

Investigation on the Primary Spins of the ^{235}U Fission Fragments

P. ARMBRUSTER, H. LABUS, and K. REICHEL

Institut für Neutronenphysik der Kernforschungsanlage Jülich, Jülich, Fed. Rep. of Germany

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Dedicated to Prof. Dr. H. Maier-Leibnitz on his 60th birthday

The prompt γ -radiation emitted in ^{235}U thermal fission has been investigated in a multiparameter experiment. γ -yield, time-distribution, energy spectrum and nuclear anisotropy of the emission have been measured, especially γ -yield and anisotropy as a function of fragment mass and γ -energy. The mass dependence of the primary spins of the fragments can be deduced from these measurements. A value of $7\hbar$ for the average primary spin and an increase of the primary spins within each fission product group is obtained. This dependence is explained within an adiabatic model of scission due to the mass dependence of the deformation of the fragments.

1. Introduction

Experiments on the mass dependence of the γ -ray yield and the γ -energy released in fission¹⁻⁴ show a remarkable increase of these quantities with increasing mass within both fission product groups. If neutron and γ -emission are independent deexcitation mechanisms with γ -emission following neutron emission, then the energy and number of γ -quanta released from a fragment are determined only by states below the neutron binding energy. Both number and total energy of the γ -quanta should not depend on the fragment mass. The experimentally observed dependence points to a competition between neutron and γ -emission. High values of the spins of the emitting nuclei make γ -emission competitive to neutron emission at excitation energies above the neutron binding energy. As the emission of neutrons cannot reduce the spin considerably, γ -emission begins in states with spin values of approximately the spin values of the virgin fragments. The higher the initial spin, the larger the γ -energy emitted by the fragments in competition with neutrons. The number and energy of γ -quanta emitted should be correlated to the primary spin of the fragments⁵.

The anisotropy of the γ -emission referred to the direction of flight of the fragments points to a

strong alignment of the spins when the fragments are created at scission⁶. The magnitude of the anisotropy gives additional information on the spin of the fragments and the multipolarity of the γ -rays emitted. From measurements of the anisotropy together with measurements of the yield of the γ -radiation the spin of the nascent fragments may be deduced.

Another quantity showing a drastic mass dependence similar to the γ -yield is the number of neutrons emitted from single fragments⁷. It has been shown that the observed saw-tooth dependence is caused by the mass dependence of the deformation energy of the fragments^{8,9}. The sharing of the total deformation energy between the fragments at scission is determined only by their nuclear structure. Soft fragments emit a high number of neutrons, more rigid ones emit a smaller number.

As neutrons and γ -quanta show the same mass dependence, a correlation between the softness of the fragments and their initial spins may exist. The final scope of this experiment is to verify such a correlation. A combined measurement of the number of γ -quanta and of the anisotropy of γ -emission has been made in order to determine the mass dependence of the spin of the fragments and to reveal the postulated correlation. Preliminary experimental results have been published at the IAEA Conference

¹ S. A. E. JOHANSSON, Nucl. Phys. **60**, 378 [1964].

² H. MAIER-LEIBNITZ, H. W. SCHMITT, and P. ARMBRUSTER, in: Physics and Chemistry of Fission (Proc. Symp. Salzburg 1965) **2**, IAEA Vienna 1966, p. 143.

³ G. V. VALSKIJ, G. A. PETROV, and Y. S. PLEVA, Sov. J. Nucl. Phys. **3**, 171 [1969].

⁴ H. ALBINSON and L. LINDOW, Rep. AE-398, 1970.

⁵ T. D. THOMAS and J. R. GROVER, Phys. Rev. **159**, 980 [1967].

⁶ V. M. STRUTINSKY, JETP **10**, 613 [1960].

⁷ J. TERRELL, in: Physics and Chemistry of Fission (Proc. Symp. Salzburg 1965) **2**, IAEA Vienna 1966, p. 3.

⁸ R. VANDENBOSCH, Nucl. Phys. **46**, 129 [1963].

⁹ F. DICKMANN and K. DIETRICH, Nucl. Phys. A **129**, 241 [1969].



at Vienna in 1969¹⁰. There our experimental technique and preliminary experimental results on mass dependent prompt γ -ray-yield and life time measurements in the psec-region have been presented.

2. Experimental Technique

Some special aspects of the experimental technique described in detail in^{10,11} shall be mentioned briefly: The experimental set up, as show in Fig. 1, consists of a multidetector arrangement combining mainly two advantages. On the one hand, the intensity is increased by a factor of 8 compared to a measurement with two detectors; the data collection time could therefore be limited to a few reactor periods. On the other hand, it is possible by making use of the symmetry of the arrangement to eliminate completel unwanted perturbations; e. g., the relativistic solid angle aberration or differences of detector efficiencies.

Two reasons made the mass determination by simultaneous energy and time of flight measurements

in our case superior to an energy-energy measurement. The use of thick targets yields high fission rates and the stable backing ensures a plane target surface. This latter property is important as the γ -rays emitted from complementary fragments must be spatially separated. This separation is made with a precise collimator (see Fig. 1), a technique introduced by JOHANSSON¹, to measure mass dependent prompt γ -ray yields of ^{252}Cf -fission products. The aperture and the distance to the target plane of such a collimator must be set within a few hundredths of a mm corresponding to a few psec, where the bulk of the prompt radiation is emitted.

The time-of-flight measurement along distances of 20 cm between target and heavy ion detectors restricts the mass resolution to 20 AMU (FWHM). Due to low counting rates this disadvantage is partly compensated by the fact that the statistical errors limit the anisotropy measurements to a few mass regions anyhow. The time resolution of the time-of-flight measurement was 1.6 nsec (FWHM). The dominating contribution to this value comes from the start signal. This is produced by the fragments recoiling into a thin Ne 102 A plastic foil which has been evaporated with

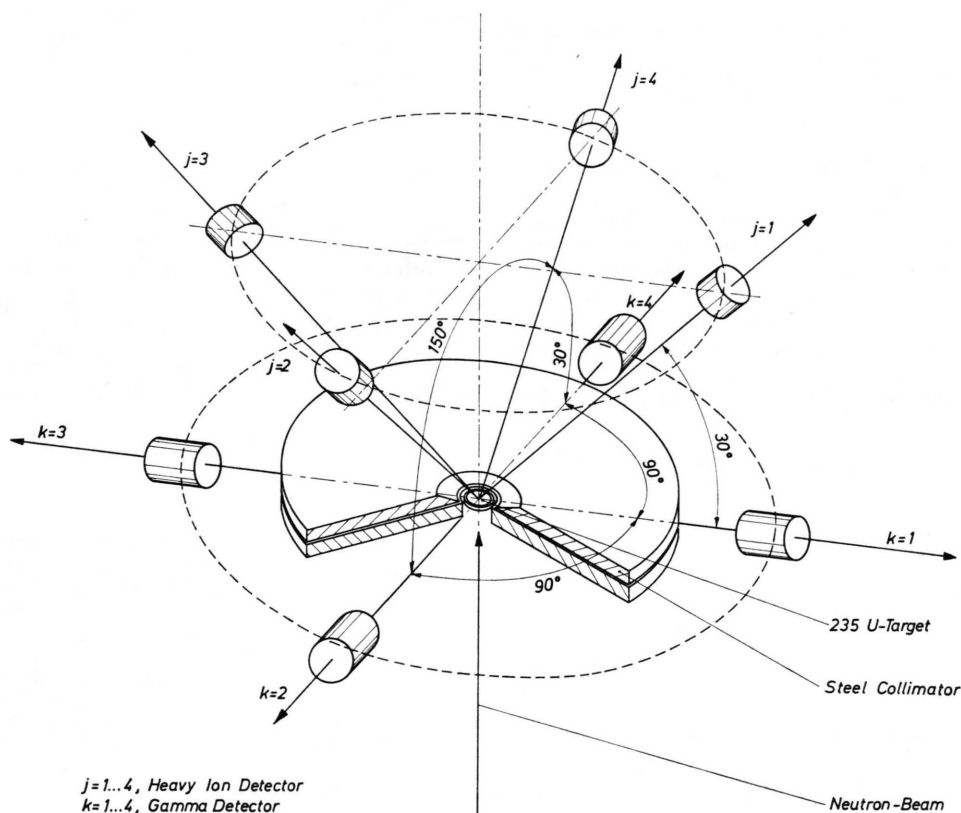


Fig. 1. Detector arrangement.

¹⁰ P. ARMBRUSTER, F. HOSSFELD, H. LABUS, and K. REICHEL, in: Physics and Chemistry of Fission (Proc. Symp. Vienna 1969) 2, IAEA Vienna, p. 545.

¹¹ H. LABUS, Rep. JÜL-643-FN, 1970.

fissionable material, $1 \text{ mg/cm}^2 \text{ UF}_4$. Each stopped fragment generates photons which reach a photo-multiplier 56-AVP via a light transmitting system. Due to radiation damage the photon production in the plastic scintillator decreases fast so that after a two days run only one photo-electron is emitted per registered fission. The time jitter of the start signal is then governed by single photon statistics and has the same magnitude as the decay time (2.2 nsec) of the scintillator. The degrading efficiency and mass-resolution is demonstrated in Fig. 2 where two mass distributions are presented as measured with a new target and after $5 \cdot 10^{11}$ fissions.

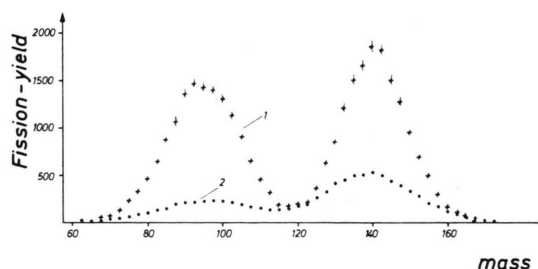


Fig. 2. Mass distributions, showing the decreasing mass resolution and efficiency with increasing radiation damage of the plastic backing of the ^{235}U target. (1) fission yield with a new target, (2) fission yield of the same target after $5 \cdot 10^{11}$ fissions.

Time-of-flight discrimination of prompt γ -quanta and neutrons between the target and the γ -ray detectors eliminates the influence of prompt fission neutrons which would otherwise give a pronounced anisotropy in the laboratory system^{3, 4, 12-15}. The γ -quanta are registered by plastic scintillators which also give a rough information on the γ -spectrum. The time of flight spectrum is shown in Fig. 3 demonstrating the suppression of γ -quanta with closed collimator. The dashed lines are the levels of a single channel analyzer

separating the prompt γ -radiation from the prompt neutrons. The signal to noise ratio was evaluated from the two areas between the dashed lines.

Due to the low counting rates of about one event per minute the data could be punched event by event on a paper tape. Each event consists of 4 binary numbers which stand for fragment energy, fragment flight-time, γ -energy in the range from 0.1 to 1.2 MeV, and for the triggered detector-pair, i. e. emission angle of the quanta referred to the flight direction. A three-fold coincidence with $0.5 \mu\text{sec}$ time resolution triggers the punch, if only one of the 4 fragment detectors and only one of the 4 γ -detectors produce a signal and if the flight time discriminator, as mentioned above, has recognized a γ -quantum in the plastic detector.

3. Measurements

The versatility of the experimental set up is shown by the fact that nearly every interesting quantity of the prompt radiation i.e. yield, anisotropy, life time can be derived from the measured coincidence rates $W(\vartheta)$, $W(\pi - \vartheta)$ and $W(\pi/2)$ between fragment detectors and γ -detectors, respectively.

Considering a fission event with fragment of mass M flying (Index 1) and the complementary fragment of mass $(A - M)$ slowed down in the plastic backing of the target (Index 2), the probability $W_{12}(\vartheta)$ for detecting a γ -quanta at the angle ϑ referred to the flight direction of the fragment with mass M may be expressed by Eq. (1). All quantities which are connected with the flying mass M have a subscript 1, the quantities of the stopped complementary mass $(A - M)$ have a subscript 2.

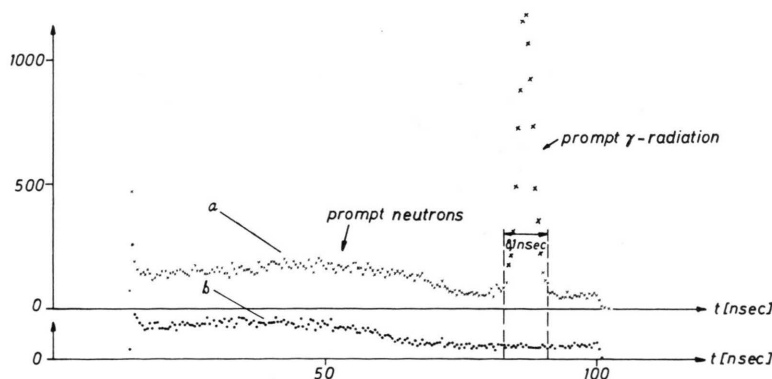


Fig. 3. Flight time distribution of prompt neutron- and γ -radiation between target and γ -detectors, (a) with open collimator: the peak of the prompt γ -radiation shows a time resolution of 8 nsec ($\text{FW}_{10} M$) for the total dynamic range of (0.1–1.0) MeV. The dashed lines are levels of a single channel analyzer to separate neutrons and γ -quanta, (b) flight time with closed collimator: the γ -peak is suppressed. The remaining area between the analyzer levels is the background.

¹² M. V. BLINOV, N. M. KAZARINOV, A. N. PROTOPOPOV, and B. M. SHIRYAEV, JETP **16**, 1159 [1963].

¹³ G. A. PETROV, D. M. KAMINKER, G. U. VALSKIJ, and L. A. POPEKO, Atomnaya Energiya **18**, 64 [1965].

¹⁴ K. SKARSVAG and I. SINGSTAD, Nucl. Phys. **62**, 103 [1965].

¹⁵ K. SKARSVAG, Nucl. Phys. A **96**, 385 [1967].

The time and γ -energy dependence is not noted explicitly for any of the quantities.

$$W_{12}(\vartheta) = K_{12} \{ p_1 G_1 (1 + \alpha_1 \sin^2 \vartheta + \varepsilon_1 \cos \vartheta) + p_2 G_2 (1 + \alpha_2 \sin^2 \vartheta - \varepsilon_2 f_2 \cos \vartheta) \}. \quad (1)$$

For the same pair of fragments two further very similar equations hold for $W_{12}(\pi - \vartheta)$ and $W_{12}(\pi/2)$. They are obtained from Eq. (1) by inserting $\pi - \vartheta$, $\pi/2$ for ϑ .

The quantities used in Eq. (1) have the following meaning:

K_{12} takes into account the fission yield of the fragment pair ($M, A-M$) as well as the solid angles and efficiencies of the detectors.

p_1, p_2 are transmission factors, giving the probability of a quantum emitted from the fragments with mass M and $A-M$, respectively, to penetrate the collimator. They mainly depend on the geometrical conditions of the collimator. The ratio $\varrho = p_1/p_2$ may be denoted as enrichment of quanta emitted from the flying fragment with mass M .

G_1, G_2 are the prompt γ -ray yields per fragment.

α_1, α_2 are the "nuclear anisotropies" of the prompt radiation emitted from the fragments with mass M and $A-M$. α is the first coefficient of the angular correlation function $P(\vartheta) = 1 + \alpha \sin^2 \vartheta + \dots$ which holds if the nuclei are aligned in a plane perpendicular to the flight direction⁶.

$\varepsilon_1, \varepsilon_2$ are the "relativistic anisotropies" of the prompt radiation emitted from fragments with mass M and ($A-M$). In a first order approximation this relativistic anisotropy is $(2+r) v(M)/c$, where $v(M)$ is the velocity of the fragment with mass M and c is the velocity of light. $2 v(M)/c$ is the relativistic solid angle aberration. The correction r takes into account the Doppler shift of the γ -energy^{14,15}. On the one hand the γ -discriminators will accept more quanta, if their energy is increased by an emission from fragments moving towards the γ -detector with the velocity $v(M) \cos \vartheta$. On the other hand the quanta with increased energy have a decreased detector efficiency. Having plastic detectors and discriminator levels of about 100 keV the correction r becomes 0.09 for the measured γ -spectrum of the fission fragments.

f_2 represents the reduction of the initial velocity v_2 of the fragment with mass ($A-M$) to an effective velocity $f_2 v_2$, during the slowing down process in the target backing. Only this reduced velocity is responsible for relativistic aberrations.

A useful expression composed from the coincidence rates W_{12} , Eq. (1), is defined as

$$A_{12}^R \equiv \frac{[W_{12}(\vartheta) - W_{12}(\pi - \vartheta)]}{[W_{12}(\vartheta) + W_{12}(\pi - \vartheta)]}. \quad (2)$$

Inserting into Eq. (2) for each quantity its average value over all masses, indicated by a dash, a mean

reduction factor \bar{f} of the initial velocity may be derived, if the coincidence rates W_{12} have been measured without collimator, that is $\varrho = 1.0$:

$$\bar{f} = 1 - \bar{A}^R \cdot c / \bar{v} \cos \vartheta \quad \text{with } r, \bar{\alpha} \sin^2 \vartheta \ll 1. \quad (3)$$

\bar{f} is related to the mean life time \bar{T} of the prompt radiation. Weighting the velocity distribution $\bar{v}(t)$ of the slowed down fragments with the decay function $p(t)$ of the prompt radiation and integrating over the whole time region the mean velocity causing relativistic aberrations of the stopped fragments is derived. Assuming an exponential decay function with time constant \bar{T} we write:

$$\bar{f} \bar{v} = \frac{1}{\bar{T}} \int_0^\infty e^{-t/\bar{T}} \cdot \bar{v}(t) dt. \quad (4)$$

$\bar{v}(t)$ is determined by a formula given by LINDHARD¹⁶ expressing the specific energy loss dE/dx of heavy ions in materials with density ϱ as a function of the ion energy E :

$$-\frac{1}{\varrho} \frac{dE}{dx} = k \sqrt{E}. \quad (5)$$

Inserting the relation $E = \frac{1}{2} M v^2$ and integrating Eq. (5) we get:

$$\bar{v}(t) = \bar{v}(0) \exp\{-t k \varrho / \sqrt{2M}\}. \quad (6)$$

The time constant $t_c = \sqrt{2M}/k \varrho$ from Eq. (6) is denoted as a characteristic slowing down time. A simple expression for the mean time constant \bar{T} of the prompt radiation is now deduced by inserting Eq. (6) into Eq. (4) and performing the time integration:

$$\bar{T} = t_c \cdot (1 - \bar{f}) / \bar{f}. \quad (7)$$

Once the value of \bar{f} is known from a measurement without collimator, Eq. (2) is used to measure the mean enrichment factor $\bar{\varrho}$ for a measurement with collimator. Inserting again for all quantities mass average values into Eq. (2) we obtain:

$$\bar{\varrho} = \frac{\bar{A}^R}{2(\bar{v}/c)(1 - \bar{f}) \cos \vartheta - \bar{A}^R}. \quad (8)$$

A second important expression, which may be composed from the coincidence rates $W_{12}(\vartheta)$, is defined as:

$$A_{12}^N \equiv \frac{W_{12}(\vartheta) + W_{12}(\pi - \vartheta) - 2 W_{12}(\pi/2)}{W_{12}(\vartheta) + W_{12}(\pi - \vartheta) + 2 W_{12}(\pi/2)}. \quad (9)$$

¹⁶ J. LINDHARD, M. SCHARFF, and M. E. SCHIOTT, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, 10 [1963].

A measurement without collimator ($\varrho = 1.0$) yields:

$$A_{12}^N = \frac{-(G_1 \alpha_1 + G_2 \alpha_2) \cos^2 \vartheta}{G_1 [2 + \alpha_1 (\sin^2 \vartheta + 1)] + G_2 [2 + \alpha_2 (\sin^2 \vartheta + 1)]}. \quad (10)$$

The value of A_{12}^N does not change, if the indices 1 and 2 are commutated. The values of A_{12}^N as a function of the fragment mass must therefore be symmetrical referred to the mass valley. This property may be used to check the correct operation of all devices. This check may easily be performed as high counting rates are available without collimator. Any systematic errors will cause a deviation from this symmetry.

From a measurement with collimator ($\varrho \rightarrow \infty$) we now derive the mass dependent nuclear anisotropy α_1 using Eq. (9)

$$\alpha_1 = \frac{-2 A_{12}^N}{A_{12}^N (\sin^2 \vartheta + 1) + \cos^2 \vartheta}. \quad (11)$$

All relativistic aberrations have cancelled completely. The existence of the only necessary condition $\varrho \rightarrow \infty$ may be checked by Eq. (8).

Concerning the γ -yield finally, a third expression combining the $W_{12}(\vartheta)$ from Eq. (1) shall be discussed.

$$N_{12} \equiv W_{12}(\vartheta) + W_{12}(\pi - \vartheta) + 2 W_{12}(\pi/2). \quad (12)$$

A measurement without collimator ($\varrho = 1.0$) yields:

$$N_{12} = K_{12} \{ G_1 [2 + \alpha_1 (\sin^2 \vartheta + 1)] + G_2 [2 + \alpha_2 (\sin^2 \vartheta + 1)] \}. \quad (13)$$

N_{12} does not change if the indices 1 and 2 are commutated. The values of N_{12} referred to the mass valley must be symmetric, as already stated for Eq. (10).

From a measurement with collimator we now derive the mass dependent yield G_1 of the prompt radiation. If $\varrho \rightarrow \infty$ Eq. (12) gives:

$$G_1 = \frac{N_{12}}{K_{12} [2 + \alpha_1 (\sin^2 \vartheta + 1)]}. \quad (14)$$

The fission yield introduced as a factor in K_{12} , has to be measured in an independent measurement to determine the prompt γ -yield per fragment.

4. Results

4.1. Decay time

The reduction factor \bar{f} of the initial fragment velocity is evaluated from Eq. (3) taking the measured value of $A^R = (2.47 \pm 0.37)\%$ from a

run without collimator, $r = 0.09$ and $\bar{v}/c = 3.75\%$. The thick target causes an energy loss of about 10 MeV of the flying fragments reducing the mean \bar{v}/c -values of 4.65% and 3.15% of the light and heavy fragment group to 4.5% and 3.0%, respectively, giving the above mentioned average (\bar{v}/c)-value of 3.75%. A value of \bar{f} (0.27 ± 0.12) is obtained.

A mean decay constant of $\bar{T} = (4.6_{-1.9}^{+5.0}) \cdot 10^{-12}$ sec is calculated from Eq. (7) with a characteristic slowing down time of $t_c = 1.7 \cdot 10^{-12}$ sec. This value holds¹⁷ for a plastic backing with

$$\bar{k} \cdot \varrho = 0.89 \cdot 10^4 \text{ MeV}^{1/2} \text{ cm}^{-2}.$$

The characteristic slowing down time t_c decreases considerably, if stopping materials with higher atomic numbers are used, for example gold with $t_c = 0.5 \cdot 10^{-12}$ sec.

The mean decay constant \bar{T} of the prompt radiation may also be derived from geometrical considerations¹. Counting the γ -quanta seen through the collimator as a function of the target position a so called "collimator transmission" is measured, depending on the intensity distribution in the vicinity of the target plane as produced by flying fragments. If the collimator transmission is also known for a target where only stopped fragments emit quanta, the mean decay time \bar{T} may be derived by an unfolding program. Assuming an exponential decay function of the prompt radiation the decay constant measured amounts to

$$\bar{T} = (1.1 \pm 0.3) \cdot 10^{-11} \text{ sec}$$

in agreement with the results of JOHANSSON¹. ALBINSON et al.⁴ found three components of the prompt radiation by a similar technique with 7 psec and 50 psec half-life times. A comparison with our result is not possible as the relative yields of the time components have not been published by the authors.

As the decay time derived from relativistic effects by Eq. (7) and Eq. (3) is smaller than all components which were found with mechanical techniques a short-time component may exist, which cannot be resolved by mechanical methods. This short-time component would have a decay constant of $0.8 \cdot 10^{-12}$ sec, if a 30% yield is assumed, corresponding shorter times will result from smaller yields.

¹⁷ J.W. GRÜTER, Diplomarbeit, Technische Hochschule Aachen 1968.

4.2. Enrichment

To get the mass dependent anisotropy α_1 and yield G_1 from Eq. (11) and Eq. (12), respectively, the collimator was set in a position to transmit quanta which were emitted in a time region from 10 psec to 100 psec after fission. The average fragment flight time was 25 psec in this time window. From a two week run in this collimator position the quantity \bar{A}^R , as defined in Eq. (2), and averaged over all masses, was found to be $(6.1 \pm 0.7)\%$. The enrichment was calculated from Eq. (8), giving that $(95 \pm 8)\%$ of the γ -quanta have been emitted from the flying fragments. A sufficient separation capability of the collimator for the radiation of the two complementary fragments has been achieved.

4.3. Mass distribution

Fig. 4 a and Fig. 5 a show the distribution of the "experimental masses M_{ex} " as computed from the raw energy and time-of-flight data using the $E = M_{\text{ex}}/2 \cdot v^2$ law and taking into account the pulse-height-energy defect for surface barrier detectors as first published by SCHMITT et al.¹⁸

The distribution shown is a measurement during a two week reactor period averaging the complete range of time resolutions which arise due to radiation damage of the target. To get the mass resolution of the system the radiochemically measured chain yields¹⁹ were folded with a Gaussian. The best fit to the experimental mass yields from Fig. 4 a was found at a standard deviation of 20 AMU.

4.4. Anisotropy and yield as a function of fragment mass

To check the experimental set up the quantity A^N as defined in Eq. (9) was measured without collimator. Then it cannot be decided which of the two complementary fragments emitted the γ -quanta registered. As a consequence the quantity A^N must be symmetrical to the mass valley as predicted by Eq. (10). This condition is fulfilled, as demonstrated by Fig. 4 b showing the mass dependent A^N . From the average of A^N an average anisotropy $\bar{\alpha} = (13.0 \pm 1.0)\%$ is evaluated in agreement with

results found by other authors^{12, 13, 15, 20}. They all measured without collimator, thus observing the whole deexcitation time of the prompt radiation. Moreover, the anisotropy as a function of the mass ratio of the fragments may be derived from Fig. 4 b. A comparison with similar measurements from IVANOV et al.^{3, 21} shows good agreement within the statistical errors.

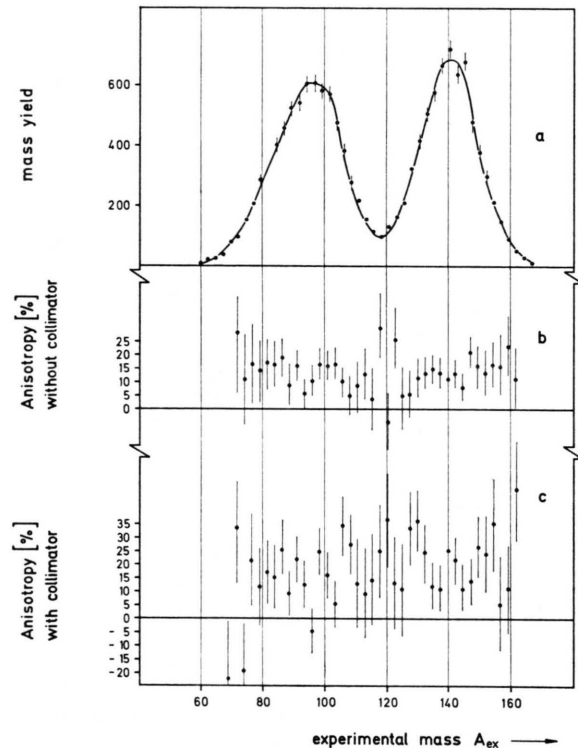


Fig. 4. Anisotropy of the prompt radiation as a function of fragment mass A_{ex} : (a) mass yield $\eta(A_{\text{ex}})$ from a two week measurement, (b) anisotropy without collimator showing the symmetry referred to the mass valley as demanded from theoretical considerations, (c) anisotropy with collimator selecting γ -quanta in the time region (10–100) psec after fission.

To get the mass dependent anisotropy α_1 from Eq. (11) no changes of the experimental set-up have to be made except to shift the collimator plates into the position characterized already in 4.2. The measured A_{12}^N have been inserted into Eq. (11) giving the values shown in Fig. 4 c. Within the statistical error no dependence of the anisotropy on

¹⁸ H. W. SCHMITT, W. M. GIBSON, J. H. NEILER, F. J. WALTER, and T. D. THOMAS, in: *Physics and Chemistry of Fission* (Proc. Symp. Salzburg 1965) 1, IAEA Vienna 1966, p. 531.

¹⁹ H. FARRAR and R. H. TOMLINSON, *Nucl. Phys.* **34**, 367 [1962].

²⁰ G. A. PETROV, *JETP* **20**, 1387 [1965].

²¹ O. I. IVANOV, Y. A. KUSHNIR, and G. N. SMIRENKIN, *Sov. J. Nucl. Phys.* **8**, 29 [1969].

the fragment mass is evident. A mean anisotropy $\bar{a} = (17.0 \pm 1.8)\%$ is evaluated from Fig. 4 c. This value holds for a time region from 10 psec to 100 psec after fission. It is larger than the anisotropy $\bar{a} = (13.0 \pm 1)\%$ found without collimator. The anisotropy increases slightly, and decreases definitely not up to emission times of 100 psec.

The data used to obtain the anisotropy could be used to calculate the yield of the prompt radiation from Eq. (13) and Eq. (14) if K_{12} would be known. As K_{12} has not been measured in these runs, the yield measurements of the prompt radiation as a function of mass have been performed separately. The experimental set up from Fig. 1 has been used similarly as done before by JOHANSSON¹ in his measurement of the mass dependent yield of ²⁵²Cf. The yield is obtained by dividing corresponding mass yields from two normalized mass distributions, one measured in coincidence with the prompt radiation, and another measured without detecting the γ -radiation. To guard against the influence of time and temperature dependent variations of the mass calibration, the measurements of the two mass distribution have been successively repeated in one hour intervals. To prevent a drift of the electronic levels due to different counting rates, both mass distributions have been measured with the same counting rates. In order to obtain equal rates the flux in the neutron beam was adjusted by thin boron carbide plates.

As done with the anisotropy measurement a check run was made without collimator. The radiation from complementary fragments cannot be distinguished. Hence the yield must be symmetrical referred to the mass valley as shown in Fig. 5 b. Most systematic errors for example, different mass scales of the two mass distributions to be divided, would give a deviation from this symmetry. To get the mass dependent γ -yield a run was made with collimator as described in 4.2. The result is shown in Fig. 5 c. A saw-tooth slope as measured by JOHANSSON¹ for ²⁵²Cf-fission products holds also for ²³⁵U-fragments, a result obtained before independently by ^{2, 3, 4} with much better mass resolution and statistics.

The summarized results of this chapter are presented in Table 1 after having reduced the data from Fig. 4 and Fig. 5 to five values for each mass group. Processing the mass resolution of the system as described in 4.3 the contribution of each radio-

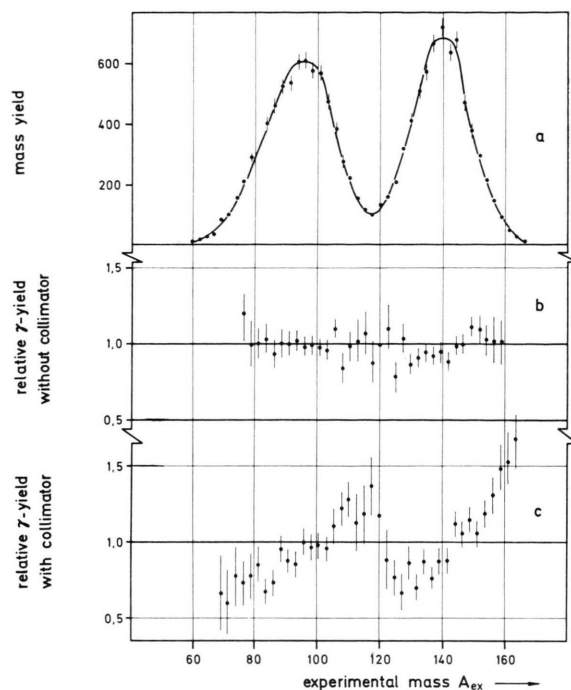


Fig. 5. Yield of the prompt γ -radiation as a function of fragment mass A_{ex} : (a) mass yield $\eta(A_{ex})$ from a two week measurement, (b) γ -yield without collimator showing the symmetry referred to the mass valley as demanded from theoretical considerations, (c) γ -yield with collimator selecting γ -quanta in the time region (10–100) psec after scission.

chemical mass M_r to the experimental mass M_{ex} can be evaluated due to its yield $\eta(M_r)$ and distance $(M_r - M_{ex})$ from the experimental value. The mean value \bar{M} from all radiochemical masses M_r which so contribute to a certain mass M_{ex} is the only physically meaningful mass. It is given in Table 1. The mass yield from Fig. 4 a is constricted to a small mass range of 10 AMU for each mass group. In a second step the raw data of anisotropy and yield have been fitted to Chebychev-polynomials, which are in this case superior to power functions. The mean values of these functions are presented in Table 1 for each of the corresponding mass regions. Yield and anisotropy do not depend on the mass ratio of the fragments, whereas the yield shows a strong increase and the anisotropy a weak increase within both the fission product groups.

4.5. Anisotropy and yield as a function of γ -energy

The yield of the prompt radiation depending on the γ -energy is shown in Fig. 6 a and Fig. 6 b for measurements with and without collimator, respectively. No difference is observed between the spectra.

Table 1.

Mass region		89.5— —91.5	91.5— —93.5	93.5— —95.5	95.5— —97.5	97.5— —99.5	184— —136	136— —138	138— —140	140— —142	142— —144	80— —150
Anisotropy %	without collimator $0 < t < \infty$	13.0 ± 1.4	13.0 ± 1.3	13.0 ± 1.2	13.0 ± 1.1	13.0 ± 1.0	13.0 ± 1.0	13.0 ± 1.1	13.0 ± 1.2	13.0 ± 1.3	13.0 ± 1.4	13.0 ± 1.0
	with collimator ($10 < t < 100$) ps	12.0 ± 1.9	13.2 ± 1.8	14.0 ± 1.7	14.8 ± 1.6	15.6 ± 1.5	17.9 ± 1.5	18.7 ± 1.6	19.5 ± 1.7	20.3 ± 1.8	21.1 ± 1.9	17.0 ± 1.5
Relative yield	without collimator $0 < t < \infty$	0.99 ± 0.015	1.02 ± 0.014	1.01 ± 0.014	0.98 ± 0.013	0.95 ± 0.013	0.87 ± 0.013	0.86 ± 0.013	0.88 ± 0.014	0.95 ± 0.014	1.06 ± 0.015	1.0 ± 0.013
	with collimator ($10 < t < 100$) ps	0.71 ± 0.019	0.76 ± 0.018	0.92 ± 0.018	1.06 ± 0.017	1.22 ± 0.017	0.73 ± 0.017	0.82 ± 0.017	1.00 ± 0.018	1.13 ± 0.018	1.42 ± 0.019	1.0 ± 0.017
Primary spin [\hbar]		5.0	5.3	6.5	7.3	8.6	5.1	5.7	7.0	7.9	10.0	7.0

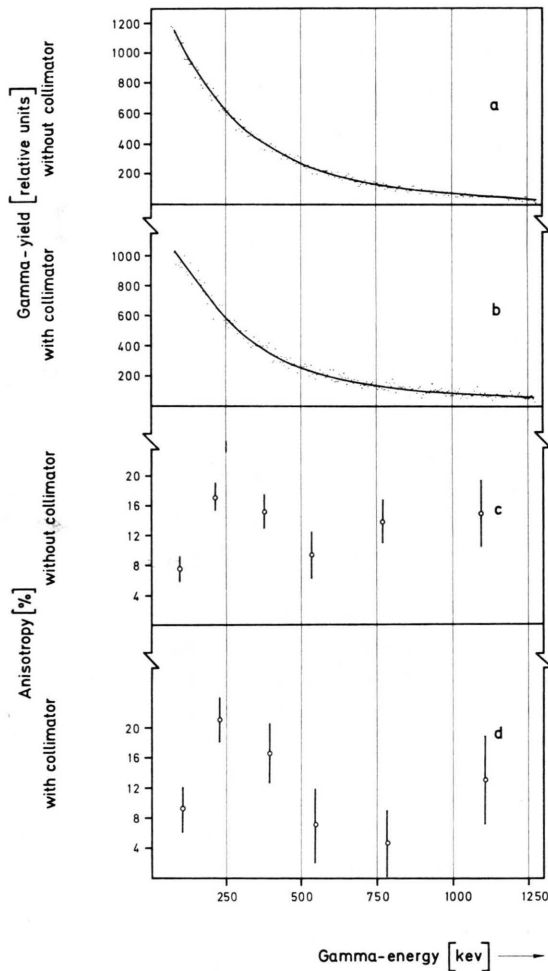


Fig. 6. Yield and anisotropy of the prompt radiation as a function of γ -energy: (a) yield without collimator, (b) yield with collimator selecting quanta in the time region (10–100) psec after scission, (c) anisotropy without collimator, (d) anisotropy with collimator selecting quanta in the time region (10–100) psec after scission.

The energy calibration of the plastic detectors has been made using the Compton edges of various sources i.e. ^{60}Co and ^{57}Co as calibration points.

The anisotropy α as a function of the γ -energy has been calculated from Eq. (10) and Eq. (11) by evaluating the quantity A^N not as a function of mass but as a function of γ -energy from the measured quantities $W(\vartheta, E\gamma, A, t)$ of Eq. (1). The result is presented in Fig. 6c and Fig. 6d showing the anisotropy as derived from measurements with and without collimator, respectively. The processed data are the same as in section 4.4. A maximum of the anisotropy exists for medium energies as already observed by PETROV²², who measured without collimator and detected the γ -radiation with a NaJ(Tl)-crystal. Furthermore we observe from the measurement without collimator that the anisotropy is larger for low and medium energies, compared to the measurements with collimator. This difference is due to our time window which suppresses quanta emitted immediately (< 10 psec) and very late (> 100 psec) after fission.

5. Discussion

5.1. Deexcitation of fission fragments

STRUTINSKY⁶ applied a statistical deexcitation mechanism, successfully used to describe the deexcitation after neutron capture, to fission fragments. The level density calculated from a Fermi gas model is assumed to govern the emission of γ -quanta from the fragments. Predictions made by the model concerning different aspects of prompt γ -emission may be tested by suitable experiments.

²² G. A. PETROV, Sov. J. Nucl. Phys. **1**, 338 [1965].

The multiplicity of the radiation emitted should be small (2–3), the average quantum energy large. Dipole radiation with life times smaller than 10^{-12} sec should strongly contribute to the total γ -spectrum.

It has been pointed out by several authors^{1, 23–25} that these predictions are not fulfilled. The average quantum energy is appreciably smaller than found after neutron capture, the multiplicity of the radiation larger. Life times below 10^{-12} sec characteristic for E1 radiation have been excluded by a Doppler shift measurement in Pt by SKARSVAG¹⁵ and the preponderance of γ -emission in the fission axis cannot be explained by dipole radiation⁶. Mainly quadrupole radiation is emitted promptly after fission.

Our results fully confirm these conclusions. The average quantum energy, as estimated from Fig. 6 a, amounts to 0.8 MeV. The shortest life times according to our time measurements amount to about 10^{-12} sec. The energy spectrum measurement without collimator including the shortest life times does not differ from the spectrum seen through the collimator. The spectrum in the time range 10^{-11} sec, where the bulk of all γ -quanta is emitted, seems to be the same as the spectrum including the shortest life times components. A strong contribution from E1-radiation should show up in the spectrum at the high energy side. No contribution is observed. Our life time and γ -energy measurements exclude the postulated strong E1-contribution. Moreover, we confirm the mean value found for the anisotropy +13%, which excludes E1-radiation. A statistical deexcitation reduces the spin of the fragments continuously leading according to STRUTINSKY⁶ to an anisotropy which decreases continuously in time. Our measurements of the mean anisotropy with and without collimator however, show an increase of the anisotropy for γ -quanta emitted in the time window defined by the collimator, that is for times which are definitely later than the emission times observed without collimator.

Within the statistical model the anisotropy should not depend on the quantum energy. Starting from a

given initial spin value all γ -transitions of the same multipolarity should have the same anisotropy. From Fig. 6 b and 6 c a pronounced peak of the anisotropy values for small quantum energies may be seen, an observation already reported by PETROV²². As dipole radiation as a major source of γ -emission has been excluded by the life time and γ -spectra arguments the variation of anisotropy with γ -energy is a further experimental fact against the predictions of the statistical deexcitation model.

There are no experimental facts which support the hypothesis that fission fragments deexcite statistically. A calculation of the initial spin values from the measured anisotropy values, applying an often used formula given by STRUTINSKY⁶, seems to be doubtful, as all other qualitative predictions of the model have not been confirmed experimentally. Using realistic values of the spin cut-off parameter²⁶ as has been done in¹⁰ and¹¹, leads to spin values of the fragments of $16\hbar$, which are much too high, as will be seen later in the discussion.

JOHANSSON¹ proposed a deexcitation of the fragments by a cascade of collective transitions. The arguments given in his paper are confirmed and supported by the results found in our experiments. The strong contribution of low energy γ -quanta (<500 keV) to the energy spectrum and the life times of the radiation between ($10^{-12} - 10^{-10}$) sec point to collective E2 transitions.

Collective E2 transitions are emitted predominantly in stretched cascades. The anisotropy is larger for the later low energy members of a collective E2 cascade than for the earlier high energy members²⁷. The increase of the mean anisotropy in our measurement with collimator may thus be understood. The lower the energy of the collective transitions, the larger their anisotropy, a trend which may be seen from Fig. 6. The life time, the average quantum energy, the time dependence of the mean anisotropy, the dependence of the anisotropy on the γ -energy, all these facts fit into the picture of a deexcitation by a collective E2 cascade.

No simple expression connecting the measured values of the anisotropy and the primary spin

²³ S. S. KAPOOR and R. RAMANNA, *Phys. Rev.* **133 B**, 598 [1964].

²⁴ S. A. E. JOHANSSON, P. KLEINHEINZ, and K. SIEGBAHN, *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, North-Holland Publ. Comp., Amsterdam 1965, p. 805.

²⁵ H. MAIER-LEIBNITZ, P. ARMBRUSTER, and H. J. SPECHT, in: *Physics and Chemistry of Fission* (Proc. Symp. Salzburg 1965) **2**, IAEA Vienna 1966, p. 113.

²⁶ U. FACCHINI and E. SAETTA-MENICHELLA, *Energia Nucleare* **15**, 54 [1968].

²⁷ J. O. NEWTON, F. S. STEPHENS, R. M. DIAMOND, K. KOTAJIMA, and E. MATTHIAS, *Nucl. Phys. A* **95**, 357 [1967].

exists. As long as the deexcitation mode does not vary strongly with the mass or the mass ratio of the fragments, no mass dependence of the anisotropy is to be expected. Indeed only a very weak dependence can be read from our results, see Fig. 4 and Table 1. This constancy of the anisotropy thus gives further support to the proposed interpretation.

The fission fragments mainly deexcite by emission of E2 quanta from a stretched collective cascade. These quanta are seen through our collimator. The mass dependence of the γ -yield observed through the collimator, Fig. 5, is the mass dependence of E2-quanta, with each quantum reducing the initial spin by about the maximum possible amount of $2\hbar$. The bulk of all quanta is emitted in the time window observed. The small number of quanta emitted earlier either is distributed as the main contribution observed or, if emitted statistically, reduces the initial spin only very little ($\sim 0.3\hbar$). The number of γ -quanta observed reflects the mass dependence of the initial spin. Upper values are calculated from:

$$I \sim 2\hbar \cdot G. \quad (15)$$

Mean values of the mean initial spin are lower and will be estimated in the following. Neutrons and early statistical γ -quanta reduce the spin of the fragment by about $1\hbar$. The average number of prompt quanta emitted per fragment amounts²⁵ to 4. An upper limit of $(8-9)\hbar$ follows for the mean primary spin of a fragment. The spin actually may be somewhat smaller as the cascade may only be partly stretched. Values between $(6-8)\hbar$ for the mean initial spin seem most realistic. In agreement with earlier conclusions by THOMAS and GROVER⁵ who obtained in their analysis $8.4\hbar$ and HUIZENGA and VANDENBOSCH²⁸ who calculated from isomer ratios $8.0\hbar$. Table 1 gives with $\bar{l} = 7\hbar$, using our data from Fig. 5, spin values in the different mass regions. The spin increases in both fission product groups from values of about $\sim 5\hbar$ to $\sim 10\hbar$.

To summarize the conclusions from the data presented:

a) The deexcitation mechanism of fission fragments is not governed by the level density following from a statistical model.

b) The fragments are mainly deexcited by a cascade of stretched E2-transitions of collective character.

c) The initial spin of the fragment remarkably depends on the mass value of the fragment. It strongly increases in both fission fragments. The spins of two complementary fragments may be different by factors of about two.

5.2. Scission of the nucleus and initial spin

The anisotropy of γ -emission follows from an alignment of the initial spins. The preponderance of emission of E2-radiation in the fission axis tells us the spins to be aligned perpendicular to the fission axis. All torques producing these spins must act perpendicular to the fission axis. The breaking of the nucleus is a process selecting states with $m=0$ in contradiction with the fundamental assumption of the statistical model of fission introduced by FONG²⁹. In this model the calculated level density of final states in the fragments calculated assuming statistical equilibrium of all states, determines the probability of finding a specific pair of complementary fragments. A preference of $m=0$ states is a violation of the fundamental assumption of the model. The statistical model of fission, moreover, predicts a ratio of the spins of complementary fragments amounting to $I_1/I_2 = V(\Theta_1/\Theta_2)$, with Θ = moment of inertia of the nascent fragments. The model demands values of the excitation energy at scission which make the moment of inertia approach its rigid body value. Light and heavy fragments should have values of the initial spin differing by a factor $(M_1/M_2)^{5/6}$. As the mean number of γ -quanta emitted from the light and heavy group is found to be equal within 10%, Fig. 5, the mean initial spins of the group, following our interpretation of the deexcitation of fission fragments above, will be equal, too, in contradiction with the statistical model of fission. The large increase of the spin within each fission product group cannot be explained by a corresponding increase of the rigid body moment of inertia.

The experimental results on prompt γ -emission do not support the statistical model of fission. The excitation energy of the fragments at scission may

²⁸ I. R. HUIZENGA and R. VANDENBOSCH, Phys. Rev. **120**, 1305 [1960].

²⁹ P. FONG, Phys. Rev. **102**, 434 [1956].

not be high enough and may not be distributed statistically.

If the excitation energy of the fragments at scission is low and concentrated in the main collective excitations, fission may be described applying the laws of statistical mechanics to these degrees of freedom, NIX and SWIATECKI³⁰, RASMUSSEN, NÖRENBERG, and MANG³¹ using an adiabatic model of scission only consider the lowest mode of collective excitation, the bending mode. They find in agreement with a result also obtained by Nix at low temperatures an alignment of the spins perpendicular to the fission axis and a ratio of the initial spins:

$$\frac{I_1}{I_2} = \left(\frac{\Theta_1 \cdot K_1}{\Theta_2 \cdot K_2} \right)^{1/4} \sim \left(\frac{\varepsilon_1}{\varepsilon_2} \right)^{3/4} \cdot \left(\frac{M_1}{M_2} \right)^{5/12}. \quad (16)$$

K is the stiffness constant of the bending mode. It is according to³¹ proportional to the deformation ε of the fragment. At low excitation energies the moment of inertia approaches its irritational fluid flow value, which is proportional to ε^2 . The mean value of the initial spins assuming equal deformations in both fission product groups differ by a factor of only 1.2. Such a difference is within the accuracy of our estimate of the spin values, Table 1.

The mass dependence of the initial spin of the fragments is reduced to the mass dependence of the deformation of the fragments. The deformations of the fragments have been calculated with different degree of sophistication in various adiabatic models⁶⁻⁹. They are determined by the nuclear structure of the fragments, mainly by the stiffness parameters against deformation c . The deformation energies of two complementary fragments are given by:

$$\frac{D_1}{D_2} = \frac{c_1 \varepsilon_1^2}{c_2 \varepsilon_2^2} = \frac{\varepsilon_1}{\varepsilon_2} \quad \text{with} \quad c \sim \varepsilon^{-1}. \quad (17)$$

A correlation tied by the deformations of the fragments between initialspin I of the fragments and

their deformation energy D follows with Eq. (16):

$$\frac{I_1}{I_2} = \left(\frac{D_1}{D_2} \right)^{3/4} \cdot \left(\frac{M_1}{M_2} \right)^{5/12}. \quad (18)$$

An analysis of a multiparameter measurement of nuclear charge Z_1 , kinetic energy E_k and neutron number ν by NIFENECKER³² leads to a positive correlation between deformation energy D and total energy E_γ emitted by γ -quanta. This correlation experimentally confirms Eq. (18), if the initial spin and the total γ -energy are assumed to be correlated.

JOHN et al.³³ pointed out the saw-tooth like mass dependence of both the number of γ -quanta and neutrons. If a proportionality between the number of neutrons and the deformation energy, and between the number of γ -quanta and the initial spin [Eq. (15)] is assumed, John's observation finds a simple explanation by Eq. (19). It follows:

$$G_1/G_2 \sim (\nu_1/\nu_2)^{3/4}. \quad (19)$$

To summarize the conclusions from our experiments concerning scission

a) The statistical model of fission is not supported by an analysis of data on prompt γ -emission.

b) Alignment and mass dependence of the initial spins are explained by models which assume low excitation energies at scission and a predominant excitation of collective degrees of freedom. Calculations of NIX et al.³⁰ and RASMUSSEN et al.³¹ are supported by the analysis.

c) A correlation between the initial spin and the deformation energies of complementary fragments exists. The deformation of the fragments ties a link between both these properties. An explanation of the striking observation of similar mass dependences of the number of prompt neutrons and γ -quanta follows.

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³⁰ J. R. NIX and W. J. SWIATECKI, Nucl. Phys. **71**, 1 [1965].

³¹ J. O. RASMUSSEN, W. NÖRENBERG, and H. J. MANG, Nucl. Phys. A **136**, 465 [1969].

³² H. NIFENECKER, Thesis, CEA Saclay 1970.

³³ W. JOHN, J. J. WESOLOWSKI, and F. GUY, Phys. Rev. **127**, 880 [1962].