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Independent isomeric yield ratio of ^{148}Pm in fission of the moderately excited ^{236}U compound nucleus as a measure of fragment angular momentum

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The independent yield ratio of 41.3-day ^{148}Pm^{m} (6−) and 5.37-day ^{148}Pm^{g} (1−) has been measured radiochemically for fission of ^{233}U and ^{235}U induced by thermal neutrons and for fission of ^{232}Th by ^{4}He ions of 26–42 MeV. The values of the ^{148}Pm isomer ratios were 3.0 ± 0.5 and $2.6^{+0.9}_{-0.5}$ for the thermal-neutron-induced fission of ^{233}U and ^{235}U, respectively, and 6.8 ± 1.2, 6.6 ± 0.7, 5.3 ± 0.5, 8.5 ± 0.9, and 8.5 ± 0.9 for fission of ^{232}Th by ^{4}He ions of 25.8, 29.1, 31.8, 37.0, and 41.4 MeV, respectively. On the basis of the measured independent yield ratios the average intrinsic angular momentum of the primary fission fragments (which lead by the emission of prompt neutrons and γ rays to the secondary fragment ^{148}Pm) was estimated by means of a simple statistical-model analysis based on the formalism developed by Huizenga and Vandenbosch. The following values for the average intrinsic spin of the primary fragment were obtained: $(10.8 \pm 0.9)\hbar$ and $(10.0^{+1.6}_{-1.1})\hbar$ for thermal-neutron-induced fission of ^{233}U and ^{235}U, respectively, and $(16.5 \pm 1.5)\hbar$, $(16.3 \pm 0.9)\hbar$, $(14.6 \pm 0.7)\hbar$, $(19.9 \pm 1.1)\hbar$, and $(19.9 \pm 1.1)\hbar$ for fission of ^{232}Th by ^{4}He ions of 25.8, 29.1, 31.8, 37.0, and 41.4 MeV, respectively. The results show that a fraction of the angular momentum of the fissioning nucleus appears as intrinsic spin of the primary fragments when the angular momentum of the fissioning nucleus is increased as in the case of the ^{4}He-ion-induced fission. A summary of all published independent isomer-yield ratio studies which were used for deducing fission fragment angular momenta is given.

[NUCLEAR REACTIONS, FISSION ^{233,235}U(n,f), E=thermal, ^{232}Th$(^{4}$He$,f)$, E= 25.8–41.4 MeV; measured ^{148}Pm independent isomer yields; deduced primary angular momenta.]

I. INTRODUCTION

Determination of the angular momentum distribution of primary fission fragments provides us with information about the properties of the fissioning nucleus between the saddle point configuration and the state shortly after the scission configuration. Information about the angular momentum of the primary fission fragment can be obtained by measurements of the number of γ rays emitted by the fission fragments[1] and by measurements of the anisotropy together with the yield of the γ radiation.[2] Measurements of the relative population of the levels of the ground-state band from the γ intensities in even-even fission products have been applied for the determination of the angular momentum of fission fragments.[3] A further method providing information about the angular momentum of primary fission fragments is based on the radiochemical determination of independent isomeric ratios of fission products, i.e., of the ratio of the independent fission yield of a long-lived isomer of a fission product to the independent fission yield of the ground state of the same fission product. The independent portion of the fission product yields arises directly from the primary fission fragments by emission of prompt neutrons and γ rays: hence, the relative population of the ground state and isomeric state (which have considerably different spin) is related to the initial angular momentum of the fission fragments. The isomeric pair must have a stable or long-lived precursor so that there is either no contribution or, at worst, only a small correctable contribution from β decay of its precursor.

Unfortunately there are only a few isomeric pairs among fission products which are shielded and which have half-lives suitable for radiochemical work. Most of the isomer-yield ratio studies reported in the literature have been carried out for fission at moderate excitation energy induced by energetic charged particles. Since the most probable charge of a mass is shifted nearer to β stability with increasing excitation energy, the independent yields of the shielded fission products are higher and consequently easier to determine. These measurements have been carried out with isomeric pairs of ^{95}Nb,[4,10,11] ^{133}Xe,[5] ^{134}Cs,[6-9,12,14] ^{90}Y,[13] ^{91}Y,[13] ^{131}Te,[15,50] ^{133}Te,[15,50] ^{130}I,[50] ^{132}I,[50] and ^{134}I.[50] Only a few measurements of independent isomer-yield ratios in low energy fission have been reported. The isomer ratio of ^{134}Cs was studied in the fission of ^{233}U induced by γ rays with an energy of up to 16 MeV.[14] The ratios of the independent yields of the isomers ^{131}Te^{m,g},[15,16] ^{133}Te^{m,g},[15,16] ^{132}Sb^{m,g},[16] ^{130}Sb^{m,g},[16] and ^{128}Sb^{m,g}[16]

were measured in thermal-neutron-induced fission of ^{235}U,[15,16] ^{233}U,[16] and ^{239}Pu.[16] The independent isomeric-yield ratio of ^{83}Se has been estimated in the fission of ^{239}Pu by thermal neutrons.[18]

In this work the independent yield ratio ^{148}Pm^{m}(6−)/^{148}Pm^{g}(1−) was determined radiochemically in thermal-neutron fission of ^{235}U and ^{233}U and in fission of ^{232}Th with ^{4}He-ion particles of energies 26–42 MeV. The two reactions ^{235}U(n_{th},f) and ^{232}Th(^{4}He,f) lead to the same compound system ^{236}U*. Excitation energy and angular momentum of the compound nucleus ^{236}U increase from the thermal-neutron-induced reaction ^{235}U(n,f) to the ^{4}He-ion-induced reaction ^{232}Th(^{4}He,f) and with increasing energy of the ^{4}He particles. Therefore a detailed study could be made of the dependence of the isomeric-yield ratio of ^{148}Pm on excitation energy and angular momentum of the compound system ^{236}U*.

The average initial angular momentum distribution of the primary fission fragments was deduced from the measured relative yields of the ^{148}Pm isomers by calculations based on the concepts of the statistical model. The formalism of these calculations was introduced by Huizenga and Vandenbosch[19] for the calculation of isomeric ratios in nonfission reactions where the angular momentum distribution of the compound nuclei can be calculated. Warhanek and Vandenbosch[14] used this formalism to obtain information about the intrinsic angular momentum of the primary fission fragments from the isomeric ratios of fission products. One asks for the angular momentum distribution which must occur in the primary fission fragments in order to explain the observed isomeric ratio. This spin distribution is modified by the neutrons and γ quanta emitted by the primary fragments which remove angular momentum from the system. Therefore knowledge of the angular momentum carried away by the prompt neutrons and γ rays is required.

An important quantity in the statistical-model calculations is the spin cutoff parameter σ. One method to gain basic information on the value of σ is the measurement of isomer ratios in nonfission reactions.[19] A better spin cutoff factor for the fission calculations might be obtained if the deexcitation process of the primary fission fragments forming the fission products under consideration is compared with the deexcitation process of the same nuclei produced through nonfission reactions. The fission product nuclei ^{148}Pm^{m} and ^{148}Pm^{g} are probably formed mainly by neutron and γ-ray emission of the primary fragments ^{149}Pm and ^{150}Pm. We have produced the same nuclei with similar excitation energies and angular momenta through the reactions ^{148}Nd(p,n)^{148}Pm^{m,g} and ^{148}Nd(d, $2n$)^{148}Pm^{m,g}.[20] Compound nuclei ^{149}Pm and ^{150}Pm, respectively, are formed in the two reactions, at least at low excitation energies. The initial spin distribution of these nuclei can be calculated when they are formed through these reactions, but not when they are formed as fission fragments. Reproducing the experimentally determined isomer ratio ^{148}Pm^{m}/^{148}Pm^{g} resulting from the ^{148}Nd(p, n) ^{148}Pm^{m,g} and ^{148}Nd(d, $2n$) ^{148}Pm^{m,g} reactions by means of statistical-model calculations results in spin cutoff parameters which were then used in the fission calculations describing the deexcitation of the same nuclei, but this time as fission fragments. In this way the results might become less dependent on specific assumptions made in the calculations.

II. EXPERIMENTAL PROCEDURES

Only a brief outline of the experimental procedure is given here. A full description is published elsewhere[21,22] along with the independent fission yields of ^{148}Pm^{m} and ^{148}Pm^{g} in thermal-neutron-induced fission of ^{233}U and ^{235}U and their formation cross sections at various ^{4}He-ion energies in the reaction ^{232}Th(^{4}He,f).

Targets of 3–5 g of enriched ^{235}U (90%) as uranium metal and 0.1 g of ^{233}U (99.7%) as U_3O_8 were irradiated for approximately 35–40 min in the graphite reflector of the Munich research reactor in a thermal-neutron flux of approximately 1.1–1.8×10^{13} cm^{-2} sec^{-1}. For the ^{4}He irradiations foils of thorium metal of thickness 111 or 24.6 mg cm^{-2} were bombarded for 10–20 h in the internal circulating ^{4}He beam of the Karlsruhe isochronous cyclotron. The reduction of the energy of the ^{4}He ions was accomplished by irradiating at a radius corresponding to the energy desired. The target assembly consisted of a target holder on a Faraday cup which contained the target foil. This Faraday cup was used to measure the current of the ^{4}He ions on the target so that absolute cross sections as well as isomer ratios could be determined. Typical beam currents were about 10–20 μA. The mean energy of the ^{4}He ions was calculated taking into consideration the energy loss in the target.

After the irradiations the targets were dissolved and the chemical separation of Pm was carried out as described in Refs. 21 and 22. The activities of the 41.3-day ^{148}Pm^{m} and 5.37-day ^{148}Pm^{g} were measured with a Ge(Li) detector with a 16.9% photopeak efficiency relative to a 7.6- × 7.6-cm NaI(Tl) crystal and a resolution of 2.1 keV for the 1.33-MeV γ ray of ^{60}Co. The detector had to be set up in a 20-cm thick lead cave because the natural ^{40}K background had to be reduced because of the low fission yields of the ^{148}Pm isomers. The γ lines used to measure the activities were

^{148}Pm^m, 629.9 and 725.6 keV[23] and ^{148}Pm^g, 1465.1 keV.[23]

The measured activities were corrected for branching ratios and counter efficiency and were used to calculate the isomer ratios. In neutron-induced fission of ^{235}U both isomers can be formed by the secondary reaction $^{147}\mathrm{Pm}(n,\gamma)^{148}\mathrm{Pm}^{m,g}$. The contribution of the secondary reaction was kept low by using short irradiation times and corrected as described in Ref. 21. The errors in the isomer ratios have been compounded from the statistical errors in the activity measurements, the uncertainties in the decay schemes, and counter efficiencies. In the error of the isomer ratio in fission of ^{235}U the uncertainty of the correction for the secondary reaction was also taken into consideration.

III. EXPERIMENTAL RESULTS

The isomer ratios of ^{148}Pm computed from the experimental data are given in Table I. The ^{4}He-ion bombarding energy given in this table refers to an average energy of the ^{4}He ions in the target. This average energy was calculated assuming a linear energy loss in the thorium foils and weighting the energy of the ^{4}He ions in different layers of the foils by the cross section for fission of ^{232}Th with ^{4}He ions[25] at this energy. The energy loss of the ^{4}He ions in the thorium foils was determined from Ref. 24.

The errors in the average ^{4}He-ion energies were estimated from the uncertainties in the energy of the ^{4}He ions before entering the thorium foils, the energy loss in the foil, and the cross section for the $^{232}\mathrm{Th}(\alpha,f)$ reaction.

IV. INTERPRETATION OF RESULTS

The experimental isomer ratios have been interpreted in terms of the initial primary fission fragment angular momentum using the method of Huizenga and Vandenbosch.[19] As suggested by Warhanek and Vandenbosch[14] the probability distribution of initial angular momentum states of the primary fragments is assumed to be represented by

$$P(J_i) \propto (2J_i+1)\exp[-J_i(J_i+1)/B^2], \tag{1}$$

where $P(J_i)$ is the probability distribution for each spin value J_i and B is a parameter which defines the width of the distribution. The root-mean-square angular momentum $(\overline{J^2})^{1/2}$ of the primary fragments is equal to B for large values of B:

$$(\overline{J^2})^{1/2} \simeq B.$$

The method of the calculation has been described extensively elsewhere.[14,15,19,26] Therefore only a brief outline of the various steps of the procedure is given here together with a discussion of the parameters used in the calculation.

The initial spin distribution of the primary fragments is modified by the emission of the prompt neutrons which carry away some angular momentum. These transitions from a specific spin level are assumed to populate residual spin levels with a probability which depends on the availability of the specific levels and given by

$$P(J) \propto (2J+1)\exp[-(J+\tfrac{1}{2})^2/2\sigma^2], \tag{2}$$

where $P(J)$ is the probability distribution of levels with spin J and σ is the spin cutoff parameter which characterizes the angular momentum distribution of the level density and is related to the moment of inertia and the temperature of the excited nucleus.

After emission of the prompt neutrons the residual nucleus deexcites by the emission of γ rays. The relative probabilities for a nucleus of spin J_i to decay to states with spin J_f are again determined by the level density factor as given in Eq. (2). The distribution obtained after the statistical emission of γ rays was assumed to divide between the two isomeric states so that the transition between the

TABLE I. Independent isomer-yield ratios of ^{148}Pm in thermal-neutron-induced fission of ^{233}U and ^{235}U and ^{4}He-ion-induced fission of ^{232}Th.

Target	Projectile	Average projectile energy (MeV)	Number of measurements	Excitation energy of compound nucleus (MeV)	$\frac{^{148}\mathrm{Pm}^m(6-)}{^{148}\mathrm{Pm}^g(1-)}$
^{233}U	n	Thermal	1	6.8	3.0 ± 0.5
^{235}U	n	Thermal	4	6.5	$2.6^{+0.9}_{-0.5}$
^{232}Th	^{4}He	25.8 ± 1.7	2	20.8 ± 1.7	6.8 ± 1.2
^{232}Th	^{4}He	29.1 ± 1.4	1	23.9 ± 1.4	6.6 ± 0.7
^{232}Th	^{4}He	31.8 ± 1.5	1	26.7 ± 1.5	5.3 ± 0.5
^{232}Th	^{4}He	37.0 ± 1.0	1	31.8 ± 1.0	8.5 ± 0.9
^{232}Th	^{4}He	41.4 ± 1.0	1	36.1 ± 1.0	8.5 ± 0.9

initial and final nuclei involves the smaller spin change. Because there exists a $J=2$ state between the $J=1$ ground state and $J=6$ metastable state it was assumed that states with $J\leq 3$ populate the ground state, all states with $J\geq 5$ populate the metastable state, and states with $J=4$ divide equally between the isomeric and ground states.

Therefore, for the calculation of the angular momentum distribution of the fission fragments after the emission of the prompt neutrons and γ rays one has to know: (a) the number, energy, and transmission coefficients of the emitted neutrons; (b) the number, energy, and multipolarity of the emitted γ rays; and (c) the spin cutoff parameters.

Prompt neutrons and γ rays

The average number of neutrons $\bar{\nu}$ emitted per fragment and their kinetic energy as a function of primary fragment mass have been measured for thermal-neutron-induced fission of ^{233}U and ^{235}U.[27-29] But one requires $\bar{\nu}$ versus the post-neutron emission mass ($\bar{\nu}_p$) to learn which primary masses give rise to the measured fission product mass. For this purpose one has to assume that the fragments with all possible charge splits for a given isobar chain lead, on the average, to the same number of neutrons in the deexcitation process. The above mentioned measurements average over the most probable charge splits for a given primary mass split. Wahl *et al.*[30] and Musgrove, Cook, and Trimble[31] have estimated the average number of neutrons $\bar{\nu}_p$ emitted in forming products of a given mass number as a function of the post-neutron emission mass. These evaluations give the average number of neutrons emitted in forming products of mass 148 as 1.69[31] (1.62[30]) and 1.66[31] for thermal-neutron-induced fission of ^{235}U and ^{233}U, respectively. Thus, taking a value of $\bar{\nu}_p=1.65$ for both fissioning nuclei, fission products of mass 148 are formed from fission fragments of average mass 149.65 in the neutron fission of ^{235}U and ^{233}U.

In Table II the Q values[32,39] for thermal-neutron-induced fission of ^{235}U into the fragment pairs ^{150}Pm/^{86}Ge, ^{149}Pm/^{87}Ge, and ^{148}Pm/^{88}Ge are given. Comparing them with the average total kinetic energy[33] for fragments of masses 148–150, which are also shown in Table II, one can doubt if their excitation energy is high enough for the emission of two or one neutrons. One has to bear in mind, however, that the total kinetic energy values are as well averaged over the most probable charge split for a given mass division. The limiting quantity is the Q value of the fission reaction resulting in Pm fragments of masses 148–150. The formation of Pm nuclei with $Z=61$ in the mass chains of $A=148$–150 are very improbable events as can be seen from the Z-Z_p values. The values for Z-Z_p (Z_p is the most probable charge for a given fragment mass chain), which are given in Table II, were calculated from Z_p data of Reisdorf *et al.*[35] The formation of Pm fragments is, however, most probable in the mass chains $A=155$–158. But these primary fragments have to emit approximately 7–10 neutrons to form the fission product ^{148}Pm which is very unlikely.

Therefore in fission events which result in an energetically unfavorable, and thus improbable, charge split (in our case forming fragments with $Z=61$ and $A=148$–150) either the total kinetic energy or the excitation energy of the fragments—the latter appears in the form of the prompt neutrons and γ rays—has to be smaller than the corresponding values for the most probable charge splits for a given mass chain. There is not much known about the dependence of the total kinetic energy release on charge division. Fragment range measurements[34] indicate that changes in the total energy release with changing charge division for a given mass split are reflected in changes of the kinetic energy of the fragments and that the excitation energy is the same for all charge splits in an isobaric chain.

Lack of knowledge of the exact excitation energy of the primary Pm fragments forming the ^{148}Pm^{m,g}

TABLE II. Q values (Refs. 32 and 39), Z_p and $Z-Z_p$ values (Ref. 35) for Pm fragments, average total kinetic energies $\bar{E}_k$ (Ref. 33), and root-mean-square width σ_{E_k} (Ref. 33) of the total kinetic energy for the most probable fragments of masses 148–150 in thermal-neutron-induced fission of ^{235}U.

Mass of Pm fragments	Q value of charge split 61/31 (MeV) Ref. 32	Ref. 39	Z_p	$Z(=61)-Z_p$	$\bar{E}_k$ (MeV)	σ_{E_k} (MeV)
148	148.6	151.0	57.18	3.82	161.7	7.2
149	154.6	156.4	57.57	3.43	160.5	7.1
150	156.5	158.1	57.95	3.05	159.5	7.0

products forces us therefore to make some assumptions. We thought it reasonable to consider the two extreme cases:

(1) The excitation energy of all fragments of a given mass division A_H/A_L is the same for all charge divisions Z_H/Z_L; also the same for very improbable charge splits as in the case of the Pm fragments of masses 148–150. The number of prompt neutrons and γ rays emitted by the fragments of mass A_H is the same for all charges Z_H of this chain. We used a value of $\bar{\nu}_p = 1.65$[30,31] as the average number of prompt neutrons emitted by primary Pm fragments leading to the $^{148}\mathrm{Pm}^{m,g}$ products. The average kinetic energy of the neutrons was taken from Ref. 27; their binding energy was calculated by means of Ref. 32. We also used an average value of 1.5 MeV for the kinetic energy of the neutrons as explained later in the text.

(2) The average total kinetic energy release $\bar{E}_K$ is the same for all charge divisions Z_H/Z_L for a given mass split A_H/A_L, although the total energy release (Q value) depends strongly on the charge division. A comparison of the Q values for charge splits 61/31, with the average total kinetic energy release $\bar{E}_K$ (Table II), shows that this assumption leads to an energy deficit of between 3.0 (1.4) and 13.7 (10.7) MeV for Pm fragments of masses 148–150 in thermal-neutron fission of ^{235}U, dependent on the nuclear-mass relation used.[32,39] It seems very unlikely that in fission events with very improbable charge splits the primary fragments are formed without any excitation energy. For that reason we have assumed that the ^{148}Pm products are formed either from ^{149}Pm fragments by the emission of one neutron only or from ^{148}Pm fragments only, which have merely enough excitation energy so that neutrons cannot be emitted, but only γ rays.

As will be seen later the results do not differ very much since the 1.65 neutrons, which are assumed to be emitted in case (1), do not take along much angular momentum but take mainly excitation energy. The average spin of the fragments is reduced by not more than one unit of angular momentum. The main difference of the two cases will be that in case (1) the calculated angular momentum distribution has to be attributed to fragments of masses 149 and 150; in case (2) to fragments of masses 149 or 148.

In the case of the medium-energy ^{4}He-ion-induced fission of ^{232}Th one could use the energy-dependent calculations of isomer ratios reported by Saha and Yaffe.[9] This model takes into account the competition between fission and neutron emission which is important in medium-energy fission, and hence the formation probability of a primary fragment of a given mass and charge from all fissioning nuclei in the multiple-chance fission cascade.

The quantities which have to be known for this model are, however, not known very well in our case. The pre-fission neutrons are not expected to lower the angular momentum of the compound nucleus to any appreciable extent. Therefore and because of the absence of more exact information we neglected the emission of pre-fission neutrons. We have treated all $^{232}\mathrm{Th} + {}^4\mathrm{He}$ results in terms of the excitation energy (and/or angular momentum) of the $^{236}\mathrm{U}^*$ compound nucleus. Thus the excitation energy of the $^{236}\mathrm{U}^*$ is the basis for comparing the results from thermal-neutron-induced fission of ^{235}U and ^{4}He-ion-induced fission of ^{232}Th. The excitation energy of the $^{236}\mathrm{U}^*$ nucleus was calculated as $E_{\mathrm{c.m.}} + Q$ where $E_{\mathrm{c.m.}}$ is the center-of-mass energy and Q is the reaction Q value.[32]

At present there are only the measurements of Fraenkel *et al.*[40] available for the number of post-fission neutrons $\bar{\nu}$ emitted as a function of fragment mass for fission of ^{232}Th by 45-MeV ^{4}He ions. Britt and Whetstone[41] give $\bar{\nu}$ for the fission of ^{230}Th by α particles of 25.7 and 29.5 MeV and mention in their paper that the $\bar{\nu}$ distribution for ^{4}He-induced fission of ^{232}Th is similar to the results for ^{4}He-induced fission of ^{230}Th. McHugh and Michel[42] obtained a $\bar{\nu}$ distribution as a function of fragment mass by an indirect method for ^{232}Th + 44-MeV α particles. They observed the same trend as Britt and Whetstone,[41] namely values for $\bar{\nu}$ in the mass region ≥ 130 which are approximately independent of the fragment mass.

Assuming again that the number of prompt neutrons as a function of fragment mass is independent of the charge split for a given fragment mass the above mentioned measurements give the number of neutrons for Pm fragments of masses 148–152 as shown in Table III.

Another way to estimate the number of neutrons which are emitted by fragments leading to the product ^{148}Pm consists of a calculation of the excita-

TABLE III. Number of neutrons for fragments of masses 148–152 in ^{4}He-induced fission of ^{232}Th (Refs. 40 and 42) and ^{230}Th (Ref. 41).

Fissioning system	E_x	$\bar{\nu}(A)$ $A = 148$	150	152
$^{232}\mathrm{Th} + 45$ MeV $^4\mathrm{He}$ [a]	39.6	2.76	2.82	2.93
$^{230}\mathrm{Th} + 29.5$ MeV $^4\mathrm{He}$ [b]	24.4	2.1	1.87	1.77
$^{230}\mathrm{Th} + 25.7$ MeV $^4\mathrm{He}$ [b]	20.7	2.1	2.1	2.0
$^{232}\mathrm{Th} + 44$ MeV $^4\mathrm{He}$ [c]	38.6	3.5	3.5	3.5

[a] Reference 40.

[b] Reference 41.

[c] Reference 42.

tion energy of the fragments by means of the excitation energy of the fissioning compound nucleus ^{236}U*, the Q value of the fission process in the two fragments under consideration, and the measured total fragment kinetic energies $\overline{E}_K$. Here again one meets the difficulty of not knowing how $\overline{E}_K$ depends on the charge split Z_H/Z_L for a given mass division A_H/A_L. Moreover, one has to make an assumption as to how the excitation energy divides among the two fragments.

Such calculations of the excitation energies of Pm fragments of masses 148–152 were performed with reasonable assumptions.[43] Because of these assumptions, however, the results of these calculations cannot be preferred to those obtained with the following simple procedure: Keeping in mind that the prompt neutrons carry away only a small amount of the angular momentum of the primary fragments, it seemed to us quite reasonable to evaluate the isomer ratios with the assumption that the number of prompt neutrons emitted by the Pm fragments in forming the fission product ^{148}Pm increases with the excitation energy of the compound nucleus ^{236}U from $\overline{\nu}=1.65$, as in the case of thermal-neutron-induced fission of ^{235}U, to $\overline{\nu}=3.0$ in the fission of ^{232}Th by 41.4-MeV ^{4}He ions.

The transmission coefficients $T_l(E)$ for neutron emission from the different Pm fragments were taken from the report of Lindner.[44] The above mentioned calculations also showed that the kinetic energies E_n of the emitted neutrons are between 0.9 and 2.0 MeV, assuming that the kinetic energies of the emitted neutrons are equal to $2T$, where T is the nuclear temperature. To simplify matters we assumed that the fragments emit neutrons of average energy 1.5 MeV. We also used this value in the calculations for thermal-neutron fission of ^{235}U (besides the values of Ref. 27) to investigate the influence of this assumption on the results.

The residual excitation energy of the ^{148}Pm fragments after neutron emission is removed by the emission of a cascade of γ rays. The average number $\overline{N}_\gamma$ and the average total energy $\overline{E}_\gamma$ of the γ rays emitted by the fragments as function of fragment mass has been measured[36,37] for thermal-neutron-induced fission of ^{235}U and ^{233}U. Again we have to assume that these values, which are obviously obtained by averaging over the most probable charge splits of each isobaric chain, are also valid for the improbable charge divisions which yield Pm isotopes of masses 148–150. No measurements are known about the number and energy of γ rays emitted by the primary fragments in fission of ^{232}Th by ^{4}He ions. Therefore the following assumption about the γ ray yield and energy of the ^{148}Pm fragments in fission of ^{232}Th by α particles seemed reasonable to us: After the emission of the prompt neutrons, the fragments are on the average in the same state of excitation whether they arise from fission of ^{236}U nuclei of 6.5-MeV excitation energy (produced through ^{235}U $+n_{\text{th}}$) or from ^{236}U nuclei of medium excitation energy (produced through ^{232}Th$+^4$He). Therefore the same average number and average total energy of the γ rays could be used as measured for thermal-neutron-induced fission of ^{235}U.[36]

Pleasanton, Ferguson, and Schmitt[36] give as average values for the number and total energy of γ rays emitted per fission in thermal-neutron-induced fission of ^{235}U $\langle \overline{N}_\gamma\rangle = 6.51 \pm 0.3$ and $\langle \overline{E}_\gamma\rangle = 6.43 \pm 0.3$ MeV. The assumption that following the emission of the prompt neutrons the fission fragments emit three γ rays before reaching the isomeric level or ground state seems reasonable in view of these results. Following the statistical emission of the three γ rays, a fourth γ ray is emitted which populates either the metastable state or the ground state.

Another assumption that was made is to estimate that E_γ, the total energy emitted in the form of prompt γ rays, is approximately one-half of the binding energy of the first neutron not emitted. The average energy and number of prompt γ rays emitted from the fragments of initial excitation energy E can be estimated by means of the formula[38]

$$\overline{E}_\gamma = 4\left(\frac{E}{a} - \frac{5}{a^2}\right)^{1/2}, \qquad (3)$$

where a is the level density parameter. The energy of each succeeding γ ray is found by computing the new excitation energy by subtracting from the residual excitation energy the average energy of the γ ray calculated by use of Eq. (3). For the excitation energy at which the isomer-deciding γ transition takes place the assumption a of Ref. 38 has been used.

There is evidence that a significant component of the prompt γ rays is quadrupole,[45] e.g., because the primary fragments are formed with a high spin[3] (see Table VII) which cannot easily be dissipated by dipole transitions only. The share of the quadrupole transitions in the deexcitation of the fission fragments is, however, not known. We have assumed a quadrupole component of 10% so as to keep the calculation parameters similar to those we have used in "calibrating" the isomer-yield ratio technique for the isomer pair ^{148}Pm^{m,g} in our studies of the reactions ^{148}Nd$(d,2n)^{148}$Pm^{m,g} and ^{148}Nd$(p,n)^{148}$Pm^{m,g} where the initial angular momentum distribution can be calculated.[20]

To be able to compare our calculations with those of other investigators[3-18] who have used dipole transitions we also assumed the γ radiation to be dipole only. The influence of pure quadrupole transitions was also investigated by assuming that three $E2$ transitions take place statistically before the emission of the isomer-deciding γ ray.

Spin cutoff parameter

In the present analysis we have used different values for the spin cutoff parameter σ. Information about the spin cutoff factor comes, among others, from isomer ratio studies in nonfission reactions.[19] We have determined σ by measurements of the isomer ratio of ^{148}Pm for the $^{148}\text{Nd}(d,2n)^{148}\text{Pm}^{m,g}$ and $^{148}\text{Nd}(p,n)^{148}\text{Pm}^{m,g}$ reactions.[20] In other words, we take the parameter for the calculation of the fission fragment isomer ratio from corresponding calculations of the same isomer ratio in nonfission reactions in which the initial angular momentum distribution of the compound nuclei ^{150}Pm and ^{149}Pm can be calculated and is, as well as their excitation energy, of about the same magnitude as in the primary fission fragments ^{150}Pm and ^{149}Pm. It is to be hoped that the results are more independent of the particular suppositions made in the calculations.

In the calculations described in our other paper[20] the spin cutoff parameter has been assumed either to be constant, independent of excitation energy, or—more realistic—energy dependent. For details see Ref. 20. Calculations of the fission isomer ratio were also done for values of $\sigma_n = 4$ for the neutron evaporation and $\sigma_\gamma = 3$ for the γ ray transitions, respectively, to be able to compare our results better with the results of other authors.[3] Table IV summarizes the values of the spin cutoff parameters used in the present analysis. The computer code of Hafner, Huizenga, and Vandenbosch[26] was used to calculate the spin distributions after the emission of neutrons and γ rays.

TABLE IV. Spin cutoff parameters used in the present analysis.

Transition	Spin cutoff parameter σ = const	 $\sigma = f(E)$	Remarks
$^{150}\text{Pm} \xrightarrow{n} {}^{149}\text{Pm}$	$\sigma_n = \sigma_\gamma = 3.9$	$\theta = 0.70\theta_r$ [a]	Ref. 20
	$\sigma_n = 4$		Ref. 3
$^{149}\text{Pm} \xrightarrow{n,\gamma} {}^{148}\text{Pm}$	$\sigma_n = \sigma_\gamma = 4.2$	$\theta = 0.65\theta_r$ [a]	Ref. 20
	$\sigma_n = 4; \sigma_\gamma = 3$		Ref. 3
$^{148}\text{Pm}^* \xrightarrow{\gamma} {}^{148}\text{Pm}$	$\sigma_\gamma = 4$		Ref. 20

[a] θ is the moment of inertia of the nucleus; θ_r is the rigid body moment of inertia.

TABLE V. Values of the fission fragment angular momentum parameter $\overline{B}$ for thermal-neutron-induced fission of ^{235}U and ^{233}U derived with different parameters for the deexcitation cascade.

Primary fragments	Number and energy of emitted neutrons N_n	 E_n (MeV) ^{235}U	 ^{233}U	Number, total energy, and multipolarity of emitted γ rays N_γ ^{235}U	 ^{233}U	 E_γ (MeV) ^{235}U	 ^{233}U	 l_γ	Spin cutoff parameter	$\overline{B}$ ($\hbar$) ^{235}U	 ^{233}U
150/149	1.65	1.1/1.3 [a]	1.3/1.1 [a]	4.3/5.5 [b]	4.4/4.5 [b]	⋯	⋯	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = 3.9/4.2$ [c]	9.3	9.9
150/149	1.65	1.1/1.3 [a]	1.3/1.1 [a]	4.3/5.5 [b]	4.4/4.5 [b]	2.9/4.5 [b]	3.2/3.7 [b]	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = f(E)$ [c]	9.9	10.5
149	1	1.3 [a]	1.1 [a]	5.5 [b]	4.5 [b]	⋯	⋯	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = 4.2$ [c]	8.7	9.1
149	1	1.3 [a]	⋯	Eq. (3)	⋯	$1/2E_{Bn}$ [d]	⋯	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = 4.2$ [c]	8.3	
149	1	1.3 [a]	⋯	5.5 [b]	⋯	$1/2E_{Bn}$	⋯	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = f(E)$ [c]	10.0	
149	1	1.3 [a]	⋯	Eq. (3)	⋯	$1/2E_{Bn}$	⋯	$E1 + 10\%\ E2$	$\sigma_n = \sigma_\gamma = f(E)$ [c]	8.9	
148	0	⋯	⋯	5.0 [b]	4.2 [b]	⋯	⋯	$E1 + 10\%\ E2$	$\sigma_\gamma = 4.0$ [c]	8.6	9.0
148	0	⋯	⋯	Eq. (3)	⋯	$1/2E_{Bn}$	⋯	$E1 + 10\%\ E2$	$\sigma_\gamma = 4.0$ [c]	8.2	
148	0	⋯	⋯	5.0 [b]	⋯	$1/2E_{Bn}$	⋯	$E1 + 10\%\ E2$	$\sigma_\gamma = f(E)$ [c]	9.7	
148	0	⋯	⋯	Eq. (3)	⋯	$1/2E_{Bn}$	⋯	$E1 + 10\%\ E2$	$\sigma_\gamma = f(E)$ [c]	8.7	
150/149	1.65	1.1/1.3 [a]	⋯	3 + 1 [e]	⋯	⋯	⋯	$E1$	$\sigma_n = 4.0; \sigma_\gamma = 3.0$	10.0	
150/149	1.65	1.1/1.3 [a]	⋯	3 + 1 [e]	⋯	⋯	⋯	$E2$	$\sigma_n = 4.0; \sigma_\gamma = 3.0$	13.3	
150/149	1.65	1.5/1.5 [d]	1.5/1.5 [d]	3 + 1 [e]	3 + 1 [e]	⋯	⋯	$E1$	$\sigma_n = 4.0; \sigma_\gamma = 3.0$	10.0	10.8
150/149	1.65	1.5/1.5 [d]	1.5/1.5 [d]	3 + 1 [e]	3 + 1 [e]	⋯	⋯	$E2$	$\sigma_n = 4.0; \sigma_\gamma = 3.0$	13.3	14.1

[a] Values taken from Ref. 27.

[b] Values taken from Refs. 36 and 37.

[c] See text and Ref. 20.

[d] See text.

[e] Three statistical γ rays plus the isomer-deciding γ ray.

TABLE VI. Derived values of the fission fragment angular momentum parameter $\overline{B}$ (calculated with the number of neutrons as given in the table, average neutron kinetic energies of 1.5 MeV, three statistical plus one isomer-deciding γ-rays, spin cutoff parameters $\sigma_n = 4.0$ and $\sigma_\gamma = 3.0$, and dipole radiation).

Fissioning system	Excitation energy of ^{236}U compound nucleus (MeV)	Experimental isomer ratio ($\sigma m/\sigma g$)	Number of emitted neutrons	$\overline{B}$ ($\hbar$)
$^{233}U + n_{th}$	6.8	3.0 ± 0.5	1.65	10.8 ± 2.0
$^{235}U + n_{th}$	6.5	$2.6^{+0.9}_{-0.5}$	1.65	10.0 ± 2.5
$^{232}Th + ^4He$	20.8 ± 1.7	6.8 ± 1.2	2.0	16.5 ± 2.5
	23.9 ± 1.4	6.6 ± 0.7	2.0	16.3 ± 2.5
	26.7 ± 1.5	5.3 ± 0.5	2.0	14.6 ± 2.5
	31.8 ± 1.0	8.5 ± 0.9	3.0	19.9 ± 2.5
	36.1 ± 1.0	8.5 ± 0.9	3.0	19.9 ± 2.5

V. THEORETICAL CALCULATION: RESULTS AND DISCUSSION

The results of our calculations of the angular momentum parameter B are presented in Table V for the isomer ratio of ^{148}Pm in thermal-neutron-induced fission of ^{235}U and, partly, of ^{233}U. The dependence of the derived angular momentum parameter B on the various values of the different parameters within reasonable limits is shown to be rather insensitive. Being unable to favor one of the different reasonable assumptions with regard to the various parameters, the results of Table V reflect the uncertainty due to the analysis. If the pure $E2$ alternative is left out, the values of B for thermal-neutron-induced fission of ^{235}U are between $B = 8.2$ and 10.0, implying an uncertainty of the calculation of the magnitude of the angular momentum from the isomer ratio of about $\pm 1\hbar$. Including the error in the experimental value of the isomer ratio the uncertainty in the average initial spin value is estimated to be about ± 2.0–$2.5\hbar$.

For the above reason we calculated the angular momentum parameter B for the isomer ratio of ^{148}Pm in ^{4}He-induced fission of ^{232}Th for one set of parameters only. The results of Table VI were calculated with 2–3 emitted neutrons, an average kinetic energy of 1.5 MeV, three statistical plus one isomer-deciding γ rays, spin cutoff parameters $\sigma_n = 4.0$ and $\sigma_\gamma = 3.0$, and dipole radiation. The resulting variation of the root-mean-square ang-

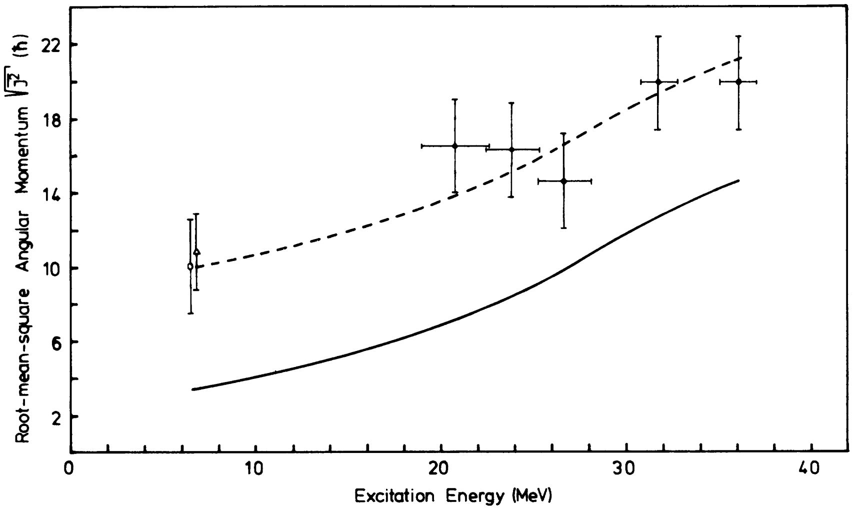

FIG. 1. A plot of the derived root-mean-square angular momentum of Pm fragments as a function of the excitation energy of the ^{236}U compound nucleus in thermal-neutron-induced fission of ^{235}U (open circle) and ^{4}He-induced fission of ^{232}Th (closed circles). The value for neutron fission of ^{233}U is also plotted (triangle). The full line gives the calculated root-mean-square angular momentum of the ^{236}U compound nucleus as a function of its excitation energy. The dashed curve is the solid curve displaced by the amount necessary to make it pass approximately through the values of $(\overline{\mathcal{J}^2})^{1/2}$ derived from the isomer ratios.

TABLE VII. Summary of the root-mean-square angular momenta of selected fission fragments as deduced from independent isomer-yield ratio studies.

Fission reaction Compound nucleus Excitation energy (MeV) Isomer ratios	^{209}Bi + ^{4}He ^{213}At 25	^{226}Ra + ^{2}H ^{228}Ac 26.7	^{232}Th + ^{2}H ^{234}Pa 28	^{233}U + γ ^{233}U 0–16	^{233}U + n_{th} ^{234}U 6.8	^{235}U + n_{th} ^{236}U 6.5	^{232}Th + ^{4}He ^{236}U 20–36	^{233}U + ^{1}H ^{234}Np 24–54	^{235}U + ^{1}H ^{236}Np 15–54	^{238}U + ^{1}H ^{239}Np 25–55	^{237}Np + ^{1}H ^{238}Pu 26–56	^{235}U + ^{4}He ^{239}Pu 21–38	^{239}Pu + n_{th} ^{240}Pu 6.5	^{238}U + ^{4}He ^{242}Pu 29.4	^{239}Pu + ^{1}H ^{240}Am 34–54
^{83}Se($\frac{5}{2}$)/^{83}Se($\frac{1}{2}$)													>8.0[a]		
^{90}Y(7)/^{90}Y(2)										3[b]					
^{91}Y($\frac{9}{2}$)/^{91}Y($\frac{1}{2}$)									3[f]	2[b]					
^{95}Nb($\frac{9}{2}$)/^{95}Nb($\frac{1}{2}$)	15[c]								12 at 15 MeV[d]						
^{128}Sb(8)/^{128}Sb(5)					11.3 ± 0.8[e]								11.0 ± 0.8[e]		
^{130}Sb(8)/^{130}Sb(5)					10.2 ± 0.5[e]	10.2 ± 1.0[e]							12.0 ± 0.9[e]		
^{132}Sb(8)/^{132}Sb(4)					7.9 ± 0.6[e]	7.1 ± 0.5[e]							8.5 ± 0.5[e]		
^{131}Te($\frac{11}{2}$)/^{131}Te($\frac{3}{2}$)					7.3 ± 0.5[e]	6.0 ± 1.5[f] 7.5 ± 0.7[e]	7.9 at 27.8 MeV[f]			10–11[g]			7.6 ± 0.5[e]		
^{133}Te($\frac{11}{2}$)/^{133}Te($\frac{3}{2}$)					5.4 ± 0.3[e]	5.9 ± 1.5[f] 6.5 ± 0.4[e]				8.5–17[g]			7.1 ± 0.3[e]		
^{130}I(5)/^{130}I(2)										10–16[g]					
^{132}I(5)/^{132}I(8)										11–17[g]					
^{134}I(5)/^{134}I(8)										11–13[g]					
^{134}Cs(8)/^{134}Cs(4)		7 ± 2[c]	13 ± 1[c]	7.4[h]				11.5–16[i]	15–16[j]		10.5–12[k]	11.5–20[l]		11 ± 1[c]	11.5–14.5[m]
^{148}Pm(6)/^{148}Pm(1)					9.9 ± 2.1[n] 10.8 ± 2.0[o]	10.0 ± 2.5[o]	16.5–19.9[o]								

[a] Reference 18.
[b] Reference 13.
[c] Reference 8.
[d] Reference 4.
[e] Reference 16.
[f] Reference 15.
[g] Reference 50.
[h] Reference 14.
[i] Reference 6.
[j] Reference 6.
[k] Reference 12.
[l] Reference 7.
[m] Reference 9.
[n] Reference 17.
[o] This work

ular momentum of the Pm fission fragments as a function of the excitation energy of the compound nucleus ^{236}U* is presented in Fig. 1.

The initial average angular momentum induced in the compound nucleus ^{236}U* as a function of its excitation energy was also calculated. Values of $(\overline{J_c^2})^{1/2}$ were calculated with the computer code of Hafner *et al.*[26] The transmission coefficients were taken from Huizenga and Igo.[46] The solid curve in Fig. 1 represents the initially expected root-mean-square angular momentum $(\overline{J_c^2})^{1/2}$ of the compound nucleus ^{236}U* as a function of its excitation energy. The dashed curve in Fig. 1 is simply the solid curve displaced by the amount necessary to make it pass approximately through the values of $B \simeq (\overline{J^2})^{1/2}$ derived from the isomer ratio.

From the apparent agreement between the dependence of the initial average angular momentum of the ^{236}U* compound nucleus and the dependence of the average angular momentum derived for the Pm fission fragments on the excitation energy of the compound nucleus, one might conclude that there is a correlation between the two. An interesting qualitative result is that in going from thermal-neutron-induced fission of ^{235}U to ^{4}He-ion-induced fission of ^{232}Th at excitation energies of up to 36 MeV the average spin of the fissioning nucleus ^{236}U* goes from 3.5 to $14.6\hbar$. The average spin of the primary Pm fragments in the considered mass region increases from 10 to $20\hbar$. Since the values of $(\overline{J^2})^{1/2}$ deduced from the ^{148}Pm isomer ratios are greater than $(\overline{J_c^2})^{1/2}$, the angular momentum brought in by the incident neutrons and α particles is not sufficient to account for the relatively large values of the root-mean-square angular momenta of the primary fission fragments. These results indicate that considerable intrinsic angular momentum can be produced in the fragments in the fission process as first predicted by Strutinsky.[47]

The absolute magnitudes of the fragment angular momentum derived from the ^{148}Pm isomer ratios are, of course, somewhat uncertain because of the uncertainty in our knowledge of the deexcitation cascade. The increase of the fragment angular momentum with excitation energy and angular momentum of the fissioning compound nucleus is, however, real, bearing in mind that the prefission neutrons can carry away only a small amount of the angular momentum from the fissioning nuclei.

In the case of ^{4}He-ion-induced fission of ^{232}Th one has some additional information about the average value of K, the projection of the spin of the fissioning nucleus I_f along the symmetry axis of the fissioning nucleus. The intrinsic spins of the fragments add vectorially to give the vector of length K along the symmetry axis. The dependence of K^2 on $(E_x - E_f)$, where E_x and E_f are the excitation energy and the fission threshold of the fissioning nucleus, has been deduced for ^{4}He-ion-induced fission by Vandenbosch, Warhanek, and Huizenga.[48] Using their functional dependence of K_0^2 the average value of $(K^2)^{1/2}$ for the excitation energies 20.8–36.1 MeV of the compound nucleus ^{236}U* used in this work (taking $E_f = 5.7$ MeV for the nucleus ^{236}U) was obtained as between 9.2 and 12.4$\hbar$. The values of $(\overline{J^2})^{1/2}$ derived for the Pm fragments from the isomer ratio are between 16.5 and 20$\hbar$. Assuming that the complementary light fragments also have similar values for the angular momentum, it seems that there is apparently some cancellation of J_H by J_L.

A summary of the root-mean-square angular momenta of fission fragments as deduced from independent isomer-yield ratio studies in thermal-neutron- and charged-particle-induced fission is given in Table VII. The data are deduced from measurements with rather different target-projectile combinations. The data can also be compared with the results of Ref. 3 in which the intrinsic angular momentum of primary fragments for spontaneous fission of ^{252}Cf was deduced for 21 different even-even fission products by statistical-model analysis of the population of ground-state bands.

The root-mean-square angular momenta of fission fragments deduced by isomer ratio experiments in thermal-neutron-induced-fission of ^{233}U ^{235}U, and ^{239}Pu are plotted in Fig. 2 as a function of fission product mass. The values from Ref. 3 for 21 even-even fission products in spontaneous fission of ^{252}Cf are plotted as well. The plot is presented such that fission products which are equidistant from symmetry of the mass distributions of the different fissioning nuclei are approximately on the same abscissa.

Upon examining Table VII and Fig. 2 one can

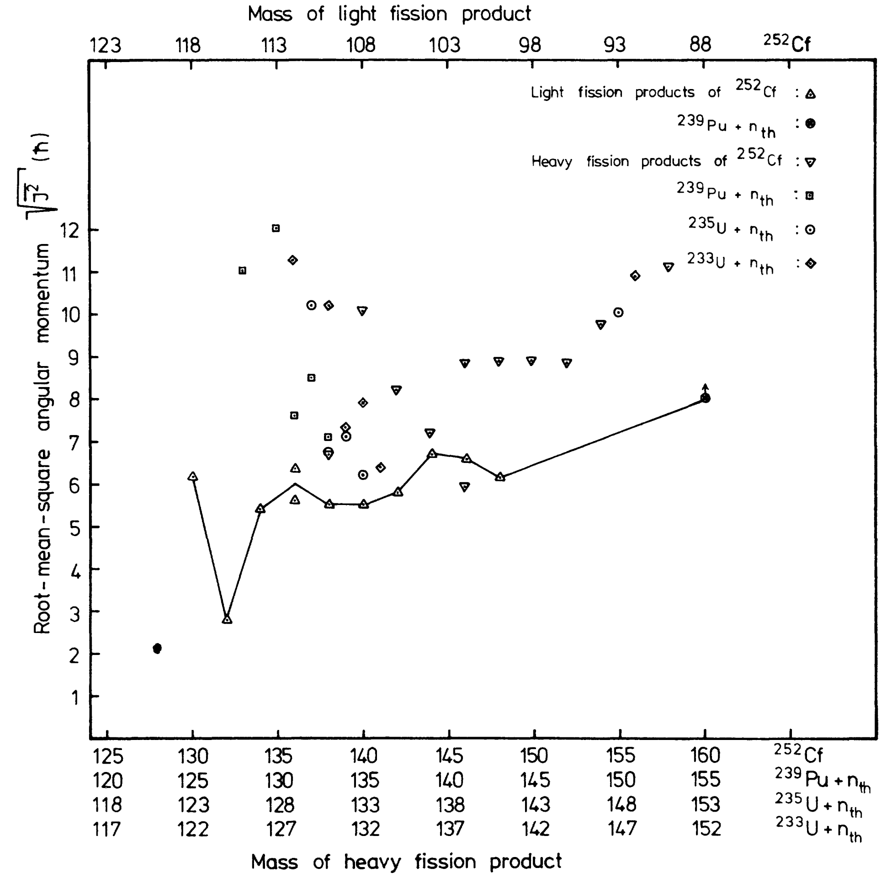

FIG. 2. A plot of the root-mean-square angular momenta of fission fragments deduced by isomer ratio experiments in thermal-neutron-induced fission of ^{233}U, ^{235}U, and ^{239}Pu as a function of fission product mass. The values from Ref. 3 for 21 even-even fission products in spontaneous fission of ^{252}Cf, which are deduced from the population of ground-state bands, are plotted as well. The plot is presented such that fission products which are equidistant from symmetry of the mass distributions of the different fissioning nuclei are approximately on the same abscissa. The solid line which connects the values for the light fission products is merely drawn to guide the eye.

conclude that (see also Ref. 3):

(1) The average angular momenta of fission fragments are higher in medium-energy-induced fission than in low-energy-induced fission.

(2) There seems to be a correlation between the angular momentum of the initial compound nucleus and the fragment angular momentum as shown clearly in this work.

(3) The fragment angular momenta in thermal-neutron-induced fission of ^{233}U, ^{235}U, and ^{239}Pu and in spontaneous fission of ^{252}Cf do not depend significantly on the fissioning nucleus.

(4) The heavy fission fragments have a somewhat greater angular momentum than the light fragments.

(5) The fragment angular momentum appears to increase slightly as symmetric and very asymmetric fission is approached.

(6) There are not enough data to find any clear trend in the fission fragment angular momenta within a larger range of the Z, A, and excitation energy of the fissioning system.

Because of the uncertainty of the absolute magnitude of the fragment angular momenta as deduced from isomer ratio measurements it is difficult to compare these data with the calculations of the average angular momentum of fission fragments by Zielinska-Pfabé and Dietrich.[49] The best agreement between our experimental data in neutron fission of ^{233}U and ^{235}U and the theoretical results in the mass region 148–150 is obtained for their adiabatic case or nonadiabatic case with a value of $kT = 1$ MeV.

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