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Study of the Stability of the Ground States and *K*-Isomeric States of ^{250}Fm and ^{254}102* Against Spontaneous Fission

Yu. A. Lazarev, Yu. V. Lobanov, R. N. Sagaidak, V. K. Utyonkov, M. Hussonnois‡, Yu, P. Kharitonov, I. V. Shirokovsky, S. P. Tretyakova and Yu. Ts. Oganessian

Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Head Post Office, P.O. Box 79, 101 000 Moscow, USSR

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Abstract

By employing the ^{249}Cf(^{4}He, 3n) and ^{208}Pb(^{48}Ca, 2n) reactions, experiments to study the stability against spontaneous fission of the nuclides ^{250}Fm and ^{254}102 as well as of the two-quasi-particle (2 q–p) *K* isomers ^{250m}Fm ($T_{1/2}$ = 1.8 ± 0.1 s) and ^{254m}102 ($T_{1/2}$ = 0.28 ± 0.04 s) have been performed. The ground-state spontaneous fission of the two nuclides has been discovered and the corresponding branching ratios b_{sf} and partial half-lives T_{sf}, respectively, have been determined to be: (6.9 ± 1.0) × 10^{-5}, 0.83 ± 0.15 yr for ^{250}Fm; (1.7 ± 0.5) × 10^{-3}, (3.2 ± 0.9) × 10^4 s for ^{254}102. As a by-product of these studies, new data about cross sections of the ^{206,208}Pb(^{48}Ca, xn) reactions have been obtained. Experiments designed to search for the spontaneous fission decay of the 2 q–p *K*-isomeric states in ^{250}Fm and ^{254}102 have not revealed the effect in question. The lower limits of the ratios of the partial spontaneous fission half-lives for the 2 q–p *K*-isomeric states to those for the respective ground states, T^*_{sf}/T_{sf}, have been established to be $\geqslant 10^{-1}$ for ^{250m}Fm/^{250}Fm and $\geqslant 5 \times 10^{-3}$ for ^{254m}102/^{254}102. This means that the stability of the 2 q–p *K*-isomeric states in ^{250}Fm and ^{254}102 against spontaneous fission is practically not inferior to that of the ground states of these nuclei. In accord with the experimental findings, the theoretical estimates of T^*_{sf}/T_{sf} made in the present paper show that, due to the influence of the specialization and blocking effects on the potential energy and the effective mass associated with fission, spontaneous fission from 2 q–p *K*-isomeric states cannot be facilitated but, on the contrary, should be essentially hindered compared with ground-state spontaneous fission.

1. Introduction and motivation

Although nearly 50 years have elapsed since the discovery of spontaneous fission made by Flerov and Petrzhak [1], the stability of heavy nuclei with respect to this unique decay mode continues to represent a challenging problem for both experimentalists and theoreticians. The reasons for this long-standing interest are easy to understand. On the one hand, the instability of heavy nuclei against spontaneous fission is considered as the main factor that limits the maximum possible number of elements in the Mendeleev Periodic Table. On the other hand, the absolute values of partial spontaneous fission half-lives, T_{sf}, and the pattern of their (Z, N) variations contain valuable information about the mechanism of large-scale cold rearrangements of nuclear matter [2].

Theoretically, the spontaneous fission process is treated as quantum-mechanical penetration through a (multidimensional) potential barrier. The problem is usually simplified by considering the probability of tunnelling through a one-dimensional potential barrier $V(q)$ along some effective trajectory L given in a multidimensional space of deformations α_i $(i = 1, 2, \ldots, m)$. Further, using the WKB approximation and the least-action principle the spontaneous fission half-life is determined [3–6] to be

$$T_{sf}[\text{years}] = \frac{\ln 2}{np} \simeq 10^{-28} \exp\,[S(L_{min})]. \tag{1}$$

Here $n \simeq 10^{20.4}\,\text{s}^{-1}$ is the number of assaults of the nucleus on the fission barrier per unit time, and p is the probability of tunnelling through the barrier for a given assault,

$$p = \{1 + \exp\,[S(L_{min})]\}^{-1} \tag{2}$$

where $S(L_{min})$ is the minimum value of the action integral

$$S(L) = 2\int_{q_1}^{q_2} \left\{\frac{2}{\hbar^2}[V(q) - E]M(q)\right\}^{1/2} dq. \tag{3}$$

In eq. (3) the parameter q specifies the position of a point on the trajectory L, with q_1 and q_2 corresponding to the classical turning points at which $V(q) = E$, and E is the total energy of the fissioning nucleus. The least-action trajectory L_{min} is determined by the variational condition

$$\delta S(L) = 0 \tag{4}$$

whereas the effective mass associated with motion along the trajectory L has the form [3, 4]

$$M(q) \equiv M_{qq}(q) = \sum_{i=1}^{m}\sum_{j=1}^{m} M_{\alpha_i\alpha_j}(\alpha_1(q), \alpha_2(q), \ldots, \alpha_m(q)) \times \frac{d\alpha_i}{dq}\cdot\frac{d\alpha_j}{dq} \tag{5}$$

where $M_{\alpha_i\alpha_j}$ are components of the (symmetric) mass tensor which correspond to the deformation parameters α_i and α_j. As for the potential energy of the fissioning nucleus, it can be expressed in the framework of the macroscopic-microscopic approach [3] as

$$V(q) = \tilde{V}(q) + \sum_{p,n}[\delta U(q) + \delta P(q)] \tag{6}$$

where $\tilde{V}$ is the macroscopic energy part, and δU and δP are, respectively, the shell correction and the pairing correction calculated for protons and neutrons separately.

It is now important to stress the dynamical traits of the fission barrier penetration problem. In fact, along with the potential energy $V(q)$ which determines the generalized forces acting in the fissioning system, the action integral (3) involves an essentially dynamical quantity – the effective mass $M(q)$. The latter characterizes the response of the system to the forces applied and, together with $V(q)$, determines the trajectory of the system's motion towards fission. The system may not follow the minimum potential energy path (i.e., the static fission trajectory) provided the effective mass on this path is too large. Therefore a kind of compromise is realized between

* Editors comment: Element 102 is also known under the name Nobelium.
‡ Institute de Physique Nucléaire, Orsay, France.

the values of $V(q)$ and $M(q)$ on the least-action trajectory (i.e., on the *dynamical* trajectory of fission). The quantities $V(q)$ and $M(q)$ enter the action integral (3) in an equivalent way so that comparatively small variations in any of them can lead to large-scale changes in T_{sf}. Therefore the problem of calculating T_{sf} and interpreting *quantitatively* the experimentally observed (Z, N) variations of T_{sf} imposes equally high requirements on the level of theoretical understanding and accuracy of calculating the two quantities – the potential energy and the effective mass. However, as compared with the potential energy surface which has been investigated by theory thoroughly enough, especially in the vicinity of its stationary points where certain properties of the surface can be checked experimentally, the effective mass is a much more complex and far less studied characteristic; furthermore, the possibilities of obtaining any empirical information concerning properties of the effective mass are very limited. On the other hand, the observable quantity T_{sf} characterizes the dynamical process of tunnelling through the fission barrier in an essentially averaged fashion. Therefore, it proves rather difficult to separate the role of conservative and inertial effects in the penetration process on the basis of empirical T_{sf} data for ground-state spontaneous fission. We believe that in this respect important information can be obtained by measuring T_{sf} values for various isomeric states in the first potential well and analyzing these results together with ground-state T_{sf} data.

By now, the partial half-lives T_{sf} have been measured for the ground-state spontaneous fission of some 70 nuclides with Z from 92 through 108. In summarizing briefly the global features of this empirical information [2] it should, first of all, be stressed that the pattern of the (Z, N) variations of T_{sf} exhibits dramatic deviations from the liquid-drop model predictions. These deviations are a direct consequence of the manifestation of the individual structure of nuclei, in the first place, of nuclear shell structure which strongly influences the landscape of the potential energy surface associated with fission. The revelation of the prominent role of the shell structure effects in strongly deformed nuclei [3] has resulted in a considerable progress in fission theory as a whole and, in particular, in the understanding and theoretical description of the patterns of change in T_{sf} with respect to Z and N (see, e.g., Refs. [2–16]). Yet a closer examination of the available T_{sf} calculations [4–16] shows that these theoretical achievements are far from exhausting the problem.

In addition to the shell structure, another essential feature of atomic nuclei is the presence of nucleon pairing correlations of superconducting type [17–22]. However, as opposed to the shell structure effects, the role of pairing correlations in subbarrier fission still remains much less clear both experimentally and theoretically. Direct experimental data in this respect are so far virtually absent, yet theory predicts quite convincingly [3, 4, 6, 7, 21–27] that the effective mass M associated with fission should depend strongly on the magnitude of the pairing gap parameter Δ. Thus, according to the adiabatic cranking model [21, 22], at $\Delta \gg G$ (where G is the pairing matrix element [17–21]), the following approximate expression is derived [3, 4, 22, 23]

$$M_{qq}(q, \Delta) \approx \frac{F(q)}{\Delta^2} + \eta \tag{7}$$

where the second term, which is approximately constant and generally very small compared to the first one, provides the correct limiting form of eq. (7) at large Δ values. It is important to note that eq. (7) (or, generally, the fact that the derivative $\partial M/\partial \Delta$ is an essentially negative and large quantity) expresses, perhaps, the most definite of all the theoretical predictions concerning properties of the effective mass. The dependence of the type (7) arises not only in the cranking model but in more advanced approaches too, for example, in the calculations [26] carried out in the framework of the generator coordinate method [21].

The question now is how Δ changes in the tunnelling process. In the standard treatment of pairing correlations in tunnelling (which we shall term also the "statical" treatment) the deformation dependence of the gap parameter Δ is determined [3, 4] by solving the BCS equations [19, 21], i.e., by requiring, at each deformation, the expectation value of the pairing Hamiltonian to be stationary (a minimum) with respect to small variations in Δ:

$$\frac{\partial\langle H\rangle}{\partial \Delta} = 0. \tag{8}$$

If the pairing matrix element G is chosen to be independent of the nuclear surface area*, then the gap parameter Δ found from eq. (8) does not show significant changes in the tunnelling process – it oscillates slightly around some average value close to the initial one (Δ_0) characterizing superfluid properties of the nucleus in the region of $q \leqslant q_1$ (see, e.g., Figs. 2 and 6 in Ref. [4]). Thus, even a slight weakening of pairing correlations in the initial state will lead to a perceptible increase in the magnitude of the action integral

$$S \sim \Delta^{-1} \tag{9}$$

and hence to a sharp increase in T_{sf} due to the exponential dependence in eq. (1).

At the same time, tunnelling through the fission barrier represents, as we have stressed above, an essentially dynamical problem. Therefore, as proposed by Moretto and Babinet [28], it would be more appropriate to determine Δ in this problem by minimizing the action rather than the expectation value of the Hamiltonian. In other words, the gap parameter Δ should be treated here as a dynamical variable similar to the deformation variables (see also Ref. [29]); hereafter this treatment based on determination of Δ by minimizing the action integral will be referred to as the "dynamical" treatment. The dynamical treatment of pairing correlations, in contrast to the standard one, predicts [28, 30] a large enhancement of superfluidity in tunnelling: while at $q \leqslant q_1$ the spontaneously fissioning nucleus is characterized by the gap value $\Delta = \Delta_0$, in deepening into the barrier the value of Δ increases, reflecting the barrier profile, and reaches its maximum $\Delta_{max} \approx 2\Delta_0$ at the saddle point deformation; after that Δ decreases down to $\Delta \approx \Delta_0$ at the turning point $q = q_2$ (see Fig. 1 in Ref. [31]). In the dynamical approach, a weakening of pairing in the initial state also leads to an increase in T_{sf} but the scale of this increase turns out to be much smaller [31] than in the standard consideration. Being somewhat surprising, the dynamical treatment of pairing correlations in tunnelling, as has been shown in Ref. [31], does not contradict any empirical evidence or generally accepted theoretical

* Only the surface-independent pairing (which seems to have the best physical justification) is considered in the present paper.

knowledge. On the contrary, it allows a more adequate explanation of some empirical facts to be given, for example, that of the typical order-of-magnitude values of the hindrance factors associated with ground-state spontaneous fission of odd-A and odd-odd nuclei (see also Section 4).

Therefore we have to conclude that the basic question as to what physical principle governs the behaviour of superfluid properties (e.g., the pairing gap Δ) of a nuclear system undergoing a large-scale subbarrier rearrangement remains open. Is the Δ behaviour in tunnelling determined by the minimum Hamiltonian condition or regulated by the least-action principle? In Ref. [31] it has been demonstrated that these two different treatments of pairing correlations yield substantially differing predictions for observable quantities: as compared to the traditional (BCS) approach, the dynamical treatment leads to a considerable weakening of the dependence of the fission barrier penetrability on the basic parameters of the problem, *viz.*, on the pairing gap in the initial state (Δ_0), on the barrier height (B_f), and on the energy of the initial state (E). This difference between predictions gives grounds to believe that the superfluidity issue can be decided on the basis of empirical data. From the analysis made in Ref. [31] it follows that the most direct information for the purpose in view can be obtained by measuring the probability of spontaneous fission from quasi-particle (q–p) isomeric states in heavy even-even nuclei since the relative change of the partial spontaneous fission half-life in going from the ground-state to a high-spin q–p isomeric* state, T^*_{sf}/T_{sf}, is predicted to be strongly dependent on whether or not the dynamically induced enhancement of superfluidity takes place in the tunnelling process. Accordingly, we have designed experiments to probe the stability of q–p isomers against spontaneous fission.

Quasi-particle or K isomers are expected to occur when breaking up of one or several pairs of nucleons in an even–even nucleus and appropriate recoupling of the spins of the unpaired nucleons lead to the formation of relatively low-lying states having high values of the total spin projection K onto the symmetry axis of the nucleus. The high K values cause a strong retardation of γ transitions, which, in turn, favours searches for a spontaneous fission branch in the decay of the K-isomeric states in the heaviest nuclei. By now, a number of K isomers has been found in the region of even–even nuclei with $Z \geqslant 92$ [32] and the occurrence of many other K isomers in this region can be expected on the basis of theoretical considerations [33] (see Table I).

Experimentally, spontaneous fission from K-isomeric q–p states in the first potential well† has never been observed. The only attempt to observe it was made by Vandenbosch *et al.* [36], for the 34-ms K-isomeric state in ^{244}Cm; however, no effect has been detected (see Section 4). As has been emphasized in Ref. [31], the most appropriate objects for searches for the spontaneous-fission decay from K-isomeric q–p states are expected to be the heaviest even–even nuclei showing spontaneous fission as a predominant or quite probable decay mode of their ground states. Accordingly, for our experimental studies we have chosen the K isomers ^{250m}Fm ($T^*_{1/2} = 1.8 \pm 0.1$ s) and ^{254m}102 ($T^*_{1/2} = 0.28 \pm 0.04$ s) detected by Ghiorso *et al.* [37, 38]. Although the energies, spins and parities of these isomers are not yet established experimentally, their interpretation [38] as 2 q–p neutron or proton states with $K^\pi = 8^-$ or 7^- (see Table I) is fully confirmed by the semimicroscopic calculations of Ivanova *et al.* [33].

Thus, the main purpose of our experiments was a search for a spontaneous fission branch in the decay of the K isomers ^{250m}Fm and ^{254m}102. In the ground state, the nuclides ^{250}Fm and ^{254}102 are known to be predominantly α-particle emitters with $T_{1/2} = 30 \pm 3$ min, $E_\alpha = 7.43$ MeV and $T_{1/2} = 55 \pm 5$ s, $E_\alpha = 8.10$ MeV, respectively [32]. As for the ground-state spontaneous fission, it has in fact never been detected either for ^{250}Fm or for ^{254}102; only a rough T_{sf} estimate for ^{250}Fm and a lower T_{sf} limit for ^{254}102 have been reported [39, 40]. Therefore we performed also direct experiments to determine the partial half-lives for the ground-state spontaneous fission of the two nuclides. The experimental technique we used and the results obtained are described in Sections 2 and 3. A discussion of the results is given in Section 4, and the main conclusions drawn are presented in Section 5.

2. Study of ^{250}Fm and ^{250m}Fm

2.1. *Experimental technique*

The ^{249}Cf(^{4}He, 3n) reaction [41] was used to produce ^{250}Fm and ^{250m}Fm. Irradiations were made at the U-200 cyclotron of the Laboratory of Nuclear Reactions, JINR (Dubna), by employing a 34-MeV ^{4}He beam with an average intensity of (1–2) $\times 10^{13}$ particles s^{-1}. Several targets of $^{249}CfO_2$ deposited onto Au backings were used in the experiments. Isotopically pure ^{249}Cf was isolated as the decay product of initially pure ^{249}Bk.

The sufficiently long half-life of ^{250}Fm (30 min) allowed us to determine its ground-state spontaneous fission branch b_{sf} in off-line measurements. In this case the reaction recoils from the target were collected on a 0.2 mg cm^{-2} Al catcher foil fixed in a vacuum chamber at a distance of 1 mm from the target. Upon the completion of an irradiation, the catcher was first brought, for an appropriate time, into contact with solid state nuclear track detectors registering spontaneous fission fragments and then placed in a semiconductor α spectrometer for determining the total yield of the ^{250}Fm nuclei by detecting their α decay. After the α-decay measurements, the catcher was put in contact with track detectors again in order to check whether there is present any long-lived spontaneous fission background. Thus, spontaneous fission and α-decay measurements were carried out alternately.

The search for spontaneous fission from the 1.8-s isomeric state of ^{250}Fm was made in on-line experiments using a tape transport system that is similar to a tape recorder with spools separated by a distance of about 1.8 m in order to form an appropriately long rectilinear path of the tape motion. In this system the reaction recoils from the target (placed near the middle of the rectilinear path) were collected on a Ni tape, ≈ 150 m long, 25 mm wide, and 0.05 mm thick, which moved with a given constant velocity. Mica fission fragment detectors were arrayed along the rectilinear path of the tape and covered a distance of 0.8 m in both directions with respect to the collection zone. The gap between the tape and the mica detectors as well as that between the tape and the target was equal to 2 mm. The tape transport assembly, the target, and

* Here and below starred quantities are those pertinent to isomeric states.
† For a discussion of the experimental information relevant to the second potential well, see Refs. [31, 34, 35]; see also Section 5.

Table I. *Properties of some 2 q–p K-isomeric states in heavy even–even nuclei*[a]

Nucleus	Energy of the isomeric state E^*, MeV	K^π	Nilsson configuration	Half-life of the isomeric state $T^*_{1/2}$
^{234}U	1.421	6^-	$\frac{5}{2}^+[633]_n$, $\frac{7}{2}^-[743]_n$	$33.5 \pm 2.0\,\mu s$
^{236}U	1.054	4^-	$\frac{7}{2}^-[743]_n$, $\frac{1}{2}^+[631]_n$	$120 \pm 20\,ns$
^{238}Pu	1.082	4^-	$\frac{7}{2}^-[743]_n$, $\frac{1}{2}^+[631]_n$	$8.5 \pm 0.5\,ns$
^{244}Cm	1.042	6^+	$\frac{5}{2}^+[622]_n$, $\frac{7}{2}^+[624]_n$	$34 \pm 2\,ms$
^{250}Fm		8^-	$\frac{7}{2}^+[624]_n$, $\frac{9}{2}^-[734]_n$	$1.8 \pm 0.1\,s$
		7^-	$\frac{7}{2}^+[633]_p$, $\frac{7}{2}^-[514]_p$	
^{256}Fm	1.3	9^-	$\frac{7}{2}^+[613]_n$, $\frac{11}{2}^-[725]_n$	
	1.3	7^-	$\frac{7}{2}^+[633]_p$, $\frac{7}{2}^-[514]_p$	
$^{254}102$	1.2	8^-	$\frac{9}{2}^-[734]_n$, $\frac{7}{2}^+[613]_n$	$0.28 \pm 0.04\,s$
	1.1	8^-	$\frac{7}{2}^-[514]_p$, $\frac{9}{2}^+[624]_p$	
$^{260}104$	1.3	9^-	$\frac{7}{2}^+[613]_n$, $\frac{11}{2}^-[725]_n$	

[a] The lines of the table which contain the $T^*_{1/2}$ values stand for the isomers observed experimentally [32, 38]; in these cases the indicated values of E^* and K^π are based on measurements (except for the isomers of ^{250}Fm and $^{254}102$ for which only the $T^*_{1/2}$ values have been measured). Other lines of the table give examples of the 2 q–p K-isomeric states expected from theoretical considerations; for further theoretical information, see Ref. [33].

the mica detectors were enclosed in a vacuum chamber. As the entire 150 m length of the tape had been reeled onto one spool of the "tape recorder", the direction of the tape motion reversed automatically (while its linear velocity remained unchanged). Thus, both halves of the rectilinear path – on both sides of the collection zone – were exploited to record spontaneous fission fragments. The corresponding track distributions were then summarized. The velocity of the tape motion was chosen in such a way that it was possible to observe, in the presence of spontaneous fission from the isomeric state, the decay of the 1.8-s fission activity on the detectors close to the collection zone while the ground-state spontaneous fission of ^{250}Fm could be observed as a "background" uniformly distributed on detectors further away. Then, by analyzing the time distribution of the recorded events it was possible to determine immediately the ratio T^*_{sf}/T_{sf} of the partial half-lives for spontaneous fission from the 1.8-s isomeric state and the 30-min ground state. Of course, with due decrease in the velocity of the tape, it was possible to measure the decay curve of 30-min ^{250}Fm by detecting its spontaneous fission fragments.

2.2. *Measurements and results*

The typical α-particle-energy spectra recorded in the off-line measurements of radioactivity on a catcher foil are shown in Fig. 1. The α-particle assignments to particular nuclides, as indicated in Fig. 1, are based on the correspondence in energy and half-life with those of well established α-emitters. From our α-decay measurements, the formation cross section of ^{250}Fm in the $^{249}Cf + {}^4He$ reaction was found to be about 0.5 mb at a bombarding energy of 32.4 MeV (see Table II), in good agreement with the previous data [41].

In the off-line spontaneous fission measurements made by using polyethylene-terephthalate ("Melinex") track detectors there were observed two fission activities – a short-lived activity with $T_{1/2} \approx 30$ min and a considerably weaker, long-lived activity with $T_{1/2} > 50$ h; a contribution from the latter was taken into account in data handling. The long-lived fission activity is likely to be due to ^{252}Cf which could be present in a tiny amount ($\lesssim 10^{-4}$%) in the target material (see also the Fig. 1 spectra showing traces of ^{249}Cf on the catcher). The spontaneous fission background from the $^{249}Cf + {}^4He$ reaction products, such as ^{252}Fm, ^{248}Cf and ^{246}Cf, was negligibly small. Table II shows that, as a result of two bombardments, a total of 126 events of the ground-state spontaneous fission of ^{250}Fm have been detected. A combined analysis of the data obtained in the off-line α-decay and spontaneous fission measurements has allowed us to determine $b_{sf} = (6.9 \pm 1.0) \times 10^{-5}$ and, correspondingly, $T_{sf} = 0.83 \pm 0.15$ yr for ^{250}Fm (see also Table V in Section 4).

The ground-state spontaneous fission of ^{250}Fm was detected also in an independent way, by using the "tape recorder" system. In that experiment 55 spontaneous fission events were observed of which the time distribution is shown in Fig. 2a. From this distribution, a half-life of 26^{+9}_{-6} min was derived using the maximum likelihood procedure [46].

Table II. *Summary of experimental results on detecting the ground-state spontaneous fission of ^{250}Fm and searching for a spontaneous fission branch in the decay of the 1.8-s K isomer ^{250m}Fm*

	W^a [$\mu g\,cm^{-2}$]	I^c	$\sigma^{d,e}_{(\alpha)}$ [mb]	N^f_{sf}	$\sigma^{e,h}_{(sf)}$ [nb]
^{250}Fm	27 ± 3	0.6	0.45 ± 0.09	29 ± 7	29 ± 10
	27 ± 3	1.4	0.53 ± 0.12	97 ± 11	38 ± 9
^{250m}Fm	11 – 29[b]	7.7	-	⩽ 8[g]	⩽ 0.3

[a] Effective target thickness calculated according to Refs. [42, 43], with experimental data of Refs. [44, 45] taken into account.
[b] Four targets of different thickness have been used in these experiments.
[c] Beam dose in units of 10^{17} incident particles.
[d] Formation cross section of ^{250}Fm obtained from α-decay measurements. On the basis of all the measurements performed in the present study (including those not mentioned in the table), the weighted average $\sigma_{(\alpha)}$ value has been determined to be 0.41 ± 0.06 mb at $E^{lab}_{^4He} = 32.4$ MeV.
[e] All cross section values given in the table correspond to the bombarding energy $E^{lab}_{^4He} = 32.4$ MeV.
[f] Number of spontaneous fission events attributed to the decay of ^{250}Fm or ^{250m}Fm. For ^{250}Fm the N_{sf} numbers are those after background subtraction (see the text); for ^{250m}Fm see Fig. 2b.
[g] This result obtained by the maximum likelihood method [46] corresponds to the ≈ 90% confidence level.
[h] Cross sections corresponding to the spontaneous fission branch of ^{250}Fm and ^{250m}Fm.

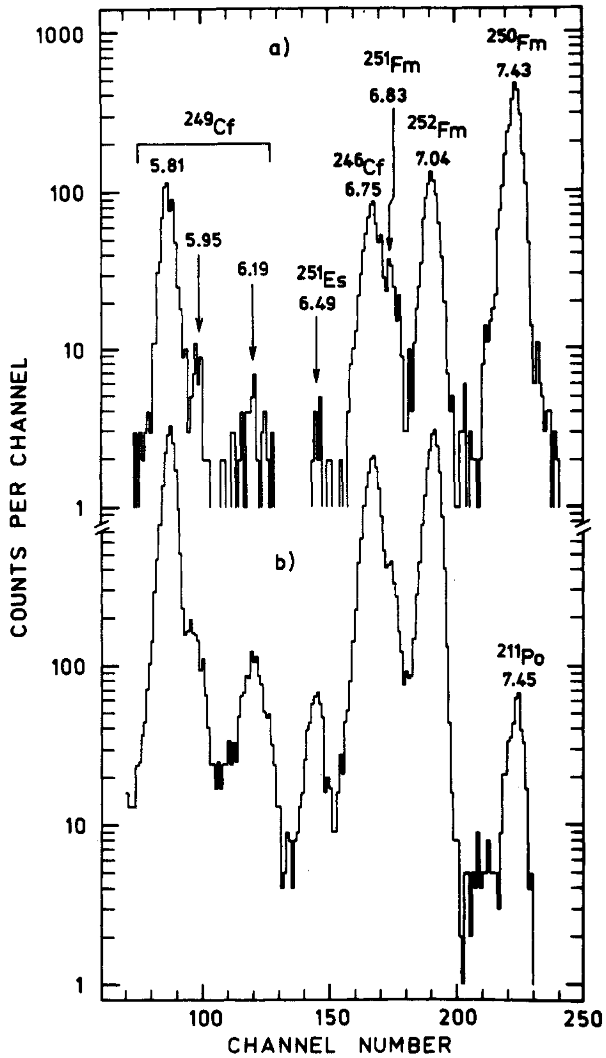

Fig. 1. Alpha-particle-energy spectra from radioactivities produced in reactions of 32.4-MeV ^{4}He projectiles with ^{249}Cf. The spectra were recorded in the time intervals 90–110 min (a) and 14.0–25.1 h (b) following bombardment.

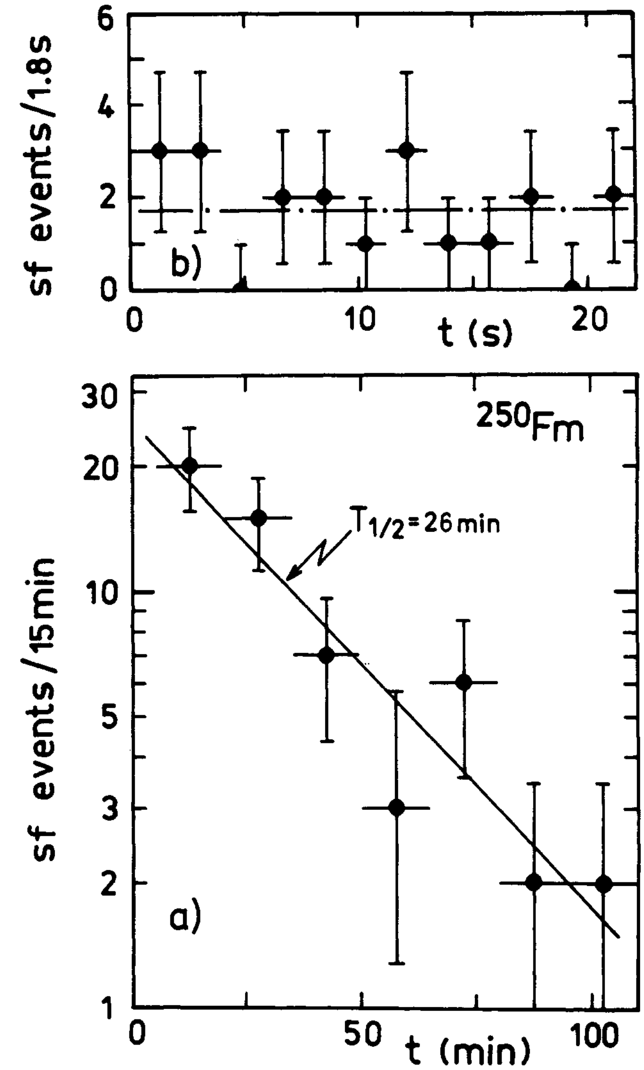

Fig. 2. Time distributions of spontaneous fission events recorded in the reaction ^{249}Cf + ^{4}He at $E^{\rm lab}_{4{\rm He}}$ = 32.4 MeV by using the "tape recorder" system: (a) the distribution obtained in detecting the ground-state spontaneous fission of ^{250}Fm; (b) the distribution obtained in searching for a spontaneous fission branch in the decay of the 1.8-s *K* isomer ^{250m}Fm.

In the search for the spontaneous fission decay of the 1.8-s isomer ^{250m}Fm, a series of bombardments was carried out at a high velocity of the tape of the "recorder" system. A summary of the results obtained is presented in Fig. 2b and in Table II. As demonstrated in Fig. 2b, the time distribution of the recorded spontaneous fission events is practically uniform, showing no evident excess in the initial part. The observed yield of fission activity corresponds to that expected from the ground-state spontaneous fission of ^{250}Fm. An analysis of the distribution of Fig. 2b within the maximum likelihood method [46] makes it possible to set the upper limit for the effect in question (see Table II) and then, taking the isomeric ratio* into account, to establish immediately the lower limit for the ratio of the partial spontaneous fission half-lives of ^{250m}Fm and ^{250}Fm, $T^*_{\rm sf}/T_{\rm sf} \geqslant 0.1$; correspondingly, $b^*_{\rm sf} \leqslant 8.2 \times 10^{-7}$ and $T^*_{\rm sf} \geqslant 0.07$ yr for the *K* isomer ^{250m}Fm (see also Table V in Section 4).

* Here we assumed that the isomeric ratio $\sigma^*_{3n}/\sigma_{3n} \simeq 1.2$ measured by Ghiorso *et al.* [38] at $E^{\rm lab}_{4{\rm He}}$ = 40 MeV has approximately the same value at $E^{\rm lab}_{4{\rm He}}$ = 32.4 MeV.

3. Study of ^{254}102 and ^{254m}102

3.1. *Experimental technique*

The ^{208}Pb(^{48}Ca, 2n) reaction was used to produce ^{254}102 and ^{254m}102. Experiments were carried out at the U-400 cyclotron of the JINR Laboratory of Nuclear Reactions (Dubna) by using the technique described in Ref. [47]. A ^{48}Ca beam struck tangentially the lateral surface of a cooled copper cylinder onto which about 3 mg cm^{-2} of the metallic target material was deposited. This cylindrical target (serving simultaneously as a recoil catcher) rotated with a constant velocity relative to the mica fission fragment detectors arranged around it. The ^{208}Pb target material used in the present study had the following isotopic composition: 99% of ^{208}Pb, 0.6% of ^{207}Pb, 0.4% of ^{206}Pb, $\lesssim$0.01% of ^{204}Pb; some control experiments were performed with a target of ^{206}Pb enriched to 94.9%. Earlier, this setup was widely used in experiments aimed at synthesizing transfermium elements (see, e.g., Refs. [47, 48]) where it permitted the detection of spontaneously fissioning nuclei produced with cross sections in the picobarn region. It was also employed in recent experiments which have led to the discovery of β-delayed nuclear fission in the region of ^{180}Hg [49].

In addition to the on-line spontaneous fission measurements, we performed off-line α-decay measurements to determine the total yield of the ^{254}102 nuclei from an irradiation via the α activity of their long-lived decay products, *viz.*, ^{246}Cf and ^{242}Cm. With this end in view, the entire layer of the ^{208}Pb target material was radiochemically treated after the irradiation in order to separate the fraction of elements from Cm to Fm (see Refs. [48, 50]). Then the prepared sources were measured using the α-activity spectrometer described in Ref. [50]. As a result of numerous experiments performed in recent years at both Dubna and Darmstadt, it has been established that in near-the-barrier bombardments of Pb or Bi target nuclei with $A \gtrsim 40$ projectiles the complete fusion reactions accompanied by the emission of charged particles (protons, α particles or heavier clusters) are strongly suppressed compared with those followed by emission of only neutrons from the composite system (see, e.g., Ref. [48]). Therefore the Cm-Fm nuclides detected in our off-line measurements should be products of the sequential radioactive decay of the $Z = 102$ isotopes formed in the ^{208}Pb(^{48}Ca, xn) reactions.

3.2. *Measurements and results*

At first we carried out control experiments to produce the well known spontaneously fissioning isotope ^{252}102 in the ^{206}Pb(^{48}Ca, 2n) reaction and to determine the dependence of its yield on the ^{48}Ca energy. The results of these experiments are presented in the first four lines of Table III. In all, several hundred spontaneous fission events have been detected of which the time distribution corresponds to a half-life of $2.25^{+0.18}_{-0.16}$ s, in complete agreement with $T_{1/2} = 2.30 \pm 0.22$ s known for ^{252}102 [32, 51]. As the target in our case is "infinitely thick", the energy dependence of the yield of the 2.25-s

Table III. *Summary of experimental results on determining cross sections of the reactions* ^{206,208}Pb(^{48}Ca, xn), *on detecting the ground-state spontaneous fission of* ^{254}102, *and on searching for a spontaneous fission brarch in the decay of the 0.28-s K isomer* ^{254m}102

No.	E^{a}_{cm}	I^{b}	Reaction channel	Detected nucleus	$T_{1/2}$	Δt^{c}_{del}	Δt^{d}_{det}	N^{e}_{sf}	N^{e}_{α}	Y^{f}
					^{206}Pb + ^{48}Ca					
1	169	1.2	2n	^{252}102	2.3 s	1.0 s	12 s	4		0.03
2	174	1.3	2n	^{252}102	2.3 s	1.0 s	12 s	87		0.6
3	179	0.6	2n	^{252}102	2.3 s	1.0 s	12 s	74		1.2
4	201	1.3	2n	^{252}102	2.3 s	1.0 s	12 s	247		1.7
					^{208}Pb + ^{48}Ca					
5	188	14	2n	^{254}102	55 s	24 s	280 s	66		0.011[g]
			2n	^{246}Cf	1.49 d	1.1 d	6.2 d		17 700	6.4
			2n	^{242}Cm	163 d	25.5 d	8.8 d		1 780	6.5
			1n	^{255}Fm	0.84 d	1.1 d	6.2 d		250	0.44[h]
			3n	^{253}Es	20.4 d	25.5 d	8.8 d		100	0.25
			4n	^{240}Cm[i]	27 d	25.5 d	8.8 d		~40	≲0.05[j]
6	212	16	2n	^{254}102	55 s	26 s	300 s	72		0.011[g]
7	177	20	2n	^{254m}102	0.28 s	0.17 s	2.0 s	≤14[k]		≤0.003[g]
			2n	^{254}102	55 s	0.17 s	2.0 s	50[l]		0.005[g,j]
8	180	24	2n	^{254m}102	0.28 s	0.15 s	1.8 s	≤15[k]		≤0.002[g]
			2n	^{254}102	55 s	0.15 s	1.8 s	119[l]		0.008[g,j]
			2n	^{246}Cf	1.49 d	1.6 d	6.1 d		9 800	4.9
			2n	^{242}Cm	163 d	26.2 d	6.9 d		630	4.6
			1n	^{255}Fm	0.84 d	1.6 d	6.1 d		160	0.35[h]
			3n	^{253}Es	20.4 d	26.2 d	6.9 d		9	0.07
			4n	^{240}Cm[i]	27 d	26.2 d	6.9 d		~6	≲0.01[j]

[a] Center-of-mass energy of incident ^{48}Ca particles, in MeV.
[b] Beam dose in units of 10^{15} incident particles.
[c] Time interval between the end of bombardment and the beginning of counting; in spontaneous fission measurements, the time delay Δt_{del} is caused by the absence of mica fission fragment detectors around the zone in which the beam hits the target (see Fig. 5 in Ref. [47]).
[d] Time interval used for counting; in spontaneous fission measurements, the counting time Δt_{det} corresponds to the interval from $\approx 007 t_{rev}$ to $\approx 0.86 t_{rev}$ (where t_{rev} is the period of revolution of the target) so that, for $T_{1/2} \gg t_{rev}$, the detected part of fission activity is equal to $\Delta t_{det}/t_{rev}$ (see also the footnote [c]).
[e] Number of events recorded in observing spontaneous fission or α decay of the detected nucleus.
[f] Relative yield (i.e., yield per one beam particle) for a given xn-deexcitation channel, in units of 10^{-12}. In determining Y, the total probability of the decay chain leading to the detected nucleus as well as the total detection efficiency of the latter have been taken into account so that Y values correspond to the primary products of the (^{48}Ca, xn) reactions. The b_{sf} value for ^{252}102 and the b_{EC} values for ^{255}102, ^{255}Md and ^{253}Fm have been taken from Ref. [32]. For ^{254}102, ^{253}102 and ^{253}Md, the values of $b_{EC} = 0.1$, 0.3 and 1.0, respectively, have been used (see Refs. [54, 58, 59] and the text).
[g] This Y value corresponds to the spontaneous fission branch.
[h] Obtained by assuming the yield of ^{252}Fm being much smaller than that of ^{255}Fm, see the text; also, a small admixture of α activity of ^{253}Fm close in α-particle energy to that of ^{255}Fm has been taken into account.
[i] Revealing the $\simeq 6.29$-MeV α activity of ^{240}Cm has proved to be rather difficult (see Fig. 4) so that Y values given for the 4n-deexcitation channel are only order-of-magnitude estimates.
[j] Obtained by taking into account the yields of the (^{48}Ca, 2–3n) reactions occurring on the ^{206}Pb and ^{207}Pb admixtures in the ^{208}Pb target.
[k] This result obtained by the maximum likelihood method [46] corresponds to the ≈ 90% confidence level.
[l] This effect involves a ≈ 20% contribution from spontaneous fission of the isotope ^{252}102 produced on ^{206}Pb and ^{207}Pb admixtures in the ^{208}Pb target, mostly in the ^{206}Pb(^{48}Ca, 2n) reaction.

fission activity has the form of a rising curve reaching a plateau. The measured yield curve provides an information about the shape and the position of the maximum of the excitation function of the ^{206}Pb(^{48}Ca, 2n) reaction. As for the maximum cross section of this reaction, our measurements give $\sigma_{2n}^{max} = 0.5\,\mu b$ taking into account $b_{sf}(^{252}102) = 0.27$ [51] (see also Table IV).

All the subsequent experiments (numbers 5–8 in Table III) were carried out using ^{208}Pb targets. The b_{sf} value for the ground-state spontaneous fission of ^{254}102 was determined in experiments 5 and 6. A total of 138 spontaneous fission events have been detected of which the time distribution is shown in Fig. 3a. A maximum likelihood analysis of this distribution gives a half-life of 54^{+8}_{-6} s, in excellent agreement with the known value $T_{1/2} = 55 \pm 5$ s [32] determined for ^{254}102 by detecting its α decay. After the termination of bombardment 5, the Cm–Fm fraction was radiochemically separated from the target material. The α-particle-energy spectra resulting from this fraction are shown in Fig. 4. Comparing now the yield of the 54-s spontaneous fission activity with that of the α emitters ^{246}Cf and ^{242}Cm we obtain $b_{sf} = (1.7 \pm 0.5) \times 10^{-3}$ and, correspondingly, $T_{sf} = (3.2 \pm 0.9) \times 10^{4}$ s for the nucleus ^{254}102 (see also Table V in Section 4).

Also, the results of bombardment 5 allow us to obtain information about the cross sections of the reactions ^{208}Pb(^{48}Ca, xn) for $x = 1$, 2 and 3. For a variety of reasons, the properties of the (^{48}Ca, xn) reactions are very important in revealing and understanding the general features of the so-called cold fusion reactions that occur in bombarding targets around Pb by projectiles with masses $A \gtrsim 40$ (see discussions in Refs. [2, 52]). During the last decade, the (^{48}Ca, xn) reactions leading to the isotopes of element 102 were studied at Dubna [53, 54], at Berkeley [55, 56] and at Darmstadt [52, 57, 58]. However, the results obtained in these experiments show considerable discrepancies and in some respects prove to be even contradictory (see, in particular, Table IV to follow). Therefore the new experimental information about the ^{208}Pb(^{48}Ca, xn) reactions appears rather helpful.

The results of our α-decay measurements given in Table III and in Fig. 4 demonstrate that in irradiating a "thick" ^{208}Pb target by ^{48}Ca projectiles at the center-of-mass bombarding energy $E_{cm} = 188$ MeV the largest yield corresponds to the (^{48}Ca, 2n) reaction. The maximum cross section of this reaction derived from the measured yields of ^{246}Cf and ^{242}Cm is $\sigma_{2n}^{max} = 1.7\,\mu b$. The α spectrum of Fig. 4a contains also a

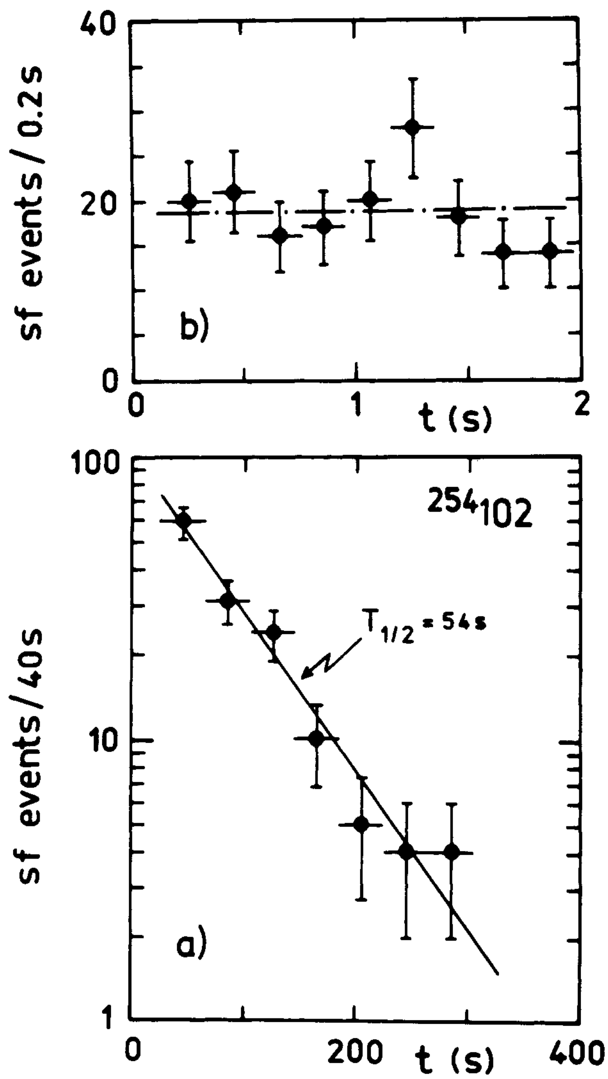

Fig. 3. Time distributions of spontaneous fission events recorded in the ^{208}Pb + ^{48}Ca reaction: (a) the distribution obtained in detecting the ground-state spontaneous fission of ^{254}102 (the net result of bombardments 5 and 6); (b) the distribution obtained in searching for a spontaneous fission branch in the decay of the 0.28-s K isomer ^{254m}102 (the net result of bombardments 7 and 8). See also Table III.

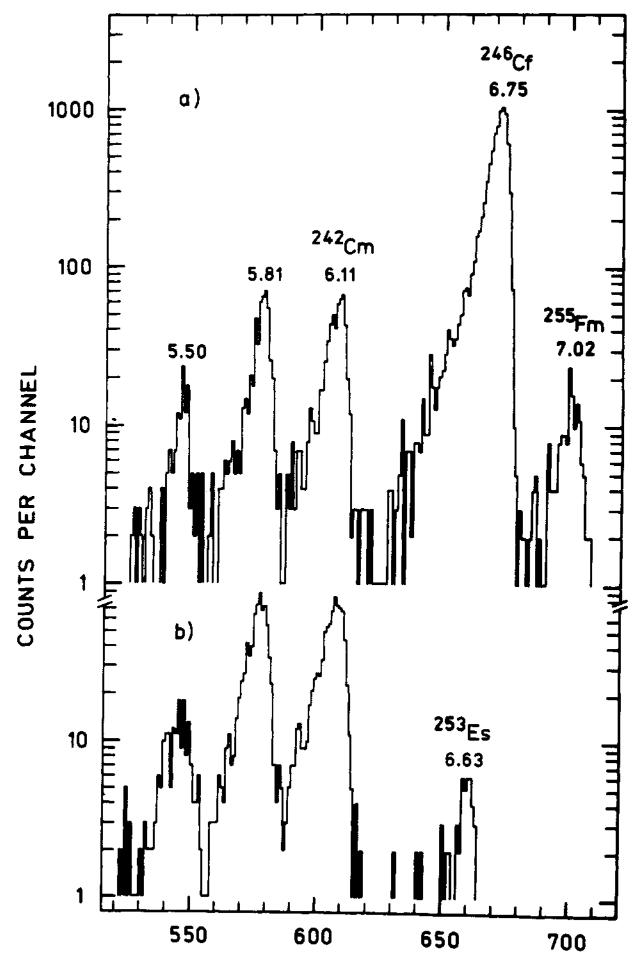

Fig. 4. Alpha-particle-energy spectra from radioactivities produced in reactions of ^{48}Ca projectiles with ^{208}Pb. These spectra recorded in the time intervals 1.1–7.3 d (a) and 25.5–34.3 d (b) after bombardment 5 (see Table III) result from one of the two Si(Au) surface-barrier detectors simultaneously used in the measurements, so that they represent only some 50% of the α events detected. The α groups at ≈ 5.50 MeV and 5.81 MeV are due to the marking activities of ^{241}Am and ^{244}Cm. As regards the 7.02-MeV α group, see the text.

visible peak at E_α just above 7.0 MeV, which we have assigned to ^{255}Fm ($E_\alpha \simeq 7.02$ MeV, $T_{1/2} = 20.1$ h) formed as a result of the ^{208}Pb(^{48}Ca, n) ^{255}102 reaction and of the subsequent decay chain $^{255}102 \xrightarrow{EC(38.4\%)} {}^{255}Md \xrightarrow{EC(93\%)} {}^{255}Fm$. It should, however, be stressed that the α-decay properties of ^{255}Fm are very similar to those of ^{252}Fm ($E_\alpha \simeq 7.04$ MeV, $T_{1/2} = 25.4$ h) [32]. On the other hand, in our case ^{252}Fm can be produced only via the rather exotic reactions (^{48}Ca, γ) and (^{48}Ca, α) whose cross sections are expected to be much lower than that of the (^{48}Ca, n) reaction. Indeed, according to the direct measurements [53], the radiative capture cross section for the system ^{204}Pb + ^{48}Ca does not exceed 0.5 nb whereas the measurements [48] demonstrate the ^{208}Pb(^{48}Ti, n) reaction cross section being at least a factor of 65 larger than that of the reaction ^{208}Pb(^{48}Ti, α). If we neglect contributions coming from the exotic channels of the ^{208}Pb + ^{48}Ca reaction, from the yield of the ≈7.02-MeV α activity we obtain $\sigma_{1n}^{max} = 0.13$ μb for the (^{48}Ca, n) channel. The cross section of the (^{48}Ca, 3n) channel can be estimated from the yield of ^{253}Es. This gives $\sigma_{3n}^{max} \sim 0.1$ μb. Unfortunately this estimate involves somewhat uncertain information [58, 59] about the electron capture branch of ^{253}102 and ^{253}Md, as well as an extrapolated value of the yield of ^{253}Es since a part of the (^{48}Ca, 3n) excitation function is expected to lie at $E_{cm} >$ 188 MeV. Finally, as to the (^{48}Ca, 4n) channel, the determination of σ_{4n}^{max} does not seem possible from the measurements carried out at $E_{cm} = 188$ MeV; we note, however, that at this energy the thick-target yield of ^{252}102 is a factor of at least 100 lower than that of ^{254}102. A comparison of our data on the ^{206,208}Pb(^{48}Ca, xn) cross sections with the results of previous measurements is given in Table IV.

Experiments 7 and 8 designed to search for the spontaneous fission decay of the isomer ^{254m}102 were carried out at an increased rotational velocity of the target, in accordance with the 0.28-s half-life of the isomer. In this case the ground-state spontaneous fission of ^{254}102 should produce a uniformly distributed background. The bombarding energy was chosen so as to cover most of the energy range corresponding to the (^{48}Ca, 2n) excitation function and yet to minimize a contribution from the (^{48}Ca, 4n) reaction leading to the isotope ^{252}102 – the source of an extra spontaneous fission background. The off-line α-decay measurements carried out after bombardment 8 (see Table III) show that the contribution from the reaction ^{208}Pb(^{48}Ca, 4n) can actually be neglected. However, the (^{48}Ca, 2–3n) reactions occurring on the ^{206}Pb and ^{207}Pb admixtures in the ^{208}Pb target give a noticeable yield of ^{252}102.

A total of 169 spontaneous fission events have been detected in bombardments 7 and 8. As seen in Fig. 3b, the time distribution of these events is practically uniform, showing no evident excess in the initial part. The observed yield of fission activity corresponds to that expected from the ground-state spontaneous fission of ^{254}102 and ^{252}102. A maximum likelihood analysis of the distribution of Fig. 3b makes it possible to set the upper limit for the effect in question – 22 events at the ≈90% confidence level. Then, assuming the isomeric ratio to be equal to 0.4*, we obtain immediately the lower limit for the ratio of the partial spontaneous fission half-lives of ^{254m}102 and ^{254}102, $T_{sf}^*/T_{sf} \geqslant 5 \times 10^{-3}$; correspondingly, $b_{sf}^* \leqslant 2.0 \times 10^{-3}$ and $T_{sf}^* \geqslant 140$ s for the K isomer ^{254m}102 (see also Table V).

* Here we employ the isomeric ratio measured by Ghiorso *et al.* [38] for the reaction ^{246}Cm(^{12}C, 4n) ^{254m}102. For the ^{208}Pb(^{48}Ca, 2n) reaction the isomeric ratio may turn out to have a somewhat different value, yet this will in no way affect our conclusion about the high stability of the isomer ^{254m}102 against spontaneous fission (see Section 4).

Table IV. *Maximum cross sections of the* ^{206,208}Pb(^{48}Ca, *x*n) *reactions (in microbarns)*

Reaction	$\sigma_{xn}^{max\,a}$	Reference
^{206}Pb(^{48}Ca, 2n)	0.5	[53]
	0.19 ± 0.03	[57, 58]
	0.5 ± 0.2	present study
^{208}Pb(^{48}Ca, n)	0.40 ± 0.15^{b,c}	[54]
	⩽0.035 (181 MeV)	[55]
	⩽0.03 (172 MeV)	[58]
	0.13 ± 0.06^{c}	present study
^{208}Pb(^{48}Ca, 2n)	4.8 ± 0.7^{d}	[54]
	3.4 ± 0.4	[55]
	0.39 ± 0.07	[57, 58]
	1.7 ± 0.7	present study
^{208}Pb(^{48}Ca, 3n)	⩽0.02 (184 MeV)	[55]
	⩽0.025 (< 181 MeV) ⩽0.133 (181–183 MeV)	[58]
	$0.10^{+0.26}_{-0.05}$	present study

[a] When a (bracketed) value of the ^{48}Ca center-of-mass bombarding energy is indicated, the respective cross section value corresponds to this particular energy rather than to the maximum of the excitation function.
[b] Our estimate obtained on the basis of the experimental data of Ref. [54] using the empirical values [32] for the electron capture branches of ^{255}102 and ^{255}Md, $b_{EC} = 0.384$ and 0.93, respectively.
[c] Obtained on the assumptions that the FWHM of the excitation function is 8 ± 2 MeV whereas the cross sections of the (^{48}Ca, α) and (^{48}Ca, γ) reactions are small compared with that of the (^{48}Ca, n) reaction; see also the text.
[d] Our estimate obtained on the basis of the experimental data of Ref. [54] using the value of 9 ± 1 MeV for the FWHM of the excitation function, in accordance with the results of Refs. [55–58] as well as the data of the present study.

4. Discussion

Our experimental data on the stability of the ground states and 2 q–p isomeric states of ^{250}Fm and ^{254}102 against spontaneous fission are summarized in Table V, together with the results of previous measurements; here one can also find the corresponding data for the ground state of ^{244}Cm as well as for the 2 q–p isomer ^{244m}Cm. As a matter of fact, the ground-state spontaneous fission of ^{250}Fm and ^{254}102 has been detected for the first time and this has allowed us to make an accurate determination of T_{sf} values for both nuclei. Although the results of our measurements do not lead to crucial changes in the T_{sf} systematics, they introduce quantitative certainty into the systematics in its essential part – near the $N = 152$ subshell.

The spontaneous fission decay of the 2 q–p isomers ^{250m}Fm and ^{254m}102 has not been revealed. Our experiments have only enabled the upper limits of T_{sf}^* to be set, which are given in Table V. It is essential to compare these limits with the ground-state T_{sf} values by determining the ratio T_{sf}^*/T_{sf}:

^{244m}Cm	$\geqslant 10^{-5}$	[36]
^{250m}Fm	$\geqslant 10^{-1}$	present study
^{254m}102	$\geqslant 5 \times 10^{-3}$	present study.

Table V. *Summary of experimental results on the stability of the ground states and 2 q–p K-isomeric states of ^{244}Cm, ^{250}Fm and ^{254}102 against spontaneous fission*

Nucleus	Total half-life	Spontaneous fission branch	Partial spontaneous fission half-life	Reference
^{244}Cm	18.1 ± 0.1 yr	$(1.4 \pm 0.1) \times 10^{-6}$	$(1.3 \pm 0.1) \times 10^{7}$ yr	[60]
^{244m}Cm	34 ± 2 ms^a	$\leqslant 8 \times 10^{-12}$	$\geqslant 1.4 \times 10^{2}$ yr	[36]
^{250}Fm	30 ± 3 min^a	$\sim 6 \times 10^{-6}$	≈ 10 yr	[39]
		$(6.9 \pm 1.0) \times 10^{-5}$	0.83 ± 0.15 yr	present study
^{250m}Fm	1.8 ± 0.1 s	< 0.2		[38]
		$\leqslant 8.2 \times 10^{-7}$	$\geqslant 0.07$ yr	present study
^{254}102	55 ± 5 s^a	$\leqslant 7 \times 10^{-4}$	$\geqslant 9 \times 10^{4}$ s	[40]
	68^{+36}_{-18} s	$\leqslant 1 \times 10^{-3}$		[57, 58]
	54^{+8}_{-6} s	$(1.7 \pm 0.5) \times 10^{-3}$	$(3.2 \pm 0.9) \times 10^{4}$ s	present study
^{254m}102	0.28 ± 0.04 s	< 0.2		[38]
		$\leqslant 2.0 \times 10^{-3}$	$\geqslant 1.4 \times 10^{2}$ s	present study

^{a} From Ref. [32].

While the ^{244m}Cm result seems to be inconclusive (see below), our data allow us to state quite positively that, despite the excitation energy $E^* \simeq 1.0$–1.3 MeV, the stability of the 2 q–p isomers against spontaneous fission is practically not inferior to that of the ground states. According to theoretical estimates (see, e.g., Fig. IX-7 in Ref. [3]), all other things being equal, a 200–300 keV change in the energy E of the initial state leads, via eqs. (2) and (3), to a factor of approximately 10^2 variation in the fission barrier penetrability. From this point of view, a factor of 10^6–10^8 decrease in the partial spontaneous fission half-life could be expected for a 2 q–p excited state, as compared with the ground state. Yet our experimental results demonstrate unambiguously that practically no decrease takes place. This means that the specific structure of the 2 q–p isomeric states strongly hinders spontaneous fission. Let us consider possible sources of this hindrance.

The ground states of even–even nuclei are known to have spin-parity 0^+ and in terms of the superfluid model of the nucleus [17–21] they correspond to a q–p vacuum (the number of quasi-particles $v = 0$). The lowest-lying non-collective excited state of an even–even nucleus is a state with one broken pair of nucleons, i.e., a $v = 2$ state with two quasi-particles located on the levels of the average field. The unpaired particles exert strong influence on the superfluid properties of the nucleus and this influence is referred to as the blocking effect [17–21]. In particular, the blocking effect leads to the pairing gap parameter Δ_0 being, *on the average*, by 20–40% smaller for $v = 2$ states than for those with $v = 0$ [17–20]. It should also be emphasized that the blocking effect has rather convincing empirical justifications (see, e.g., Refs. [17–20]).

Thus, a broken pair in the neutron (n) or proton (p) subsystem of the nucleus not only leads to an excitation energy $E^* \simeq 1.0$–1.3 MeV but also entails a significant weakening of nuclear superfluidity. This, in turn, may strongly affect both the potential energy V and the effective mass M associated with fission. In addition, the isomeric states in question possess rather high spins. Therefore, in discussing the stability of q–p isomers against spontaneous fission it is necessary also to take into account the influence exerted on V and M by the spin (quantum number K) of the initial state. Other effects are also possible, for example, some difference between deformation of the ground state and 2 q–p state [19]. On the whole, considerable changes in all the ingredients of the action integral (3) are expected to occur in going from the ground state to a K-isomeric 2 q–p state. It is the combined effect of these changes that will determine the difference between the minimum values of the action integrals S^* and S and thus the value of T^*_{sf}/T_{sf}. If we neglect possible changes in the pre-exponential factor in eq. (1), the logarithm of the T^*_{sf}/T_{sf} ratio can be defined as follows:

$$\delta T_{sf} \equiv \lg \frac{T^*_{sf}}{T_{sf}} = 0.434(S^* - S) = 0.434 S_{emp}\left(\frac{S^*}{S} - 1\right) \tag{10}$$

where S_{emp} is the empirical value of the action integral for ground-state spontaneous fission, which can be found from the experimental T_{sf} value by means of eq. (1).

A further consideration, as we emphasized in Section I, will essentially depend on the approach adopted for treating pairing correlations in the tunnelling process. Let us first discuss the problem in terms of the statical (BCS) approach. In this case changes in the gap parameter in tunnelling are expected to be comparatively small [3, 4, 23], so that a weakening of the pairing in the initial state will proportionally decrease the superfluidity of the system in the subbarrier region of deformations.

Now we shall estimate the partial spontaneous fission hindrance factor due to an increase in the effective mass M in the presence of q–p excitations in the fissioning nucleus; although this problem was considered earlier by Urin and Zaretsky [23], not all of their conclusions proved to be justified. Let us assume for a moment that in eq. (3) the average value of the quantity $[V(q, \Delta) - E]$ does not change in going from the ground state to a 2 q–p isomeric state and that the ratio $[M^*(q, \Delta^*)/M(q, \Delta)] \approx [\bar{M}^*(\Delta^*)/\bar{M}(\Delta)]$ is independent of deformation. Then

$$\frac{S^*}{S} \simeq \left[\frac{\bar{M}^*(\Delta^*)}{\bar{M}(\Delta)}\right]^{1/2}. \quad (11)$$

Since

$$M = M_n + M_p \quad (12)$$

and

$$\frac{\bar{M}_n}{\bar{M}} \simeq \frac{N}{A}, \quad (13)$$

for the relative change in the effective mass, caused by the appearance of a 2 q–p excitation, e.g., in the neutron subsystem of a nucleus, we obtain

$$\frac{\bar{M}^*}{\bar{M}} \approx \frac{Z + N\beta_n^{-2}}{A} \quad (14)$$

where $\beta_n = \Delta^*_{on}/\Delta_{on}$ is the blocking factor. For $\beta_n = 0.7$, eq. (14) leads to $\bar{M}^*/\bar{M} \simeq 1.62$. Hence, by using eqs. (10) and (11) we obtain δT_{sf} estimates of 7.6 and 6.8 for ^{250}Fm ($S_{emp} = 64.3$) and ^{254}102 ($S_{emp} = 57.6$), respectively. Similar estimates can be derived also for the case of 2 q–p excitations in the proton subsystems of these nuclei: $\delta T_{sf} = 5.3$ and 4.8, respectively, for $\beta_p = 0.7$.

As the q–p number v increases, $v > 2$, the average magnitude of the pairing gap should decrease according to theoretical predictions (see, e.g., Refs. [19, 20, 61, 62]) and this effect*, in turn, is expected to cause a further increase in the effective mass associated with fission. For example, for $v = 4$ excitations of the (2n, 2p) type the following estimate can be made:

$$\frac{\bar{M}^*}{\bar{M}} \approx \frac{Z\beta_p^{-2} + N\beta_n^{-2}}{A} \quad (15)$$

where $\beta_{n(p)} \approx 0.6$–0.8. For $\beta_n = \beta_p = 0.7$ it follows from eq. (15) that $\bar{M}^*/\bar{M} \approx 2$. Calculating average pairing gap values for $v = 4$ excited states of the (4n) or (4p) type represents quite a complicated problem; some examples of such calculations can be found, e.g., in Refs. [61, 62]. Finally, if superfluidity of one of the subsystems of a nucleus is destroyed completely, the effective mass for this subsystem should decrease to the independent-particle value $M^{ip}_{n(p)}$ which is known to be several tens of times smaller than the $M_{n(p)}$ value for a superfluid subsystem [3, 4, 23] (remember that for $\Delta \to 0$ eq. (7) is invalid). For example, if superfluidity of the neutron subsystem vanishes, then

$$M^* = M_p + M_n^{ip} \approx M_p \quad (16)$$

and

$$\frac{\bar{M}^*}{\bar{M}} \approx \frac{Z}{A}. \quad (17)$$

Such a situation for, say, ^{250}Fm would lead to $\delta T_{sf} \simeq -10.3$, i.e., to a strong *increase* in the probability for spontaneous fission, which results from a factor of 2.5 *decrease* in the effective mass. In 1966 Urin and Zaretsky [23] suggested that this kind of effect might explain the origin of the spontaneously fissioning isomers of the actinide nuclei, in particular, ^{242f}Am. Surprisingly enough, nowadays some attempts are still being made to relate spontaneously fissioning isomers to q–p excitations at deformation $\varepsilon_0 \approx 0.25$ characteristic of the ground-state potential well of the actinide nuclei (see, e.g., Ref. [63]). At the same time, our experimental results unambiguously show that no acceleration of spontaneous fission takes place for 2 q–p isomeric states in the first potential well; the acceleration can hardly be expected also for 4 q–p isomeric states of the (2n, 2p) type.

The attenuation of pairing correlations not only strongly influences the magnitude of the effective mass but also can lead to some changes in the potential barrier. As the shell correction δU and the correction to the pairing energy, δP, oscillate out of phase with increasing deformation [3], then the attenuation of pairing will generally lead to an increase in the total microscopic correction ($\delta U + \delta P$), see eq. (6). Some idea of the influence of the blocking effect on the potential barrier can be obtained from comparison of the theoretical fission barrier heights calculated for odd-A and odd–odd nuclei (whose ground states are $v = 1$ and $v = 2$ states) with those calculated for neighbouring even–even nuclei ($v = 0$), if the calculations for odd species are performed under the assumption that, during the whole tunnelling process, the odd particle occupies the lowest available orbital near the Fermi surface, irrespective of its spin and parity. Such fission barrier calculations for odd-A, odd–odd and even–even nuclei have been made by Howard and Möller [64] and by Čwiok *et al.* [65, 66] in the framework of the shell correction method [3], with non-axial variations of nuclear shape taken into account. The results of our analysis of these calculations are presented in Table VI. Hence it follows that the blocking effect can lead to a noticeable increase in the fission barrier heights of odd nuclei.

At the same time, an isomeric 2 q–p state is characterized by specific values of quantum numbers related to spins of unpaired nucleons. The quantum numbers Ω_1 and Ω_2 – the projections of spins of unpaired nucleons onto the symmetry axis of the nucleus – can play an especially important role in determining stability against spontaneous fission. If it is required that Ω_1 and Ω_2 (or their sum $K = \Omega_1 + \Omega_2$) should be conserved during the tunnelling motion, then an extra increase in the fission barrier* will arise due to the "specialization energy" [67–69]. In this respect the situation under consideration is similar to the ground-state spontaneous fission of an odd–odd nucleus. Earlier [6–8, 15, 22, 23, 31, 34, 35, 65, 67–74] the specialization effect was repeatedly discussed in connection with the analysis of hindrance factors for ground-state spontaneous fission of odd nuclei. Therefore, without going into details, we shall conclude that, on the average, the concurrent influence of the blocking and specialization effects on the potential energy of deformation should at least cancel the effect of the initial energy gain $\delta E = E^* \simeq 1.0$–$1.3$ MeV associated with passing from the

* Note, however, that there may take place also some other effects capable of changing the effective mass with increasing excitation energy.

* Theoretically, the effective mass can also show some dependence on the quantum numbers of unpaired nucleons (see Refs. [7, 70, 71]). However this effect is expected to play a minor role compared with the influence of unpaired particles on the effective mass through the blocking effect.

Table VI. *Increments (in MeV) of the calculated fission barrier heights for odd-A and odd-odd nuclei due to the blocking effect*

	Howard and Möller [64] $96 \leqslant Z \leqslant 100$, $140 \leqslant N \leqslant 160$			Čwiok et al. [65, 66] $100 \leqslant Z \leqslant 104$, $142 \leqslant N \leqslant 162$		
Odd–even character	$\Delta\bar{B}_f$ [a]	ΔB_f^{min} [b]	ΔB_f^{max}	$\Delta\bar{B}_f$	ΔB_f^{min}	ΔB_f^{max}
even Z, odd N	0.25	0.15	0.45	0.5	0	1.1
odd Z, even N	0.15	0	0.3	0.2	0	0.6
odd Z, odd N	0.4	0.2	0.6	0.5	0	1.1

[a] $\Delta\bar{B}_f$ is the average increment obtained on the basis of 15–20 individual ΔB_f values for nuclei with a given odd–even character. In turn, the increment ΔB_f, e.g., for an even-Z, odd-N nucleus is defined as $\Delta B_f(Z, N) = B_f(Z, N) - \frac{1}{2}[B_f(Z, N-1) + B_f(Z, N+1)]$; for nuclei of other odd–even characters, similar interpolation formulae have been used.

[b] ΔB_f^{min} and ΔB_f^{max} indicate minimum and maximum values of ΔB_f appearing in the calculations.

ground state to a 2 q–p K-isomeric state. In other words, one can hardly expect that the fission barrier for a K-isomeric state will be lower than that for the ground state. On the contrary, the barriers for K-isomeric fission are expected to be increased, by some 1 MeV or even 2 MeV, as suggested by the simplified estimates for ^{250m}Fm and ^{254m}102 made in Ref. [73] where the fission barriers for these isomers were constructed by adding the energy of 2 q–p excitation to the ground-state deformation energy. In Ref. [73] the logarithmic hindrance factors for spontaneous fission of ^{250m}Fm and ^{254m}102 were estimated to be $\delta T_{sf} \simeq$ 3–8 (see also Ref. [74]). We emphasize that these hindrance factors are due *only* to the potential barrier increase. As might be expected, this increase turns out to be considerably different for different assumptions concerning quantum numbers of an isomeric state and their conservation during the tunnelling motion.

All in all, due to the blocking and specialization effects causing a considerable increase in the effective inertia and a noticeable augmentation in the potential barrier, spontaneous fission from 2 q–p K-isomeric states is predicted to be strongly hindered compared to ground-state spontaneous fission. As for the hindrance factors, T_{sf}^{*}/T_{sf} values of the order of 10^5–10^{10} and even greater would not be surprising despite all the uncertainties involved in the quantitative estimates and their sensitivity to assumptions concerning properties of a particular nucleus and structure of a particular isomeric state.

Now let us discuss the problem for the case of the dynamical treatment of pairing correlations [28, 30, 31] which predicts a large enhancement of nuclear superfluidity in the tunnelling process thus considerably changing the hindrance factors for spontaneous fission from q–p isomeric states. The discussion will be done in terms of an analytically solvable model a detailed description and substantiation of which are given in Refs. [28, 31]. In this model the penetrability of a one-humped parabolic barrier depends solely on the magnitude of the dimensionless parameter

$$\kappa = \left[\frac{B_f - E}{g\Delta_0^2}\right]^{1/2} = \left[\frac{B_f - E}{2E_{cond}}\right]^{1/2} \tag{18}$$

where B_f is the barrier height, E is the energy of the initial state ($E = 0$ for ground-state spontaneous fission), $E_{cond} = \frac{1}{2}g\Delta_0^2$ is the condensation energy [20] associated with the presence of the monopole pairing interaction in nuclei, $g = \frac{1}{2}(6/\pi^2)\tilde{a} \simeq A/33$ is the total density of the uniformly distributed, doubly degenerated single-particle levels inclusive of neutrons and protons, and $\tilde{a} = A/10$ is the "macroscopic" level density parameter [20]. In aspects relevant to pairing the model makes no difference between neutrons and protons: the nucleus is considered as a one-component system characterized by a single effective pairing gap parameter. In particular, for the ground state use is made of the mere parametrization $\Delta_0 = 12.84\,\mathrm{MeV}/A^{1/2}$ (see Ref. [75]). As demonstrated in Ref. [31], in the dynamical treatment of pairing correlations the minimum value of the action integral associated with ground-state spontaneous fission is given by the following expression:

$$S_{dyn}(\kappa_0) = S_0\kappa_0 f(\kappa_0) \tag{19}$$

where

$$S_0 = \pi(q_2 - q_1)(F_0 g/2\hbar^2)^{1/2} \tag{20}$$

with $F_0 \equiv \langle F(q)\rangle_q$ and $\kappa_0 = (B_f/g\Delta_0^2)^{1/2}$. The universal function $f(\kappa)$ is defined as

$$f(\kappa) = \frac{4}{\pi}\cdot\frac{(1+\kappa^2)^{1/2}}{\kappa^2}\left[\boldsymbol{E}(k) - \frac{\boldsymbol{K}(k)}{1+\kappa^2}\right] \tag{21}$$

where $\boldsymbol{K}(k)$ and $\boldsymbol{E}(k)$ are the complete elliptic integrals of the 1st and 2nd kind, respectively [76]. The modulus of the elliptic integrals is

$$k = [\kappa^2/(1+\kappa^2)]^{1/2}. \tag{22}$$

We note that in the framework of the accepted model [28, 31] the statical treatment of pairing correlations leads to the well-known formula

$$S_{stat}(\kappa_0) = S_0\kappa_0 = \pi(q_2 - q_1)(B_f F_0/2\hbar^2\Delta_0^2)^{1/2}. \tag{23}$$

Thus

$$\delta T_{sf}^{dyn} = 0.434 S_{emp}\left[\frac{S_0^*\kappa_0^* f(\kappa_0^*)}{S_0\kappa_0 f(\kappa_0)} - 1\right] \tag{24}$$

whereas δT_{sf}^{stat} is given by the same formula for $f(\kappa_0^*) = f(\kappa_0) = 1$.

To make numerical estimates we assume $B_f = 6\,\mathrm{MeV}$ and $g\Delta_0^2 = 5.5\,\mathrm{MeV}$ (see the footnote on page 256 in Ref. [31]). For the blocking factor $\beta = \Delta_0^*/\Delta_0$ we take the value $\beta = 0.82$ which characterizes [20, 77] the one-component $A = 250$ system having a 2 q–p excitation. At first we suppose that the potential barrier is the same for both the isomeric state and the ground state. Then $\kappa_0^* = \kappa_0/\beta$ and eq. (24) gives $\delta T_{sf}^{dyn} \approx 2.9$ and $\delta T_{sf}^{stat} \approx 6.1$ for ^{250m}Fm. However, if we assume the barrier to be, say, 1.5 MeV higher for the isomer, then estimates will give $\delta T_{sf}^{dyn} \approx 4.6$ and $\delta T_{sf}^{stat} \approx 10.1$ for $S_0^* = S_0$. In reality the values of δT_{sf}^{dyn} and δT_{sf}^{stat} can be still larger since one should expect that $S_0^* > S_0$.

Now we see that the dynamical treatment of pairing correlations also leads to a hindrance for spontaneous fission from 2 q–p isomeric states. However, it is an important finding that in the dynamical approach the hindrance factors turn out to be 3–5 orders of magnitude lower than in the statical one. A similar situation takes place also for hindrance factors associated with ground-state spontaneous fission of odd nuclei. This situation was discussed in detail in Ref. [31] where it was shown that the dynamical approach to pairing provides a more adequate solution of the problem since in this case the correct order of magnitude of the hindrance factors can be obtained only if *all* the reasons for hindering spontaneous fission, namely those due to both the blocking and specialization effects, are taken into account simultaneously. By contrast to this, in the statical approach the hindrance factors calculated taking into account all the essential effects turn out to be unreasonably large, exceeding by many orders of magnitude the empirical hindrance factors. Then, in order to fit the statical version of theory to the experiment it is necessary either to neglect completely one of the strong effects (for example, the blocking effect on the effective mass, as is often done) or to weaken several effects simultaneously by making rather artificial assumptions. The possibility of avoiding such manipulations represents an important advantage of the dynamical treatment of pairing correlations in tunnelling.

5. Conclusions

The experimental results obtained in the present study demonstrate that the stability of the 2 q–p *K*-isomeric states in ^{250}Fm and ^{254}102 against spontaneous fission is rather high – it actually is not inferior to that of the ground states of these nuclei. Again, the principal outcome of our theoretical considerations presented in Section 4 lies in that, irrespective of the approach used to treat pairing correlations in tunnelling, spontaneous fission from 2 q–p *K*-isomeric states is predicted to be essentially hindered rather than facilitated compared with ground-state spontaneous fission; quantitatively, the corresponding hindrance factors, T^*_{sf}/T_{sf}, can be expected to vary in a very wide range, say, from 10^2–10^3 to 10^8–10^{10} and more. Thus, we ought to note a good qualitative agreement between the theory and experiment. Remember now that 2 q–p isomeric states can occur not only in the first but also in the second potential well which gives rise to the existence of the spontaneously fissioning shape isomers of the actinide nuclei. In fact, such states lying at an energy of ≈ 1.3 MeV above the bottom of the second well have been observed for a number of even–even Pu and Cm nuclides [34, 35]. For spontaneous fission from these "doubly" isomeric states, the empirical values of the logarithmic hindrance factors $\delta T^{(f)}_{sf} \equiv \lg (T^{*(f)}_{sf}/T^{(f)}_{sf})$ range from 1.1 to 4.3 whereas theoretical estimates similar to those made for ^{250m}Fm in Section 4 give $\delta T^{(f)\mathrm{dyn}}_{sf} \approx 1.6$–3.5 and $\delta T^{(f)\mathrm{stat}}_{sf} \approx$ 2.3–5.7 (see also Ref. [31]).

Confirming the theoretical prediction about the high stability of 2 q–p *K*-isomeric states against spontaneous fission, the available experimental data do not so far allow one to make a decision between the two alternative treatments of pairing correlations in tunnelling. An attempt could be made to decide the issue on the basis of the empirical data for *K*-isomeric states in the second potential well yet it seems to be a difficult task since in this case the difference between the theoretical hindrance factors $(T^{*(f)}_{sf}/T^{(f)}_{sf})^{\mathrm{stat}}$ and $(T^{*(f)}_{sf}/T^{(f)}_{sf})^{\mathrm{dyn}}$ is expected to be not sufficiently large ($\sim 10^1$–10^2) so that it may prove to be obscured by inaccuracies which cannot be avoided even in most realistic calculations of the hindrance factors within each of the two alternative treatments of pairing. For 2 q–p *K*-isomeric states in the first potential well, the difference between the "statical" and "dynamical" hindrance factors is expected to be of the order of 10^3 to 10^5, which is auspicious for solving the problem. However, a substantial increase in experimental sensitivity is required here which would permit observation of spontaneous fission from 2 q–p *K*-isomeric states despite the considerable hindrance. As has been demonstrated in Ref. [31] and emphasized in Section I of the present paper, removal of the ambiguity in treating pairing correlations in tunnelling would be of great importance for a deeper insight into the physics of large-scale subbarrier rearrangements of complex nuclei in fission and fusion. Therefore undoubtedly justified seem to be any efforts to increase the sensitivity of experimental searches for spontaneous fission from q–p isomeric states as well as attempts to perform thorough realistic calculations of the corresponding hindrance factors to replace the order-of-magnitude estimates.

It is extremely difficult to increase the experimental sensitivity to the required level in dealing with q–p isomers in relatively long-lived nuclei for which the probability of ground-state spontaneous fission is low. Thus, for ^{250m}Fm or ^{254m}102 one could try, by using a different technique, to enhance the sensitivity of searches for the spontaneous fission branch by several tens of times relative to the one achieved so far; however, one can hardly achieve more than that. At the same time, the required sensitivity can sooner be obtained in the region of nuclei for which spontaneous fission is the predominant mode of decay whereas partial spontaneous fission half-lives are so short that they fall into the range of characteristic lifetimes for *K*-forbidden γ transitions. Such situations are possible for, say, the known even–even isotopes of kurchatovium – element 104 – which are characterized by $b_{sf} \approx 1$ and $T_{1/2} \approx T_{sf} \sim 10^{-3}$–$10^{-1}$ s; theoretically, the occurrence of *K*-isomeric states in these nuclei is quite probable (see, e.g., Ref. [33]). Detailed theoretical predictions for the occurrence of spin isomeric states in the region of short-lived spontaneously fissioning nuclides as well as the performance of experiments designed specially to search for such states are topical issues. Finally, we would like to emphasize that the existence of a variety of spin isomeric states in heavy nuclei not only opens up new prospects for studies of diverse effects of nuclear structure in cold fission but also may have important implications for the work aimed to synthesize and identify new transactinide nuclei.

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