr>
<tr>
<td>$^{97}Zr - 97\text{Nb}$</td>
<td>0.0595 (+ 3.0%)</td>
<td>0.0603 (+ 2.4%)</td>
<td>0.0588 (+ 1.8%)</td>
<td>0.0598 (+ 0.5%)</td>
</tr>
<tr>
<td>$^{99}Mo$</td>
<td>0.0609 (+ 3.2%)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{103}Ru$</td>
<td>0.0332 (+ 3.0%)</td>
<td>0.0333 (+ 2.3%)</td>
<td>0.0329 (+ 2.6%)</td>
<td>0.0324 (+ 1.0%)</td>
</tr>
<tr>
<td>$^{131}I$</td>
<td>0.0346 (+ 3.3%)</td>
<td>0.0331 (+ 4.3%)</td>
<td>0.0323 (+ 1.9%)</td>
<td>0.0321 (+ 0.7%)</td>
</tr>
<tr>
<td>$^{132}Te - 132I$</td>
<td>0.0470 (+ 2.9%)</td>
<td>0.0483 (+ 6.1%)</td>
<td>0.0463 (+ 3.0%)</td>
<td>0.0465 (+ 0.7%)</td>
</tr>
<tr>
<td>$^{133}I$</td>
<td>0.0709 (+ 3.0%)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{140}Ba - 140La$</td>
<td>0.0612 (+ 2.9%)</td>
<td>0.0610 (+ 1.9%)</td>
<td>0.0595 (+ 1.5%)</td>
<td>0.0595 (+ 0.7%)</td>
</tr>
<tr>
<td>$^{143}Ce$</td>
<td>0.0581 (+ 5.8%)</td>
<td>0.0517 (+ 8.7%)</td>
<td>0.0574 (+ 2.1%)</td>
<td>0.0572 (+ 0.5%)</td>
</tr>
<tr>
<td>$^{147}Nd$</td>
<td>0.0224 (+ 4.7%)</td>
<td></td>
<td></td>
<td>0.0222 (+ 3.1%)</td>
</tr>
</tbody>
</table>

(a) European compilation.

(b) Most recent (1983) U.S. compilation.
The obtained yields are generally in good agreement with other reported values within the quoted uncertainties, which gives confidence in our absolute efficiency calibration of the Ge-detector system as well as to the data set used for calculation of the absolute reaction rate $\sigma_{f}\phi$. From this reaction rate value, the flux is than obtained by division by the environmental neutron spectrum averaged cross-section value.

II. NEUTRON FLUX DETERMINATION BY DETECTION OF THE DELAYED NEUTRONS EMITTED BY A FISSIONABLE SAMPLE

This technique has been thoroughly studied at SCK/CEN, Mol, during the last few years. Its objective consisted in developing a high-sensitivity neutron yield diagnostics monitor for JET which is amenable to automatic control with on-line data acquisition and analysis. The principle of this diagnostic consists in irradiating a fissile target during the plasma pulse, transferring it to a delayed neutron counting assembly, measuring the delayed neutron emission within a time span of $\sim 10$ to $100$ s and recycling the target for the next pulse.

The delayed neutron counter consists of 6 one inch diameter $^3$He counters, with a sensitive length of 30 cm, embedded in a cylindrical polyethylene block of 40 cm diameter, at a radial distance of 8 cm from a central hole which fits the counter end of the pneumatic conveyor. The whole system is protected against background neutrons by means of a 1 mm Cd screen and a 10 cm thick polyethylene shield.

Two fission reactions, $^{238}$U(n,f) and $^{232}$Th(n,f) have been identified as ideal fusion plasma neutron yield diagnostics monitors since both are little perturbed, due to their effective energy thresholds of $\sim 1.5$ MeV, by neutron scattering effects on structural materials under the 2.45 MeV spectral peak conditions prevalent in D,D regime.

The calibration of the system is based on irradiations performed at the BRL fission spectrum 4), since its average neutron energy is comparable with the average neutron energy of the fusion plasma neutron spectrum in the D,D phase; actually corrections due to the variation of delayed-neutron yields with incident neutron energy are only a few percent between these two neutron spectra (superposition, calculations).

The relation, between the number of counts in the delayed neutron counter and the neutron flux $\phi$, based on the empirical KEEPIN six-group representation of the delayed neutron data is given by:

$$N_c = c \cdot P \cdot \frac{N^0}{A} \cdot \sigma_f \cdot \phi \cdot t_m \cdot a_t \cdot \sum_{i} a_i \left(1 - e^{-\lambda_i t_d}\right) e^{-\lambda_i t_m} \left(1 - e^{-\lambda_i t_m}\right)$$
with \( \varepsilon \) : efficiency of the detector

\( P \) : weight of the target

\( a_t \) : total absolute delayed neutron yield

\( a_i \) : fractional delayed neutron yield for group \( i \)

\( \lambda_i \) : decay constant for group \( i \).

the other symbols have the same meaning as in section I.

Again as in section I, one could determine the absolute efficiency of the delayed neutron counter using calibrated neutron sources and use literature data for the delayed neutron characteristics to determine the neutron flux \( \Phi \). To show which problems occur when using this method, we have represented in table 2 the total absolute delayed neutron yield for the fast reactor neutron induced fission in \( ^{232}\text{Th} \) and \( ^{238}\text{U} \) from recent compilation work of R.J. Tuttle 5) and R.W. Waldo et al. 6) and compared to the earlier Keepin et al. data 7).

<table>
<thead>
<tr>
<th></th>
<th>Keepin et al. 7)</th>
<th>R.J. Tuttle 5)</th>
<th>R.W. Waldo 6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{232}\text{Th} )</td>
<td>0.0496 ± 0.0020</td>
<td>0.0545 ± 0.0011</td>
<td>0.05577 ± 0.0040</td>
</tr>
<tr>
<td>( ^{238}\text{U} )</td>
<td>0.0412 ± 0.0017</td>
<td>0.04508 ± 0.00060</td>
<td>0.04435 ± 0.0023</td>
</tr>
</tbody>
</table>

Although no differences in fractional group yield data nor in decay data were found between the listed references, one sees that important differences in absolute yields occur. Even, if accepting the most recent Waldo-evaluation, again the accuracy of the flux determination will be mainly governed by the accuracy of the absolute delayed neutron yield (5 to 7 %).

This mentioned difficulty is again, as in section I, bypassed by calibration of the detector system against absolute fission chambers using \( ^{232}\text{Th} \) and \( ^{238}\text{U} \) deposits. The accuracy is now contained in some calibration factor \( K \) (in first approximation equal to \( \varepsilon \cdot a_t \)) linking directly the countrate in the delayed neutron counter to the absolute fission chamber measurements. An accuracy of typically 1.5 % to 2 % is reached.

More details on the delayed neutron counting systems as well as on the verification of constancy of the calibration factor with varying irradiation, decay and measuring times are given in 8).
III. CONCLUSIONS

As a general conclusion we can stress that the two neutron dosimetry techniques, the first being applicable in fission reactor devices and the second in fusion reactor devices, can yield, using the mentioned calibration procedures, reaction rate values with an accuracy of about 1.5% to 2%. As the link with the neutron flux is determined through an average cross-section value at the irradiation position, the accuracy of the neutron flux will mainly be governed by the accuracy of the fission cross-section data available. It is not the aim of this paper to report on the accuracy of fission cross-sections, but in order to give the differential cross-section specialist some indication to this respect in terms of reactor applications we refer to table 3: this shows the results of integral fission cross-sections measured at MOL in the benchmark $^{235}\text{U}$ fission neutron spectra and they are compared with values calculated from ENDF/B-V cross-section data for various evaluations of the $^{235}\text{U}$ fission neutron spectrum.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Experimental cross-section (mb)</th>
<th>ENDF/B-V Watt Spectrum</th>
<th>Madland-Nix &quot;Exact&quot; Spectrum</th>
<th>NBS Evaluated $^{235}\text{U}$ Fission Spectrum</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}\text{Th}(n,f)$</td>
<td>$83 \pm 3.1%$</td>
<td>0.904</td>
<td>0.907</td>
<td>0.872</td>
</tr>
<tr>
<td>$^{233}\text{U}(n,f)$</td>
<td>$1949 \pm 3.1%$.</td>
<td>0.978</td>
<td>0.978</td>
<td>0.981</td>
</tr>
<tr>
<td>$^{235}\text{U}(n,f)$</td>
<td>$1200 \pm 1.9%$.</td>
<td>1.030</td>
<td>1.029</td>
<td>1.030</td>
</tr>
<tr>
<td>$^{238}\text{U}(n,f)$</td>
<td>$312 \pm 2.3%$.</td>
<td>0.978</td>
<td>0.984</td>
<td>0.945</td>
</tr>
<tr>
<td>$^{237}\text{Np}(n,f)$</td>
<td>$1359 \pm 2.1%$.</td>
<td>0.991</td>
<td>0.973</td>
<td>0.973</td>
</tr>
<tr>
<td>$^{239}\text{Np}(n,f)$</td>
<td>$1818 \pm 1.9%$.</td>
<td>0.985</td>
<td>0.982</td>
<td>0.982</td>
</tr>
<tr>
<td>$^{240}\text{Pu}(n,f)$</td>
<td>$1332 \pm 2.1%$.</td>
<td>1.015</td>
<td>1.021</td>
<td>0.995</td>
</tr>
</tbody>
</table>

(a) These are macroscopic cross-sections averaged over the thermal-neutron induced uranium-235 fission neutron spectrum.

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SESSION II

LIGHT CHARGED PARTICLE AND NEUTRON EMISSION IN FISSION

Chairman: E. Jacobs
R.U. Gent
THEORETICAL APPROACHES TO THE EMISSION OF α PARTICLES DURING NUCLEAR FISSION

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Abstract
Recent models as well as new trajectory calculations used to interpret the experimental data on α-accompanied fission are presented and experimental hints towards the emission mechanism are pointed out. The important question of the distribution of the emission points and moments is brought up: are these particles coming only from the region between the fragments (the neck) or, are they coming with comparable probability, from any point of the surface of the fissioning system? and: are they emitted just at scission or at any sufficiently deformed stage of the fission process?

The simultaneous division of the nucleus into three fragments (the true ternary fission) is discussed in terms of the dynamical liquid-drop model. It is suggested that this process occurs in the tail (corresponding to overelongated shapes) of the distribution of the fission probability in the multidimensional space of the collective coordinates.

I. INTRODUCTION
The alpha-accompanied fission i.e., the detection of α particles in coincidence with the two fission fragments (fig. 1), has been constantly considered with a particular interest mainly due to the observed angular distribution of these particles which is strongly peaked perpendicular to the fission axes. This feature implies that the α particles are emitted from an extremely deformed nucleus and therefore they could supply information about the least known fission stage: the scission.
II. STANDARD TRAJECTORY CALCULATIONS AND THEIR INHERENT AMBIGUITIES

How has one tried to extract this information, about the manner in which the nucleus breaks, which these α particles might carry along? One has usually assumed simple distributions, e.g., gaussian, for the quantities defining the nuclear configuration at the moment of the alpha particle emission and scaled them to the experimental distributions via the calculation of the trajectories of the α particle and of the two fragments in their mutual (mainly Coulomb) field.

The initial distributions on which the time evolution of the system depends are listed below:

1) $V(M_1)$ is the experimental mass distribution corrected for neutron emission. From this, one can deduce $V(Z_1)$ assuming $N/Z = \text{const}$ during fission and $V(M_2)$ from the conservation of the total mass $A = M_1 + M_2 + 4$.

2) $V(D) = \exp (-\frac{(D-D_0)^2}{2\sigma^2})$. There are also the multipole deformations of the fragments $\epsilon_2, \epsilon_4, \ldots$ but usually this quantities are not treated explicitly.

3) $V(v_{F_1}) = \exp (-\frac{(v_{F_1}-\overline{v}_{F_1})^2}{2\sigma^2_{v_{F_1}}})$, $V(v_{F_2})$ being determined from the conservation of the linear momentum.

4) $V(x) = \exp (-\frac{(x-x_0)^2}{2\sigma^2_x})$

5) $V(y) = \exp (-\frac{(y-y_0)^2}{2\sigma^2_y})$

6) $V(px) = \exp (-\frac{(px-p_{x_0})^2}{2\sigma^2_{px}})$

7) $V(py) = \exp (-\frac{(py-p_{y_0})^2}{2\sigma^2_{py}})$

![Figure 1 Experimental configuration and initial parameters](image)
From 6) and 7)

\[ W(E_d) = W(p_x) W(p_y) = e^\sqrt{-E_d/(c_p^2/m_\nu)} \]

i.e., Maxwellian with \( T = \sigma_p^2/m_\nu \)

and

\[ W(\theta_d) = |p_x/p| = p_x/\sqrt{p_x^2 + p_y^2} \]

can be inferred.

Even without assuming anything about the nature of the process (i.e., the emission mechanism) the number of the initial parameters (which is at least 12)

can be slightly reduced from general considerations like energy conservation and uncertainty principles.

Supposing there is no way to transform Coulomb energy into internal fragment excitation and precession excitation into fragment kinetic energy:

\[ E_F + E_d + \sum_{i,j} Z_i Z_j e^2 \left[ (x_i - x_j)^2 + (y_i - y_j)^2 \right]^{1/2} = E_F + E_d \]

which means that \( D \) and \( \nu_\nu \) are not independent and we are left with 11 parameters.

There are two quantum mechanical uncertainties with completely different significances, which can be used:

a) the Heisenberg uncertainty principle:

\[ \Delta x \Delta p_x = \frac{\hbar}{2} \]

\[ \Delta y \Delta p_y = \frac{\hbar}{2} \]

which tells that two dynamical variables, the observables of which do not commute, like position and linear momentum, can not be determined simultaneously with infinite precision. \( \Delta \) means standard deviation

b) the energy-time uncertainty relation:

\[ \Delta E \Delta t = \hbar/2 \]

which tells that fluctuations appear in the energy of a system with very short lifetime.

The magnitudes of these effects are calculated in the Appendix.

Remembering that, the total width of the momentum components has also a contribution not determined by the uncertainty principle but by the statistical (random) nature of the fission process:

\[ \sigma_{p_x} = \sqrt{(\Delta p_x)^2 + \sigma_p^2} \]

\[ \sigma_{p_y} = \sqrt{(\Delta p_y)^2 + \sigma_p^2} \]
one can further reduce the number of initial parameters to 9 only if $v_0 = 0$
which is unjustified a priori.

These at least 9 parameters have to reproduce at most 9 measured quantities:
the mean values $\langle \theta \rangle$, $\langle \alpha \rangle$ and the widths $\Delta \phi_\theta$, $\Delta \phi_\alpha$ of the angular and energy distributions of the $\alpha$ particles, the total kinetic energy of the fragments $E_f$ and
the slopes of the following correlations:

$$E_F(\theta_\alpha), \quad E_\alpha(\phi_\alpha), \quad \Delta E_\alpha(\theta_\alpha) \quad \text{and} \quad \Delta \phi_\alpha(E_\alpha)$$

One should not count: a) correlations like $E_\alpha(\phi_\alpha)$ and, they are therefore redundant with the previous ones; or
b) inverse correlations like $\phi_\alpha(\theta_\alpha)$ since they are also not independent on the
direct ones. They are consequences of the same features of the initial
distributions. In this respect it would be worth using a realistic model to
determine the initial parameters on which each observable most strongly
depend along the lines of ref. 2.

It seems that, inspite of their large amount, the available experimental
results are neither totally independent nor have enough structures to provide
the amount of information necessary for the determination of the initial
distributions. The first to have pointed out that it is impossible, with the
standard procedure, to obtain unique values of the initial parameters was A.
Gavron 5 for the spontaneous fission of $^{252}$Cf. He was able to fit the data
with any initial fragment kinetic energy between 8 and 15 MeV and even with
larger values allowing $\gamma = 0$.

In conclusion, to remove these ambiguities one should try
- to increase the number of observables by looking for and exploiting possible
fine structures and
- to provide theoretical estimates for some of the initial parameters by
understanding the emission mechanism of the scission light particles.

Some progress made in the latter direction is presented in the next section.

III. THE PRECISION $\alpha$ CLUSTER EMISSION MODEL

Assuming that the $\alpha$ cluster existing in nuclei are the source of the fission
$\alpha$-particles (like in the $\alpha$ decay of the ground state), the distribution $W(x,y)$
is given by the probability \( |\psi_\alpha(x,y,D')|^2 \) ds of forming an \( \alpha \) cluster in the ds vicinity of a point \((x,y)\) on the nuclear surface at a fission stage defined by \( D' \). This probability was estimated by the overlap between the wave function of the emitting nucleus and that of the \( \alpha \) channel like in the R-matrix theory and a typical result is plotted in fig. 2. The emission points are in this model continuously distributed all over the surface of the fissioning system with a preference, only at scission, for the neck region.

The time available for the emission being less than \( 10^{-23} \)s, the particles must leave the nucleus with energies around the top of the barrier through penetration or overtransmission like in fig. 3. This implies there is an efficient mechanism, not well known at present, through which the distortion energy liberated between saddle and scission is transferred to the \( \alpha \) cluster till it escapes.

Figure 2: Distribution of the \( \alpha \) particle formation probability on the surface of \(^{238}\text{U}\) at two stages between saddle and scission.

Figure 4 shows the probability of escaping as a function of the \( \alpha \)-energy with respect to the top of the barrier. As far as there is enough energy to reach the barrier height, this probability is independent on the emission point. It is for this reason that the model does not favor, in agreement with the experiment, emissions from the
Figure 3: Barriers seen by an α particle in the neck of $^{238}$U.

Figure 4: Alpha spectrum at the moment of emission.

The escaping probability from fig. 4 corrected for the extra broadening due to quantum mechanical uncertainties (for an estimate see appendix) provides theoretical evaluations for two other initial distributions:

- the part corresponding to positive energies is identical with $W(E^+_{\alpha})$
- the other part corresponding to energies below the barrier can be related to the distribution of the emission points outside the ridge of the α-nucleus potential.

Figure 5: The sum of nuclear and Coulomb potential for an α particle in $^{238}$U immediately after its most probable scission. The value in MeV of the equipotential line are written above the right corner.
An important progress in the interpretation of the α-accompanied fission experimental data was the observation of the inaccuracy of the point-charge approximation in the calculation of the trajectories\(^2,3\)). The consideration of the real sizes of the fragments has two effects: 1) it restricts the available initial phase-space to the region outside the nuclear absorption. This region is limited by the ridge of α-nucleus potential which is marked by thick points in fig.5 One sees that even after the separation of the fragment surfaces (solid curves) the interfragment region is absorptive (i.e., forbidden) due to the nuclear diffusivity. 2) It allows the inclusion of the nuclear forces and improves the Coulomb forces.

The result of these finite-size calculations deserving most attention is the oscillating shape of the deflection function

Figure 6: Final angle \(\theta_{\alpha L}\) between α-particle and light fragment direction (top) for different initial α-positions all around the fissioning \(\alpha^\text{-emitters}\) (bottom)
\( \theta_{\alpha L} (z') \) as exemplified in fig. 6. It indicates that in addition to quantal interference effects, the resulting angular distribution could present structures of the type shown in fig. 7. \( \alpha \) emission with comparable probability from all the points around the fissioning system being the necessary condition for the appearance of these structures in the angular distribution, their experimental observation would definitively clear up the crucial question of the distribution of the emission points. Besides excellent angular resolution such an experimental requires good mass and energy resolutions to allow a certain selection of the initial configuration at scission. To avoid the blurring of the angle \( \theta_{\alpha L} \) between \( \alpha \) particle and the light fragment by neutron emission one should also select cold events i.e., with \( Q = (E_{\alpha} + E_{n})/Bn \).

The recently measured angular distributions for events selected according to the above mentioned criteria seem to present such structures as can be seen in fig. 8. Unfortunately even these most detailed and complete measurements...
have not the required precision to confirm or disprove this still attractive possibility.

V - REDUCING EXPERIMENTAL DATA TO DISTRIBUTIONS OF INITIAL PARAMETERS

We have seen in SecII that trajectory calculations have been generally used to obtain initial distributions (of about 6 independent quantities) which give, through the Monte Carlo method, the best fit to the experimental results.

Another, less common, use of the calculated trajectories is to find the correspondence between each experimental point (i.e., a set of measured quantities) and all possible trajectories (i.e., sets of values for the initial parameters) which lead asymptotically to the respective point. By subsequently weighting these initial values with the number of measured counts over the number of trajectories one can obtain distributions (of interdependent initial quantities) compatible with each experimental point.

For exemplification, the distributions obtained for two of the initial parameters leading to the same experimental point are shown in figure 9. Each
of the 40 rectangles represents a trajectory and trajectories with the same value of a given parameter ($D^*$, for instance) must have different values for at least one of the other parameters ($E_\omega^*$, for instance). Such plots show only the range of the possible values, giving an idea of the expected ambiguities arising from the finite resolution of the experimental points and from the number of initial parameters which is larger than the number of measured quantities. To further infer the distributions, really occurring during alpha-accompanied fission, these partial distributions depending whether the process is selective or not, should be multiplied or summed.

One should stress that this procedure is not a fit but rather another way of presenting the data and the results are therefore affected by the experimental errors.

VI - TERNARY FISSION AND THE DYNAMICAL LIQUID-DROP MODEL

As recent dynamical calculations in the frame of the liquid-drop model have shown', when sufficiently heavy nuclei $A > 300$ undergo fission they do not lead to compact binary shapes but they are rather forming a relatively long cylindrical neck which subsequently contracts at the extremities to divide the nucleus into three fragments (fig. 10). This behaviour can be understood in terms of the well known hydrodynamical instability $^{12,13}$ of a fluid cylinder of radius against perturbation with $\lambda > 27\pi^2$ (Plateau-Rayleigh).

![Diagram](Figure 10: Time evolution of nuclear shapes leading to true ternary fission)
Lighter nuclei, for which the maximum of the fission probability evolves in the binary valley of the multidimensional space of the collective coordinates, have a certain chance however (given by the tail of the distribution extending to shapes with unstable necks) to fission into three or more fragments.

For low energy fission, the shell effects will modify the above mentioned liquid-drop behaviour by favoring the α particles as middle fragments and therefore this represents an alternative possible origin for the fission α particles.

Acknowledgements

I am very grateful to all my friends and colleagues, members of the collaboration between I.K. Darmstadt and C.E.N. Bordeaux-Gradignan on light-particle accompanied fission, for allowing me to use some of their unpublished data and for numerous stimulating discussions.

APPENDIX

Let us try to estimate the minimum possible value for the width of $W(E_{\alpha})$ compatible with the uncertainty relations.

Using eq. (2) and assuming emission along y axes,

$$\Delta E_{\alpha} = \frac{h}{m_{\alpha}} \Delta p_{y} = \sqrt{2 \frac{2 E_{\alpha}}{m_{\alpha}}} \rho \Delta y_{0}$$

or

$$\Delta E_{\alpha} \left( \frac{\text{MeV}}{E_{\alpha}} \right) = 2.1 \left[ \frac{E_{\alpha} \left( \text{MeV} \right)}{m_{\alpha} \text{MeV}} \right]^{1/2} / \Delta y_{0} (\text{fm})$$

$\Delta y$ is model dependent and has two contributions:
- the radial extension of the preformation probability in the surface region which is about $35 \text{ fm}^2$;
- the extension of the emission points beyond the ridge of the barrier (due to penetration) which is about $1 \text{ fm}$.

From this, one obtains $\Delta E_{\alpha} = 2.3 \text{ MeV}$ for $E_{\alpha} = 1 \text{ MeV}$.

Eq. (3) can be written as $\Delta E (\text{MeV}) = 3.3 / \Delta t \left( 10^{-22} \text{s} \right)$. The time $\Delta t$ is:
- the penetration time in the precission cluster emission model $\tau_{p} = 5 \times 10^{-22} \text{s}$;
- the characteristic time for break-up ($\tau_{c}$) in the Rayleigh-unstable neck model $\tau_{c} = 6 \times 10^{-22} \text{s}$;
- one quarter of a period for a quadrupole oscillation in the sudden-snap picture $\tau_{q} = 4 \times 10^{-22} \text{s}$.

Although the definition of $\Delta t$ depends on the assumed mechanism, its value seems to be model independent and it gives $\Delta E_{\alpha} = 0.7 \text{ MeV}$.

In conclusion, the standard deviation should be at least $2.4 \text{ MeV}$ for $E_{\alpha} = 1 \text{ MeV}$. 
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Ternary Fission

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Abstract: This is a report on experimental results obtained with the double torus ionization chamber "DIOGENES", which has been developed for the investigation of particle associated fission. The role of ternary fission for the understanding of the fission process will be discussed.

1. Introduction

Since the discovery of charged particle accompanied or, as it is sometimes called, ternary fission in 1947, it has been the hope of physicists, that these particles, which are emitted in space and time close to the scission point, carry information on scission point parameters, i.e. nuclear shapes, precession fission fragment energies, fragment excitation energies and initial particle energies.

In order to investigate this complicated process it has turned out that it is inevitable to perform multiparameter measurements, i.e. to determine all relevant observables for each ternary fission event: fragment kinetic energies, fragment specific energy losses, particle kinetic energies and their specific energy losses together with all relative angles between the three reaction products. From these data main fragment masses, nuclear charges and the species of the third light charged particles can be derived.

For a detailed multiparameter measurement Manfred MUTTERER, Joachim PANNICKE and Klaus SCHEELE together with several students have developed the double torus ionization chamber "DIOGENES", a cross section of which is shown in figure 1 and a short description given in ref. 5).

For its first experiment on $^{235}\text{U}$ the detector has been operated at the high flux reactor of the Institut v. Laue - Langevin in Grenoble for the study of alpha particle accompanied fission. In the past, ternary fission experiments have been evaluated on the basis of particle trajectory calculations and most conclusions about scission point parameters depend on assumptions, on which these calculations are based.
Fig. 1
Cross Section through the detector system "DIOGENES"

Trajectory Calculation for Alpha Particles Emitted in Fission

VARYING EMISSION CONFIGURATIONS

\[ \rho_{cm} = 15.0 \text{ fm} \]
\[ \rho_{cm} = 23.5 \text{ fm} \]

Alpha particle angular and kinetic energy distributions for two different scission point configurations

Fig. 2
An example of trajectory calculations by Willy FLASSIG\(^7\) for well defined "frozen" scission point configurations with different nuclear elongations shows (figure 2) that different scission point shapes yield different alpha particle final angular and kinetic energy distributions. The question is, if the distributions of experimental observables like the alpha particle kinetic energy, the emission angle and the kinetic energy of the associated main fission fragments carry "fingerprints" of their corresponding scission point parameters. This question arises another one on the relevance of these scission point parameters for the binary fission process. Before I elaborate these questions in chapter 4, in the next chapter experimental results will be presented. In chapter 3 I shall introduce a new technique to deduce scission point parameters from multiparameter experimental data.

2. Experimental Results

The experimental results can be presented in different ways. The way chosen here shows yields as functions of two observables, figure 3 alpha particle yields as function of their emission angles and kinetic energies. The lower plot is a projection onto the angular axis.

Polar emitted alpha particles are easily separable from those of equatorial emission. Most probable values of particle observables are listed in table 1. Zero degree is defined by the direction of the light fragments flight path. Polar emission yield is integrated for "polar-light" from \(0^\circ - 25^\circ\) and for "polar - heavy" from \(155^\circ - 180^\circ\) respectively.

Figure 4 is a sequence of plots of main fragment intensities as functions of their masses and mean kinetic energies for the three emission types. In the two cases of polar emission the fragment, which determines the emission direction absorbs nearly all recoil momentum, the second main fragment receives also a small push. This can better be seen in figure 5.

The figures 6 and 7 are essentially representations of particle fragment correlations. They will be discussed in the last chapter in some detail.

3. Trajectory Calculations

In order to introduce our method elaborated by Frank KRASKE and Joachim PANNICKE to derive scission point parameter distributions from corresponding observables, the following notations will be used: \(E_\alpha^0\), \(\theta_\alpha^0\) and \(E_F^0\) for the initial alpha particle kinetic energy, emission angle relative to the light fragment and fragments' kinetic energies at the instant of particle emission, \(E_\alpha\), \(\theta_\alpha\) and \(E_F\) stand for the corresponding final observables.
Alpha particle yields as function of emission angles and kinetic energies with a projection onto the angular axis.

Fig. 3
Fig. 4
Fragment intensities as functions of their masses and mean kinetic energies

Fig. 5
Most probable fission fragment kinetic energies and widths of fragment kinetic energy distributions compared to the same observables in binary fission
Fig. 6
Correlation plots for \( (E^\alpha, E_F) \) and \( (E^\alpha, E_F + E^\alpha) \)

Fig. 7
Correlation plots between the fragment mass ratio and \( E^\alpha, \theta^\alpha \) and \( E_F \) resp.
These definitions are valid for the alpha particles trajectory calculations. If experimental quantities are considered, $E_\alpha^\text{exp}$ and $E_F$ get the index (exp.).

The origin of the alpha particles is expressed by spatial coordinates, for which we use the symbols of two dimensional polar coordinates $(r, \theta)$. The distance of the fragment surfaces on the scission axis is denoted by $d$, which can also be interpreted as neck elongation between the fragments, the deformation of which is described by $\beta_1$ and $\beta_2$.

The evaluation method is characterized by the following steps:

1) From the experimental list mode data 3 dimensional distributions of the observables $E_\alpha^{\text{exp}}$, $\theta_\alpha^{\text{exp}}$ and $E_F^{\text{exp}}$ are generated for each fragment mass split $R$ in the three dimensional space $S_{\text{exp}}$, shown in figure 8.

![Diagram](image-url)

Fig. 8 Illustration of our data evaluation method
ii) For each mass split more than $10^6$ trajectories have been calculated. Each trajectory is determined by an alpha particle start point $(r, \theta)$ at a time, at which the fragments have a distance $d$; the deformation $\beta_1, \beta_2$ and by the initial parameter set $E_\alpha^0, \theta_\alpha^0, E_F^0$ as well as the final observable set $E_\alpha', \theta_\alpha', E_F'$. During the trajectory calculations windows limit the range of initial parameter values. The sizes of these windows have been defined according to previous experiences $2,3$)

\begin{align*}
0 \leq E_\alpha^0 &\leq 10 \text{ MeV}, & 0 \leq E_F^0 &\leq 50 \text{ MeV} \\
0 \leq \theta_\alpha^0 &\leq 180, & 0 \leq d &\leq 30 \text{ fm}
\end{align*}

The spatial coordinates $(r, \theta)$ follow the potential ridge of the Coulomb plus nuclear potential (of Krappe - Nix type) along the nuclear surface for alpha particles with non zero initial energies.

iii) The initial $(E_\alpha^0, \theta_\alpha^0, E_F^0)$ and final $(E_\alpha', \theta_\alpha', E_F')$ distributions are generated in space $S_i$ and $S_f$ of figure 8 respectively. Trajectories with the parameters $(\beta_1, \beta_2, r, \theta, d)$ transform for each mass split $R$ points of space $S_i$ into those of $S_f$.

iv) The experimental distribution $(E_\alpha^{\text{exp}}, \theta_\alpha^{\text{exp}}, E_F^{\text{exp}})$ in space $S_{\text{exp}}$ is divided channel by channel through the corresponding distribution $(E_\alpha', \theta_\alpha', E_F')$ in space $S_f$. Channels are symbolized in figure 8 by cubes. By this procedure weight factors are obtained which are applied to the initial parameter distributions $E_\alpha^0, \theta_\alpha^0, E_F^0, (r, \theta), (\beta_1, \beta_2)$ and $d$.

Figure 9 shows initial parameter distributions for a few arbitrarily selected channels (cubes).

From the trajectory calculations we can draw the following conclusions:

Initial alpha particle energies $E_\alpha^0$ range up to 8 MeV with a dominance of small energies. The most probable neck elongation $d$ is $9 \pm 1$ fm. The emission of alpha particles is not exclusively occurring at the point of rupture $8$). Pure neck emission does not cover all the range of measured observables. Emission occurs also from the fragment bodies close to the neck due to the reduced potential there.

Fragment initial kinetic energies have almost a "white" spectrum up to 50 MeV, see also ref. $2$). As trajectory calculations do not give any information about the instant of particle emission, it is not evident that $E_F^0$ is identical with the so called prescission kinetic energy.
Fig. 9 Initial parameter distributions for 5 arbitrary channels (cubes) a) inter­
fragment distances $d$, b) initial fragment kin. energies $E_F$, c) $E_F$ as function of $d$,  
d) initial alpha particle energies, e) alpha particle origins
An essential deficiency of trajectory calculations is the fact that initial alpha particle energy spectra are not available from a quantum mechanical theory of alpha decay for time dependent nuclear potentials describing the nuclear potential from saddle to scission.

4. Discussion beyond the trajectory calculations

The spectra, which we obtain from our trajectory calculations for the scission point parameters are a result of a subtle interplay of these quantities among each other. They depend also on our parametrisation of the nuclear shapes. Figure 10 is an illustration of what has still to be done to understand the interdependences of the initial parameters.

In order to get an understanding of ternary fission and its relevance for the fission process in general, we have tried a more pedestrian's way to proceed.

We compare the average excitation energies $E_x$ of fission fragments as functions of their mass splits for binary and ternary fission respectively. They are obtained as differences of the Q-values and the total kinetic energies $(E_p + E_{\alpha})^{(\text{exp})}$ from data displayed in figures 6 and 7. In figure 11 maximum Q-values for each mass split are plotted together with $(E_p + E_{\alpha})^{(\text{exp})}$ - distributions and average $(E_p + E_{\alpha})^{(\text{exp})}$ values. As is seen from figure 11 the average excitation energy of the ternary main fragments is about half that of the binary ones. This is more clearly shown in figure 12, where $E_x$ and the width of the $E_x$-distribution are plotted as functions of light fragment masses for binary and ternary fission. In this figure the nearly equal width of the energy distributions for both fission modes is surprising.

From these observations we can conclude

i) Ternary main fragments are less deformed than binary ones for comparable masses.

ii) As the width of energy distributions are determined on the way from saddle to scission, binary and ternary fission are similar up to the scission point. The decision to emit an alpha particle is taken in the last stages of nuclear fission.
Fig. 11
Total kinetic energy distributions as function of light fragment masses for ternary and binary fission. The two curves are maximum Q values\(^9\) and average \(E_F + E_\alpha\) values.
Fig. 12
Fragment excitation energies and energy distribution widths as function of light fragment masses

Fig. 13
The same as function of alpha particle energy
iii) Alpha particle and fragment excitation energies are anticorrelated. This is shown in figure 13. Also as function of alpha particle energy the widths of the excitation energies for the three particle emission types are within the experimental errors equal. This supports our conclusion ii).

iv) From figure 11 it is evident that cold fission\textsuperscript{10}, in which the fragment kinetic energies exhaust the Q-values, occurs for ternary fission at a higher yield level than for binary fragmentation. Figure 14 shows a fragment mass spectrum for $E_F + E_\alpha > 200$ MeV, which can be qualitatively explained by the ternary fission Q-value spectrum.

Finally I would like to say a few words about polar emitted particles. Preliminary calculations based on an evaporation theory reproduce intensities and average kinetic particle energies quite well, if the nuclear temperatures are calibrated with the number of emitted neutrons. However, the sharp focussing into polar emission directions cannot be explained, if the alpha particles are assumed to be evaporated isotropically by the accelerated fission fragments.
A way out of these difficulty could be the application of curvature dependent potential penetrabilities for alpha emission, which would favour narrow polar angular distributions.

Acknowledgement

I have to thank M. MUTTERER, J. PANNICKE, F. KRASKE, B. LEROUX, N. CARJAN, P. KOCZON W. FLASSIG, R. HASSE and F. GÖNNENWEIN for a successful collaboration and/or many interesting discussions. The ILL, Grenoble has supported the experiments and the GSI, Darmstadt the computer calculations. We are greatly indebted to both institutions.

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8) N. Carjan
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C. Wagemans, P. Schillebeeckx, P. D'hondt, J.P. Bocquet, A. D'Eer

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Abstract

Results are reported of a systematic investigation of the emission probabilities and the energy distributions of the charged light particles emitted during the thermal neutron induced ternary fission of the actinides. The data are discussed in terms of the liquid drop model, the scission point model of Wilkins et al. and the emission mechanism proposed by Cărjan.

1. INTRODUCTION

During the last ten years, a systematic investigation of the emission probabilities and the energy distributions of the charged light particles emitted during the thermal neutron induced ternary fission of the actinides has been performed at the ILL (Grenoble). The measurements are done at the end of the 87 m curved neutron guide installed at the high flux reactor. Here a thermal neutron flux of $6 \times 10^8$ neutrons/cm$^2$ sec is available with a ratio of slow neutrons to epithermal and fast neutrons of about $10^6$. Since also the direct $\gamma$-ray flux from the reactor is reduced by a factor of about $10^6$, the background due to fast neutron and photon induced reactions is strongly reduced. The charged particles are identified by means of surface barrier $\Delta E-E$ telescope detectors.

So far results have been obtained for $^{231}$Pa (1), $^{233}$U (2), $^{235}$U (2,3,4), $^{237}$Np (5), $^{239}$Pu and $^{241}$Pu (2). Measurements on $^{241}$Am and $^{243}$Am are being performed.

2. RESULTS AND DISCUSSION

Since detailed results on the individual nuclei have been reported in detail in the references mentioned above, we will concentrate here on the systematic aspects which we will try to interpret.

A very important result was the demonstration of a non-Gaussian low-energy tail in the ternary $\alpha$ energy distribution. This is illustrated in fig. 1, where we combined the $^{235}$U($n_{th},f$) data of D'hondt et al. (3) with those of Caïtucoli et al. (6). This non-Gaussian tail gives rise to an increase of 6% with respect to the integrated Gaussian spectrum. Since our results on $^{235}$U and $^{239}$Pu are very similar, a correction of +6% to the Gaussian extrapolated ternary $\alpha$ yields seems to be justified for all nuclei considered in the present work. Such a correction has
generally not been applied in older ternary α measurements, which did not go down low enough in energy. The non-Gaussian tail on the lower side of the ternary α energy distribution is in contrast to the Gaussian shape of the p,d,t and ⁶He energy distributions(4). The difference could be explained by the formation of low-energy α-particles during the decay of ⁵He and of excited ⁶He particles (3).

For the fissioning systems studied, the mean α and triton energies are compatible with a unique value of $\bar{E}_α = 15.9$ MeV and $\bar{E}_t = 8.3$ MeV. This is however not the case for the widths of the energy distributions nor for the α and triton emission probabilities $L_{RA}/B$ (number of Long Range Alpha-particles per Binary Fission) and $t/B$. This is illustrated in fig. 2, which shows the widths and the yields mentioned above as a function of $Z^2/A$ and of $\log \lambda$ ($\lambda$ being the radioactive α-decay constant) of the fissioning system. The general trend for $L_{RA}/B$, $t/B$ and the FWHM of the triton and LRA energy distributions is an increase with increasing $Z^2/A$ and a decrease with increasing $-\log \lambda$. In the following alinéas we will try to interpret these correlations.

A correlation between $L_{RA}/B$-values and $Z^2/A$ was already observed by Nobles (7) in 1962. The quantity $Z^2/A$ appears in the liquid drop model as the ratio of the electrostatic to the surface energy of the drop. So $Z^2/A$ is a measure of the fissility of the fissioning system considered. Hence it has been widely used to be correlated with all sort of fission observables.

The observed increase of the light particle yield with increasing $Z^2/A$ can be understood as follows: Both the neutron yield and the mass yield data show that α-particle emission occurs at the expense of the excitation energy of those fragments having the largest deformation energy at scission. So the light particle yield increases with increasing deformation at scission. Since liquid drop model calculations show an increase in the deformation energy at scission with increasing $Z^2/A$, the observed yield increase with $Z^2/A$ is expected.

Cărjan (8) on the other hand interpreted ternary fission as an α (or t,d,p,...) decay of the fissioning system during the last phase of the process. In such a picture, the ternary α-emission probability is expected to be correlated with the Coulomb barrier penetrability $P$ and with the reduced α-emission width $δ^2$ (i.e. the
probability of having an alpha-particle inside the nucleus). Since $\lambda = \delta^2 P/h$ (h being Planck's constant), a correlation between $\lambda$ and the ternary alpha-emission probability is likely to occur in the framework of Cărjan's model, although the $\lambda$-values used in fig. 2 are for ground-state transitions and the $(n_{th},f)$ reactions considered are leading to a fissioning system in an excited state.

Fig. 2 Absolute ternary alpha and triton yields and FWHM of the alpha and triton. alpha energy distribution as a function of $Z^2/A$ and of -log $\lambda$ of the fissioning system. Here $\lambda$ is given in year$^{-1}$.

However, the parameters $Z^2/A$ and $\lambda$ are not fully uncorrelated. From a least-squares adjustment in the region $84 \leq Z \leq 98$, Viola and Seaborg (9) determined a semi-empirical relation between $\lambda$ and $Z$: $\log \lambda = (2.11 Z - 48.99) Q_a^{-1/2} - (0.39 Z + 16.95)$, $\lambda$ being in sec$^{-1}$ and $Q_a$ being in MeV. So clearly $Z$ is a common parameter in $Z^2/A$ as well as in -log $\lambda$.

Before trying to interpret these observations, we will enlarge the data base for the present discussion. In fig. 3 T/B-values (i.e. the total number of ternary particles emitted per binary fission) are represented as a function of $Z^2/A$ of the fissioning system. Our published $(n_{th},f)$ data (1,2,5) are completed with preliminary values for $^{241}$Am and $^{243}$Am$(n_{th},f)$ (*) and with the spontaneous fission results given in the paper of Wild et al. (10) (**), except for $^{242}$Pu (s.f.) where the value of ref. 11 is used.

In fig. 4 the same spontaneous fission data are plotted as a function of -log $\lambda$. In contrast to the $(n_{th},f)$ results shown in fig. 3, here ternary fission and $\lambda$ are both ground-state data. The (s.f.) as well as the $(n_{th},f)$ data considered correlate reasonably well with $\lambda$ and hence do not contradict Cărjan's model. For the $(n_{th},f)$ data there remains however one problem, i.e. $^{232}$Pa, $^{238}$Np and $^{244}$Am not being alpha-emitters it is not obvious which $\lambda$-value to use.
Fig. 3 Total number of ternary particles emitted per binary fission \( (T/B) \) as a function of \( Z^2/A \) of the fissioning system.

Fig. 4. \( T/B \) as a function of \(-\log \lambda (\lambda \text{ in year}^{-1})\) for the spontaneous fission data of refs. 10 and 11.
The \((n_{th,f})\) data in fig. 3 show as a general trend an increased ternary particle emission probability with increasing \(Z^2/A\)-values. The same can be said for the \((s.f.)\) data. There is however one striking observation to make, i.e. the heavier isotope of the same nucleus always has a lower T/B-value than its lighter brother. In other words, the smooth "liquid drop"-increase of T/B with \(Z^2/A\) apparently needs

In fig. 5, the total fission fragment kinetic energy is shown as a function of \(Z^2/A^{1/3}\). Also here the liquid drop model predicts a smooth increase, but also here isotopes of the same nucleus systematically deviate from the average curve. As a more detailed example, we plotted our results on the plutonium isotopes \((12)\) as a function of \(Z^2/A^{1/3}\) (fig. 6). As explained in \((12)\), the observed decrease of \(E_k\) with \(Z^2/A^{1/3}\) is a consequence of the interplay between several neutron shells (mainly the spherical \(N=82\) and the deformed \(N=88\) shell) described in the static scission point model of Wilkens et al. \((13)\) in which shell corrections are applied to the liquid drop model (cfr. also the paper of Moreau et al. \((14)\) at this conference). In other words, when going from \(^{244}\)Pu to \(^{238}\)Pu, the influence of the deformed \(N=88\) strongly increases compared to that of the spherical \(N=82\) shell. This explains the reduction of \(E_k\) with increasing \(Z^2/A^{1/3}\).
These neutron shells may also explain the observed difference in T/B-values for spontaneous and neutron induced fission (fig. 7). The reduced ternary particle emission probability could be a consequence of the washing out of the shell effects due to the ~ 6.5 MeV neutron binding energy get free in the \( (n_{th},f) \) reaction.

**Fig. 7** Total number of ternary particles emitted per binary fission (T/B) as a function of \( Z^2/A \) for the fissioning systems \( ^{240}\text{Pu} \) and \( ^{242}\text{Pu} \).

Finally, shell effects probably also explain the steep increase in T/B for Cf and Fm isotopes. As explained by Wilkins et al. (13) and shown in fig. 8, the average deformation of the most probable light fragment changes abruptly for masses above 244. This strongly increased deformation is likely to result in a significant enhancement of the ternary particle emission.

**Fig. 8** The open circles represent the deformation of the light fragment complementary to mass 142 at its potential energy minimum (ref. 13).

### 3. CONCLUSIONS

In the present paper our results on thermal neutron induced ternary fission have been combined with spontaneous fission data. All these data have been interpreted with the help of the static scission point model of Wilkins et al. (13), in which the liquid drop model is corrected for shell effects.

**Acknowledgements**

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EMISSION OF LIGHT CHARGED PARTICLES IN THE PHOTOFISSION OF ACTINIDES.

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Abstract: Light charged particles accompanied photofission of $^{232}$Th, $^{233}$U, $^{235}$U, $^{238}$U, $^{237}$Np and $^{242}$Pu was studied with a telescope set-up. Energy distributions and emission probabilities for $\alpha$ and $^3$H particles were obtained. The emission probabilities show an increasing trend with increasing deformation energy of the compound nucleus.

1 Introduction

Although the spontaneous and thermal neutron induced ternary fission of fissile isotopes has been studied since forty years, information on light charged particle accompanied fission induced by $\gamma$-rays is very scarce 1,2). We report here on the first extensive study of the ternary photofission of actinides. For the nuclei $^{232}$Th, $^{233}$U, $^{235}$U, $^{238}$U, $^{237}$Np and $^{242}$Pu we measured the relative emission probabilities (LRA/B ratio) and the energy distribution characteristics ($<E>$ and FWHM) of the ternary $\alpha$ particles and the triton to $\alpha$ emission probability ($t/LRA$) as a function of the excitation energy of the compound nucleus.

2 Experimental set up and data handling

Ternary photofission experiments were performed with bremsstrahlung, produced by 12, 15 and 20 MeV electrons in a 0.1 mm thick gold foil. The detector system consisted of 8 telescopes. An identification of the light charged particles was necessary to separate them from the high proton background, which was produced predominantly by ($\gamma$,p) reactions in the target and in the target backing. The identification of the light particles is based on the difference in energy loss in the $\Delta E$ detector for the different light particles with the same initial energy. The detector telescopes consisted of two Au-Si surface barrier detectors: a totally depleted $\Delta E$ detector of ± 30 $\mu$m thickness, followed by an E detector depleted to a depth of 500 $\mu$m, each with an active area of 150 mm$^2$. The telescopes were shielded with a 20 $\mu$m Al foil. The telescopes are placed circularly around the $\gamma$-beam axis at an angle of 45 degrees with the target.

The fission fragments were counted simultaneously in a separate detector with an active area of 600 mm$^2$, placed on the opposite side of the target. The geometry factor between this detector and the telescope
set-up was determined for each experiment by counting in both detector systems the number of fission events. A correction for coincidence losses in the telescopes was obtained by measuring the natural α-particle countrate from a $^{235}\text{U}$ target in the E- and the ΔE-detector separately. The amplified pulses from the detectors were coded and stored event by event on a PDP11/10 system. The γ pile-up during the pulse of the linac (γ-flash) was measured continuously and an online subtraction of an average of it was performed. The detectors were calibrated using well known natural α lines from an $^{235}\text{U}$ target and from a $^{228}\text{Th}$ source. The two-dimensional ΔE-E data are handled off-line with our VAX-11/780 system. The measured coincident ΔE and E values are combined via the relation $T/\alpha = (E + \Delta E)^{1.73} - E^{1.73}$ (with T the thickness of the ΔE detector) to obtain an identification spectrum. Fig.1 gives an example of such an identification spectrum.

![Fig.1 Typical identification spectrum.](image1)

![Fig.2 Typical LRA energy distribution and associated Gaussian fit.](image2)

3 Results and discussion

A typical energy spectrum of the LRA-particles is shown in fig.2. The parameters of the energy distribution are obtained by fitting a Gaussian through the data points above 12.5 MeV. As a consequence of this procedure the non-Gaussian low-energy tail of the energy distribution is not taken into account 3). In table 1 we summarize our results. The parameters of the energy distribution and the emission probabilities of the LRA-particles relative to the binary fission probability (LRA/B) are given together with the triton to α and triton to binary fission emission probability ratios (t/LRA and t/B). The quoted errors are only statistical ones. The energy of the electrons producing the bremsstrahlung is indicated by $E_e$. As a consequence of the high proton background for measurements with 20 MeV bremsstrahlung, it's impossible to distinguish the tritons.
Table 1

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<th>Target E_e (MeV)</th>
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<th>t/B (10^-4)</th>
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Although the <E>-values show some dispersion, the average LRA energy seems to be nearly constant over the whole range of nuclei and excitation energies (the average value for our measurements is 15.8 ± 0.1 MeV). This is consistent with the general trend observed in ternary fission. There is a considerable amount of dispersion in the full width at half maximum values. These FWHM values show a tendency of a systematic increase in going to a higher excited nucleus but the uncertainties do not permit any further conclusions. Plotted as a function of Z^2/A, our values show an increasing trend with increasing Z^2/A.

Due to the availability of more data, compared to our previous report \(^4\), an increase of the emission probability of the \(^3\)H-particles, relative to the LRA emission probability, (t/LRA) in going to a heavier nucleus can be observed. Although \(^237\)Np is an odd-Z nucleus and the existence of an unpaired proton in the compound nucleus could perhaps enhance the yield of H-particles, our measurements indicate that there is no significant increase in the triton yield for \(^237\)Np.

As generally observed our results show an increased LRA yield with increasing Z^2/A \(^5,\6\). The triton emission yields (t/B) show the same trend. This is interpreted as a confirmation of the idea that the LCP-yield is correlated with the amount of deformation at scission. A better way to control this hypothesis is probably to plot the LRA-yield as a function of Q-<TKE> \(^7\). The difference between the calculated reaction Q-value and the measured average total kinetic energy of the fragments gives the deformation energy plus the internal heating. As
for spontaneous fission the internal heating is probably low, the difference \( Q - \langle TKE \rangle \) represents an estimate of the deformation energy at scission \(^7\). The \( Q \)-values for our photofission experiments were obtained by averaging the ground-state atomic masses, using the tables of Moller and Nix \(^8\) over the experimental massdistributions. These were however only available for \( ^{232}\text{Th}, \, ^{235}\text{U}, \, ^{238}\text{U} \) and \( ^{242}\text{Pu} \) \(^9\text{--}^{12}\). Our LRA/B values were converted to LCP/B values, using the same method as in ref. \(^7\). For photofission the calculated difference \( Q - \langle TKE \rangle \) should include the average excitation energy of the compound nucleus. When we plot our results together with those for spontaneously fissioning nuclei, given in a very recent survey by Wild et al. \(^7\) a significant different trend between induced and spontaneous fission is obvious (fig.3). However, if we do not include the extra excitation energy contributed by the photons, there's agreement between our data and the sf data (fig.4). This can be explained by assuming that most of the extra excitation energy remains as internal excitation and only a small fraction of it is converted into deformation energy. As a consequence an increase of the excitation energy of the compound nucleus should have only a small influence on the LRA emission probability, as we have observed (see table 1). Together with our photofission values we plotted some results from thermal neutron induced ternary fission \(^6\). The trend of nearly excitation energy independence of the LCP emission probability is confirmed. The full lines represent a linear least squares fit (taking into account the uncertainties on both x and y coordinates) for the three different systems. It's remarkable that the slope for our \((\gamma,F)\) data is the same as for the \((n,f)\) data, although there is an observable displacement.

![Graph showing LCP emission probabilities versus \( Q - \langle TKE \rangle + E_{\text{exc}} \) (MeV).

\( E_{\text{exc}} \) is the average excitation energy of the compound nucleus induced by the photons.](image-url)
However, when comparing with other systems, possible systematic errors and the fact that Q-<TKE> is only a rough approximation of the deformation energy, should be kept in mind. Our conclusion can be that also our results on the emission of light charged particles during photon induced fission support the idea that the emission probability of these light charged particles is correlated with the deformation energy available at the moment of scission.

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Fragmentation and neutron emission for $^{252}\text{Cf}(\text{SF})$

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ABSTRACT

At present a measurement of the $^{252}\text{Cf}(\text{SF})$ fragment and fission neutron correlation is being performed at C.B.N.M. It is the main purpose of this experiment to clarify the nature of the fission neutron emission processes, i.e. to determine the fraction of neutrons evaporated from the accelerated fragments versus the fraction of neutrons emitted dynamically at scission. In contradiction to previous measurements the presently found neutron-fragment-angular-distributions are conformable to the assumption that all neutrons are emitted from the fully accelerated fragments. The unique properties of the employed fission chamber made it interesting to (re)determine the fragment distributions, especially at high TKE values where the Q-values are nearly exhausted. The presented results are based on $5 \times 10^7$ analyzed fission events and show that cold fragmentation for $^{252}\text{Cf}(\text{SF})$ is reached for fragment splits for which the light fragments are in the mass range $A_L = 102-112$ and $A_L = 120$.

I. INTRODUCTION

Nuclear fission has been a longstanding theme of scientific investigation and here the spontaneous fission of $^{252}\text{Cf}$ gives a relatively easy opportunity to study the correlations between neutron emission and fission fragment parameters. The measurement of such correlations can contribute to a better understanding of the fission process. On $^{252}\text{Cf}$ much more effort than on other nuclei was devoted to the measurement and interpretation of the prompt fission neutron spectrum, because this spectrum is used also as a neutron spectrum shape standard. Several attempts \(^1\)\(^2\) have recently been made to give a theoretical description of the prompt fission neutron spectrum of $^{252}\text{Cf}$. These models are based on the assumption that the mechanism of neutron emission is the evaporation from the fully accelerated fragments. However, the comprehensive measurements of Bowman et al. \(^3\) of the prompt neutron anisotropy have led to the conclusion that a fraction ($\sim 10-20 \%$) of the total number of fission neutrons is emitted isotropically in the laboratory frame of reference. In spite of many further investigations the knowledge of the so-called scission neutron emission is poor and partially contradictory. Therefore it is of much interest for the basic understanding of the neutron emission process, not only to measure precisely the integral prompt fission neutron spectrum, but to obtain
also in multiple-dimensional measurements the correlations between the neutron emission and the different fission fragment parameters which can help to clarify the nature of the scission neutrons.

The experimental technique employed in the present investigations offered an excellent tool for studying fragmentation for 252Cf(SF). Interesting here are the rare so called "Cold Fragmentation" events for which the total excitation energy of the two fragments is so low that neutron emission is unlikely. Detection of these events gives unique information about the roles of nuclear-pairing, shell and liquid-drop effects as well as ground-state deformations in the fission process.

II EXPERIMENTAL METHOD

Fission fragment detection is done using the gridded ion chamber developed at our lab with which fragment type, kinetic energy and angle can be determined simultaneously.

The set-up is shown in fig. 1. The fission fragment kinetic energies \((E_{\text{kin}})\) are determined using the anode pulses \((q_a)\) from the twin chamber. The excellent energy and mass resolution of this detector is illustrated in fig. 2 which shows a part of a measured 252Cf light fragment mass distribution.

The grid signal \((q_g)\) is a function of \(E\) and \(\cos \theta\) where \(\theta\) is the angle between the normal of the cathode and the ion trace made by the fragment:

\[
q_g = E \cdot \frac{\overline{X}}{D} \cdot \cos \theta
\]

\(D\) is the cathode grid distance and \(\overline{X}(E,A,Z)\) is the distance from the origin of the track to the centre of the ionization charge, and is in general a function of fragment energy \((E)\), mass \((A)\) and charge \((Z)\).

For fixed values of \(E\) and \(A\) the ratio \(q_g/q_a\) is distributed between 0 and \(\overline{X}(E,A,Z(A))/D\). \(X/D\) can therefore be determined from measured \(q_g/q_a\) distributions and eq. (1) can be solved with respect to \(\cos \theta\). The resolution on the \(\cos \theta\) values
found this way is < 0.05 and is partly due to the dispersion of the isobaric charge yield. The above method makes it possible to measure fragment angular distributions, however using the 4π angular efficiency of the ionization chamber. Moreover the angle information is essential for the use of the ionization chamber, e.g. corrections for angle dependent energy losses in the sample and the backing can be applied.

Recently we have attempted also to determine nuclear charges with the ion chamber. Forming the ratio:

$$R = \frac{(q_g/q_a)_L}{(q_g/q_a)_H} = \frac{\overline{x}_L(E_L,A_L,Z_L)}{\overline{x}_H(E_H,A_H,Z_H)}$$

(2)

for complementary light (L) and heavy (H) fragments a quantity is obtained, where the only unknown parameter is the nuclear charge split \(Z_L/Z_H\). Fig. 3 shows the distribution of \(R\) obtained for \(^{252}\text{Cf}\)(SF). Only fragments with \(\text{TKE} = \text{TKE} + 30\ \text{MeV}\) have been selected. The distribution shows a multipeaked structure where each peak is assumed to arise from even charge splits. Large proton even-odd effects at high TKE values have been observed for other nuclei\(^6\,^7\). The fractional charge yields for fixed mass split \((A_L/A_H)\) were found from fits to the \(R\)-distribution with Gaussian functions as exemplified in fig. 4. The fit function parameters were determined from those mass splits \((A_L = 96, 102, 106, 112, 117, 121)\) where the charge distribution is dominated by only one \(Z\)-value. Although the charge resolution of the method \((\Delta Z/Z = 1/40, Z = 46)\) does not allow separation of individual nuclear charges it is however sufficient for determination of charge distributions in fission.

For timing the pulses from the common cathode are used together with the neutron detector signals giving a resolution < 0.7 ns FWHM. The neutron detector, a 4"x1" NE 213 scintillator, is located on the axis of the ionization chamber. The distance between the \(^{252}\text{Cf}\)-source and the neutron detector was 0.51 m. Both, the pulse height and the pulse shape for n/\(\gamma\) discrimination are recorded. Neutron energies are determined using conventional time-of-flight technique. All 7 parameters are digitized, each allocated 8192 channels, and stored sequentially on tape for off-line analysis. At present a \(^{252}\text{Cf}\) source
prepared by vacuum evaporation onto a 120 μg·cm⁻² thick Ni-foil is mounted in the chamber. The source activity is ~3·10² fiss·s⁻¹, yielding a fragment-neutron coincidence rate of 1 s⁻¹ for the present geometry of the experiment.

III  FRAGMENT-NEUTRON CORRELATIONS

So far 1.5·10⁶ coincidences have been recorded on which the present analysis is based. The results should be regarded as preliminary since corrections such as the recoil correction for the determination of masses and kinetic energies of the fragments have not been applied. The results are uncorrected for mass resolution. The experimental data contain the needed information for a determination of these corrections but a more complex analysis code has to be written. However, the effect on the result presented here is expected to be minor. The mass integrated prompt fission neutron spectrum of ²⁵²Cf(SF) was evaluated in some detail. For the present preliminary evaluation the neutron detection efficiency, which is needed to obtain the spectrum shape, was calculated using the Monte Carlo-code from the thorough detector efficiency investigation of Dietze and Klein. The neutron spectrum divided by √E is plotted in Fig. 5 logarithmically versus the incident neutron energy. The full line in Fig. 5 represents the result of a least squares fit through the experimental data with a Maxwellian energy distribution. The temperature parameter obtained by the fit is T=1.41 MeV. An error for this value was not yet

![Fig. 5](image-url)  
*Fig. 5: Preliminary neutron energy spectrum divided by √E versus the neutron energy.*
evaluated, also since the experimental determination of the detector efficiency is not yet done. This preliminary neutron energy spectrum shows no major deviations from the Maxwell distribution in the neutron energy range from 0.8 MeV to 20 MeV. The deviations from the Maxwell distribution are in general less than 5% in the above mentioned range. The dashed line corresponds to the experimental spectrum measurements of Märten et al. \(^9,^{10}\), who found a large excess of neutrons above 20 MeV. With the presently evaluated data we can not confirm nor contradict the measured excess of neutrons. From a total of about \(2 \times 10^6\) events the statistics obtained in the high energy region above 20 MeV is not sufficient.

The measured fragment-neutron angular distributions integrated over all fragments versus the neutron energy are shown in a bi-parametric representation in fig. 6. The neutron emission angular distributions as function of neutron energy for each mass split are also available. The present data agree fairly well with the results of Bowman et al. \(^3\) below 4 MeV. However, at higher neutron energies our data are much more anisotropic with intensity ratios \(N(90^\circ)/N(0^\circ)\) more than one order of magnitude smaller than those of Bowman above 8 MeV. The comparison between the present angular anisotropy measurements as function of fission neutron energy and those of Bowman et al. \(^3\) is made in fig. 7. The full line in fig 7 represents calculations of the angular anisotropy as function of the neutron energy with the assumption that all neutrons are emitted from the
fully accelerated fragments. The according equations and the needed numerical values were taken from Terrell. The energy dependence of the present N(90°)/N(0°) intensity ratio is conformable to the assumption that all neutrons are emitted from the fully accelerated fragments and the existence of a hard (T=2.0-2.5 MeV) scission neutron component which Märtens concluded from the Bowman angular distributions must be refuted. A more thorough analysis of our angular distributions is needed in order to decide whether perhaps a soft component is present. Fig. 8 shows the measured neutron emission multiplicities v(A) from the individual fragments compared to those of Walsh. The minor differences in these two data sets might be due to the neglect of recoil effects in the present analysis. However, our data seem not to confirm the fine structures in v(A) seen by Walsh.

Since the measurement of Bowman in 1962 very little experimental information has been gained on the average neutron emission energies \( \bar{\eta}(A) \) in the center-of-mass frame. Fig. 9 displays the measured dependence of the average \( \bar{\eta} \) on \( A \) as compared to the results of Bowman. Some striking differences may be observed. The present data show a pronounced dip at \( A=130 \), also seen in \( v(A) \), followed by a broad hump extending to \( A=145 \). This behaviour is also reproduced in theoretical calculations of \( \bar{\eta}(A) \).
The unique properties of the fission chamber made it interesting to (re)determine the fragment distributions, especially at high TKE where the Q-values are nearly exhausted. Cold fragmentation has previously been studied for thermal neutron induced fission of $^{233}\text{U}$, $^{235}\text{U}$ and $^{239}\text{Pu}$.

![Image of mass distribution](image1)

Fig. 10 shows the present yield/fission for TKE close to the maximum Q-values. The latter quantity is shown as a thick line calculated from the mass tables. In the range $A_L = 102-112$ and for $A_L = 120$ the highest measured TKE lie within ±1 MeV around the $Q_{\text{max}}$-line. In the $A_L=102-112$ range the light and heavy fragments are stabilized by the $N \sim 66$, $N \sim 86$ deformed neutron shells, respectively. The mass split 120/132 is favoured by the double magic 132Sn50 heavy fragment. Neglecting pre-scission kinetic energy the TKE is given by the coulomb interaction $V_C$ between the nascent fragment:

$$TKE = V_C = Z_L \cdot Z_H \cdot e^2 / D$$  \hspace{1cm} (3) 

where $D$ is the distance between the charge centres at scission. In the static scission point model $D$ is given by $D = a_L (8L) + a_H (8H) + d$. $a_L, a_H$ are the major axis of the spheroids representing the fragments with deformations $8L$, $8H$. The parameter $d = 1.4 - 2.0 \text{ fm}$ is the distance between the tips of the spheroids. In the limit of "true" cold fragmentation $TKE = Q$ and the deformations take the value of the fragment groundstate deformations. From eq. (3) it follows that this energy condition only can be met if at least one fragment has a strong prolate deformation. This is indeed the case for 252CF(SF) splits with $A_L = 100-109$.
for the fragments yielding the highest Q-values. However, in the range $\Delta L = 110-119$ the heavy fragment is nearly spherical and the light fragments have even oblate groundstate deformations. The TKE-values calculated from eq. (3) thus exceed $Q_{\text{max}}$ by more than 20 MeV. It is therefore remarkable that the measured TKE-lines come close to $Q_{\text{max}}$ for this mass range. A possible explanation is that the light fragments have low-lying prolate levels and that these can be populated in cold fragmentation. This will also explain why the TKE-lines for the lowest yields are 1-3 MeV below the $Q_{\text{max}}$ line here.

The TKE-lines show clearly fine structures with a period of $\sim 5$ mass units which can be attributed to favoured fragmentation into even charged fragments. Using the above described method the fragment nuclear charges could be determined for all fission events for which the TKE is larger than the values given by the dash-dotted line in fig. 10. The lower part of the figure shows the independent fractional charge yields $P(Z_L)$ as function of $\Delta L$. Yields belonging to even charge splits are connected with full lines whereas odd splits are connected with dashed lines. The structures of the TKE lines are clearly in phase with the even charge yields, showing that the TKE is relatively higher for even splits than for odd ones. Even charged nuclei may be less deformable and be characterized by a more compact scission configuration. From the charge yield, fig. 10, it can also be seen that even charge splits are 2-3 times more probable than odd splits in this TKE range. This suggests that the nuclear charge split is determined at an early stage of the fission process where the internal excitation of the fissioning system is so low that breaking of proton pairs is unlikely. Contrary, neutron pairing is generally not conserved in cold fragmentation. Only for the lowest yield levels $\sim 10^{-7}$ MeV$^{-1}$ does the TKE-line partly reproduce the neutron even-odd effects as given by the $Q_{\text{max}}$-line. At these low yield levels the "true" cold fragmentation limit may be reached, where fragments are born without internal excitation. For fragmentation with lower TKE the neutron number seems then to be determined at a stage where the internal excitation energy of the fissioning system is so high that one or more neutron pairs are broken.

### IV CONCLUSIONS

Although most of the results of the ongoing multi-parameter neutron-fragment correlation experiment are preliminary, some conclusions might already be drawn now:

- The more pronounced angular anisotropies measured in the present experiment compared to those of Bowman et al. at higher neutron energies lead to at least a much smaller portion of scission neutrons if their existence has not even to be refuted.
- With the presently calculated neutron detector efficiency no major deviation from a Maxwellian distribution between $0.8 \text{ MeV} \leq \text{En} \leq 20$ MeV is observed.
- Pronounced structure as function of mass was observed for the average c.m. fission neutron energy, in relative agreement with theoretical calculations.
At a yield of ~10^{-7} MeV^{-1} the TKE exhausts the Q-value for fragmentations for which the light fragments are in the mass range A_L = 102-112 and for A_L = 120.

In the mass range A_L = 109-119, where oblate groundstate deformations are predicted, the light fragments must be formed in low lying prolate states.

Proton pairing effects plays an important role in cold and near cold fragmentation. Whereas neutron pairing effects are only seen at the lowest measured yield levels, where fragments may be born without internal excitations.

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SESSION III

FISSION FRAGMENT PROPERTIES

Chairman: A.J. Deruytter
CBNM, Geel
The scission-point model: possibilities and deficiencies

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Abstract

Three theoretical models for the description of nuclear fission distributions are compared: the fragmentation theory, the statistical theory and the scission-point model. These models, although developed from different starting points, yield results which are all mainly governed by the potential energy at the scission point and it can be shown that they are equivalent. In each model however, the relation of the scission-point to the collective degrees of freedom (particularly to the mass fragmentation coordinate) has been put forward arbitrarily. Within the three models a dynamical study of the ratio of the collective kinetic energy to the intrinsic excitation energy has to be performed. Suchlike extended calculations are introduced in the framework of the scission-point model.

1. Comparison between the scission-point model and other prescriptions

Within the last decades, extensive efforts have been pursued in order to explain both the systematics and the details in the structure of the various distributions associated with the fission phenomenon. Three models seem to be quite successful:

1) The fragmentation theory\(^1\) of the Frankfurt group introduces collective mass and charge fragmentation coordinates \( \xi \). Only these degrees of freedom and the elongation parameter \( z \) of the relative motion are treated dynamically, whereas all other important collective degrees of freedom \( \beta \) (deformation coordinates, necking-in parameter, \ldots) are determined by requiring minimal potential energy for a given \((\xi,z)\)-pair. Furthermore, only the fragmentation coordinates \( \xi \) are quantized, yielding a simplified Schrödinger oscillator wave equation:

\[
\{ \frac{-\hbar^2}{2\sqrt{B}\xi} \frac{\partial}{\partial \xi} + \frac{1}{\sqrt{B}\xi} \frac{\partial}{\partial \xi} \} + V(\xi,z) \psi_n(\xi,z) = \epsilon_n(z) \psi_n(\xi,z) \quad (1)
\]

Herewith, \( B \) stands for the mass parameter in the \( \xi \)-direction, while \( z \) is regarded as a time-dependent parameter: \( z = z(t) \). In a first approximation, the eigenfunctions \( \psi_n(\xi,z) \) are superimposed with a Boltzmann factor of nuclear temperature \( \theta \),

\[
\psi(\xi,z) = \sum_n \psi_n(\xi,z) e^{\epsilon_n(z)/\theta} \quad (2)
\]

and the probability of a particular mass fragmentation is finally obtained by calcula-
ting the square of the absolute value of the wavefunction \( \Psi(\xi, z_{\text{scission}}) \), i.e.

\[ \text{Probability}(\xi) \sim |\Psi(\xi, z_{\text{scission}})|^2 \quad (3) \]

2) The **statistical theory**\(^2\) of Fong states that the mass distribution is determined by the density of quantum states, calculated (mostly for convenience) at the moment of scission. Using the appropriate level-density formulas\(^3\), one obtains:

\[ \text{Probability}(\xi) \sim \sum_{\beta} \eta_{\text{levels}}(E_{\text{exc}}(\beta, \xi, z_{\text{scission}})) \quad (4) \]

with \( E_{\text{exc}}(\beta, \xi, z) \) the internal excitation energy at a particular deformation configuration \((\beta, \xi, z)\). The latter can easily be calculated due to the additional assumption that the difference in potential energy, \( E_{\text{pot}} \), between saddle point and scission point is nearly entirely converted into \( E_{\text{exc}} \), rather than in collective kinetic energy \( E_{\text{kin}} \):

\[ \text{Probability}(\xi) \sim \sum_{\beta} \eta_{\text{levels}}(E_{\text{tot}}(\beta, \xi, z_{\text{scission}}) - E_{\text{pot}}(\beta, \xi, z_{\text{scission}})) \quad (5) \]

3) The **scission-point model**\(^4\) assumes an equilibrium among all collective degrees of freedom at the scission point. Thereby, one is able to obtain the various fission probabilities by calculating the sum of the collective potential and of the collective kinetic energy at the scission point:

\[ \text{Probability}(\xi) \sim \sum_{\beta} e^{-E_{\text{pot}}(\beta, \xi, z_{\text{scission}}) - E_{\text{kin}}(\beta, \xi, z_{\text{scission}})}/T \quad (6) \]

Herewith the nuclear "collective temperature" \( T \) is approximately 1 MeV. Very simple numerical calculations\(^4\) become possible because of the additional assumption that, for a particular distance between the fragments, the collective kinetic energy is independent of the collective variables and therefore can be omitted in the fission probability calculations:

\[ \text{Probability}(\xi) \sim \sum_{\beta} e^{-E_{\text{pot}}(\beta, \xi, z_{\text{scission}})}/T \quad (7) \]

These three theories are developed from completely different approaches. However, the final results obtained are all governed mainly by the potential energy at the scission point, and are consequently very similar, from a **qualitative** point of view. The correspondence between scission-point model and statistical theory is very clear, comparing equations (5) and (7), and taking into account the principal factor in level density formulas being proportional to

\[ e^{\sqrt{a_0 E_{\text{exc}}}} = e^{\sqrt{a_0 (E_{\text{tot}} - E_{\text{pot}})}} \quad (8) \]

The equivalence of the fragmentation theory with the two other models can be explained by three facts: First, the behaviour of the wavefunctions \( \Psi_n(\xi, z) \), appearing in equation (4), is obviously mainly determined by the potential energy \( V(\xi, z) \). The overall magnitude of the mass parameter \( B \) influences the peak-to-valley ratio and the spread of the wavefunction, while the oscillations of \( B \) only determine the fine structure to
some extent. Secondly, in fission problems which exhibits several minima in the potential energy $V(\xi, z_{scission})$ as a function of the fragmentation coordinates $\xi$, the low-lying wavefunctions $\psi_n(\xi, z)$ are ever peaked around one minimum. Finally, the eigenvalues $\psi_n$ of the lowest-lying wavefunctions of each minimum differ precisely by the difference in the corresponding potential energy. So, when calculating fission probabilities using (2) and (3), the principal determining factor is again the potential energy and the functional dependence is the same as in (5) for the scission-point model.

The three theories, being mathematically equivalent to each other, are also subject to the same fundamental criticism: in each model, the choice of the scission point, i.e. the value of the elongation parameter $z$ versus the other collective variables (mass fragmentation $\xi$, fragment deformations $\beta$, ...) is put forward in one way or another. By this, the quantitative difference between the results of the three models in discussion, can be ascribed exclusively to different forms of the prescription for the scission-point. In the scission-point model for instance, the hypothesis of equal collective kinetic energy at equal distances between the tips of the fragments is explicitly assumed. On the contrary, in the most recent statistical theory calculations, the potential energy surface calculations according to an asymmetric two-center shell model$^5$ were performed with a constant value of the neck constriction radius. These calculations yield results which are incompatible with earlier statistical theory calculations which assumed equal distances between the mass centers of the nascent fragments. Finally, in the fragmentation theory calculations, the total length of the nucleus at scission is held constant. Various discrepancies between the theoretical calculations and experimental results on mass and kinetic energy distributions, indicate that these choices are not justified, nor optimal. In the fragmentation theory, this fundamental shortcoming is somewhat met by partly taking into account the dynamic aspects of the elongation parameter $z$: a classical equation of motion for $z$ may be obtained$^6$ and, instead of equation (2), the eigenfunctions of the stationary Schrödinger equation (1) are superimposed with coefficients which are obtained by inserting the wave function into a time-dependent Schrödinger equation.

2. Extended scission-point model calculations

In the perspective of the scission-point model, we study explicitly the dynamic aspects of the fission process and the associated intrinsic excitation energy, in order to obtain the precise relation between the collective kinetic energy and the collective variables. The various fission probability distributions will then be calculated using equation (6), rather than using the more simple equation (7), based upon questionable assumptions. This dynamic study has to be reiterated for each possible boundary condition at the scission point and consists in three major steps.
1) The potential energy surface has to be calculated in the six-dimensional deformation space of the asymmetric two center shell model. Taking into account the extremely large set of geometrical configurations, we necessarily have to use modified prescriptions for the Strutinsky shell correction and the B.C.S. pairing correction calculations: we calculate both of them as a sum of two contributions (corresponding to the two parts of the scissioning nucleus), keeping in mind that the well known classical prescriptions for the limiting cases of both ground-state deformations and separated nuclear shapes have to be reproduced exactly. This potential energy surface can be calculated at an arbitrary value for the nuclear temperature.

2) Starting from the saddle point, the classical Hamilton equations of motion have to be integrated. Herewith, the collective kinetic energy is calculated provisionally for incompressible, nearly irrotational hydrodynamical flow by use of the Werner-Wheeler method, whereas the Raleigh dissipation function, which describes the transfer of energy from collective motion into internal excitation energy, is calculated according to two-body viscosity formulas. Different values for the direction in parameter space of the deformation velocity at the saddle point result in scission paths towards different boundary conditions at the scission point. In order to be able to perform the enormous calculations of steps 1) and 2) in a reduced amount of time, we developed computer codes which consistently calculate the derivatives of all physical quantities (single-particle levels, macroscopic and microscopic potential energy, mass and viscosity tensors ...) towards all external parameters (deformation coordinates & elongation z and fragmentation ξ).

3) Along the various paths between saddle and scission point, as obtained in step 2), we use the cranking model equations, derived from the T.D.H.F.-theory, in order to obtain the deformation velocity and the intrinsic excitation energy, consistent with energy conservation. We avoid to use adiabatic approaches (although this prevents us from performing self-consistent calculations, because of computer-time limitations) since these contain contradictory assumptions: following the first assumption, the nucleus remains in its lowest potential energy state so that the difference in potential energy between saddle point and scission point is nearly entirely converted into collective kinetic energy, rather than into intrinsic excitation energy; this however opposes the second assumption which demands a small deformation velocity. In the framework of the cranking model, the nucleus starts in the B.C.S.-groundstate, just beyond the saddle point. The excitation mechanism consists in the fact that one-pair excitations get occupied at each crossing of a single-particle level with the Fermi level. This process stops after scission, whereas the single-particle energies become nearly constant as a function of the elongation parameter z. Like in adiabatic approaches, the collective kinetic energy is described by the time-dependence of the coupling between single-particle states. However, over close pseudo-crossings, a Lan-
dau-Zener transition will be made, which acquires internal excitation energy, rather than contribute to the collective kinetic energy. The mass parameter in the cranking model differs from that in the adiabatic Inglis approximation by the exclusion of pair excitations. In a first approximation however, we do not include the corrections to the mass parameter value due to the excitation of the nucleus during deformation. Finally, starting from the ratio of the collective kinetic to the intrinsic excitation energy, as a function of the collective degrees of freedom at the scission point, we will be able to obtain the various fission probabilities.

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COLD FRAGMENTATION: EXPERIMENTS AND MODELS

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ABSTRACT

Cold fragmentation is a limiting case of nuclear fission, where the fragments carry no excitation energy and hence all of the available energy is converted into the total kinetic energy of the primary fragments. It is argued that cold fragmentation corresponds to the most compact scission configuration being compatible with energy conservation. Experimental methods and results for thermal neutron induced and spontaneous fission are reviewed. Characteristic experimental features of cold fragmentation are discussed in terms of a static scission point model. While these features may be interpreted on rather simple physical grounds, it is pointed out that the mere fact of cold fragmentation being observable at all is a challenge to any theory of nuclear fission.

1. INTRODUCTION

The energy, Q, being set free in the fission process is due to the difference in mass between the fission prone nucleus and the two primary fragments. This energy will show up in the total kinetic energy, TKE, and the total excitation energy, TXE, of both fragments:

\[ Q = TKE + TXE. \]  

In a typical reaction, e.g. thermal neutron induced fission of $^{235}$U, the primary fragments will carry some 170 MeV as TKE and some 25 MeV as TXE. The limiting case of fission, with TXE tending towards zero within a few MeV, has been dubbed "Cold Fragmentation" (CF), since evidently the fragments then have to be borne in - or at least close to - their ground states. If CF is indeed attained, the fragments will emit no neutrons, and eventually even no gammas. It follows from eq. (1) that, alternatively, CF may be characterized by stating that the Q-value of the reaction is exhausted by the TKE. In fact, so far all experiments studying CF have been searching for high TKE events.

The interest in CF, even though being a limiting and rare process,
may be justified by arguing in terms of a scission point model. At scission, TKE will be made up by the Coulomb repulsion, $V_C$, of the nascent fragments and the prescission kinetic energy, $\varepsilon$, the fragments already have at this stage of the process:

$$\text{TKE} = V_C + \varepsilon.$$  \hfill (2)

Similarly, TXE may be decomposed into the total deformation energy $V_D$ and the intrinsic excitation energy $E^*$ of the fragments right at scission:

$$\text{TXE} = V_D + E^*.$$  \hfill (3)

In eqs. (2) and (3) the energies $V_C$ and $V_D$ are tied up as potential energy, $V_P$:

$$V_P = V_C + V_D.$$  \hfill (4)

The sum of $\varepsilon$ and $E^*$ may be called the "free energy".

The energy balance vs. deformation is illustrated schematically in Fig. 1 for a given mass fragmentation. In a scission point model, where the scission configuration is visualised as two collinear spheroidal fragments separated by a fixed tip distance, the least possible deformation corresponds to two spherical fragments. In Fig. 1 it has been assumed that both fragments have ground states being spherical. Starting with $V_D = 0$ at this point, $V_D$ will therefore increase with deformation. On the other hand, the Coulomb potential $V_C$ slopes downwards with deformation. In liquid drop model calculations with realistic parameters one shows that for small deformations $V_C$ decreases faster than $V_D$ increases. From eqs. (1) to (3) and energy conservation it follows that for any fixed mass fragmenta-
tion the total available energy

\[ Q = V_C + V_D + \epsilon + E^* \]  

stays constant, i.e. is independent from deformation. Therefore, scission configurations with deformations less than the one marked "Cold Fragmentation" in Fig. 1 are physically outruled. Stated otherwise, the free energy has necessarily to be non-negative.

Heading for CF, one has to look for the conditions under which TXE = V_D + E^* may be minimised. For any model of nuclear dissipation the internal excitation energy E^* of the fragments will increase monotonically with the available free energy. Hence, E^* will be minimised, i.e. E^* = 0, for vanishing free energy viz. least accessible deformation. Under the assumptions of Fig. 1 for V_D vs. deformation, also V_D will reach a minimum for the smallest feasible deformation. Therefore, CF is attained at the least possible deformation, defined by the deformation where the V_p - curve intersects the Q-line (s.Fig.1). For this limiting scission configuration, and since E^* = 0, the minimum attainable total excitation energy

\[ \text{TXE}_{\text{min}} = V_D \]  

is to be observed. For this same configuration, by conservation of energy, the total kinetic energy will be maximised, and since with vanishing free energy also \( \epsilon \) approaches zero, one has

\[ \text{TKE}_{\text{max}} = V_C. \]  

Cold Fragmentation is thus seen to correspond to the limiting case of nuclear fission with the most compact scission configuration. In addition, in CF both TXE and TKE are uniquely linked to the potential energies of deformation, V_D, and Coulomb repulsion, V_C. One may, therefore, learn something on scission configurations by studying Cold Fragmentation.

2. EXPERIMENTAL TECHNIQUES

The first experimental device having been specifically designed to study CF was invented about 10 years ago by C. Signarbieux [1]. The basic idea is that in fission mass and momentum conserva-
tion strictly apply to the primary fragments, and that, indeed, in CF no neutrons are emitted, i.e. there are only primary fragments. The setup proposed is ingeniously simple, making use of two surface barrier detectors only, which are positioned on opposite sides of a thin high quality fission target. Rather surprisingly it is possible to determine fragment masses one by one by taking just the difference in time of flight of the two complementary fragments, provided the two flight paths are of sufficient length and chosen in the appropriate ratio. The energy signal of the detectors serves to fix the kinetic energy $E_K$ of the fragments. The results obtained with this technique [1,2] already exhibit all the main features of CF, which were confirmed and complemented later on by other methods.

A comprehensive program studying CF is underway on the fission fragment spectrometers LOHENGRIN and COSI FAN TUTTE of the Institut Laue-Langevin in Grenoble. The isotopes studies with thermal neutrons range from $^{229}$Th, $^{233}$U, $^{235}$U, $^{237}$Np, $^{239}$Pu, $^{241}$Pu, $^{245}$Cm up to $^{249}$Cf. The advantage with these spectrometers is that, along with the fragment masses $M$ and kinetic energies $E_K$, also the nuclear charges may be identified [3,4,5].

More recently the "twin" or "back-to-back" ionization chambers have been pushed to take CF data. For fission fragments the intrinsic energy resolving power $E_K/\delta E_K$ of gas ionization chambers supersedes the resolving power of solid state detectors by at least a factor of 5. In fact, the performance of ionization chambers may approach $E_K/\delta E_K = 10^3$ [6]. In a twin chamber, with the fission target placed inside the sensitive gas volume, $E_K/\delta E_K$ is mainly limited by the energy straggling of the fragments in the target and its backing. From the two energy signals of the complementary fragments and from momentum conservation one has $E_{K1}/E_{K2} = M_2/M_1$. In a contour plot in the $(E_{K1}, E_{K2})$ plane well separated lines corresponding to individual mass ratios $M_2/M_1$ and hence individual masses are to be observed in the CF regime. Note that in CF the masses $M_1$ and $M_2$ unambiguously add up to the mass of the fissioning nucleus. Moreover, variants of the
Bragg curve spectroscopy have been developed, allowing to measure also nuclear charge numbers \( Z \) of fragments in a twin chamber assembly \([7, 8, 9]\). The power of the chamber technique is the large solid angle of detection which compensates for the low yield of events in CF. First results obtained by this method in CF are already available \([9, 10]\).

3. EXPERIMENTAL RESULTS

Rather comprehensive experimental results on Cold Fragmentation are now available for the following reactions: thermal neutron induced fission of \(^{233}\text{U}\), \(^{235}\text{U}\) and \(^{239}\text{Pu}\), and spontaneous fission of \(^{252}\text{Cf}\). In all of the above cases, CF has been identified by looking for very high TKE events. Fig. 2 shows the example \(^{233}\text{U}(n,f)\). Plotted vs. the ratio of mass numbers of light to heavy fragments \(A_L/A_H\) are

a) the maximum TKE as obtained in ref. \([1]\) by extrapolating the distribution of TKE to zero yield,

b) the TKE at a fixed yield level of \(10^{-6}\)/MeV from ref. \([3, 4]\) and

c) the highest Q-value, i.e. the Q-value for a given mass ratio \(A_L/A_H\), but with the ratio \(Z_L/Z_H\) chosen in such a way that Q is maximised; the even charge numbers \(Z_L\) yielding the highest Q are indicated in the figure.

Whenever possible the \(Z\)-values were calculated from experimental data on nuclide masses \([11]\). In some cases these data had to be complemented by resorting to a mass table \([12]\).

It is seen from Fig. 2 that the two different ways of evaluating high TKE data essentially yield the same result. Therefore, in
the following the two sets of experimental data will be treated together.

The CF data exhibit three distinct features:
1) True Cold Fragmentation, with TKE$_{\text{max}}$ being equal to the respective Q$_{\text{max}}$ within the experimental uncertainty of about 1 MeV, is only attained for some specific mass fragmentations.
2) While Q$_{\text{max}}$ shows a pronounced odd-even staggering, there is no such effect for TKE$_{\text{max}}$, i.e. TKE$_{\text{max}}$ is smoothed as compared to Q$_{\text{max}}$.
3) Still, some structure seems to remain with TKE$_{\text{max}}$. It is observed from Fig. 2 that whenever the charge numbers change, there is a kink in an otherwise smooth variation of TKE$_{\text{max}}$ with mass ratio A_L/A_H. These kinks have been traced back to a "Coulomb effect" in CF [13].

The above features will be discussed in turn in the next sections.

4. TRUE COLD FRAGMENTATION
As reasoned in the Introduction, CF corresponds to the most compact scission configuration with Q = V_C + V_D, and with the potential energies at scission, V_C and V_D, being converted one to one into the two observable energies, TKE$_{\text{max}}$ and TXE$_{\text{min}}$ respectively (see eqs. (6) and (7)). Evidently, true cold fragmentation is only feasible, if - by chance - at the most compact scission configuration V_D = 0 obtains. Only in this case all of the available energy, Q, will show up as total kinetic energy, TKE$_{\text{max}}$.

The above condition is visualised in Fig. 3. There it is postulated that one or both fragments are deformed in their ground states. For this deformation the deformation energy V_D will vanish. If for this same deformation it so happens that the Coulomb repulsion V_C is equal to the Q-value, then all energy is exhausted by TKE and true cold fragmentation should be observed.

The arguments may be put in a more quantitative form by a simple
In the spirit of the static scission point model of B.D. Wilkins et al [15], the scission configuration is parametrised by two coaxial spheroids, with the tips of the spheroids a distance \(d\) apart. The distance \(d\) is kept fixed for all mass ratios of the nascent fragments. The shape of the spheroids is given by the major and minor semiaxis, \(R\) and \(r\) respectively. Closely following B. D. Wilkins et al., the major semiaxis, \(R\), is written in terms of a deformation parameter:

\[
R = k(\delta)r_0A^{1/3}(1+2\delta/3). \quad (8)
\]

In eq. (8) the factor \(k(\delta)\) is inserted to ensure volume conservation. The nuclear radius constant \(r_0\) has been taken to be \(r_0 = 1.16\) fm. The Coulomb repulsion potential \(V_C\) between the two spheroids 1 and 2 is then calculated from

\[
V_C = \frac{e^2Z_1Z_2}{D} F. \quad (9)
\]

The distance between the charge centers, \(D\), is given by \(D = R_1 + R_2 + d\), \(Z_1\) and \(Z_2\) are the charge numbers of the two fragments maximising \(Q\), while \(F\) is a correction factor for the Coulomb interaction of two spheroids as compared to spheres.

In accordance with the basic idea of the model expounded in Fig. 3, the deformations are chosen to be those of the ground state deformations of the respective fragments. Unfortunately, only a few experimental data are available [16]. Most of the ground state deformations were taken from a table prepared by P. Moller and J. R. Nix [17]. The table is based on an elaborate nuclear model calculation [14].

![Diagram](image)

**Fig. 3:** Repartition of energies at scission in case of true cold fragmentation. The fragments are assumed to be deformed in their ground states.
mass formula. Upon relying on this table, a difficulty for the above calculation is encountered whenever oblate ground state deformations are predicted. In a more detailed publication by the same authors 18 examples of energy vs. deformation are given for some nuclei. It appears that, even in those cases where an oblate deformation yields an absolute energy minimum, there are also favored prolate deformations lying close in energy to the absolute minimum. It is conjectured that in the process of nuclear fission only these prolate fragment deformations are relevant. Still, for reasons of consistency only the tabulated deformations have been kept for the calculations.

In addition to the Coulomb repulsion, a nuclear attractive potential has been included in the total interaction energy, $V_{\text{scission}}$, at scission. This rather small correction term has been modelled following J. Krappe and J. R. Nix [19].

Finally, the tip distance $d$ between the spheroids was treated as a free parameter. One readily realizes that for touching spheroids with $d = 0$ the interaction energy by far exceeds the available energy $Q$. This configuration is therefore outruled. Consequently, the parameter $d$ was increased until the interaction energy $V_{\text{scission}}$ is equal (within 1 MeV) to the maximum $Q$-value, $Q_{\text{max}}$, for at least one mass fragmentation. It should be stressed that $d$ was kept constant for all mass ratios of a given fission reaction.

The results, both from experiment and the model, are summarized in the figures 4 to 6 for thermal neutron induced fission of $^{233}\text{U}$, $^{235}\text{U}$ and $^{239}\text{Pu}$. The heavy line represents $Q_{\text{max}}$ vs. the mass ratio $A_L/A_H$. The points give the experimental total kinetic energy TKE at a fixed yield level $Y = 10^{-6}/\text{MeV}$ from [3,4]. The crosses show the experimental extrapolated maximum TKE$_{\text{max}}$ from [1,2]. On the other hand, the triangles correspond to the interaction energy $V_{\text{scission}}$ at scission calculated in the above model. The tip distance $d$ emerging from the model is indicated in the
figures. The spikes in the curve $V_{\text{sci}}$ vs. $A_L/A_H$ occur whenever at least one of the fragments has an oblate ground state deformation. A similar situation holds for the extremely high $V_{\text{sci}}$ values (out of scale of the drawings) close to symmetric mass splits. Triangles marked by an arrow pointing downwards sort out those nuclei having an oblate deformation, but where a prolate deformation is known to be energetically almost as favored. A prolate deformation would drastically reduce the calculated $V_{\text{sci}}$.

It is observed from the Figs. 4 to 6 that, both the experimental $TKE_{\text{max}}$ and the calculated $V_{\text{sci}}$ values come close to the respective $Q_{\text{max}}$ values for the same mass fragmentations (there is one exception for $A_L/A_H = 96/144$ in $^{239}\text{Pu}(n,f)$). This corresponds to the situation sketched in Fig. 3, with the interaction energy being equal to the $Q$-value and quantitatively converted into kinetic energy. The model thus correctly predicts those mass fragmentations where true cold fragmentation is to be expected. For the
mass combinations where $V_{\text{sci}}$ stays larger than $Q$, the fissioning system has to be deformed beyond its ground state in order not to violate energy conservation. The fragments will therefore acquire deformation energy and since $TXE$ then will be non-zero, the maximum possible TKE will lag behind $Q_{\text{max}}$. This compares well with the experimental data.

The tip distances $d$ which have been obtained for thermal neutron induced fission of U- and Pu-isotopes decrease from $d = 2.15$ fm for $^{233}\text{U}(n,f)$ down to $d = 1.90$ fm for $^{239}\text{Pu}(n,f)$. These values for $d$ are consistent with the findings of B. Wilkins et al. [15]. The trend of $d$ to decrease with increasing fissility is not unexpected: in cold fragmentation the scission point is pushed as close as possible towards the saddle point in deformation space and liquid drop model calculations show that the saddle point configuration is the less deformed the higher the fissility.

In Fig. 7 are plotted the calculated energies $Q_{\text{max}}$ and $V_{\text{sci}}$ as a function of the mass ratio $A_L/A_H$ for the spontaneous fission of $^{252}\text{Cf}$. For a large fraction of all masses the $V_{\text{sci}}$ values are excessively and unrealistically high. This is again due to oblate ground state deformations of the fragments. Conclusions should therefore be drawn with due caution. Still, the tip distance parameter $d = 1.80$ fm seems reasonable.
Experimental results on the cold fragmentation of $^{252}$Cf(sf) are presented in Fig. 8 (taken from ref. [9]). TKE values at fixed yield levels per MeV are compared to $Q_{\text{max}}$. In the insert independent charge yields are given.

The yields have been averaged for TKE lying between $TKE_{\text{max}}$ and the TKE indicated by the dot-dashed line. The agreement between the predicted masses being candidates for true CF in Fig. 7 and the experimentally observed cases of true CF in Fig. 8 is not as good as in the former instances, though the general trends are not at variance. It should be pointed out, however, that the $Q_{\text{max}}$ values in Fig. 7 are mainly based on experimental mass data [11], while those in Fig. 8 were taken from a mass table [17].

The discrepancies are largely removed, if in Fig. 8 use is made of the experimental $Q$ values. An intriguing special case obtains for the mass ratio $A_L/A_H = 120/132$, where from the experiment true CF shows up for an isolated fragment pair.

The model calculation at
least gives a hint to this behaviour. Also the well known dip in kinetic energy for symmetric mass splits seems to be reproduced by the calculations.

Comparing neutron induced and spontaneous fission it is remarkable that for the latter reaction one has to go down to much lower yields before reaching the CF regime. Without further systematic studies it is not possible to state whether this behaviour is due to the higher fissility of $^{252}$Cf or to a different behaviour of induced and spontaneous fission. In any case, it is the outstanding power of the twin ionization chamber method which made these studies of CF feasible.

A final remark concerning true CF should stress once again that one is probing here a compact scission configuration close to the saddle point. The mere fact that fragment properties seem to be decisive for the outcome of CF, points to the presence of fragments at a rather early stage of fission. Amongst others, the cluster model of fission provides a natural basis for this observation [20].

5. SMOOTH DEPENDENCE OF TKE$_{\text{max}}$ ON MASS RATIO
In all studies of CF being reviewed here, the fissioning compound nucleus is of the (even, even) type. As borne out by experiment, odd-A and even-A mass splits have comparable probabilities, up to the highest TKE. This means that the breaking of a neutron or a proton pair of the compound nucleus is not hindered in fission, even down to the lowest feasible excitation energies of the fragments. A closer inspection of Fig. 2 reveals that even Z charge numbers dominate in the maximum Q-values. Hence, the odd-even staggering of Q$_{\text{max}}$ in Fig. 2 is brought about mainly by neutron pairing. The smooth dependence of TKE$_{\text{max}}$ on mass ratio of the fragments is then readily understood, by assuming that at least one neutron pair is always broken, independent from the odd or even character of the mass split. By this conjecture, the potential gain in energy through neutron pairing can not be realised, and TKE$_{\text{max}}$ should not show any odd-even effect, in full agreement with experiment.
The high probability for neutron pair breaking, contrasted to a rather low probability for proton pair breaking, is supported by independent Z yield measurements in the CF regime. For example, at a fixed high kinetic energy of the light fragment $E_L = 110.5\text{MeV}$, ruling out prompt neutron emission in the reaction $^{233}\text{U}(n,f)$, the odd-even effect is much more pronounced for protons as compared to neutrons [3]. Quantifying the odd-even effect in the yields, $\delta$, by the difference between the even and the odd yields (with the sum being normalised to 100%), one finds for the protons $\delta = 46\%$ and for the neutrons $\delta = 11\%$. Based on the statistics of pair breaking, a model has been put forward by H. Nifenecker et al [21], which traces the above observation back to the preponderance of neutrons in heavy nuclei.

It is worthwhile to rephrase the arguments in more cautious and more general terms. The experimental message is that odd mass fragments come closer with TKE to their respective $Q_{\text{max}}$ than even mass ones. This means that even mass fragments stay with a higher excitation energy $TXE$. As reasoned in the introduction, in the limiting case of CF all of the final $TXE$ is tied up as deformation energy, i.e. $TXE = V_D$. But, let us now envisage the possibility that experimentally it might be very difficult, if not impossible, to reach this point exactly. In this case, besides being deformed, the fragments will be created with some internal excitation $E^*$. Admittedly, $E^*$ will be small for those TKE values which have been identified as CF by experiment. Of course, one can increase $E^*$ deliberately by looking for events at lower TKE. Again nuclear charge measurements may be invoked to give a clue. An example from the reaction $^{235}\text{U}(n,f)$ is given in Fig. 9 [5,22].

For a light fragment of fixed even mass $M_L = 102$, the independent relative nuclear charge yields are plotted as a function of the kinetic energy $E_{KL}$ of the light fragment. Open points are LOHENGRIN data [22], while full points stem from COSI FAN TUTTE [5]. At low $E_{KL}$ the two sets of data are at variance, but more credit should be given here to the measurement on LOHENGRIN, where a thin $^{235}\text{U}$ target was used. The target employed on COSI FAN TUTTE was rather thick and, therefore, only the data points at
higher $E_{KL}$ are reliable. In the present context, however, we are only interested in charge yield data at high kinetic energies. It is observed from Fig. 9 that, rather surprisingly, odd $Z$ charge numbers compete quite successfully with even $Z$ numbers up to fairly high kinetic energies. It is only for the very last data point at the highest TKE that the even $Z = 40$, maximising the Q-value, predominates.

The above findings for the even mass $M_L = 102$ can be immediately interpreted by invoking a statistical level density argument. For an even mass split of an even compound nucleus the two fragments are both either (even, even) or (odd, odd). With the conjecture that, at least for a narrow range of masses, charges and energies, the yields are governed by statistics, the higher level density of an even mass (odd, odd) nucleus will compensate its lower Q-value and give it the same yield as for an (even, even) nucleus. Stated otherwise, the energy gain for even-even nuclei is just balanced upon introducing effective energies, i.e. corrected for pairing, in a level density formula. It is only in case the TKE can be pushed high enough to leave behind less favorable Q-values that one is finally left with one unique (even-even) fragmentation corresponding to the maximum Q, like in Fig. 9.

Applying this same phase space argument now to all mass splits, even and odd, at the same yield all fragmentations should have the same total level density. The crucial point is that for even mass fragments also the charge numbers $Z$ maximising Q are even and one is, hence, faced with (even, even) nuclei with a low
level density; on the other hand, the odd mass fragments will show a comparatively high level density. This then explains why at very high TKE even mass fragments, with a dominant contribution by (even, even) nuclei, will stay with a higher excitation energy, i.e. further away from their optimum $Q_{\text{max}}$ as compared to odd mass fragments. In other words, the odd-even staggering of $Q_{\text{max}}$ is washed out in the TKE evaluated at fixed yield levels.

A last comment addresses the $^{252}$Cf(sf) experiment, where the accessible yield level was by more than a factor of ten lower than in the induced fission studies. It is seen from Fig. 8 that, in those mass regions where true CF obtains, an odd-even structure is indicated for TKE, contrary to the discussion above. However, if at the yield levels of the Cf-experiment it is indeed feasible to reach the individual true ground states of the fragments, then evidently the odd-even staggering of $Q_{\text{max}}$ should also become visible in the limiting TKE values. As yet it is not clear whether this feature is specific to spontaneous as contrasted to induced fission, or whether it is just due to the lower yield levels becoming accessible with the twin chamber technique.

6. COULOMB EFFECT

The experimental TKE data vs. mass in Figs. 4 to 6 and in Fig. 8 exhibit a structure which is spaced at about 5 mass units. The same spacing is observed in the bottom part of Fig. 8 for the distribution of independent charge yields: every 5 masses the leading even charge numbers switch to a neighboring even charge. This seems to imply that the kinks in TKE vs. mass occur at those masses where the charge numbers $Z$ of the fragments change.

The interpretation, which has been given by M. Montoya and R. W. Hasse in terms of a Coulomb effect, is illustrated in Fig. 10. The diagram, showing the repartition of energies as a function of deformation in a scission point model, is again invoked. Fig. 10 displays the energies vs. deformation for two neighboring mass fragmentations, with the charge numbers differing by one unit, viz. $Z_L$ and $(Z_L + 1)$ for the light fragment (and correspon-
Cold Fragmentation

Energy

\[ V_C(Z_L+1) \]

Cold Fragmentation

\[ V_C(Z_L) \]

Free Energy

\[ V_P \]

\[ V_D \]

TKEmax

TKEmax

Q

0 0 0

Deformation

0 0

Fig. 10: Coulomb effect in cold fragmentation:
upon shifting the charge of the light
fragment from \( Z_L \) to \( (Z_L + 1) \) the
maximum possible TKEmax decreases
dingly \( Z_H \) and \( (Z_H - 1) \) for
the heavy one. It is assumed
that both, the available energy \( Q \) and the deformation
potential \( V_D \), are identical
for the two mass and charge
splits. These simplifying
assumptions ought to be better
justified from case to case,
but let us take them for
granted in the following.

In any case, the Coulomb
repulsion \( V_C \) at a given deforma-
tion will be larger for the
combination of charges \( (Z_L + 1, \]
\( Z_H - 1) \) as compared to the
combination \( (Z_L', Z_H') \). This
is simply due to the fact
that \( Z_L < Z_H \) and that the
product \( Z_L Z_H \) in the Coulomb potential will reach a maximum for
\( Z_L = Z_H \) at charge symmetric splits. Note that the sum of \( Z_L \) and
\( Z_H \) equals the charge number of the fissioning nucleus. The shift
of \( V_C \) will cause a displacement of the cold fragmentation point
in deformation space. As a consequence, the maximum possible
kinetic energy release TKEmax will also change. As shown in Fig.
10, upon moving from the charge combination \( (Z_L', Z_H') \) to \( (Z_L + 1, \]
\( Z_H - 1) \), the TKEmax decreases. This then explains, why at those masses where the
light fragment nuclear charge switches to the next higher one,
the limiting TKEmax has a tendency to drop and thereby to induce
a kink in an otherwise uniformly varying total kinetic energy.

7. CONCLUDING REMARKS

From an experimental point of view, studies in cold fragmentation
are difficult because CF is such an improbable fission process.
This means that the measuring times have to be very long. In
addition, it has proven crucial not only to measure with high
accuracy kinetic energies and masses of fragments, but also nuclear
charges. To take nuclear charge data at low statistics is a delicate task. Since the first pioneering experiment, a big effort has been put into technological developments, and studies have now become feasible which, even a few years ago, just could not be performed.

On the other hand, from a physical point of view, the study of CF is felt to be rewarding. Even though CF is not a typical, but rather an exotic limiting case of nuclear fission, much can be learnt on this process. This is mainly due to the fact that in CF simplifying conditions obtain, with the energetics being largely governed by the potential energies at the scission point. Therefore, even simple models can give some insight. In the present review some distinct features of CF, like true cold fragmentation and the structure of $TKE_{\text{max}}$ as a function of fragment mass, have been shown to be understood by these models. Certainly, many questions are left open and more systematic and more accurate data are still needed.

One last but very important point should be made. Although characteristic features of CF may be explained in simple terms, it is a challenge for any general theory of fission to cope with cold fragmentation as an observable process, i.e. a process with non-vanishing yield. Most theories of fission have difficulties in predicting correctly the width of the distribution of total kinetic energies. From theory the widths of TKE distributions are narrower than from experiment. This means that cold fragmentation is wholly unexpected. However, encouraging results have been presented by J. F. Berger et al. [23]. In a sophisticated microscopic model of fission being based on the HFB-method, these authors first calculated the potential energy landscape for the $^{240}\text{Pu}$ compound nucleus. In a second step the dynamics of the descent from saddle to scission was investigated. It could be shown, and this is a considerable success, that at least the order of magnitude for the yield of cold fragmentation may be understood. It is hoped and should be encouraged that further studies will cover a variety of fissioning systems.
REFERENCES

FISSION FRAGMENT ENERGY AND MASS DISTRIBUTIONS IN THE SPONTANEOUS FISSION OF THE PLUTONIUM ISOTOPES

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Abstract
The fission fragments energy and mass characteristics and their correlations have been studied for the spontaneous fission of $^{238}$Pu, $^{240}$Pu and $^{242}$Pu and the thermal neutron induced fission of $^{239}$Pu and $^{241}$Pu. The results are interpreted in terms of the static scission point model.

1. INTRODUCTION

In the frame of a systematic study of the fission fragment mass and energy characteristics, the spontaneous fission of $^{238}$, $^{240}$, $^{242}$Pu and the thermal neutron induced fission of $^{239}$Pu has been studied. The measurements were performed at the BR1-reactor of the Nuclear Energy Center at Mol. The energy and mass characteristics of the fission fragments were determined with the so-called double-energy method. In this method the fission fragments are detected by two colinear surface barrier detectors. The analysis was based on the mass and momentum conservation relations and the Schmitt Neiler calibration procedure, in which the detector constants can be determined from the pulse-height spectra of the $^{239}$Pu($n_{th}, f$) reaction. For the measurements a mixture of a spontaneously fissioning Pu-isotope and $^{239}$Pu was evaporated on a very thin polyimide backing. In this way, previous measurements on $^{242}$Pu(sf) and $^{240}$Pu(sf) could be improved, as far as their resolution concerns. This is illustrated in fig. 1, which shows the better mass resolution obtained with an evaporated $^{242}$Pu target instead of a target prepared by electrospraying. In the present experiments, only evaporated samples have been used, applying the same calibration method and the same $^{239}$Pu($n_{th}, f$) calibration reaction for the three measurements. So an accurate comparative study could be performed. The BR1-reactor was operated during the day and shut down at night and during the weekends. Such an operation allows one to measure a sequence of separate (sf)($n_{th}, f$) runs, which can be analysed individually. In this way a very careful detector calibration with the $^{239}$Pu($n_{th}, f$) reaction can be obtained.

2. RESULTS AND DISCUSSION

The results can be interpreted in terms of the static scission point model of Wilkins and analogous calculations performed by Moreau and Heyde. In these calculations the probability for the formation of a complementary pair of fission...
Neutron number

Fig. 2. Neutron shell corrections as a function of the deformation and the neutron number

Additional effects may result from neutron shell corrections in the regions C-B in the light fragment. The neutron shell corrections play an important role when the formation of both fragments is favoured by shell effects. For $^{240}$Pu(sf) and $^{242}$Pu(sf) this leads to a large peak in the mass region $m_A \approx 135$, due to the combined influence of the shell G in the heavy and the shell C in the light fragment. For $^{238}$Pu(sf) on the other hand, only the neutron shell G plays a role in this mass region, which explains the smaller peak yield observed in our measurements. However for $^{238}$Pu(sf) we notice a high peak yield around mass 142, which can be explained by the preferential formation of a heavy fragment with $N_H \approx 86$ and a light fragment with $N_L \approx 58$, due to the simultaneous influence of the shells H resp. B. The enhanced influence of the shell B in the light fragment is less pronounced for $^{240}$Pu(sf) but still the mass distribution shows a shoulder around mass 142.

For $^{242}$Pu(sf) both shells do not coincide, but each shell on its own causes a structure in the mass distribution: shell H in the mass region 140 and shell B in the mass region 144. The strong decrease of the mass yields in the extreme asymmetric mass region reflects the lack of strong negative neutron shell corrections in this region.

2.2. Kinetic energy distributions for $^{238}$, $^{240}$, $^{242}$Pu(sf)

A comparison of the total kinetic energy as a function of the heavy fragment mass (fig. 4) shows the typical maximum in the mass region 130-135. This maximum can be explained by the formation of spherical fragments due to the influence of the spherical neutron shell $N \approx 82$, which is enhanced by the spherical proton shell $Z \approx 50$. 
fragments is determined by the potential energy of the system at the scission point. This potential energy is a sum of liquid drop terms and shell correction terms. It appears that strong negative neutron shell corrections as shown in fig. 2 play an important role in the formation of the fragments. Taking into account that the fission fragments conserve the charge/mass ratio of the compound nucleus the most probable neutron number for a given mass can be calculated.

**TABLE 1: Main energy and mass characteristics of the spontaneously fissioning plutonium isotopes, all calibrated relative to $^{239}$Pu(n$_{th}$,f)**

<table>
<thead>
<tr>
<th></th>
<th>$^{238}$Pu(sf)</th>
<th>$^{240}$Pu(sf)</th>
<th>$^{242}$Pu(sf)</th>
<th>$^{239}$Pu(n$_{th}$,f)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_k^*$ (MeV)</td>
<td>176.5</td>
<td>179.1</td>
<td>180.4</td>
<td>177.7</td>
</tr>
<tr>
<td>$\sigma_{E_k^*}$</td>
<td>11.9</td>
<td>12.5</td>
<td>12.1</td>
<td>12.5</td>
</tr>
<tr>
<td>$E_f^*$ (MeV)</td>
<td>103.1</td>
<td>103.4</td>
<td>102.9</td>
<td>103.3</td>
</tr>
<tr>
<td>$E_A^*$ (MeV)</td>
<td>73.4</td>
<td>75.7</td>
<td>77.5</td>
<td>74.4</td>
</tr>
<tr>
<td>$m_A^*$ (u)</td>
<td>139.2</td>
<td>138.7</td>
<td>138.2</td>
<td>139.7</td>
</tr>
<tr>
<td>$\bar{m}_f^*$ (u)</td>
<td>98.8</td>
<td>101.3</td>
<td>103.8</td>
<td>100.3</td>
</tr>
<tr>
<td>$\sigma_{m_f^*}$</td>
<td>6.0</td>
<td>5.7</td>
<td>5.4</td>
<td>6.0</td>
</tr>
<tr>
<td>$N_a^*$</td>
<td>1972</td>
<td>14842</td>
<td>11020</td>
<td>1.9x10^6</td>
</tr>
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</table>

2.1. Comparison of the mass distributions for $^{238,240,242}$Pu(sf).

Fig. 3 shows the almost constant position of the heavy fragment mass peak, so that the increase of the mass of the compound nucleus results in a shift of the light fragment peak. This is demonstrated numerically by the $<m_f^*>$ and $<m_A^*>$ values given in table 1. Fig. 3 also shows that many structures appear in the mass distributions. These observations can be explained by the shell effects mentioned above. In the symmetric mass region there are no shell corrections available which can influence the formation of the fragments. For this region one observes indeed an almost zero yield in the mass distributions. For more asymmetric fission, the formation of the fragments will be mainly influenced by the spherical neutron shell G and the deformed neutron shell H in the heavy fragment. These shells cause the con-
The coefficient of dissymmetry (c.d.) of $<E_k*> (m_h^*)$ is given in fig. 5. One observes the highest dissymmetry in the mass region 130-135, and oscillations around low dissymmetry for the other mass regions. This low dissymmetry values can be explained by the presence of more than one configuration with different deformations for a given mass split $m_h^* / m_i^*$. This is illustrated in fig. 6 showing the total kinetic energy distributions for mass split 135-105 for $^{240}$Pu(sf) and for mass split 135-107 for $^{242}$Pu(sf) which have been decomposed in a sum of two distributions. Indeed, for a given mass split two configurations with different total deformation can appear: for mass $m_h^* ~ 135$ spherical fragments can be formed due to the influence of the spherical neutron shell G, but also deformed fragments can be formed under the influence of an offshoot of region H. The formation of the light fragment can only be influenced by the presence of region C, which means that the light fragment has an equal deformation for both configurations of the heavy fragment. Since the total kinetic energy is mainly due to the Coulomb repulsion of the two fragments, the total kinetic energy can be calculated as:

$$E_k^* = \frac{Z_i^*Z_h^*e^2F}{D} \quad (a)$$

with $Z_i^*$ and $Z_h^*$ the proton number, D the distance between the charge centers and F a
shape factor. As in ref.3, D is calculated as:

\[ D = d + r_m \left( \frac{1 + \frac{1}{3} \varepsilon_2^i}{1 - \frac{2}{3} \varepsilon_2^i} \right) + r_m \left( \frac{1 + \frac{1}{3} \varepsilon_2^h}{1 - \frac{2}{3} \varepsilon_2^h} \right) \]  

with \( d \) a constant equal to 1.4 fm and \( \varepsilon_2^i \) and \( \varepsilon_2^h \) deformation parameters. From (a) and (b) the total kinetic energy for a given configuration \( (m_i^*, \varepsilon_2^i) \) and \( (m_h^*, \varepsilon_2^h) \) can be deduced taking into account the deformations calculated by Moreau and Heyde (fig.1).

For both mass splits one obtains in this way a total kinetic energy \( E_k^* \approx 190 \text{ MeV} \) for the configurations dominated by the neutron shell \( G \), and \( E_k^* \approx 170 \text{ MeV} \) for the configurations influenced by the region \( H \). These calculated values are in good agreement with the average values from the Gaussian fits (fig.5).

2.3. Influence of the excitation energy on the shell effects.

A comparison of the mass distribution of \( ^{240}\text{Pu}(sf) \) with \( ^{239}\text{Pu}(n_{th},f) \) (fig. 6) shows a narrower mass distribution, a much higher peak yield and more pronounced fine structures for the spontaneous fission as compared to the neutron induced fission, which reveals a decrease of the influence of the shell corrections with increasing excitation energy. For the above mentioned systems, the difference of more than 6 MeV excitation energy does not show up, the kinetic energy being even higher for the spontaneous fission (cfr. fig.8 and table 1). This points to the formation of more compact configurations for the spontaneous as compared to the neutron induced fission and can be attributed to a decrease of the
shell corrections with increasing excitation energy of the compound nucleus. The same conclusion can be drawn from a comparison of $^{242}\text{Pu}(sf)$ and $^{241}\text{Pu}(nth,f)$.

Fig. 8. Total fission fragment kinetic energy for $^{240}\text{Pu}(sf)$ and $^{239}\text{Pu}(nth,f)$ as a function of the heavy fragment mass

3. Conclusions

The present results clearly illustrate that the formation of the fission fragments is very much influenced by the shell effects, especially by the strong spherical neutron shell $N \sim 82$ but also by the deformed shell $N \sim 86$. The influence is enhanced when both fragments have a stability caused by a neutron shell. It decreases however with increasing excitation energy of the fissioning system.

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STUDY OF THE $^{241}$Pu(n$_{th}$,f)-FRAGMENTS WITH COSI FAN TUTTE

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Abstract
Correlated mass and energy distributions have been obtained for $^{235}$U(n$_{th}$,f) and $^{241}$Pu(n$_{th}$,f) fragments, using the COSI FAN TUTTE spectrometer of the I.L.L. in Grenoble. Fine structures due to neutron shell and pairing effects have been observed.

1. INTRODUCTION
To complete our systematic study of the fission fragments mass and energy characteristics for several Pu-isotopes$^1$, the high resolution spectrometer COSI FAN TUTTE$^2$ was used to obtain correlated mass and energy distributions of the fission fragments of $^{241}$Pu(n$_{th}$,f). With the COSI FAN TUTTE spectrometer the energy and velocity of the fission fragments are measured in coincidence with such a high resolution that one almost gets a one-by-one mass separation.

2. METHOD
The experimental set up comprises a start and stop TOF system and an energy detector to measure the velocities and kinetic energies of single fission fragments in coincidence. The start and stop detectors are of the same type$^3$. One obtains for the TOF-system a resolution of $\delta T \leq 110$ ps for the light fragment group. The energy detector is an axial ionization chamber with a resolution of $\delta E \leq 500$ keV for the light fragment group$^4$. Since $m = 2E/v^2$, the mass resolution can be calculated as follows:

$$(\delta m/m)^2 = (\delta E/E)^2 + (2\delta v/v)^2.$$  

If we consider a light fragment with energy $E = 100$ MeV, one has $\delta E/E = 0.5 \%$. With a flight path $L = 107$ cm and $v = 1.4$ cm/s one gets from $v = L/T$, $\delta v/v = 0.2 \%$. This results in a calculated mass resolution $\delta m/m = 0.6 \%$.

Fig. 1 shows the TOF spectrum for fission fragments from a 10 µg/cm$^2$ evapo-
Fig. 2. The energy pulse-height spectrum for $^{235}$U(n$_{th}$,f).

Fig. 4 The $^{235}$U(n$_{th}$,f) mass distribution.

3. RESULTS

For the $^{241}$Pu(n$_{th}$,f) measurements a 7 $\mu$g/cm$^2$ electrosprayed plutonium acetate-layer on a 30 $\mu$g/cm$^2$ thin poly-imide backing was used. About $1.5 \times 10^5$ fission events were recorded. The post-neutron emission mass distribution for the light fragment peak is given in fig. 5. In the same figure, the present results are compared to the radiochemical data of Cuninghame$^{71}$. Both distributions are in fair agreement. This is also the case for the average mass, for which we obtain 100.2 u compared to 100.2 u for Cuninghame.

rated $^{235}$UF$_4$ layer. This measurement was used to calibrate the TOF-system with the mean velocity values obtained by Geltenbort et al$^{15}$. The corresponding energy spectrum is given in fig. 2. From the correlated TOF and energy pulse-height data one can construct a (T,E)-matrix (fig. 3), which clearly shows lines with constant mass. Using the energy pulse-height relation:

$$E = 0.5 m v^2 = (a+a'm) P + (b+b'm)$$

determined by Weissenberger et al$^{16}$ for the same ionization chamber, an intrinsic calibration for the fragment masses can be obtained. The constants were determined by a $^{235}$U(n$_{th}$,f) measurement using a 350 $\mu$g/cm$^2$ evaporated $^{235}$UF$_4$ target to obtain a good statistical accuracy. From the $^{235}$U(n$_{th}$,f) mass distribution shown in fig. 4, one calculates an experimental mass resolution $\delta m/m = 0.7\%$. 

Fig. 5 Comparison of the $^{241}$Pu(n$_{th}$,f) mass distribution obtained in this work (•) and by Cuninhame et al$^{71}$(o).
Contour plot of correlated time-of-flight and energy pulse-height data for $^{235}$U(n$_{th}$,f). The matrix cells with more than 17 counts are given in red (the maximum number of counts in one cell is 30).
Fig. 6 The $^{241}$Pu($n_{th},f$) post-neutron mass distribution for light fragments with a kinetic energy higher than 110 MeV.

In fig. 6 a preliminary result is shown for the light fragment post-neutron mass distribution corresponding to light fragment kinetic energies above 110 MeV. Here the influence of the neutron shells $N \sim 58, N \sim 64, N \sim 82$, and $N \sim 88$ and the odd-even effect becomes apparent.

Fig. 7. Highest total kinetic energy (*) for $^{241}$Pu($n_{th},f$) as a function of the light fragment mass. For each mass split the highest Q-value (a) and the one immediately below (b) are indicated.

In fig. 7, the maximum total kinetic energy for each mass split is displayed as a function of the light fragment mass, together with the highest Q-value ($Q_{\text{max}}$) and the Q-value closest to $Q_{\text{max}}$, but obtained with a different $Z_{1}-Z_{2}$ combination. One observes two separate regions where the fragments are formed nearly in the ground state. These mass splits can be interpreted as cold fragmentations.

Furthermore, the energy-mass correlation data confirm the strong influence of the $N \sim 50$ shell, as already observed by Càitucoli et al. During these measurements, also information on the nuclear charges has been obtained using the method described by Oed et al. The analysis of these data has not yet been finalised.
4. CONCLUSION

During a first measuring campaign on the COSI FAN TUTTE spectrometer, correlated energy- and mass distributions have been obtained for the $^{241}$Pu$(n_{th},f)$-fragments. Structures due to neutron shell and pairing effects have been observed. More detailed and more complete information is expected to be obtained from a coming second campaign.

Acknowledgments

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Fission fragment properties of 235U(n,f) in the neutron energy range from thermal to 1 MeV

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ABSTRACT

The fission fragment energies and emission angles of both fragments have been measured for 235U(n,f) in the neutron energy range from thermal to 1 MeV. The experimental detection technique used together with the pulsed white neutron source GELINA is described. The fission fragment angular anisotropies show, in agreement with earlier measurements, negative values below 0.3 MeV. This leads to the conclusion, that either R- (eigenvalue \( r = +1 \)) or S- (s = +1) invariance of the fissioning system at the saddle is fulfilled. The average total kinetic energy shows structure in the resolved resonance neutron energy region below 120 eV. This structure is anticorrelated with \( \bar{N}(E) \), the average number of prompt fission neutrons. The average total kinetic energy changes are mass dependent.

1. INTRODUCTION

Fission properties like fragment mass-, and kinetic energy distributions of thermal and high energy (>0.5 MeV) neutron induced fission of 235U are rather well known \(^1,2\). For the understanding of the fission process in the low and medium neutron energy range (eV to ~ 0.5 MeV) the changes of these properties with the excitation energy of the compound nucleus 236U and with the resonance spin (\( J=3^+, 4^+ \) for s-wave neutrons), are of interest. No systematic measurement has been done in this energy region in the past. Therefore the fragment mass, kinetic energy and angular distribution of 235U(n,f) were measured at GELINA, as function of incident neutron energy from thermal to 1 MeV.

2. EXPERIMENT

The experiment was setup at about 9 m from the GELINA target. The experimental arrangement and the electronic equipment is shown in fig. 1. The fission fragment detector is a twin ionization chamber consisting of a common cathode and two Frisch grids and anodes \(^3,4\). The 235UF\(_4\)-layer is evaporated on a thin polyimide backing (~30 \( \mu \)g/cm\(^2\)) covered by ~80 \( \mu \)g/cm\(^2\) Au. This to ensure electrical conductivity
since the sample is positioned in the centre of the common cathode of the two chamber parts. The thickness of the 235UF4-layer is ~ 47 µg/cm² and has a diameter of 7.0 cm. The neutron energy was determined by a time-of-flight technique using a time-coder with 4 ns resolution per channel. The timing signal was obtained from the cathode of the ionization chamber which was operated as a flow chamber with a gas pressure of 10^5 Pa 90 % Ar, 10 % CH₄. From the anode signals the energies of the two fragments are obtained 2,4), whereas the sum signals of the respective grids and anodes are energy and angle dependent. Both signals together allow the determination of the cosine of the fragment emission angle with respect to the neutron beam direction.

The GELINA γ-flash induces a signal of almost the same height in the ionization chamber as the fission fragments do. This disturbs the measurements of fission events during the period of the γ-flash signals. This time period correspond to the neutron energy range above 3 keV. Since this is one of the energy regions of interest, a compensation technique was applied to reduce the disturbing influence of the γ-flash. The γ-flash signals measured in an unloaded but otherwise identical ionization chamber were subtracted from the signals of the chamber which contains the 235UF4-layer. This way the final response of the ionization chamber to the γ-flash could be reduced to less than 2 MeV.

The four analog signals from the detector were digitized and stored on magnetic tape together with the neutron flight time information from the time-coder for later off-line evaluation. Approximately 8·10⁶ fission events were recorded in the neutron energy range from 1 eV to 1 MeV. The measurements were regularly interrupted for measurements with thermal neutrons which served as a reference. These neutrons were produced by thermalization in a paraffin (CH₂)n block positioned in front of the detector. Approximately 5·10⁶ thermal events were recorded.

3. ANGULAR DISTRIBUTION

Angular distributions are important for the application of the 235U(n,f) standard cross section and have been investigated in several previous measurements 5,6).
Those measurements have shown, that for incident neutron energies below 300 keV the anisotropy \( W(0°)/W(90°) - 1 \) of the fragment angular distribution becomes negative. Theoretical calculations of the anisotropy based on statistical theory in this neutron energy region \(^2\) can only reproduce these negative anisotropies if in the calculation axial symmetry at the saddle point, but with restrictions (\( R^-\), \( S^-\) invariances) on the possible quantum states in which the compound nucleus can be formed, is assumed. Unfortunately the difference between these two invariances is small and it is not possible to distinguish between the two invariances by a fit to the existing data. In ref. \(^2\) it is shown in detail how \( \cos \theta \) values for fission fragments can be found from the ionization chamber signals. The same procedure is used in the present experiment. Figure 2 shows the measured anisotropies in comparison with the above mentioned previous measurements. For the first time the complete neutron energy range from about 3 keV to 0.5 MeV has been covered. The measured anisotropies are in good agreement with the earlier data, whereas the theoretical calculations, even if they reproduce the negative anisotropies, can not reproduce for a fixed \( K \) quantum number the tendency of the experimental data.

4. TOTAL KINETIC ENERGY AS FUNCTION OF \( E_n \) IN THE RESOLVED RESONANCE REGION.

According to the "channel theory" of fission from Bohr \(^7\) it is expected to find differences in the fragment mass- and total kinetic energy distributions depending on the resonance spin (\( 3^- \) or \( 4^- \)), which is the vector sum of the ground state spin of the fissioning nucleus (\( 235u, J_0 = 7/2^- \)) and the spin of the absorbed \( s^-\) wave neutron (\( s = 1/2, l = 0 \)). Experiments performed in the resonance region were measurements of the average number of prompt fission neutrons \( \overline{\nu}_p(E) \) \(^8\) with rather discrepant results and radiochemical measurements to determine asymmetric to symmetric fission fragment mass ratios \(^9\).

In the lower part of fig. 3 the relative number of prompt fission neutrons \( \overline{\nu}_p \) measured by R.E. Howe et al. \(^8\) is shown as function of the incident neutron energy. In the upper part of fig. 3 the presently measured average total kinetic energy (TKE) relative to the thermal value is plotted for the same energy intervals. In the neutron energy region from 1 eV to about 25 eV a significant positive structure of the TKE relative to the thermal value is seen. The relative \( \overline{\nu}_p \)-values show the same variation but anticorrelated as expected from energy balance consideration.
Fig. 3: Upper part: relative total kinetic energy Lower part: Relative number of prompt fission neutrons $\nu_p$ as function of incident neutron energy from ref.8).

Fig. 4: Relative total kinetic energy for $4^-$ and $3^-$ resonances as function of incident neutron energy.

Fig. 5: Relative kinetic energy as function of mass split
a) for $3^-$ resonances below 20 eV
b) for 19.3 eV, $4^-$ resonance.

Structures in the relative TKE are also visible around 55 eV and 100 eV. They are, however, not as pronounced in the $\nu_p$-values.

According to the resonance spin assignments of M.S. Moore et al. 10), a selection of the completely resolved resonances below 35 eV into $4^-$ and $3^-$ groups is made. Fig. 4 shows the relative TKE-values of these resonances. It is seen that the relative TKE-variation is followed only by the $4^-$-resonances, whereas the $3^-$-resonances stay on a constant level but significantly higher than the thermal value. Furthermore, the relative TKE-variation as function of mass-split for the $4^-$ and $3^-$ resonances show nearly the same dependence. Figure 5a shows the relative TKE as function of mass-split for the sum of the three $3^-$-resonances below 20 eV. Figure 5b gives the same for the 19.3 eV, $J = 4^-$-resonance. For both spin states the TKE increases for the more symmetric mass-splits. From mass ~ 134 to ~ 150 the TKE stays around the thermal value. For the extreme asymmetric mass-splits the TKE-dependence becomes different for the two spin states. The TKE for the $4^-$-spin state decreases below the thermal value, whereas the TKE for the $3^-$-spin state increases. Up to now it
is assumed that thermal neutron induced fission is a superposition of $3^-$- and $4^-$-spin states. But then it is not understood, why only the $4^-$-resonances show structure in the relative TKE as function of neutron energy. Also the dependence of the TKE as function of mass-split can not be understood. Another interpretation is not yet found.

5. CONCLUSIONS

The present results for the fission fragment angular anisotropy as function of incident neutron energy confirm the negative anisotropies found in previous measurements below $E_n \approx 300$ keV. Also theoretical calculations can reproduce the negative anisotropies under the assumptions that either $R-$ (eigenvalue $r = +1$) or $S-$ ($s = +1$) invariance of the fissioning system at the saddle point is fulfilled. $S$-invariance would be in agreement with a suggestion of Bohr 7) that the nucleus at saddle would be pear-shaped. The average total kinetic energy shows structure below 120 eV incident neutron energy which is anticorrelated with structures in $\bar{\nu}(E)$. The TKE changes are mass dependent but up to now a theoretical interpretation is not found.

6. REFERENCES

Fission fragment energies and the emission angles with respect to the incident neutron beam were measured for $^{235}\text{U}(n,f)$ using a twin ionization chamber. A neutron energy range from thermal to 6 MeV in steps of 0.5 MeV was covered. The pre-neutron emission mass distributions were evaluated and described by a five-Gaussian representation. The parametrization made visible a sudden change in the incident neutron energy dependence for all but the symmetric fission parameters between 1.5 and 2.0 MeV, which might be due to pair breaking. From the total kinetic energy as function of mass split the sum of the deformations of the two fragments was evaluated in the frame of Wilkins static scission point model. Here clear proton pairing effects became visible which seem to make the even-proton fragments less deformed than neighbouring odd-proton fragments. The present data show also that the observed drop in TKE averaged over all fragments above 4 MeV is due to changes in TKE for different mass splits with excitation energy. At a yield level of $10^{-6}$ cold fragmentation was observed for light fragment masses 87 to 110. In this mass region the light fragments are prolate and have a ground state electric quadropole moment larger than $\approx 1$ barn. Angular distributions for the different fragment masses showed no mass dependence.

1. INTRODUCTION

For the thermal neutron-induced fission of $^{235}\text{U}$ several measurements of fragment mass and kinetic energy distributions exist. For the understanding especially of the fission dynamics the changes of these distributions with the excitation energy of the compound nucleus $^{236}\text{U}$ are of interest. They have not yet been measured systematically. Therefore, the mass, kinetic energy, and angular distributions of fission fragments from $^{235}\text{U}(n,f)$ with incident neutron energies from thermal to 6 MeV were measured in steps of 0.5 MeV using the Van de Graaff accelerator of CBNM as a neutron source. About $1.5\times10^7$ fission events were recorded for thermal fission and $\sim 10^5$ events for each measurement with neutron energies higher than thermal.
2. EXPERIMENTAL SETUP

The experimental setup is shown schematically in fig. 1. The $^{235}\text{UF}_4$-layer is positioned on a thin polyimide backing which was covered by about 20 μg/cm$^2$ Au in order to make it electrically conducting. This sample is mounted in the centre of the cathode of a twin ionization chamber as proposed in 1). The two fragments of each fission event are then detected simultaneously. The inserted Frisch-grids ensure that the anode signals are proportional to the fragment energies $E_1$ and $E_2$. The grid signal is a function of the $E_i$ and $\cos \phi_i$ where $\phi_i$ is the angle between the normal of the cathode and the ion trace made by the fragment $i$ in the counter gas 2). Therefore the detector gives four signals for one fission event which determine the two fission fragment energies and the emission angles of the fragments with respect to the incident neutron beam. The acquired data were saved event by event on magnetic tape for further evaluation.

Fig. 1 Experimental setup.

3. MASS DISTRIBUTIONS

From the measured fragment kinetic energies the pre-neutron emission mass distributions were calculated. Corrections were applied for the energy losses in the sample, for the neutron emission as function of fragment mass and incident neutron energy, and for the laboratory to centre-of-mass system transformation at incident energies larger than thermal. Fig. 2 shows as examples the mass distributions as measured at thermal and 6 MeV incident neutron energies.
The measured mass distributions are fitted with five Gauss distributions as indicated by the dashed curves in fig. 2:

\[ Y(M) = \sum_{i=1}^{5} W_i \left( \frac{1}{(2\pi)^{1/2}} \sigma_i \right) \exp \left[ -\frac{(M - M_{i0})^2}{2\sigma_i^2} \right] \]

where

- \( M \) is the fragment mass
- \( M_{i0} \) is the center of Gauss function \( i \)
- \( W_i \) is the weight of Gauss function \( i \)
- \( \sigma_i \) is the variance of Gauss function \( i \)

It is assumed that \( M_{30} = 118, \ M_{50} = 236 - M_{10}, \ M_{40} = 236 - M_{20}, \ W_5 = W_1, \)

\[ W_4 = W_2 \sum_{i=1}^{5} W_i = 200, \quad \sigma_5 = \sigma_1 \quad \text{and} \quad \sigma_2 = \sigma_4. \]

The full line in fig. 2 represents the result of a least squares fit. The seven parameters \( W_1, W_2, W_3, M_1, M_2, \sigma_1 \) and \( \sigma_2 \) are shown as function of the incident neutron energy in fig. 3. The increase of the symmetric yield is exponential.

The other parameters are expected to vary linearly with the nuclear temperature, \( \Theta \), at the saddle point \( 4 \) and according to \( 4 \) \( \Theta \) is at the saddle proportional to the square root of \( \) the excitation energy above the barrier \( \Theta \alpha (E_n + B_n - E_d)^{1/2} = (E_n [\text{MeV}] + 1)^{1/2} \). Since all the parameters, but \( W_3 \), show a discontinuity at a neutron energy between 1.5 and 2.0 MeV, these data were fitted separately above and below this boundary with the expression
The results are shown as full lines in fig. 3. The most striking behaviour of these parameters is the almost constant value for \( E_n \leq 1.5 \text{ MeV} \) and much stronger dependence on excitation energy for \( E_n \geq 2 \text{ MeV} \). This sudden change might be connected with the onset of pair breaking. No previous mention of this sudden change in the mass distribution was found in the literature.

4. **TOTAL KINETIC ENERGY AND FRAGMENT DEFORMATION**

The fragment total kinetic energy is closely connected to the fragment deformations \( \beta_1 \) and \( \beta_2 \), since the deformations determine the average distance between the charge of the fragments at the moment of scission and therefore the coulomb repulsion energy. The total kinetic energy is thus given by:

\[
\text{TKE} = Z_1 \cdot Z_2 \cdot e^2 / D (\beta_1, \beta_2)
\]

where \( Z_1 \) and \( Z_2 \) are the proton numbers of the fragments 1 and 2 respectively, and \( D \) is the average distance between the charge of the two fragments at scission. In the static scission point model, \( D \) is given by \( a_1 (\beta_1) + a_2 (\beta_2) + d \); \( a_1 \) and \( a_2 \) are the major axis of the prolate spheroids representing the deformed fragments; \( d \) is chosen as in 5) to be 1.4 fm.

The total kinetic energy and its variations with incident neutron energy can then give information on changes in the deformation of the fragments at the scission point, and hence...
give a measure of the potential energy surface as function of mass split and excitation energy. Assuming that both fragments are equally deformed at the scission point, it is then possible to find the total deformation $\beta_{\text{tot}} = 2\beta$ for a given mass split by solving the above equation for TKE with respect to $\beta$.

![Diagram](image-url)

**Fig. 4** Sum of quadrupole deformations of both fragments as function of mass split for $^{235}\text{U}(n,f)$. Region of more than 75% yield with even proton number, position of fragments with even proton number (4).

Fig. 4 shows the sum of the quadrupole deformations of the two fragments as function of mass split for thermal neutron induced fission of $^{235}\text{U}$. The deformations are calculated with the above formula from the average TKE-values measured as function of mass split. The full triangles are the values from 5) who calculated $\beta_1$ and $\beta_2$ for the mass splits 118/118, 102/134 and 96/140. The hatched areas and the vertical lines indicate masses with high predominance of fragments with even proton numbers. Fig. 4 shows clearly the local dips in the total deformation caused by the proton pairing effect. The deformation as function of mass split can be understood from the proton and neutron shell corrections as given in 5). The different shell, pairing and liquid drop terms interplay of course to give the prefixed deformation also as function of the incident neutron energy. However, these details cannot be discussed here 2).

The total kinetic energy averaged over all fragments compared to the thermal value of $(170.6 \pm 1.0) \text{ MeV}$, as found in the present experiment, is plotted as function of the incident neutron energy in fig. 5 as full line. The open circles indicate the $\Delta$TKE-values which are calculated if the change were caused by a change in the mass distribution only. The sudden drop at 4 to 4.5 MeV in the experimental TKE which was already observed earlier 6), is clearly due to changes of TKE for different mass splits. This can be seen from fig. 6 which gives an example for $[\text{TKE}(E_n) - \text{TKE(thermal})]$ as function of mass split. In this comparison to thermal energy a clear drop is seen in the region of the mass splits around 102/134 and some smaller structures at other mass splits and changes in the wings of the mass distribution.

Fig. 7 shows some examples for the changes of the total deformation of the fragments as function of energy. The comparison is made relative to the thermal incident neutron energy. For the symmetric mass split a large decrease of fragment deformation is observed. This observation is in agreement with the static scission point model 5) which predicts an approach
towards the smaller deformation which one calculates when shell and pairing corrections are set to zero. For the mass splits around 102/134 an increase in deformation at 6 MeV is observed compared to the thermal value. Also this is qualitatively in agreement with the static scission point model.

For thermal neutron induced fission of $^{235}$U 1.5·10$^7$ events in total were registered. The total kinetic energy for a yield level of 10$^{-6}$ found in this experiment was determined as function of mass split. In the lower part of fig. 8 these total kinetic energies are plotted with full triangles as differences to the maximum Q-values of the specific splits ($Q_{\text{max}} - \text{TKE}$) versus the light or heavy fragment mass numbers. The Q-values were obtained from the tables of ref. 7). For light fragment masses from 87 to 110 the energy values scatter by about ±1.2 MeV around the zero line. This scatter reflects the experimental uncertainty in determining TKE, but it might also contain an uncertainty component of the theoretical values for $Q_{\text{max}}$. For the mass regions 87 to 110 cold fragmentation seems to be realized. The yield curve for masses of all kinetic energies is plotted for orientation as a full line in the lower part of fig. 8. Cold fragmentation e.g. is realized for the masses 109 and 110, although their yield in the whole mass distribution is very small. On the other hand no cold fragments were found for splits with $M < 87$, where the yield in the mass distribution is still fairly high. The open triangles in the lower part of fig. 8 are the results of ref. 8) at a yield level of 10$^{-5}$. The latter measurements extend only up to mass 106, so that the steep rise beyond mass 110 cannot be seen.

In the upper part of fig. 8 the ground state electric quadrupole moments of the light and heavy fragments, ref. 5), are plotted for those isobaric splits which have the maximum Q-values. The open circles at the masses 88, 91, and 110 are quadrupole moments of the isobars with the second highest Q-value. Their difference to the maximum Q-value is 4.3, 0.19, and 0.13 MeV, respectively. The numerical values for the ground state electric quadrupole moments were taken from the tables in ref. 7). From the present results it seems that cold fragmentation in $^{235}$U(n,f) for thermal neutrons can only occur when the light fragment has a positive quadrupole moment of $\geq 1$ barn. In order to prove this a nuclear charge measurement for the masses 91 and 110 would be interesting. If the predicted quadrupole moments are
correct, then one should find the isobars $^{91}$Kr and $^{110}$Mo with the second highest Q-values which are prolate in shape. One exception in this picture is the light fragment with mass 88. The split with the maximum Q-value $^{88}$Kr/$^{148}$Ba has according to ref. 5) a slightly negative ground state electric quadrupole moment and the closest isobaric neighbour $^{88}$Se/$^{148}$Ce has a distance of 4.3 MeV with respect to the maximum Q-value. The steep rise of $(Q_{\text{max}} - \text{TKE})$ for the mass split $^{111}$Te/$^{125}$In and $^{112}$Ru/$^{124}$Cd in fig. 8 is a support for the predicted sharp change from prolate to oblate nuclear shapes with mass.

![Fig. 8. In the lower part the full triangles represent the difference between the maximum Q-value and the observed kinetic energy for a yield level of $10^{-6}$. The open triangles are the same differences for a yield level of $10^{-5}$ from ref. 8). The full line represents the yield distribution as function of light or heavy fragment mass measured for all fragment kinetic energies. In the upper part the ground-state electrical quadrupole moments of the heavy □ and light ● fragments are plotted for the isobaric mass splits where the maximum Q-value is predicted, ref. 7). The open circles are light fragment quadrupoles of splits with second highest Q-value.](image)

6. ANGULAR DISTRIBUTIONS

Angular distributions were investigated in several previous measurements. The measurements are mostly averaged over all fragments. The present experiment gives the angular distributions also as function of mass split. However, the present angular distributions of fission fragments showed no fragment mass dependence.

7. CONCLUSIONS

The experimental mass distributions as function of the incident neutron energy between thermal and 6 MeV revealed, when fitted with a five Gaussian representation, that there is only a very small or no dependence on neutron energy below 2 MeV. From 2 MeV on the shape of the mass distributions changes markedly as is seen on the energy dependence of the mass distribution fit parameters. This change in the energy dependence of the shape of the mass
distributions between 1.5 and 2 MeV incident neutron energy might be due to pair breaking at saddle.

The differential measurement of the total kinetic energy as function of mass and incident neutron energy allow three main conclusions:

- The drop of the total kinetic energy averaged over all fragments with neutron energy above 4 MeV is not due to the change of the mass distributions, but due to drastic changes of the average total kinetic energy for individual mass splits with neutron energy.

- At neutron energies below 2 MeV the average total kinetic energy rises slightly for all mass splits. This can mean that the additional energy brought into the compound system by the kinetic energy of the neutron is mainly transferred to pre-scission kinetic energy. When pair breaking at saddle becomes feasible other energy dissipation processes come into play.

- The dependence of the average fragment deformation on mass split and compound nucleus excitation energy follows at least qualitatively the prediction of the static scission point model 1), as shown for the mass splits 118/118, 102/134, and 90/146.

From the kinetic energy at a yield level of $10^{-6}$ for the different mass splits at thermal fission it is observed that cold fragmentation for the isobaric splits which have the maximum Q-value is realized only when the light fragment ground-state electric quadrupole moment is positive and larger than 1 b about. The steep rise of $(Q_{\text{max}} - \text{TKE})$ at a yield level of $10^{-6}$ for light fragment masses larger 110 supports the predicted sharp change from prolate to oblate nuclear shapes in this mass region. In this context it is of great interest to measure the nuclear charge distributions of the mass split 91/145 and 110/126 as function of TKE, since there are two isobaric splits which have about the same Q-value but a very different ground state electric quadrupole moment.

The measured angular distributions of the fission fragments with respect to the incident neutron beam show no mass dependence, in accordance with the channel theory of fission.

For further study of the fission process it seems to be of interest if cold fragmentation could be studied also at higher compound nucleus excitation energies. One could then check e.g. if cold fragmentation is possible also for fragments with a negative ground-state quadrupole moment. Also correlation measurements with the neutrons and gammas would be of high interest.

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PHOTON INDUCED FISSION OF $^{232}$Th WITH 10, 12 AND 20 MeV BREMSSTRAHLUNG.

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Abstract

For 10, 12 and 20 MeV bremsstrahlung induced fission of $^{232}$Th the postneutron mass distribution was determined by γ-ray spectrometry of fission product catcher foils. For symmetric fission a small distinct third peak is observed. The light and heavy mass wings show strong finestructures probably due to odd-even effects. The mass distribution for 10 MeV bremsstrahlung induced fission is considerably more asymmetric compared to 12 and 20 MeV. For 12 and 20 MeV bremsstrahlung induced fission energy correlation measurements were performed. The comparison of the total preneutron kinetic energy as a function of the heavy fragmentmass shows the usual decrease of $E_k^*(M_H)$ with increasing excitation energy in the vicinity of $M_H=132$. In the symmetric fission region the tendency of increasing kinetic energy with increasing excitation energy is observed.

1. INTRODUCTION

The results presented here are part of an excitation energy dependence study of several fission characteristics for the photon induced fission of $^{232}$Th. The results for 12 and 20 MeV bremsstrahlung induced fission obtained with the 70 MeV linac of our laboratory will be discussed. Recently measurements with the newly installed low energy linac ($E_{max}=10$ MeV) were started. The first results for 10 MeV bremsstrahlung induced fission are also included.

2. EXPERIMENTAL PROCEDURE

The postneutron mass distributions for 10, 12 and 20 MeV bremsstrahlung induced fission of $^{232}$Th were determined by γ-ray spectrometry of fission product catcher foils. To investigate the massdistribution in the valley region chemical separations of the Ru- and Sb-fractions of irradiated Th-nitrate were carried out for 12 and 20 MeV bremsstrahlung. This allowed the determination of the mass yield ratios 105/103, 106/103 and 125/127. The yields of masses 103 and 127 were well defined from the catcherfoil method.
To increase the yields for 10 and 12 MeV bremsstrahlung induced fission, thorium targets with a thickness exceeding several times the range of fission fragments in it were used. As a consequence a correction to the observed mass distribution, which is for each fragment mass proportional to its range was necessary. These range corrections were determined experimentally by comparing the photopeak area of catcherfoil spectra obtained with these thick targets and those obtained with very thin targets (0.5 mg/cm$^2$), for which no range correction is needed. These experiments were performed with 20 MeV bremsstrahlung. Although it is sufficient to know the relative ranges a simple additional experiment did provide the absolute values. For a target thickness equal to the range of a specific fission product one can calculate that one fourth of these fission products produced in the target leaves the target and is captured in the catcherfoil. It is clear that with the knowledge of the thickness of the thorium target the range of each fission product can be determined from the ratio of the activity in the catcherfoil and the remaining activity in the thorium target of that specific fission product. This experiment is limited however by the disturbing $\alpha$-decay of $^{232}$Th in the target. In fig.1a the deduced ranges for thorium fragments are represented by the squares and compared with the measured ranges for uranium fragments represented by the crosses $^1)$. These distributions show an analogous behaviour as the fragment kinetic energy distributions for the same fissioning systems (see fig.1.b).

For 12 and 20 MeV bremsstrahlung induced fission energy correlation measurements were performed. From the pulse heights measured in two oppositely placed surface barrier detectors the provisional mass and postneutron kinetic energy of the complementary fragments can be deduced using the calibration method and constants of Schmitt et al. $^2$).

Combination of the postneutron mass distribution and the results of the energy correlation measurements provides the preneutron mass and kinetic energy distributions and correlations and the neutron emission curve.
3. RESULTS AND DISCUSSION

The postneutron massdistributions for 10, 12 and 20 MeV bremsstrahlung induced fission are shown in fig. 2. For the fission with 10 MeV bremsstrahlung the mass yields in the valley region are not yet determined except the yield of mass 115 which was calculated from a small photopeak in the catcherfoil spectra and which provides an indication of the upper limit of the symmetric fission component. Striking in fig. 2 is the appearance of a distinct third peak for symmetric fission. This third peak is less pronounced for 20 MeV compared to 12 MeV bremsstrahlung. This is in agreement with the results of Glendenin et al. for $^{232}$Th(n,f) with 2 MeV < $E_n$ < 8 MeV who observed also a small third peak which is washed out with increasing excitation energy. This third peak may be caused by shell effects in the fragments: calculations in the framework of Wilkins et al. predict indeed that thorium is situated in the transition region where the symmetric fission component disappears for heavier nuclei. Another more probable explanation is the possible existence of an additional mass symmetric saddle point besides the ordinary mass asymmetric saddle points. Calculations of fission cross sections by Takaaki Ohsawa for thorium isotopes are in better agreement with the experimental data when they assume the existence of a supplementary symmetric barrier which is about 1 MeV higher than the asymmetric barrier.

![Fig. 2. The postneutron massdistribution for $^{232}$Th(γ,F) with 10 (---), 12(----) and 20 MeV (------) bremsstrahlung.](image-url)
The mass distribution for 10 MeV bremsstrahlung induced fission is more asymmetric compared to 12 MeV. This seems to be in contradiction with the expectations in the framework of Wilkins et al. where the contribution of the strong spherical $N=82$ neutron shell associated with fragment mass 134 is more important for lower excitation energies.

Some characteristics of the mass distributions for 10, 12 and 20 MeV bremsstrahlung are summarized in Table I.

<table>
<thead>
<tr>
<th>$E_0$ (MeV)</th>
<th>$&lt;M_L&gt;$ (u)</th>
<th>$&lt;M_H&gt;$ (u)</th>
<th>$&lt;V&gt;$</th>
<th>A/S</th>
<th>$&lt;E_{exc}(E_0)&gt;$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>89.48 ± 0.10</td>
<td>140.44 ± 0.10</td>
<td>2.08 ± 0.15</td>
<td>&gt;250</td>
<td>7.2</td>
</tr>
<tr>
<td>12</td>
<td>90.58 ± 0.09</td>
<td>139.39 ± 0.09</td>
<td>2.03 ± 0.13</td>
<td>38 ± 5</td>
<td>8.8</td>
</tr>
<tr>
<td>20</td>
<td>91.25 ± 0.09</td>
<td>138.16 ± 0.09</td>
<td>2.59 ± 0.13</td>
<td>11 ± 1.5</td>
<td>12.4</td>
</tr>
</tbody>
</table>

The $<V>$-values are calculated from the conservation of mass. These values are in agreement with the measurements of Caldwell et al. The parameter A/S indicates the ratio of the asymmetric to the symmetric fission yield. For 12 and 20 MeV bremsstrahlung the average excitation energy of the compound nucleus $<E_{exc}(E_0)>$ was calculated from the fission cross section of $^{232}$Th measured by Caldwell et al., assuming that the bremsstrahlung spectrum produced in a thin Au-target has the Schiff shape. 10 MeV bremsstrahlung was produced in a thick carbon target and the value for $<E_{exc}(10)>$ in Table I. is a preliminary estimation based on calculations of Tanaki et al.

The mass distributions in Fig. 2 show strong fine structures in the light and heavy mass wings. They are more clearly visible on a linear yield scale as presented in Fig. 3 for 10 MeV bremsstrahlung induced fission. The arrows indicate the positions of fragment masses associated with the even Z-values. The enhanced yields in the light and heavy fragments can indeed be explained by proton pairing effects. Mariolopoulos et al. observed strong odd even effects (about 35%) in the charge distributions for $^{229}$Th(n$_{th}$,f). Their deduced mass yields from the charge yields were in good agreement with the measured values of Unik et al. The mass distribution for $^{229}$Th(n$_{th}$,f) measured by Unik et al. is included in Fig. 3. It is striking that the enhanced yields
for the two compared mass distributions almost coincide.

The calculated neutron emission curves for 12 and 20 MeV bremsstrahlung show the usual saw-tooth behaviour. The increase of the total number of emitted neutrons for 20 MeV compared to 12 MeV bremsstrahlung induced fission can practically completely be attributed to the heavy fragments.

The total preneutron kinetic energy as a function of the heavy fragment mass shows a completely identical behaviour for fragment masses larger than 135 for 12 and 20 MeV bremsstrahlung induced fission. In the vicinity of mass 132 the usual decrease of the kinetic energy with increasing excitation energy is observed, due to the diminishing influence of the spherical N=82 neutron shell. For the symmetric fission region the kinetic energy seems to increase for 20 MeV compared to 12 MeV. This is a tendency that has also been observed for other fissioning systems (see f.i. ref. 12-13). It is not clear whether this behaviour can be attributed to shell effects in the fragments.

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SESSION IV

HEAVY ION INDUCED FISSION

Chairman: Y. El Masri
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I. INTRODUCTION

At projectile energies below 10 MeV/n, if one excludes elastic and quasi-elastic channels, heavy ion (H.I.) induced reactions can be divided into three main classes of dissipative mechanisms:

- deep inelastic collisions (D.I.C.) which correspond to the highest angular momenta 1-waves in the entrance channel and where the clutching of partners occurs. However the intermediate system keeps a two-centre potential so that disruption into two fragments follows shortly after a large kinetic-energy loss and few nucleons exchange. The main characteristic of this binary phenomenon is its input channel mass memory;

- compound nucleus formation (C.N.) which is followed by light particle evaporation (p, n, α,...) and subsequent heavy residue formation which mass approaches the total mass of the colliding system;

- fusion-fission (F.F.) mainly characterizing heavy colliding nuclei. The most probable masses of the residual fragments correspond nearly to the half mass of the total system. During several years, this third class of dissipative mechanism has been only associated to fission after compound nucleus formation. Such a concept corresponds to a mononuclear configuration with complete thermodynamic equilibrium. It also implies that the lifetime of the total intrinsic nucleon excitation is much longer than the nucleus single rotation period.

Figure 1 displays a schematic diagram of the integrated mass distribution of the reaction products for a medium mass colliding system. This figure clearly illustrates the conventional classes of the dissipative mechanisms in H.I. induced reactions.

However, fission process following complete fusion is not the unique mechanism where fission fragments can be seen in these reactions. Indeed, depending on projectile-target and energy combinations, fission fragments can be encountered as primary or secondary products in almost all H.I. interactions. This fact can explain the very large amount of fission like data collected during the last twenty years where this process has been studied either for its own or to inlight other types of dissipa-
2. H.I. INTERACTIONS INDUCING FISSION PROCESSES

A. Coulomb fission

Coulombinduced fission has been originally studied with medium mass projectiles\(^1\) (Kr, Xe, ...). It was recently brought into clear evidence\(^2\) with heavier projectile and target combination such as \(^{152}\text{Sm},^{184}\text{W}\), \(^{232}\text{Th,}^{238}\text{U}\) or \(^{232}\text{Th,}^{248}\text{Cm}\) at incident energies ranging between 0.85 and 1.03 times their respective coulomb barrier \(E_C\). Experimentally, coulomb fission was identified by measuring the fission fragment energies in coincidence with the conversion electrons of the simultaneously coulomb excited scattered projectile, thus excluding any nuclear transfer reaction contribution. The measured fission probabilities \(P_f\) are shown in figure 2 (extracted from reference \(^1\)) as a function of the bombarding energy expressed as \((E/E_C)\). The exponential increase of these probabilities is fairly reproduced by Oberacker et al.\(^2\) calculations assuming a pure coulomb interaction. As pointed out by G. Himmeler et al.\(^1\) the projectile dependence of \(P_f\) behaving like \(Z_p^{7.4}\) indicates a multistep coulomb excitation for the initiating reaction in contrast to a single step process characterized by a \(Z_p^2\) behaviour.

B. H.I. sequential fission

In heavy systems, sequential fission can be observed as a second step process following either quasi-elastic or deep inelastic collisions. This particular feature has been exploited in order to obtain informations on the angular momentum transfer and the sharing of the excitation energy in deep inelastic collisions. Indeed, large amount of angular momentum can be transferred from the relative motion into the intrinsic spin of the D.I.C. products. If one of these fragments is heavy enough (Au,
Bi, U for instance), it may undergo sequential fission. For nuclei like Au or Bi, the intrinsic spin must be large enough to lower the effective fission barrier inducing thus a large fission probability. Then, the study of the angular correlation in and out of the reaction plane gives detailed informations on the intrinsic angular momentum of the fissioning nucleus and consequently on the transferred angular momentum in the D.I.C. process.

In fact when compound nuclei are produced with large angular momenta the angular distribution of the nascent fission fragments approaches a $1/\sin \theta$ - dependence since the spin of the decaying system lies in a plane perpendicular to the beam axis. In the framework of simple models describing D.I.C. the spin is expected to be completely aligned along the Z axis perpendicular to the reaction plane defined by the beam direction and the non-fissioning light fragment. Figure 3 schematically shows the related parameter to this process. Usually, one introduces the quantum number $K$ which describes the projection of the heavy fragment spin (I.) on the direction of fission, and one admits that the angular distribution $W_I(\theta)$ of the subsequent fission fragments is completely determined by the orientation in the laboratory system of the nuclear symmetry axis of the fissioning nucleus at the saddle point. Then $W_I(\theta)$ can be written:

$$ W_I(\theta) = N_K 0 \frac{1}{4} (2I+1) \frac{1}{2} \sin \theta)^2 I \sum_{K=-I}^{I} \exp(-\frac{K^2}{2K_0^2}) \frac{(2I)!}{(I+K)!(I-K)!} \left[ \cotg \frac{2K \theta}{2} + \tan^2 \frac{K \theta}{2} \right] , $$

\[ \text{Figure 2.} \]
Sequential fission of the heavy fragment recoiling along the \( \Gamma \)-axis. \( L_z \) is the orbital angular momentum perpendicular to the reaction plane in the entrance channel. \( I \) is the intrinsic angular momentum of the fragment. \( M_z \) is the \( z \)-component of \( I \). \( M_z \) is assumed to be equal to zero in this picture.

Figure 3.

where

\[
N_{K0} = \frac{1}{\sum_{K=-1}^{1} \exp(-\frac{K^2}{2K_0})}
\]
and

\[
K_0^2 = \frac{1}{\hbar^2} J_{\text{eff}}
\]

with \( J_{\text{eff}} \) defined such as:

\[
\frac{1}{J_{\text{eff}}} = \frac{1}{J_\|} - \frac{1}{J_\perp}
\]

\( J_\| \) and \( J_\perp \) are respectively, the parallel and perpendicular component of the nuclear moment of inertia calculated with respect to the symmetry axis. Two difficulties appear when the experimental results are compared to this expression: i) the \( K_0 \) evaluation; it is generally extracted either from compound nucleus fission or from liquid drop calculations; ii) the integration of the above relation over the whole range of angular momenta involved in the sequential fission. On the other side, if the spin \( I \) of the fissioning nucleus is perpendicular to the reaction plane (\( M = M_z = 1 \)), the in-plane correlation is isotropic and only the out-of-plane correlation is sensitive to the \( I \) population. However if the \( I \) direction fluctuates (\( M_x, M_y \) and \( M_z \neq 0 \)), one then expects anisotropy in the reaction plane. Therefore, the degree of the spin alignment may be extracted from the analysis of both the in-plane and out-of-plane angular distributions. After a long controversy\(^3\), sequential fis-
sion data clearly showed that the simple classical models are unable to describe the experimental features: the angular momentum of the fissioning nucleus is not completely aligned along the orbital angular momentum of the entrance channel but fluctuates around this direction. Similar conclusions were also drawn from the y-ray multiplicity measurements associated to these reactions. Presently, the angular momentum depolarization is thought to result from the combination effects of i) nuclear exchange as calculated by Randrup; ii) collective mode excitations as described by Moretto et al. and iii) statistical fluctuations and quantal diffractive effects as described for example by the recent Van Geertruyden-Willain model.

Sequential fission fragments properties were recently used to answer one of the main and open questions on D.I. collisions: what about the excitation energy sharing between the primary fragments? Up to now, this problem was mainly investigated by the analysis of the subsequent evaporated neutron spectra. Indeed in addition to the nuclear temperatures extracted from these spectra for both fragments, the ratio of the light and heavy partners neutron multiplicities were used to deduce informations about the excitation energy sharing between them. The interpretation of these data supported thermal equilibrium for the whole range of energy losses down to 30 MeV. In reference 8 the relative yields of the symmetric and asymmetric mass distribution components of the sequential fission fragments were measured. As these yields are very sensitive functions of the excitation energies (below 60 MeV), they were used to extract the heavy fragment excitation energy in the D.I. collision of the 480 MeV $^{56}$Fe + $^{238}$U system, where in this reaction the total excitation energy is calculated from the kinetic energy of the projectile-like fragment assuming two-body kinematics. Figure 4 shows samples of the fission-fragment mass distributions at several values of the total kinetic energy loss TKEL. The results, interpreted in terms of the two fragment excitation energy ratios ($E^H/E^L$), are plotted.
as a function of TKEL in figure 5. In this figure, the result of a transport-model calculation by Randrup is also shown. It is clearly found that the thermal equilibration of the system is not achieved below 100 MeV energy loss. Such an effect was previously observed in the Ar + Au reaction by Dakowski et al. measuring the neutron multiplicity. It was then interpreted in terms of deformation energy effects.

C. N/Z properties of the fission fragments

However, sometimes, studies conducted in order to obtain informations about D.I.C. process, can enlighten a blocked situation in fission. Fission charge distribution were interpreted using two main approaches: the equal charge displacement (ECD) and minimal potential energy. They were no clear experimental evidences to choose between these two interpretations. It was stressed out that the evolution of N/Z ratio is associated to a short relaxation time, short enough to allow a marked influence of the properties of the nascent fragments. Owing to the large target-projectile combination set, the characteristic time of the N/Z relaxation for deep inelastic collisions can be studied. This time is very short, so short that the N/Z relaxation has completed as soon as the energy dissipation has begun. The interaction time for fission is much longer and allows to believe that this ratio must also be completely equilibrated for this slower mechanism. It has also been shown that, in deep inelastic collisions, the equilibrated N/Z ratio corresponds to the one which minimizes the composite system potential energy. The situation was made clearer in deep inelastic collisions because in this process, the fragments are not both neutron deficient as it is always in fission.

D. Light system binary fragmentation

Heavy ion induced reactions have also enlarged the range of nuclei for which fission channel properties could be studied. In addition to very heavy systems, light systems can also undergo fission or binary symmetric fragmentation. This can be due to the large amount of excitation energy and angular momenta damped in such systems enhancing thus the fission probability. The fission channel is already open for a compound nucleus such as $^{108}$Sn. Fission like fragments have been recently detected in system as light as $^{32}$S + $^{12}$C $\rightarrow$ $^{44}$Ti.

3. SOME CHARACTERISTICS OF FISSION PROCESS

Among the other fission characteristics which were exclusively studied one must mention the evolution of the total energy of fission fragments as a function of the fissility parameter ($X$) and the masses of the fissioning nuclei ($A$). Thus the ranges of $0.475 \leq X \leq 0.965$ and $136 \leq A \leq 322$ have been investigated using mainly Ar and
Kr beams as displayed in figures 6 and 7. In these figures the average total kinetic energy of fission (TKE) at symmetry is respectively plotted as a function of fissility parameter \( X \) and fissioning nucleus mass \( A \). The experimental results are confronted to the theoretical expectations based on the liquid drop model (LDM)\(^\text{14}\) and the two-center model\(^\text{15}\). The shell effects, predicted by the two-center model, are not observed probably due to the high excitation energies involved in these reactions (50 to 160 MeV). One must remind that shell effects were originally introduced to describe the experimental results observed in spontaneous and neutron induced fission\(^\text{16}\) of \(^{257}\)Fm where the TKE were much higher than the predictions of the liquid drop model or the extrapolation of the previously available data. Nevertheless the TKE variation with the parameter \( X \) is rather well described by NIX\(^\text{14}\) even if the calculated absolute values are somewhat lower. It consequently appears that the fissility parameter (used in the LDM) rather than the fissioning nucleus mass (used in the two-center model) is the most important parameter governing the total fragment kinetic energies of highly excited nuclei.
4. FUSION AND FISSION CORRELATIONS

Under large excitation energy and high angular momentum effects, the fission channel dominates very often in the deexcitation of fusion-like nuclei induced in medium and heavy mass ranges. This fact allowed to see fission as a tracer of the fusion process. Therefore it has been largely used to investigate the fusion reaction mechanism and more precisely the limitation of compound nucleus formation. Indeed if for instance one considers the collisions of two heavy nuclei such as Ar + U and Kr + W leading to a composite system with similar $Z(110)$ at approximately same excitation energy and initial $l$-wave ranges, one easily sees in figure 8 that the total kinetic energy maps, as a function of the fragment masses, are quite different for both systems. In the Argon induced reaction (at 300 MeV), the

- Comparison of a system (Ar+U) in which fission-like products are observed and a system (Kr+W) leading to the same $Z$ for the composite where nearly all the cross-section is in deep inelastic transfers: a) $^{40}$Ar+$^{238}$U $\rightarrow ^{29}$9110, $E^* = 125$ MeV, $E_{lab} = 300$ MeV, $E_{c.m.} = 257$ MeV, $l_{max} = 160$; b) $^{84}$Kr+$^{144}$W $\rightarrow ^{29}$9110, $E^* = 100$ MeV, $E_{lab} = 492$ MeV, $E_{c.m.} = 339$ MeV, $l_{max} = 173$

Figure 8.
Fission-like fragments are clearly observed around the mass-symmetry splitting whereas such events are practically absent in the Kr induced reaction (at 492 MeV). In the latter case only two peaks are clearly seen in the mass distribution of the residual fragments, one situated around the light fragment mass (Kr) and the second around the complementary heavy residue (186). The lack of symmetric fission fragments (A=135) in this reaction and consequently of the fusion process is presently well established for H.I. collisions with projectiles heavier than copper and very heavy targets. In those systems, the entire set of 1-waves from 0 to l_{\text{max}} contributes only to the D.I. process. One must also remark that in the Ar + U system the fission-like mass distribution around symmetry corresponds to full momentum transfer (A_{CN} = 270) followed by a coulomb repulsion after scission (TKE = 200 MeV) and a typical 1/sin θ angular distribution recorded experimentally.

At this stage an important question has to be pointed out: is it possible, as in such systems, to form a well defined compound nucleus when, under the influence of centrifugal forces, the fission barrier (B_{f}) has already disappeared? Can we, under these circumstances, still admit concepts such as deformation, saddle point and fission? In fact the angular momentum effect on B_{f} has been described by the rotating liquid drop model (RLDM) which tells us that, depending on the choice of the moment of inertia of the fissile nucleus (J), the highest value corresponding to B_{f} = 0 can be obtained by

\[ B_{FR} = B_{F}(1=0) - \frac{1(1+1)h^2}{2} \left( \frac{1}{J_{R0}} - \frac{1}{J_{RS}} \right) \]

where J_{R0} and J_{RS} are respectively the moment of inertia of the nucleus described as a spherical shape and its value at the saddle point. The results of different calculations are shown in figure 9. For instance, following

![Figure 9](image-url)
these predictions, a $\lambda$-value of the order of 90 $\hbar$ appears to be the limit of stability of medium mass nuclei, whereas these limits range between 10 and 40 $\hbar$ for heavy nuclei depending on the considered model.

It was also known, since Kowalski et al. work\textsuperscript{22}, that high angular momenta inhibit complete fusion. This has been expressed by a so-called low $\lambda$-wave cut-off for complete fusion, above which centrifugal forces are so large that they prevent attractive potential between the two colliding nuclei. In sharp cut-off approximation, one defines the critical angular momentum ($\lambda_{cr}$) as the highest initial $\lambda$-wave leading to fusion. However, for many heavy systems in which fusion cross sections and critical angular momenta ($\lambda_{cr}$) were deduced from fission-like data\textsuperscript{20}, $\lambda_{cr}$ largely exceeded the corresponding $\lambda_{Bf=0}$ value.

Therefore, for $\lambda$-waves above $\lambda_{Bf=0}$, one must still expect reaction processes allowing: i) symmetric-mass scission with complete energy relaxation, ii) very large mass transfer needing long interacting time and iii) $1/\sin \theta$ residual angular distribution.

5. FRICTION FORCE EFFECTS

An answer to this question was already proposed in the Bass model\textsuperscript{21} for fusion where it was postulated that fusion process can only occur when the total potential energy ($V(r)$) of the two interacting nuclei displays a pocket as a function of their separation distance ($R$). Then, the system can be trapped under the action of friction forces or nuclear viscosity. For very heavy systems ($Z_1Z_2$ product $> 2700$), no pocket exists in $V(r)$ even at $\lambda = 0$ and only deep inelastic reactions can occur.

In figure 10 the evolution of $V(r)$ as a function of the internucleus separation distance $R$ for different $\lambda$-values for two systems $^{40}$Ar + $^{165}$Ho and $^{24}$Mg + $^{181}$Ta leading to the same composite nucleus ($^{205}$At). In these calculations sudden nuclear potential approximation is used as described in reference 23. In this figure, one first has to remark that, due to tangential friction force, the orbital angular momentum in the output channel ($\lambda_{out}$) is always lower than the entrance channel ($\lambda_{i}$). Depending on the cohesion achieved by the composite system, the angular momentum loss ($\Delta \lambda$) can drastically vary. Thus $\Delta \lambda$ is $\frac{2}{7} \lambda_{i}$ in the case of rolling condition and reaches even larger value in the case of sticking mode for asymmetric systems ($A_p \neq A_t$). For relatively small centrifugal forces, a pocket remains in the potential energy curve. For increasing $\lambda$-values the pocket vanishes and completely disappears for a given $\lambda_{limit}$ value; for instance, assuming rolling condition $\lambda_{out} \lambda_{limit} = 90 \hbar$ for Ar + Ho reaction (corresponding to $\lambda_{in} \lambda_{limit} = 130 \hbar$) and $\lambda_{out} \lambda_{limit} = 70 \hbar$ for Mg + Ta ($\lambda_{in} \lambda_{limit} = 100 \hbar$) as shown in figure 10. Above those limits the complete system cannot be trapped and so no chance exists for complete fusion and even for a large mass transfer. At this stage of our understanding two different situations appear to happen: i) if $\lambda_{crit}$ or $\lambda_{limit}$ is smaller than $\lambda_{Bf=0}$
the system evolves towards compound nucleus formation; $l_{\text{crit}}$ being always limited by $l_{\text{im}}$; ii) if $l_{\text{im}}$ exceeds $l_{B_f=0}$, the system cannot reach a compound nucleus formation because such a stable configuration does not exist for $l$ ranging between $l_{B_f=0}$ and $l_{\text{im}}$ or $l_{\text{crit}}$. In this case, the two incident partners tend to equilibrate their mass degree of freedom and when a nearly symmetric configuration is reached, they reseparate as the fission barrier has completely vanished. This reaction channel has been tentatively named \(^{24)}\) "fission without barrier" or quasi-fission and is presently better known as "fast fission". This phenomenon could be essentially observed for systems where $l_{\text{im}} > l_{B_f=0}$. The contour diagram of figure 11 clearly illustrates this process and displays the potential energy minima as a function of projectile and target masses. Thus combining the two concepts of $l_{B_f=0}$ and $l_{\text{im}}$ one can easily distinguish between compound nucleus formation mechanism and fusion reaction where in this case $l_{\text{crit}}$ will be limited to $l_{\text{im}}$ value.

![Diagram showing potential energies vs. separation distance for two systems forming the same composite nucleus](image)

Potential energies vs. separation distance for two systems forming the same composite nucleus: $^{48}\text{Ar} +^{144}\text{Ho}$ and $^{24}\text{Mg} +^{181}\text{Ta}$. Nuclear potentials are calculated with the energy density method \(^7\). Centrifugal potentials are taken for several $l$-values, assuming rigid-body moments of inertia for two spheres. The drop of $\mathcal{F}$ in the region of close contact is expected for orbital-angular-momentum dissipation in rolling conditions. The limiting $l$-value corresponds to the disappearance of a pocket in the exit channel curve ($d^2\mathcal{F}/dl^2 = 0$).

Figure 10.
Masses of projectiles and targets for which the quasi-fusion process occurs, i.e., fission from composite nucleus where $l'_{ci} > l_{\beta, i} = 0$. The limiting angular momentum corresponds to a pocket depth lower than 5 MeV.

Figure 11.

THE NUCLEAR CONFIGURATION AS SPECIFIED BY 3 DEGREES OF FREEDOM

ASYMMETRY $\Delta = \frac{R_1 - R_2}{R_1 + R_2}$

DISTANCE $\rho = \frac{r}{R_1 + R_2}$

WINDOW OPENING $\alpha = \frac{\sin \theta}{\sin \theta_{max}}$

The nuclear configuration is parametrized by two spheres connected by a conical neck. The asymmetry is specified by $\Delta$, the center-separation by $\rho$, and the degree of window opening by $\alpha$. The distance between the tips of the spheres may be positive or negative.

Figure 12.
6. FAST FISSION PROCESS

A. Theoretical models - Swiatecki's approach

The preceding considerations did not include dynamic concepts which can be simply introduced as recently described by Swiatecki\(^{(25)}\). In this model the main idea is to allow the two colliding partners (considered as spheric shapes until they touch) to develop a neck which decreases the potential energy. Nucleon exchange is then allowed when the neck or window opening (\(\alpha\)) is large enough (\(\alpha = 1/2\)). This value (\(\alpha = 1/2\)) corresponds to the boundary condition between dinuclear and mononuclear regimes. This model, depending on three variables or degrees of freedom, has no adjustable parameter (see figure 12 for the definition of these variables). Also one body-dissipation is assumed in these calculations and various asymmetries are treated. The main predictions of this model are shown in figure 13, they concern calculations for head-on collisions of a symmetric system at 3 kinetic energies 0.3, 6 and 10 MeV above the interaction barrier and take into account only two degrees of freedom (\(\Delta_{in} = 0\)).

For geometric reasons, the entire (\(\alpha, \rho\)) plane is not accessible. A saddle point for fission is clearly displayed. Trajectories start from a two tangent spheres (\(\rho = \frac{r}{R_1 + R_2} = 1\)) configuration. Along these trajectories each point corresponds to a \(\frac{1}{2} \times 10^{-22}\) sec. Two classes of trajectories are shown depending on the available energy: if it is small, non-trapping trajectories lead to a dinuclear system; if it reaches 10 MeV, capture occurs and the trajectory enters the spherical mononuclear configuration (line 1)\(^{(2)}\)). In order to reach and overcome the saddle point, the system has to spend energy: this is the so-called "extra-push" energy (\(E_x\)). For the capture trajectory, the system spends a long time in the vicinity of the saddle point. Swiatecki calculations showed that \(E_x\) is zero for light systems and increases with the total mass of the system following the relation:

\[
E_x (\text{MeV}) = 2000 \left( X_e - 0.7 \right)^2
\]

where

\[
X_e = \frac{Z^2/A_{\text{crit}}}{(Z^2/A)_{\text{crit}}} \quad \text{the fissility parameter with } \frac{Z^2}{A_{\text{crit}}} = 50.1.
\]

This relation is also valid for asymmetric systems where in these cases \(X_e\) is defined as:

\[
X_e = \frac{(Z^2/A)_{\text{eff}}}{(Z^2/A)_{\text{crit}}} \quad \text{with } \frac{Z^2}{A_{\text{eff}}} = \frac{4Z_T Z_T}{A_p^{1/3} A_T^{1/3} (A_p^{1/3} + A_T^{1/3})}
\]
If fusion occurs, the time evolution of the system is long ($\approx 10^{-20}$ sec) and then the mass asymmetry degree of freedom ($\Delta$) has time to relax and, in particular, the saddle point position moves in the $(\alpha, \rho)$ plane. This case is calculated in figure 14 when an asymmetric system is studied ($A_1 = 217, A_2 = 64, \Delta_{\text{in}} = 0.2$). There one can observe a trajectory entering the mononuclear regime, but as the mass asymmetry degree of freedom decreases, the coulomb repulsion increases, the saddle point moves, and the trajectory, instead of going further towards capture, leaks out again even with 67 MeV above the barrier (see line 1)(4)(5)(6)(7)(8)).

![Figure 13](image)

More recent calculations for head-on collisions of a symmetric system at 3 kinetic energies 0.3, 6 and 10 MeV above the interaction barrier. Capture occurs for trajectory 1) corresponding to 10 MeV. Parameters are the window opening related to $\nu$ and the separation distance: a) $\Delta \approx 208, Z = 82, A_1 = A_2 \approx 104, Z = Z_0 \approx 41, PE \approx 583$ MeV, time unit $\approx 12 \cdot 10^{-12}$ s; b) $X_0 = X = 0.7, \Delta = 0, S' = 0, -0.6, -1$.

Figure 13.

However, the trajectory spends a very long time in position 1) to 7) and mass transfer towards symmetry have time to occur (at $\rho > 1.5$ the fraction target mass has reached 0.5).
Same as Fig. 13, but, because of asymmetry, capture is not achieved even with 67 MeV above the barrier. However, the trajectory spends a very long time in positions 1) to 7) and mass transfer towards symmetry have time to occur. This would be a typical quasi-fusion with $l = 0$, $A = 282$, $Z = 106$, $A_1 = 217$, $Z_1 = 83$, $A_2 = 64$, $Z_2 = 23$, $PE = 690\frac{3}{4}$ MeV, time unit $\approx 15 \times 10^{-22}$ s, $A_0 = 0.5$, $d = 0.2$, $X = 0.7406$, $S' = -3$.

Figure 14.

This figure illustrates three types of reactions: (a) deep-inelastic (lowest trajectory) following injection with zero energy above the barrier, (b) quasifission, following injection with 7.2 MeV (the other reseparating trajectory), and (c) a compound-nucleus reaction (trajectory on the left, with 65.7 MeV injection energy). The fourth curve corresponds to a critical injection energy close to 16.4 MeV, chosen so that the system would come to rest (in unstable equilibrium) at the unconditional saddle point (indicated by a crossed semicircle).

Figure 15.
In Swiatecki model the frozen $\Delta$ saddle point (figure 13) and the equilibrated $\Delta$ saddle point (figure 14) are called conditional and unconditional saddle points respectively. In this framework, to overcome the unconditionnal saddle point is not always sufficient to overcome the unconditionnal one. In some cases, especially for heavy systems, an extra energy is needed to reach the unconditionnal saddle point, it is called the "extra-extra-push" energy.

Angular momentum effect can also be introduced assuming that its role, and consequently the role of the centrifugal forces, are similar to the coulomb repulsion effect, both inhibiting the fusion process. The preceeding relations remained valid if $X_e$ is defined using a $(Z^2/A)_{\text{eff}}$ given now by:

$$ (Z^2/A)_{\text{eff}} = (Z^2/A)_{\text{eff}} + (f_1)^2 $$

where $f_1$ is the fraction of the initial orbital angular momentum $I$ involved and

$$ f_1^2 = 0.0105 \left( \frac{A_1}{A_T} \right)^{4/3} \left( \frac{A_1^{1/3}}{A_T^{1/3}} \right)^2 $$

In other words, one can consider that the rotational energy decreases the available energy of the system. Typically this model predicts three different types of trajectories: deep inelastic collision (the system does not overcome the conditionnal saddle point), compound nucleus formation (the system overcomes the two saddle points) and fission-like reactions where the system only overcomes the conditionnal saddle point and has enough time for mass transfer towards symmetry, but it reseparates before reaching the compound nucleus configuration. These reactions are the so-called quasi-fission or fast fission.

Similar conclusions can also be drawn from other more sophisticated models including hydrodynamic calculations, transport theory and statistical fluctuations. In particular, concepts similar to the extra-push and the extra-extra-push are obtained and named respectively first and second compression effects in reference 28.

B. Experimental data for fast fission

In fact, the experimental results, supporting the existence of fast fission process as an intermediate H.I. mechanism between D.I.C. and C.N. fusion-fission, were known before the development of these models. Simple calculations based on
Bass model and the RDLM already allowed the predictions of the main characteristics of this new process and the mass range where it could occur (see ref. 24 and fig. 11). Experimental evidences for fast fission were extracted from i) the critical angular momentum values for fusion reactions with \(l_{\text{crit}} \geq l_{B_f=0}\) in many systems; ii) the evolution of the mass distribution for fission-like fragments as a function of the excitation energy or the critical angular distributions and later iii) on angular distributions (see ref. 20,24,29,30 and 31).

Three systems leading to approximately the same fused nucleus \(^{205}\text{At} \text{and} ^{213}\text{At}\) have been studied at Orsay by the \(^{40}\text{Ar} + ^{165}\text{Ho}, ^{24}\text{Mg} + ^{181}\text{Ta}\) and \(^{20}\text{Ne} + ^{N_{\text{a}}\text{Re}}\) reactions. Following the above considerations, fast fission had to occur in the \(^{40}\text{Ar} + ^{165}\text{Ho}\) system and is unexpected in the two other reactions. In fact in the case of the argon projectile induced reaction the critical angular momentum for fusion (C.N. + fast fission) largely exceeds \(l_{B_f=0}\) limit.

Consequently the corresponding mass distribution of the fission-like fragments has to be wider in the angular momentum window leading to the fast fission process because of the incomplete mass equilibration. But for an equal value of angular momentum, this mass distribution must be similar for the three above systems. The experimental results, displayed in figures 16, 17 and 18, are in good agreement with these predictions. These results have been confirmed by data derived from \(^{40}\text{Ar} + \text{Au or Bi or U reactions}\)\(^{29}\). These data do not constitute unambiguous evidences for the occurrence of the fast fission process. Other signatures remain thus needed and in particular it would be of great interest to confirm that different ranges (or windows) of the initial \(l\)-waves do lead to the two types of fission mechanisms. As presently, mass relaxation seems to be a good tracer of the competition between both mechanisms, the transferred angular momentum behaviour as a function of the mass ratios of the outgoing fragments can also bring relevant and conclusive informations. As the fast fission cannot be clearly observed alone, it is worthwhile to compare the data obtained on almost the same fission nucleus, at similar excitation energy. Therefore we proposed to study the following two reactions: \(^{20}\text{Ne} + ^{N_{\text{a}}\text{Re}}\) at 220 MeV incident energy and \(^{40}\text{Ar} + ^{160}\text{Ho}\) at 315 MeV. In the first case only compound nucleus-fission process can occur because \(l_{\text{crit}} = l_{B_f=0} \approx 80\ \hbar\) and \(<l_i> \approx 53\ \hbar\). In the argon induced reaction \(<l_i> \approx 100\ \hbar\) and \(l_{\text{crit}} = 120\ \hbar \gg l_{B_f=0}\), thus both C.N. - fission and fast fission compete.

In these reactions the angular momentum transfer can be calculated as a function of the fragment masses assuming rigid rotors with or without the contributions of low-energy fission collective modes\(^{32}\) such as bending, wriggling or twisting (see figures 19 and 20). For rigid rotation the transferred angular momentum increases with the fragment mass ratios and allows thus a clear distinction between different
Experimental and calculated (with the critical distance concept) values of the fission cross section for the fusion system $^{203}$At formed by $^{40}$Ar+$^{165}$Ho and $^{24}$Mg+$^{181}$Ta respectively. Also shown are the limiting values of $I$ contributing to fusion, as calculated by Bass.

**Figure 16.**

Fission fragment mass distribution width versus the fissioning nucleus excitation energy, for four systems leading to nearly identical compound nuclei ($^{203}$At for Ar+Ho and Mg+Ta, $^{210}$At for Ne+Re, $^{213}$At for He+Bi). $I_{\text{crit}}$ is reached for the system Ar+Ho at an excitation energy of 98 MeV.

**Figure 17.**

Fission fragment mass distribution width versus the maximum angular momentum of the fissioning nucleus $^{203}$At formed either by $^{40}$Ar+$^{165}$Ho or by $^{24}$Mg+$^{181}$Ta.

**Figure 18.**
Figure 19.

Figure 20.

Figure 21.
initial 1-wave ranges. However, the collective modes also contribute to the spin of the fission fragments as it has been proved in many experiments covering spontaneous particle and H.I. induced fission\textsuperscript{33}). The influence of these collective modes increases with decreasing initial 1-waves and decreases with the mass ratio of the fragments.

From figure 21 showing the total spin of the two fragments, one can notice that the distinction between the two processes is not easy at symmetric mass splitting ($M_1 = M_2 = 100$) while it remains possible when this mass ratio increases ($M_2$ increasing). In addition to this, the shape of the fragment spin distribution (its first three moments) are of some help to disentangle the angular momentum transfer and collective mode distributions.

Experimentally, $\gamma$-ray multiplicity data associated to the mass fission fragments are recorded. From the 1, 2 and 3-folds coincidence data\textsuperscript{34}) one obtains $\gamma$-ray multiplicity distribution and its first moments ($\bar{M}_\gamma$, $\sigma_\gamma$ and $s_\gamma$ its skewness), these values could be associated to the fragment spin distribution.

Data collection has been done using Ne and Ar beams delivered by the Louvain-la-Neuve cyclotron. The data reduction and analysis exactly confirm the previously measured mass distribution for each reaction. The $\gamma$-ray data reduction are in progress\textsuperscript{36}), the results being confronted to the available theoretical models\textsuperscript{32,28,35}).

7. CONCLUSIONS

Heavy ion induced reactions, and in particular heavy ion induced fission-like reactions have been widely studied during the last twenty years. Their basic properties are presently known in some details. The growth of this field has been possible thanks to the always increasing performances of heavy ion accelerators mainly located in France, U.S.A., U.S.S.R. and Germany. In Belgium, the heavy ion facilities of the Louvain-la-Neuve-cyclotron are now widely opened. H.I. production by Electron Cyclotron Resonance Source already assures high $E/n$ ratios with large intensities.\textsuperscript{12}C to \textsuperscript{40}Ar beams, with energies ranging up to 30 MeV/n and 10 MeV/n respectively, are available with a particularly excellent fiability and month to month improvements (see figures 22 and 23).
Figure 22.
Maximum energy per nucleon as a function of the projectile mass.

Figure 23.
Maximum energy delivered by CYCLONE as a function of the projectile mass. The curve Bc shows the evolution of the coulomb barrier for Pb + projectile.
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ABSTRACT
Heavy ion induced fission between 10 and 100 MeV/u is discussed. It is shown that one can obtain information on fusion limits and on typical times characterizing nuclear matter.

INTRODUCTION
For few years, intermediate energy (10-100 MeV/u) heavy ion beams are available in few laboratories. In this paper, we will describe results concerning fission which have been obtained with such beams. They can be mainly divided in two classes.

In the first case, fission has been used as a tool to study reaction mechanisms; the main question is: what are the limits concerning fusion between two nuclei?

The second class of results concern competition studies between fission and evaporation when a very hot fusion nucleus has been built. Indeed intermediate energy heavy ions are the only projectiles which can be used to build very excited nuclei and this is a new field of study.

These two classes of results are discussed in the two parts of this contribution.

I - FISSION AS A TOOL TO UNDERSTAND HEAVY ION PHYSICS
Among the different observables that can be measured in heavy ion induced reactions, the linear momentum transfer from the projectile to the target is of particular interest for the understanding of the reaction mechanisms governing the nucleus-nucleus collisions.

A very common way to obtain information on the linear momentum transfer is the measurement of the separation angle between the two fission fragments emitted by the fused system after it has decayed by fission. Most of the measurements which are the subject of this paper have been made using light ions such as $^4$He, $^{12}$C, $^{14}$N, $^{16}$O and $^{20}$Ne (1-14). More recently, experiments have been performed with $^{40}$Ar projectiles at intermediate bombarding energies (15-19).

Below 10 MeV/nucleon, at low bombarding energies, the folding angle distribution of the fission fragments is characterized by a nearly symmetric peak as shown in fig. 1 and 2. The maximum of the distribution is centered on a mean value corresponding to the complete linear momentum transfer from the projectile to the target. In this low energy domain, typically
FIGURE 1: Evolution of the fission fragment angular correlation measured in the $^{14}N + ^{238}U$ system as a function of the bombarding energy between 7.4 and 45 MeV/nucleon. From ref. 14.

FIGURE 2: Same as in fig. 1 for the $^{40}Ar + ^{238}U$ system between 8.5 and 44 MeV/nucleon. From ref. 4, 10, 16 and 17.
As the bombarding energy increases, the folding angle distribution exhibits two components. At the smaller correlation angles, an intense peak is associated with large mass and momentum transfers to the target occurring in rather central collisions. At larger angles, a broad contribution shows up which corresponds to relatively low momentum transfers taking place in more peripheral collisions. This component originates in transfer reactions or inelastic scattering of the two ions, followed in a second step by the sequential fission of the quasi target. Only a few nucleons are involved in that process.

When increasing the incident energy, the component associated with the large momentum transfers decreases while the component associated with the low momentum transfers increases. This is nicely shown in fig. 1 for the \(^{14}\text{N} + ^{238}\text{U}\) system measured at various bombarding energies\(^{(14)}\). With the increase of the bombarding energy, the most probable value of the folding angle distribution for the fusion-like component is no more longer centered on the \(\theta_{\text{FMT}}\) value, corresponding to the full linear momentum transfer from the projectile to the target, but is shifted towards larger correlation angles (see fig. 2), indicating that we have to deal with an incomplete linear momentum transfer. Assuming a symmetric splitting of the heavy nucleus and using as the fission fragments kinetic energies the values deduced from fission systematics\(^{(21)}\), the folding angle scale can be converted into a linear momentum transfer scale, as shown in fig. 1. By plotting the most probable value of linear momentum transfers measured in central collisions, we obtain the universal curve, displayed in fig. 3, showing, as a function of the relative velocity of the two ions at contact, the behaviour of the fraction of the incident momentum transferred in fusion-fission reactions. The same trends are observed when are plotted transfer values issued from evaporation residue measurements. This shows that the general features noted in figure 3 are due to entrance channel (fusion) properties but not to exit channel characteristics. Two regimes are clearly distinguished in fig. 3, at low energy and high energy, respectively. As can be seen, the experimental data exhibit a limitation of the linear momentum transfers above the Coulomb barrier, with incomplete transfers down to 40% at energies as high as 84 MeV/nucleon, indicating that the projectile has not completely transferred its linear momentum to the compound system.

**FIGURE 3**: Fraction of the incident momentum transferred to the compound system in fusion-fission reactions as a function of the relative velocity of the two nuclei.
What is the mechanism responsible for that missing linear momentum transfer? Experimental results have shown that projectile-like fragments cannot account for the whole missing linear momentum observed in central collisions, and that some prompt light particles are likely ejected in the forward direction before the fusion-like nucleus undergoes fission. Other results show that light nuclei with the beam velocity are detected in very central collisions in the forward direction with two coincident fission fragments. A possible interpretation of these observations is that nucleons can escape from the mean field of the target if their kinetic energy is much larger than the Fermi energy ("Fermi jet"). Other possible explanations are pre-equilibrium or pre-compound particles, emission from a hot spot, break up of the projectile, etc... All of these theories can account for the fact that the fast ejected particles carry away a sizeable fraction of the linear momentum, but at the present time it is not clear which model prevails at these intermediate energies. Furthermore, with the onset of the nucleon nucleon collisions as the bombarding energy increases, linear momentum is less and less easily deposited in the target. Such a picture is expected to occur around the Fermi energy (\(E_F \sim 35\) MeV) as observed experimentally.

By looking in more detail at the spectra in figs. 1 and 2, we can see that coincident fission fragments are recorded with a folding angle greater than 180° or with a folding angle smaller than the \(\Theta_{FMT}\) value corresponding to the full momentum transfer from the projectile to the compound nucleus. Indeed, the velocity and mass dispersions of the fission fragments, as well as the effects due to the particle evaporation (mainly neutron emission for the heavy targets under consideration in this study) are responsible for such events in broadening the angular correlation of the fission fragments.

![FIGURE 4: Folding angle distributions measured in the \(^{40}\text{Ar}+^{232}\text{Th}\) system at different bombarding energies (from ref. (18)). The fusion like events disappear at an incident energy between 35 and 44 MeV/nucleon.](image)

Let us come back to the evolution of the folding angle distribution with the incident energy. As it can be seen in fig. 4, the low momentum transfer component increases steadily with the bombarding energy meanwhile the fusion-like events fall off and progressively vanish. In case of heavy targets with very low fission threshold such as U or Th nuclei, peripheral collisions account for most of the total reaction cross section for energies beyond 30 MeV/nucleon, as shown in fig. 5. The disappearance of the fusion-like
events occurs at an energy depending upon the projectile and target masses. With uranium targets, the fusion-like component vanishes at an incident energy of 35 MeV/nucleon with Ar projectiles (17), but is still observed at 45 MeV/nucleon with 14 N projectiles (14) and at 84 MeV/nucleon with 12 C projectiles (9).

A similar disappearance of the fusion-like component is seen by looking at the evaporation residues formed in the 40 Ar + 124 Sn collisions (30) or 40 Ar + Ag (43). Then, this effect is an entrance channel (fusion) effect and it is not due to the fact that fission would no longer be binary when the fissioning nuclei is very excited. As the fusion process is still observed with lighter projectiles even at higher bombarding energy, it has been suggested that the vanishing of the fusion-like events is linked with the importance of the projectile mass. More excitation energy is brought into the composite system with a heavy projectile rather than with a light one. These observations are interpreted as an evidence that the disappearance of the fusion-like events is related to the maximum excitation energy that the composite nucleus can carry (17, 31). Calculations have been done on the statistical properties of highly excited nuclei (32-34) in order to estimate the limit of stability of such heated nuclei. Due to the Coulomb repulsion, a hot nucleus becomes unstable when the temperature goes beyond a limiting value $T_{\text{lim}}$ depending on the charge and the mass of the nucleus. From this temperature, maximum energy that a system can sustain can be derived, and using the universal curve given in fig. 3 to deduce the linear momentum transfers, we can roughly predict for any system the laboratory energy at which the fusion-like component is expected to disappear. Such estimations have been performed (35) and, for the heavier systems, the fusion mechanism should no more longer be observed beyond a laboratory energy of 30-35 MeV/nucleon, in relative agreement with the experimental data (fig. 6). Above these energies, it is no more possible to build an equilibrated nucleus and other mechanisms take place such as multifragmentation (36) or liquid-gas phase transition (37). Nevertheless up to now, the basic mechanism responsible for the vanishing of the fusion-like component is not well understood, neither the decay mode of these very hot nuclei.

An interesting question concerns the fission properties when a very hot nucleus has been built. Very few data are available and experimental efforts have to be done. One knows only that fission remains binary even

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**FIGURE 5**: Evolution of the cross-sections of the fusion-like events (large transfers) and peripheral collisions (small transfers) as a function of the bombarding energy, in the 40 Ar + 232 Th system (18).
for extreme excitation energies (see above). Concerning the fragments kinetic energies they seem to be slightly larger than the Viola systematics predicts, but the deviation is quite small, thus indicating that the shapes of the nascent fragments do not depend strongly on the available excitation energy. If one considers the mass distributions, no clear conclusion can be drawn because the primary distributions are very much perturbed by sequential fragment decay. A careful analysis of these decay steps has to be performed in order to conclude.

II - FISSION DECAY AT HIGH TEMPERATURE

Heavy ions are the only projectiles which can be used to build very hot nuclei. In this section, we will discuss the evolution and the qualitative changes of nuclear fission probability when the fissioning nucleus temperature is strongly increased.

II-1 : The limits of the static statistical theory

An excited nucleus may deexcite either by fission or by particle evaporation. The theory describing this competition is the statistical theory where the basic feature (microcanonical representation) is that the probability $P$ of a given process is proportional to the corresponding final state density (or to the state density at the saddle point for fission). By expressing with a model these state densities as a function of the excitation energy $E^*$, one is led to the well known expression:

$$
\frac{P_F}{P_1} = \frac{k(E^*) \exp \left( \frac{2a_F}{2a_F(E^*-B_F)} \right)}{\exp \left( \frac{2a_1}{2a_1(E^*-B_1)} \right)}
$$

where $B_F$ and $B_1$ are respectively the fission barrier and the minimum energy required to evaporate the particle $i$; $a_F$ and $a_1$ are the so-called level density parameters for deformed (saddle point shape) and spherical nuclei respectively.

When the temperature of the nucleus is increased, several features have to be considered.

a) the parameters of relation (1) evolve;

b) the basic assumptions of the statistical theory quoted above have to be reconsidered: indeed, the above theory is essentially static since the probability of a given process is simply proportional to the state density.
This may be valid only if a given process (evaporation or fission) may be considered as instantaneous. At large temperature, this no longer true because time between two successive evaporation becomes also very small.

These two aspects are developed in the following sections.

II-2-a : Evolution at high temperature of the statistical theory parameters

When the temperature of a nucleus is increased, several parameters of relation (1) ($\alpha_f$, $F$ and $B_F$) are expected to vary. In both cases, there is no direct experimental way to observe these changes and one is led to believe theoretical predictions. In the case of the level density parameters, the variations are predicted rather weak as it is shown in figure 7: it is shown that several theoretical approaches (Hartree Fock, Extended Thomas Fermi) agree rather well one with the other and one expects similar nuclei properties at low and high temperatures as far as the influence of the level density parameters is concerned.

The situation is clearly different in the case of fission barriers. Figure 8 is a clear illustration of this feature. It appears that for increasing temperatures, the fission barrier of a $^{240}$Pu nucleus is strongly decreasing and becomes negligible for a temperature of 4 MeV. Very hot nuclei would then undergo fission rather easily whatever their mass number.

From this point of view, the nuclear temperature effect is quite similar to an angular momentum effect and can be used to extend fission studies over all
the periodic table at variance with what is possible in neutron induced fission.

From an experimental point of view, the main difficulty is here to disentangle between temperature and angular momentum effects since it is rather difficult to heat strongly a nucleus without increasing too its angular momentum; the experimental situation is then still unclear and fission properties of very hot nuclei are still unknown.

II-2-b: Dynamical competition between fission and evaporation

The statistical theory is a static theory in which one neglects the typical durations needed to evaporate or to fission. This is valid if the time between two successive decay steps is long. But this last time is a very fast decreasing function of the temperature \( T \) of the decaying nucleus. In figure 9\(^\text{40} \) which is an example for a \(^{208}\)Pb nucleus, it can be seen that this time is shorter than \( 10^{-22} \) s for \( T \) values exceeding 5 MeV. Now, if the evaporation duration may be much smaller than \( 10^{-22} \) s, the fission duration is about \( 4 \times 10^{-21} \) s\(^\text{41} \). It is then clear that the competition evaporation-fission will be perturbed by time scale considerations as soon as the decaying nucleus temperature exceeds 2 to 3 MeV. In such cases, evaporation will become possible during the fission process itself.

\[ \text{FIGURE 9: Evolution with temperature of the neutron evaporation half life for a }^{208}\text{Pb nucleus. From ref. 40.} \]

From an experimental point of view, such an analysis may be used to understand why the \( P_F/P_I \) values (relation 1 are often found much smaller than expected.

The first results of this kind have been obtained by the J. Alexander group\(^\text{10} \) slightly below 10 MeV/u. But similar behaviours have been observed at 27 MeV/u\(^\text{16} \), 35 MeV/u\(^\text{19} \) or 60 MeV/u\(^\text{42} \). Figure 10 is a typical example concerning the C+Th system at 60 MeV/u. Light charged particles were detected at 135 degree to the beam in coincidence with fission fragments resulting from fission following fusion. It appears in figure 10 that they exhibit identical kinetic energy spectra shapes whatever the direction of the coincident fragments. This feature shows that they are emitted before (or during) fission by the composite system rather than from the fission fragments. The multiplicity of these pre-fission charged particles is a very fast increasing function of the excitation energy in agreement with the qualitative arguments discussed above.
All these behaviours have been very well quantitatively understood in dynamical statistical calculations\(^{40,43}\) where the fission is assumed to be a Markov process which is then described by a Fokker Planck equation. Fission is assumed to be sufficiently slow to be decomposed in a succession of steps characterized by given collective variables values (elongation variable for instance). At each step the intrinsic degrees of freedom are assumed to be equilibrated and the evaporation time is small enough to allow an evaporation process to take place at a given step. In such an approach, the number \(S_n\)

\[ \text{FIGURE 10 : Backward emitted light charged particle spectra when the coincident fission fragments are detected at various angles. See text. From ref. 42.} \]

sees that the calculated fission width (full line) is negligible for small \(S_n\) values. This feature is simply due to the relatively long time needed for fission. When this time scale constraint is relaxed (longer decay times, i.e. larger \(S_n\)), the fission width increases. For large \(S_n\) values, it decreases again because the excitation energy has been drastically reduced by previous evaporation. For \(S_n\) values larger than 8, the full line curve of figure 11 is equivalent to the static statistical model prediction (dashed line) but the difference between both approaches is huge for very small decay time values.

This aspect of evaporation-fission competition was completely unknown ten years ago and its main features are now rather well understood. Intermediate energy-heavy ions have been quite successful tolls to progress.

\[ \text{FIGURE 11 : See text} \]
Another aspect of fission evaporation competition can be understood in looking again at relation (1). $B_i$ is the minimum energy required to evaporate particle $i$. If the particle is charged, $B_i$ is the sum of the binding and coulomb energies. Coulomb energy is responsible for the fact that only light charged particles (H and He isotopes) are usually evaporated. However, for very large excitation energies, the relative influence of $B_i$ decreases and evaporation may concern clusters as Li or Be isotopes. It seems that intermediate energy heavy ions are able to open this new decay channel because very hot nuclei can be build. This feature can again dramatically modify the fission probability. For instance, in the case of the Ar+Au system at 35 MeV/u where incomplete fusion reaction lead to Z=90 nuclei with huge excitation energies ($\sim$600 MeV), one has recently detected evaporation residues which had not undergone fission. Their mass is about 130 a.m.u. which means that they have emitted about 100 nucleons. The only way to understand it is to assume that very fast cluster emission occurred.

From a theoretical point of view, cluster emission has been recently successfully described in the static statistical model. An interesting feature lies in the fact that evaporation may then be considered as a very asymmetrical fission and one is led to try to unify evaporation and fission theory as it was already suggested by Moretto ten years ago.

CONCLUSION

Intermediate energy heavy ions can be used to build very excited fusion nuclei. We have seen in section I that fission can then be used as a tool to test the fusion mechanism and to discover what are the extreme limits concerning fusion and hot nuclei formation.

In section II, we have seen that when very hot nuclei are built, fission evaporation competition cannot any longer be fully described in the usual way by the statistical model. New features as dynamical aspects or cluster evaporation modify dramatically the landscape. Concerning the detailed fission properties of very hot nuclei (for instance fragments properties), no strong deviations from the already know systematics has been obtained. However, very few detailed studies are yet available and a clear experimental programm has to be developed in order to progress. From a theoretical point of view, it is rather necessary to described fission and evaporation is an unified way.

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We show that a Fokker-Planck transport equation can describe continuously in a natural way, the deep inelastic reactions (D.I.R.), the fast fission (F.F.) and the fusion (C.N.) cross sections, for heavy ion collisions at energies of 5 to 10 MeV/A\(^1\). The D.I.R. deal essentially with the energy and angular momentum dissipation, the F.F. with the transfer of mass.

We generalize the statistical transport concepts previously developed for D.I.R.\(^2\). Owing to the general characteristics of heavy ion collisions, a classical treatment is possible. One has to solve a classical transport equation describing the time evolution of a density distribution function \(d(q_1, p_1, t)\) in the phase space of a set of collective variables \(\{q_i\}\). In the present analysis, we have to deal with a 10-dimensional phase space; 5 collective variables are explicitly treated: the relative motion distance \(r\), the scattering angle \(\theta\), the rotation degrees of freedom for each ion \((\theta_1, \theta_2)\), the mass drift \((x = \frac{A_2 - A_1}{A})\). The deformations of the ions are treated on a phenomenological way by including their effects in the conservative and dissipative forces.

Using the local harmonic approximation and initial delta-conditions
\[
d(q_1, p_1, t)_{t=t_0} = \int (q_1 - q_1^0) \cdot (p_1 - p_1^0),
\]
the density distribution \(d(q_1, p_1, t)\) has a multigaussian form. The first moments \(\langle q_1 \rangle\), \(\langle p_1 \rangle\) are the mean or classical values \(q_1^c\), \(p_1^c\); they are the solutions of the Newton equations of motion with conservative and dissipative forces. Our approach assumes a dynamical transition between a sudden potential in the entrance channel and an adiabatic potential to describe the phase of formation and breaking of the intermediate composite system.

The second moments \(\chi_{qq}\), \(\chi_{pp}\) are solutions of a system of coupled differential equations which are themselves coupled to the Newton equations for the first moments. These equations contain a term in the temperature \(T(t)\) of the heat bath for the intrinsic degrees of freedom which are assumed to be in equilibrium at each time \(t\).

This model allows to describe:

1) the dissipation i.e. it defines the mean values of the macroscopic observables as a function of the impact angular momentum \(L_0\): the total kinetic energy \(E(L_0)\), the scattering angle \(\Theta(L_0)\), the final angular momentum \(L_f(L_0)\), the mass transfer \(\Delta A / A\).

---

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\(\dagger\dagger\) Chercheur agréé I.I.S.N.
ii) the statistical fluctuations i.e it defines the multidifferential cross-sections $d^4\sigma/dE\ d\theta\ d\Omega\ da$, any integrated ones $d^2\sigma/dE\ da$, $d^2\sigma/d\theta\ da$ ... described as contour diagrams in the 2-dimensional plane of the associated macroscopic observables and finally any distributions like $d\sigma/da$ ... 

iii) quantum diffractive effects due to cut-off in the initial angular momentum range.

We have chosen to study the systems (Ar, Ho) and (Mg, Ta) leading to the same compound nucleus $^{205}$At for two mass asymmetries, respectively .61 for the (Ar, Ho) system and .77 for the (Mg, Ta) one. Both collisions are studied at several initial energies.

The results focus attention on the three configurations which the colliding ions system may be faced with; they are given at an incident energy, for different $l_0$ values of the entrance channel angular momentum. The D.I.R. are controlled by $l_0$ waves lower than the grazing value. At more lower $l_0$, two situations can develop:

i) the composite system does not involve the compound nucleus formation but separates into almost symmetric fragments ($\neq 0$), this is typical of F.F. processes

ii) the composite system evolves the C.N. formation.

<table>
<thead>
<tr>
<th>System</th>
<th>$l_0$</th>
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<tr>
<td>(Ar, Ho)</td>
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<tr>
<td>170</td>
<td>C/F</td>
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<tr>
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<td>359</td>
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</tbody>
</table>

Table 1

In Table 1, we report the $l_0$ values leading to C.N., F.F. and D.I.C. at several identical centre of mass energies for the systems (Ar, Ho) and (Mg, Ta). The critical limit to fusion $l_{CN}$, the $l_0$ limits $l_{20\text{uma}}$ and $l_{up}$ associated respectively to the transfer of 20 uma and to the maximum of mass relaxation are indicated. The results show that the $l_0$ angular momentum range to F.F. events grows with incident
energy and for lower initial mass asymmetry. In Fig. 1, we have drawn $\theta (l_o)$ and $A (l_o)$ for both systems at the different selected initial energies.

Using the sharp cut-off expressions for the F.F., the C.N. and the total reaction cross-sections, we obtain the curves drawn in Fig. 2.

Connecting the $l_o$ values to the energy by its centrifugal energy component, the above results are in agreement with the conditions of extra-push and extra-extra push energies in the model of Swiatecki (Fig. 3).
The mass distributions $d\sigma/dA$ reported in Fig. 4 for (Ar, Ho) and (Mg, Ta) at the various selected incident energies show that D.I.R. and F.F. appear to be quite well separated. In this model, we do not treat the fission of the C.N. so that the symmetric peak at $\frac{A}{2}$ comes only from F.F. events. We also calculate the 2-dimensional cross-sections $d^2\sigma/dE\, dA$, $d^2\sigma/d\theta\, dA$, $d^2\sigma/d\Delta \, dA$ ....

In Fig. 5 we report the contour diagrams of $d^2\sigma/dE\, dA$ in the (E,A) plane. The general pattern of the distributions reproduces clearly two typical regimes: the D.I.R. characterized by narrow peaks at the projectile ($A_1$) and the target ($A_2$) mass, reflecting the kinetic energy loss mode; the fully relaxed events centred at $A/2$, reflecting the energy loss mode and the mass drift towards symmetry.
In Fig 6, we report the \((\Delta l, \theta)\) contour diagram. For fragments scattering at \(\theta < \theta_g\), the angular momentum transfer grows from \(\circ\) to 30 or 40 \(\circ\) when \(\theta\) proceeds from the grazing to the forward direction. At \(\theta > \theta_g\), a large angular momentum transfer is observed which is nearly independent of the scattering angle. The value \(\Delta l\) is lower than the value accessible in the D.I.C., due to the fact that a different range of \(l_o\) waves is involved in the D.I.C. compared to the F.F.
Conclusion

This statistical dynamical model describes continuously the D.I.C., the F.F. process and the fusion cross-sections. The results obtained are in good qualitative agreement with the experimental data. We show how to separate the ranges of angular momentum in the entrance channel which contribute to each process.

The results obtained by using different parametrisations of the conservative and dissipative forces are qualitatively comparable. However, to have a better test, we need precise experimental signatures for the different mechanisms; there is also a clear need of more systematic experimental results.

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ABSTRACT

Inclusive and coincidence measurements have been performed to study symmetric fragmentation of $^{44}$Ti binary decay from the $^{32}$S + $^{12}$C reaction at 280-MeV incident energy. Element distributions after binary decay were measured. Fragment correlations are presented. Total CM kinetic energy for the symmetric products is extracted from our data and from Monte-Carlo model calculations including Q-value fluctuations. This result was compared to liquid drop model calculations and standard fission systematics. Comparison between the experimental value of TKE and the RLDM predictions locates the angular momentum window for symmetric splitting of $^{44}$Ti between 33 and 38 $\hbar$. It also showed that 50% of the corresponding rotational energy contributes to the TKE values. The dominant reaction mechanism was found to be symmetric splitting followed by evaporation.

1. INTRODUCTION

The present talk summarizes some of the experimental work performed by the Louvain-Krakow collaboration on binary fission of a light nucleus $^1$. Recently, the existence of symmetric or nearly symmetric decay of three light systems has been established in a coincidence experiment $^2$.

These data suggest a center-of-mass (CM) angular distribution $\propto 1/\sin \theta_{CM}$, which is expected from the decay of a system with a life time at least equal or greater than a rotation period $^3$.

In the present experiment $^1$ we study the fission channel for the 280 MeV $^{32}$S + $^{12}$C $\rightarrow$ $^{44}$Ti$^*$ reaction. Due to reversed kinematics fragment energies are high (no energy threshold limitations) and measurements could cover a wide range of CM angles, 30°-170°.

At 280 MeV incident energy the primary decay fragments from $^{44}$Ti are excited up to about 20 MeV, and lose a significant fraction of mass, and charge ($\approx 10\%$) by sequential particle emission. Therefore the resulting recoil changes in the primary velocity and direction induce broadening in the measured energy distributions and correlation curves. Another important effect is the fluctuation of the Q value and the scission point.

In this work both recoil and Q value fluctuation effects are treated in a systematic way. Consequently it was possible to get information on relative probabilities of symmetric and asymmetric decays of the $^{44}$Ti nucleus.
The most probable CM total kinetic energy (TKE) of decay fragments measured in this work provides a new value for the Viola fission systematics 7,8).

The beam of $^{32}_{\text{S}}9^+$ ions was delivered by an ECR source and accelerated up to 280 Mev in the variable energy cyclotron CYCLONE of the University of Louvain. The reaction products were detected by two ΔE-E gas-silicon telescopes positioned on rotatable arms inside a 100 cm diameter scattering chamber.

The ΔE, E, and time correlation signal between the telescopes (TAC) were registered on magnetic tapes in an event by event mode for off-line analysis.

2. MISSING CHARGE ΔZ

When one looks in an inclusive measurement for a nearly symmetric binary splitting of a light system such as $^{32}_{\text{S}} + ^{12}_{\text{C}} \rightarrow ^{44}_{\text{Ti}}^*$, the presence of products with about one-half the mass of the compound nucleus does not prove the existence of a fission process. Also a model independent distinction between symmetric and asymmetric decay modes is possible due to secondary evaporation. In order to avoid most of the above difficulties the main emphasis was laid in this work in coincidence measurements.

In an energy and angle integrated charge distribution for $Z_1-Z_2$ coincidences, binary reactions without charged particle evaporation should correspond to $Z_1+Z_2 = Z_p+Z_T = Z_{\text{CN}} = 22$. Here $Z_p$, $Z_T$, $Z_{\text{CN}}$, $Z_1$, and $Z_2$ denote, respectively, the atomic numbers of the projectile, target, composite system, and of the fragments detected in the two telescopes. However, it appears that the main bulk of the events are spread around lines $Z_1+Z_2 = 20$ for nearly symmetric events, and $Z_1+Z_2 = 18$ for projectile and target like pairs. Thus the most probable missing charge ΔZ is 2 to 3, which is most likely lost through particle emission from the excited composite system and/or from any of the reaction partners.

3. ANGULAR CORRELATIONS

In Fig. 1 are shown the in-plane laboratory angular correlations of different $Z_1$, $Z_2$ pairs measured with two telescopes placed at equal angles but on opposite sides of the beam direction, $\theta_1 = \theta_2$.

It contains cases classified later as symmetric and asymmetric binary decay followed by light particle evaporation from fully accelerated fragments. The following paragraphs will explain and justify how this classification was made.

For the moment we adopt in what follows the post-scission evaporation picture. We shall denote in this work all reaction parameters and cross sections before evaporation by primed symbols and after evaporation by unprimed ones.

The first row of data in Fig. 1 presents laboratory angular correlations for fragment pairs with $Z_{1}' = Z_1$, and $Z_{2}' = Z_2$, (ΔZ = 0). The case $Z_{1}' = 11$, $Z_{2}' = 11$ are classified as the symmetric decay, the $Z_{1}' = 10$, $Z_{2}' = 12$, and $Z_{1}' = 9$, $Z_{2}' = 13$ cases are classified as asymmetric decays.
The angular correlations are peaked at LAB angles around 20°.
For a binary process the mean angle of correlation $\theta_0 = \theta_1 = \theta_2$ is related to TKE by

$$
tg^2 \theta_S = \frac{4A_1^2A_2^2}{A_p(A_T+A_p)E_p} TKE - \left(\frac{A_1^2-A_2^2}{A_p+A_T}\right)^2$$

where $A_p, A_T, A_1, A_2$ denote the mass numbers of the projectile, target and primary fragments nuclei, respectively, and $E_p$ is the LAB incident energy. In this work we assume $A_i = 2Z_i$, where $i = 1$ or 2.

Because the total kinetic energy release is practically the same for symmetric and slightly asymmetric splittings of light systems (see Sec. 4), the mean correlation angle should decrease with increasing fragmentation asymmetry. Fig. 2 shows the experimental $\theta_S$ versus $\Delta Z$ dependence for all correlation curves from Fig. 1. Values of $\theta_S$ were extracted from centers of gravity of correlation data. Inspection of Fig. 2 shows that the $\theta_S - \Delta Z$ points are grouped around 3 different lines labelled a, b, c. For $\Delta Z = 0$ corresponding to the data shown in the first row of Fig. 1 the $\theta_S$ dependence on asymmetry agrees quite well with the predictions of formula (1) within the uncertainties in determining $\theta_S$. An average value of TKE (32.3 MeV) was extracted from the points at $\Delta Z = 0$. The corresponding values of $\theta_S$ are shown by arrows. For each group of points $\theta_S$ decreases with increasing $\Delta Z$. The $\theta_S$ vs $\Delta Z$ dependence is a consequence of particle evaporation from primary decay fragments (see Ref. 1). Therefore we classify all points corresponding to line a as the symmetric decay and all points corresponding to line b and c as the asymmetric decays.

3.1. Monte Carlo calculations

In order to gain a quantitative understanding of the correlation curves, Monte Carlo calculations have been performed. Application of this method enabled us to avoid difficulties in the LAB to CM transformation which are expected.
for binary reactions of light systems in the presence of particle evaporation 2).

The calculations were performed in velocity space (see Fig. 3) where \( \dot{V}_0 \) is the velocity of the composite system, \( \dot{V}_1 \) and \( \dot{V}_2 \) denote the initial CM velocities of fragments 1 and 2, respectively. \( \dot{V}_1 \) and \( \dot{V}_2 \) were calculated for a given TKE and primary mass asymmetry. The Q-value fluctuations at the scission point give a distribution of the CM kinetic energies around the average value TKE. This distribution was approximated by a Gaussian function with a variance \( S_0^2 = 25.8 \pm 3.2 \text{ MeV}^2 \). The value of \( S_0^2 \) was calculated from the RLDM (see Sec. 4). A random number generator gave the initial directions of the fragments according to an assumed CM angular distribution

\[
\frac{1}{\sin \theta_1} \sim \text{Gaussian} \quad \text{where } \theta_1 \text{ is the angle of fragment 1 in the CM system.}
\]

To determine the CM average recoil velocity for a given fragment \( \dot{V}_i^{\text{recoil}} \), \( i = 1 \) or 2, a Gaussian probability distribution was assumed for each Cartesian component of \( \dot{V}_i^{\text{recoil}} \) with a variance \( S_i \) (see for more details Ref. 9). The variance \( S_i^2 \) should increase with increasing number of evaporated nucleons \( \Delta A \) and with decreasing mass \( A_i^* \) of the evaporating fragment (larger recoil). Both dependences are combined in a formula derived by Lide 10,11:

\[
S_i^2 = K \frac{\Delta A_i}{(A_i - \Delta A_i)^2} T_i
\]

where \( \Delta A_i \) denotes the total mass evaporated by a fragment with an initial mass \( A_i^* \) and a nuclear temperature \( T_i \) defined by 11

\[
T_i^2 = \frac{E_i^*}{a}
\]

where \( E_i^* \) is the excitation energy of a fragment assumed to be proportional to its mass \( A_i^* \), and \( a \) is the level density parameter. Due to Q-value fluctuations at the scission point the TKE and consequently fragment excitation energies are given by separate distributions. The excitation energy distribution was replaced in calculations by an average value \( E_i^* \). The resulting additional broadening is presented in formula (2) by a phenomenological factor \( K \). The angular distribution of recoil velocities \( \dot{V}_i^{\text{recoil}} \) was assumed to be isotropic. The final velocity of a fragment in the LAB system is given by \( \dot{V}_i = \dot{V}_0 + \dot{V}_i + \dot{V}_i^{\text{recoil}} \).

This Monte Carlo model has three free parameters: the primary mass asymmetry, \( A_i^*/A_j^* \), the total CM kinetic energy of fragments, TKE, and the parameter \( K \). Asymmetry \( A_i^*/A_j^* \) was assigned for each case according to the classification given in Figs. 1 and 2. Values of the remaining two parameters: TKE = 32.5 ± 1.5 MeV and \( K = 2.2 \pm 0.4 \) were obtained from fits to the correlation curve \( Z_1 = Z_2 = 10 (\Delta Z = 2) \) which has the largest cross section in the group of symmetric splitting.

Angular correlations calculated from this model are plotted by solid lines in Fig. 1. The overall agreement is good. For some cases it can be improved by adding neutron evaporation (Fig. 1, dotted and dashed lines).
4. ROTATING LIQUID DROP MODEL CALCULATIONS

Results of this work for the $^{32}$S + $^{12}$C binary decay reaction together with observations made for other light systems $^2$, $46 \leq A \leq 52$, may be indicative of a fusion-fission like mechanism conventionally described in terms of the rotating liquid drop model.

In our RLDM calculations we use a conventional parametrization of nuclear shapes following Ref. 4 (the $p$, $\lambda$, $\Delta$ parametrization). In this model, the nuclear shapes are axially symmetric and correspond to two spheres modified by a smoothly fitted portion of a quadratic surface of revolution. Such simplified parametrization gives a reasonable agreement with the liquid drop model of Cohen, Plasil and Swiatecki $^5,6$).

The model predicts that there are no stable $^{44}$Ti nuclei with angular momenta higher than 43 $\hbar$.

The RLDM energies of the nucleus at saddle and scission points determine the effective reaction Q-value and the total kinetic energy of fission fragments. The TKE is given as:

$$\text{TKE} = E^{\text{SC}}_{C} \text{(int)} + E^{\text{SC}}_{\text{rot,rel}} + E_{\text{kin}},$$

where $E^{\text{SC}}_{C}$ \text{(int)} is the Coulomb interaction energy of fragments at scission point, and $E^{\text{SC}}_{\text{rot,rel}}$ is the rotation energy of relative motion at scission. The translation kinetic energy gained by the system between saddle and scission points is denoted $E_{\text{kin}}$ (see, for more details, Ref. 1).

In Fig. 4 the calculated total CM kinetic energy of fragments from symmetric fission of $^{44}$Ti is presented as a function of the total angular momentum L. Since the saddle-to-scission energies are here negligible the TKE is mainly the sum of the Coulomb, $E^{\text{SC}}_{C}$ \text{(int)} and the relative motion, $E^{\text{SA}}_{\text{rot,rel}}$ energies which are also shown. The hatched band in Fig. 4 represents the measured TKE = 32.5 $\pm$ 1.5 MeV. The liquid drop total kinetic energy curves increase with L and cross the experimental band suggesting an angular momentum window for fission from 33 $\hbar$ to 38 $\hbar$. Inside of this window the Coulomb and rotation components of the total kinetic energy are of equal importance.

The total kinetic energy depends very little on the initial asymmetry. For $^{A_1}_{A_2} = 22 \ 22$, and $^{18}_{26}$ the difference in TKE is smaller than 0.2 per cent.

The RLDM provides not only the mean value but also the distribution of the TKE, which is related to the fission Q-value fluctuations. According to a method described in Ref. 12, the variance of the distribution of TKE was found to be $\sigma^2_Q = 25.8 \pm 3.2 \text{MeV}^2$ for $L = 36 \hbar$. This value was used in our Monte Carlo calculations.
5. SUMMARY AND CONCLUSION

The coincidence measurement presented and discussed in this study clearly established the symmetric and nearly asymmetric binary fragmentation of the $^{44}$Ti nucleus. It was found that the dominant reaction mechanism is the postscission evaporation in agreement with previous results obtained for similar light systems \(^2\). The most probable total CM kinetic energy $TKE = 32.5 \pm 1.5$ MeV of decay fragments provides a new value for the standard fission systematics. The first Viola paper \(^7\) provides for our case the value 36.9 MeV and the recent value of Viola et al. \(^8\) is 23.6 MeV. The RLDM predicts that most of the initial angular momentum in the reaction is converted to the orbital motion of the nascent fragments. Consequently the rotation energy contributes in about 50% to the total kinetic energy $TKE$ of fragments. This may explain the discrepancy observed between the experimental $TKE$ value and predicted by the Viola systematics. However, it should be stressed out that all quantitative conclusions of our calculations are likely to be affected to some extend by refinements to the RLDM. Beside the shell effects which seem to be not important at such excitation energy, these refinements include both the finite nuclear interaction and the temperature dependence of the liquid drop model parameters.

REFERENCES

CONTENTS

Programme
Welcome address
C. Wagemans
List of participants

SESSION I
Introductory Session

Fission revisited
A.J. Deruytter
How fission was discovered
P. Van Assche
Photofission
E. Jacobs, D. De Frenne, A. De Clercq, M. Verboven, M. Piessens, G. De Smet
Neutron induced fission below and at the barrier
H. Weigmann
Application of some fission properties to neutron dosimetry
P. D'hondt and A. Fabry

SESSION II
Light charged particle and neutron emission in fission

Review of theoretical approaches to the emission of α-particles during nuclear fission
N. Cârjan
Ternary fission
J. Theobald
Neutron induced ternary fission
C. Wagemans, P. Schillebeeckx, P. D'hondt, J.P. Bocquet, A. D'Eer
Emission of light charged particles in the photofission of actinides
M. Verboven, E. Jacobs, P. D'hondt, A. De Clercq, D. De Frenne, M. Piessens, G. De Smet
Fragmentation and neutron emission for 252Cf(s.f.)
C. Budtz-Jørgensen and H.-H. Knitter

SESSION III
Fission fragment properties

The scission point model: possibilities and deficiencies
J. Moreau, K. Heyde, M. Waroquier, J. Jolie, J. Ryckebusch
Cold fragmentation: experiments and models
F. Gönnenwein
Fission fragment energy and mass distributions in the spontaneous fission of the Pu-isotopes

P. Schillebeeckx, C. Wagemans, A. Deruytter, R. Barthélemy

Study of the $^{241}\text{Pu}(n_{\text{th}},f)$ fragments with Cosi Fan Tutte

P. Schillebeeckx, C. Wagemans, P. Gellenbort, A. Oed, F. Gönnenwein

Fission fragment properties of $^{235}\text{U}(n_{\text{th}},f)$ in the neutron energy range from thermal to 1 MeV


$^{235}\text{U}(n,f)$ fragment mass-, kinetic energy- and angular distributions for incident energies between thermal and 6 MeV

C. Straede, C. Budtz-Jørgensen, H.-H. Knitter

Photon induced fission of $^{232}\text{Th}$ with 12 and 20 MeV bremsstrahlung

M. Piessens, E. Jacobs, D. De Frenne, A. De Clercq, M. Verboven, G. De Smet

SESSION IV

Heavy ion induced fission

Heavy ion induced fission at $E_1 \leq 10$ MeV/u

F. Hanappe and Y. El Masri

Heavy ion induced fission above 10 MeV/u

B. Tamain

Dynamical and statistical theory of deep inelastic, fast fission and fusion processes

A. Van Geertruyden and C. Le Clercq-Willain

Binary fission of $^{44}\text{Ti}$

P. Cohilis and the UCL-Krakow collaboration