Observation of Delayed Nuclear Fission in the Region of ^{180}Hg

1987 Europhys. Lett. 4 893

(http://iopscience.iop.org/0295-5075/4/8/006)

EUROPHYSICS LETTERS 15 October 1987
Europhys. Lett., 4 (8), pp. 893-898 (1987)

Observation of Delayed Nuclear Fission in the Region of ^{180}Hg.

YU. A. LAZAREV, YU. TS. OGANESSIAN, I. V. SHIROKOVSKY, S. P. TRETYAKOVA, V. K. UTYONKOV and G. V. BUKLANOV

Laboratory of Nuclear Reactions Joint Institute for Nuclear Research
Head Post Office, P.O. Box 79, 101000 Moscow, USSR

(received 4 March 1987; accepted in final form 21 July 1987)

PACS. 25.85. – Fission reactions.
PACS. 23.40. – Beta decay; electron and muon capture.
PACS. 27.70. – $150 \leq A \leq 189$.

Abstract. – The formation of nuclides undergoing delayed fission with half-lives in the range of $T_{1/2} \geq 1$ s has been observed in fusion-evaporation reactions induced by 230 MeV ^{40}Ca and ^{40}Ar projectiles on isotopically enriched targets of ^{144}Sm, ^{147}Sm and ^{150}Sm. The fission activity with $T_{1/2} = 0.70^{+0.12}_{-0.09}$ s produced in the ^{144}Sm + ^{40}Ca reaction has been found to be the most striking one. The origin of this activity is explained as being due to the β-delayed fission occurring in the chain $^{180}\text{Tl} \xrightarrow[0.7\,\text{s}]{\beta^+(\text{EC})} {}^{180}\text{Hg}^*$. The discovery of β-delayed fission in the Hg region opens up valuable prospects for extending low-energy ($E^* \leq Q_{\beta^+(\text{EC})}$) fission studies to neutron-deficient nuclei of many elements belonging to the translanthanide part of the Mendeleev periodic table.

It is an undoubted fact that the most precise and detailed experimental information about the probability and mechanism of nuclear fission is that gained at low excitation energies that are comparable to, say, the nucleon binding energy. However, until recently low-energy fission, such as neutron-induced and spontaneous fission, remained the «privilege» of the actinide nuclei. At present low-energy fission properties of the actinide nuclei are explored thoroughly enough, so that the experimental results obtained just in the region of Ra to Fm provide both a general basis for understanding the fission mechanism and a source of data for testing fission theories and models. A challenging task now is to extend low-energy fission studies to nuclei outside the well-explored region. In this respect valuable possibilities are offered by the β-delayed fission, *i.e.* fission from excited states populated in the $\beta^{\pm}$-decay or electron capture (EC) [1, 2]. The phenomenon of the β-delayed fission was discovered in 1965 by Flerov and coworkers [3] at Dubna and was subsequently studied also in a number of other laboratories (for a recent review, see ref. [1]). Although until now the β-delayed fission has been observed only for the actinide nuclei, there are good reasons to believe [2] that this phenomenon should also take place for nuclei of considerably lighter elements.

Beta-delayed fission is expected to occur with a detectable probability when the total β-decay energy of a precursor, $Q_{\beta(\text{EC})}$, is comparable to or greater than the fission barrier height of the daughter nucleus, $B_f^{\max}$. Then a certain branch of the β (EC) decay can lead to the population of rather high-lying states of the daughter nucleus which possess large fission widths. Although fission from the states near the barrier top occurs «instantaneously» — on a time scale of $\tau_f \sim (10^{-14} \div 10^{-15})$ s, the population of the fissioning states takes place according to the half-life of the precursor, $T_{1/2} \gg \tau_f$, which in turn determines the time distribution of fission events. Thus there appears a «delay» — the factor that essentially facilitates the performance of experimental studies. As regards the probability of β-delayed fission, $P_{\beta\text{-df}}$ ([1]), it is a very sensitive function of the difference $(B_f^{max} - Q_{\beta(EC)})$: the systematics of empirical $P_{\beta\text{-df}}$ values for the subbarrier β-delayed fission of the actinide nuclei (see table 2 and fig. 19 in ref. [1]) demonstrates that a 1 MeV increase in $(B_f^{max} - Q_{\beta(EC)})$ results in a decrease by at least two orders of magnitude in $P_{\beta\text{-df}}$. This yields a basis for obtaining information on the fission barrier from empirical data on a probability of the β-delayed fission.

As follows from our previous examinations [1, 2] the appropriate conditions for β-delayed fission to occur can be realized in a wide range of neutron-deficient nuclei that can be produced with high yields in heavy-ion reactions, not only in the interesting region of near-magic precursors with $Z = 87 \div 91$ and $N < 126$ such as, *e.g.*, ^{208}Ac, ^{212}Pa or the lighter

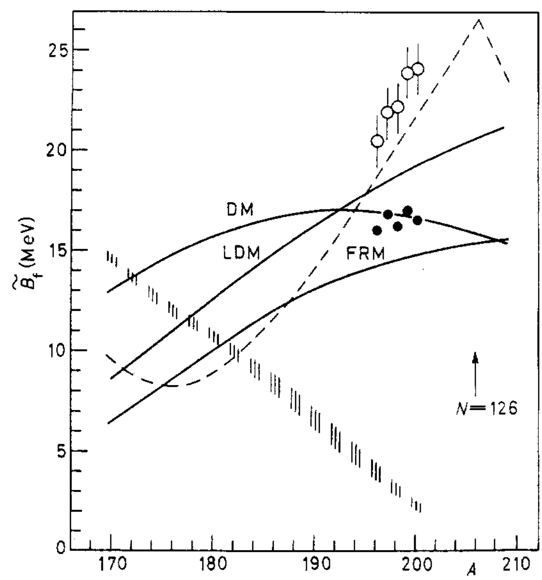

Fig. 1. – The isospin-dependence of the macroscopic part $\tilde{B}_f$ of the fission barrier heights for Hg isotopes, as predicted by the droplet model of Myers (DM) [4], the liquid-drop model of Myers and Swiatecki (LDM) [5], and the finite-range model of Sierk (FRM) [6]. The dashed curve shows the fission barriers $B_f = \tilde{B}_f - \delta U_{g.s.}$ composed of the $\tilde{B}_f$ values by Sierk [6] and of the ground-state microscopic corrections $\delta U_{g.s.}$ by Möller and Nix [7]; note that around ^{180}Hg the $\delta U_{g.s.}$ corrections are small. The open circles represent the empirical fission barrier heights obtained in ref. [8] from statistical-model analyses of the measured excitation functions for fission induced by light charged particles with $A \leq 4$; the closed circles show the macroscopic barrier heights $\tilde{B}_f$ obtained by removing the calculated $\delta U_{g.s}$ corrections of Möller and Nix [7] from the empirical B_f values. The triplets of vertical bars drawn for each even A outline, according to the mass tables [5, 7, 9], the «corridor» of the $Q_{\beta^+(EC)}$ values for the odd-odd Tl isotopes; the bar heights thus reflect a scatter of the $Q_{\beta^+(EC)}$ predictions.

([1]) By definition, $P_{\beta\text{-df}}$ is the probability of fission of the daughter nucleus per one event of the β (EC) decay of the precursor; $P_{\beta\text{-df}} = \sigma_{\beta\text{-df}}/(\sigma_\beta \cdot b_\beta)$, where $\sigma_{\beta\text{-df}}$ is the cross-section corresponding to th measured yield of β-delayed fission events, whereas σ_β and b_β are, respectively, the production cross-section and the β (EC) branch for the precursor.

isotopes of Ac and Pa, but also in the region of *preactinide* elements, in particular, for the ultra neutron-deficient nuclei from Tl to Bi. Figure 1 shows an example of the case of Tl $\xrightarrow{\beta^+(EC)}$ Hg. We see that the β-delayed fission of Hg isotopes becomes possible, if one moves by 15-20 neutrons off the β-stability line. Yet the actual A value at which the β-delayed fission will come into play depends critically on the pattern of change in the macroscopic part $\tilde{B}_f$ of the fission barrier height with respect to the number of neutrons in the Hg nuclei. Therefore, as we have stressed previously [1, 2], the β-delayed fission provides a unique probe for studying the Z, N variations of the fission-associated macroscopic properties of nuclei as well as for determining the fundamental parameters characterizing a drop of nuclear liquid, such as the fissility parameter x or the surface-asymmetry parameter k_s [5] which governs the isospin dependence of the surface energy of a nuclear drop. Under condition of sufficiently high yields, the β-delayed fission could furnish a unique chance also for more detailed studies of the low-energy fission properties of exotic nuclei — for measuring, *e.g.*, the total kinetic energy of fragments or probing gross features of the fragment mass distributions.

In this letter we report on experiments which have led to the detection of β-delayed fission of nuclei in the region of ^{180}Hg.

For producing ultra neutron-deficient Pb, Tl and Hg nuclei, isotopically enriched targets of ^{144}Sm (88.8 %), ^{147}Sm (96.4 %) and ^{150}Sm (95.0 %) were irradiated with 230 MeV ^{40}Ca and ^{40}Ar beams provided by the U-400 cyclotron at JINR, Dubna. The U or Th contamination in the targets did not exceed $2 \cdot 10^{-8}$ by weight. The experiments were carried out by using the technique described in ref. [10]. The ^{40}Ca (or ^{40}Ar) beam with an average intensity of about $5 \cdot 10^{12}$ particles/s struck the lateral surface of a cooled copper cylinder onto which about 2 mg/cm^2 of the target substance 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. Earlier, this technique was widely used in experiments aimed at synthesizing transfermium elements (see, *e.g*, refs. [10, 11] where it permitted the detection of spontaneously fissioning nuclei produced with cross-sections in the picobarn region (1 pb = 10^{-36} cm^2).

In the present exploratory experiments searches were carried out for delayed-fission activities with half-lives $T_{1/2} \geqslant 0.1$ s. The relevant experimental data are presented in table I and fig. 2 from which it follows that the delayed fission has been detected in all the four reactions studied. The most striking fission activity, with $T_{1/2} = 0.70^{+0.12}_{-0.09}$ s, was observed in the ^{144}Sm + ^{40}Ca reaction (fig. 2). The measured yield of the 0.7 s activity corresponds to a cross-section of some 50 pb (with an accuracy within a factor of 2-3). In the reactions on targets of ^{147}Sm and ^{150}Sm, as well as in the ^{40}Ar-induced reaction, the yields of delayed-fission fragments decrease substantially, whereas the time distributions of the fission events recorded indicate a considerable increase in half-lives.

Evidently the origin of any delayed-fission activities should primarily be associated with nuclear-decay processes occurring at the lowest excitation energies. Although at present we *cannot* completely exclude a possible manifestation of more exotic processes, the most probable source of the fission activities observed seems to be the β-delayed fission. It is this phenomenon that has been predicted [1, 2] for the ultra neutron-deficient nuclei of Bi-Tl. If that is the case, an examination of the data of table I in the light of the radioactive properties of the residual nuclei formed after particle emission from the compound system ^{184}Pb with the initial excitation energy $E^* \approx (40 \div 75)$ MeV leads to the assumption that fission with $T_{1/2} = 0.7$ s occurs in the decay chain $^{180}\text{Tl} \xrightarrow{\beta^+(EC)} {}^{180}\text{Hg}^*$. In fact, the radioactive properties of the majority of nuclei from the region of $A \sim 174 \div 184$ and $Z \leqslant 82$ are well known, whereas the properties of unknown nuclides can be estimated reliably by an extrapolation of the empirical data. Again, the value of $T_{1/2} = 0.70^{+0.12}_{-0.09}$ s determined rather accurately, sharply

TABLE I. – *Delayed fission activities detected in the reactions of* ^{40}Ca *and* ^{40}Ar *projectiles with targets of* ^{144}Sm, ^{147}Sm *and* ^{150}Sm

Reaction	I (^{a})	t (^{b})	N_f (^{c})	$T_{1/2}$ (^{d})	Y (^{e})
^{144}Sm + ^{40}Ca	0.42	1.4	87	$0.9^{+\infty}_{-0.3}$	4.0
	0.46	4.9	93	$0.70^{+0.12}_{-0.09}$	4.7
^{147}Sm + ^{40}Ca	0.6	4.8	26	{ $0.3^{+0.7}_{-0.2}$	0.4
				{ >5	0.5
	0.7	19.0	26	—	0.7
	0.8	200.0	16	28^{+14}_{-8}	0.5
^{150}Sm + ^{40}Ca	0.7	5.1	23	—	0.6
	0.2	210.0	5	$\geqslant 30$	0.5
^{144}Sm + ^{40}Ar	0.5	5.2	6	$\geqslant 1.4$	0.3 (^{f})

(^{a}) Beam dose in units of 10^{18} incident particles.
(^{b}) Period of revolution of the target in s.
(^{c}) Number of fission tracks recorded. It corresponds to the time interval of $0.06t$ to $0.85t$, with t being the period of revolution of the target (note that there are no mica detectors around the zone in which the beam hits the target; see fig. 5 in ref. [10]).
(^{d}) Half-life conforming to fission tracks recorded. A dash in this column means that no decay is observed at the given speed of revolution of the target. For all reactions except for ^{144}Sm + ^{40}Ca, the indicated $T_{1/2}$ values should be considered to be effective ones, since the recorded tracks may originate from complex rather than single fission activities.
(^{e}) Relative yield (number of delayed fission events per beam particle), in units of 10^{-16}. The total detection efficiency is taken into account in the determination of Y.
(^{f}) In this reaction the relative yield Y of the 0.7s fission activity does not exceed 0.1.

restricts the range of possible precursors of the β-delayed fission. Thus Pb isotopes with $A < 182$ should predominantly undergo α-decay with half-lives that are expected to be noticeably shorter than 0.7 s. All the neutron-deficient Hg isotopes with $A \geqslant 175$ as well as Au isotopes with $A \geqslant 173$ are presently well known (see ref. [12] and references therein); none of them can serve as a precursor of the β-delayed fission with $T_{1/2} = 0.7$ s. As regards Tl, the lightest known thallium isotope is ^{179}Tl with $T_{1/2} = 0.16^{+0.09}_{-0.04}$ s, which has been observed by detecting its α-decay [12], whereas the isotopes ^{184}Tl and ^{186}Tl are known to undergo β^+ (EC) decay with $T_{1/2} = 11$ s and 26 s, respectively. The Tl isotopes with $A = 180 \div 183$ have so far not been observed (only short-lived isomeric states in ^{181}Tl and ^{183}Tl are known [12]). At the same time, from the above-mentioned data it is clear that the half-life of ^{180}Tl should be close to 1 s, with comparable branches for α and β^+(EC) decay; Takahashi *et al.* [13] predict $T_{1/2\beta^+(\mathrm{EC})} \approx 2$ s for this nucleus. Therefore ^{180}Tl is expected to be the most probable precursor of the β-delayed fission with $T_{1/2} = 0.7$ s. Finally, the fission activities detected in the ^{147}Sm + ^{40}Ca and ^{150}Sm + ^{40}Ca reactions seem to be due to the β-delayed fission in the isobaric chains Tl $\xrightarrow{\beta^+(\mathrm{EC})}$ Hg and/or Pb $\xrightarrow{\beta^+(\mathrm{EC})}$ Tl with larger A values (more probably the even ones); appropriate chains can also be indicated in the case of the ^{144}Sm + ^{40}Ar reaction. In fact, as A increases, the $Q_{\beta^+(\mathrm{EC})}$ values decrease, the half-lives become longer, and the difference $(B_f^{\max} - Q_{\beta^+(\mathrm{EC})})$ grows steeply, see fig. 1. On the other hand, a move towards less neutron-deficient nuclei is accompanied by a considerable increase in their production cross-sections. In addition, branching ratios for ground-state α-decay as well as those for β-delayed proton and α-particle emission decrease sharply in shifting toward the valley of β-stability. As a net result of concurrent but opposite actions of the two above-mentioned groups of factors, the yield on targets of ^{147}Sm and ^{150}Sm or with an ^{40}Ar beam is expected to diminish, compared to the ^{144}Sm + ^{40}Ca case, by one order of magnitude or more, but the cross-sections still remain in the order of several pb.

Thus all the experimental data presented in table I and in fig. 2 can be explained naturally in terms of β-delayed fission.

In bombarding ^{144}Sm by ^{40}Ca ions, ^{180}Tl is formed via the p3n evaporation channel which is expected to have a cross-section of the order of $(0.1 \div 1)$ mb. Then the probability of β-

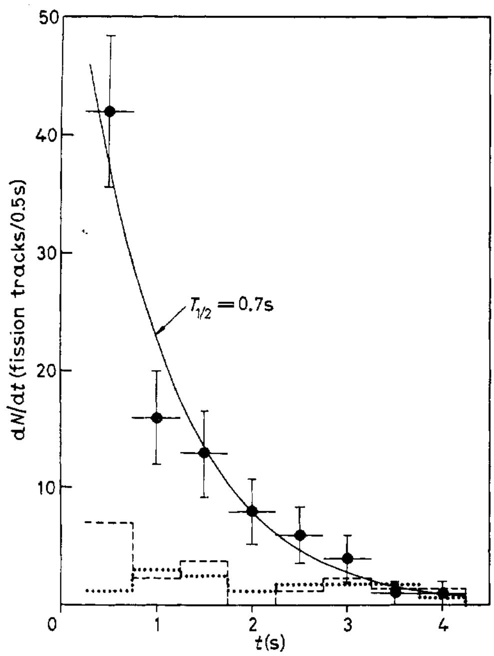

Fig. 2. - The time distributions of the delayed-fission events detected in the reactions of ^{40}Ca projectiles with targets of ^{144}Sm (——) ^{147}Sm (---) and ^{150}Sm (···). For the reactions on ^{147}Sm and ^{150}Sm, the distributions (histograms) are shown after renormalization to the beam dose of $4.6 \cdot 10^{17}$ for which the ^{144}Sm + ^{40}Ca data are presented. Note the linear scale of ordinates.

delayed fission in the $^{180}\text{Tl} \xrightarrow{\beta^+(\text{EC})} {}^{180}\text{Hg}$ chain can be estimated to be $P_{\beta\text{-df}} \sim 10^{-6}$, with an uncertainty of 1-1.5 orders of magnitude. For a more accurate determination of $P_{\beta\text{-df}}$ experimental data are needed on the excitation function of the ^{144}Sm + ^{40}Ca reaction as well as on the β^+(EC) branch for ^{180}Tl. Postponing a quantitative analysis of $P_{\beta\text{-df}}$ until the necessary data have been obtained, we will discuss *qualitatively* what $P_{\beta\text{-df}} \sim 10^{-6}$ means in the context of information about the fission barrier height of the ultra neutron-deficient *cold* nucleus ^{180}Hg. First of all the comparatively low value of $P_{\beta\text{-df}}$ indicates that the β-delayed fission observed is a subbarrier process. Yet this process appears to be only slightly subbarrier. In fact, using as a rough guide the systematics of empirical $P_{\beta\text{-df}}$ values for the subbarrier β-delayed fission of the actinide nuclei (see fig. 19 and table 2 in ref. [1]), one can conclude that the fission barrier height for ^{180}Hg is unlikely to exceed by more than $(0.5 \div 1)$ MeV the $Q_{\beta^+(\text{EC})}$ value for ^{180}Tl which is predicted to be $(10.5 \div 11)$ MeV [5, 7, 9]. One should also bear in mind that, in contrast to the actinides, a much steeper dependence of $P_{\beta\text{-df}}$ upon $(B_f^{\max} - Q_{\beta^+(\text{EC})})$ is expected for the mercury region. Firstly, for the Hg isotopes fission barriers are anticipated to be appreciably «broader» than, say, for the Cm-Cf isotopes for which, furthermore, the $P_{\beta\text{-df}}$ values seem to be determined by the penetrability of only one (inner) peak of the double-humped barrier. Secondly, in the vicinity of the proton drip line, where the $Q_{\beta^+(\text{EC})}$ values are rather large, β-delayed proton and/or α-particle emission may compete with the β-delayed fission and thus reduce the probability of the latter. Bearing in mind all the above-mentioned points, we believe that a reasonable estimate for the fission barrier amplitude of the cold nucleus ^{180}Hg is given by the value $B_f^{\max} \approx (11 \div 12)$ MeV.

Returning now to fig. 1 one can confront this $B_f^{\max}$ estimate with theoretical predictions for the macroscopic fission barrier heights $\tilde{B}_f$ of the ultra neutron-deficient Hg isotopes. As is clear from fig. 1, the very fact of occurrence of the β-delayed fission of ^{180}Hg with a detectable probability makes it possible to conclude that the droplet model [4] fails to properly predict both the magnitude and the isospin dependence of B_f. With respect to the liquid drop model [5] and the finite range model [6], their predictions do not contradict our empirical data for ^{180}Hg; however, more definitive conclusions about the degree of adequacy of these versions of the macroscopic theory will become possible only after more quantitative information about the fission barrier height of ^{180}Hg and of nearby ultra neutron-deficient Hg nuclei has been extracted from supplemental data on the probability of β-delayed fission. Obviously, for extracting such information it will be necessary to know the beta strength function [14] and, in addition, to have certain (*a priori*) notions about the landscape of the potential-energy surface associated with fission of the nuclei under study.

In conclusion, the detection of β-delayed fission in the region of ^{180}Hg signifies that low-energy fission is not a privilege of the actinide species. Judging from our findings in the Hg region, ultra neutron-deficient nuclei of many other preactinide elements should also undergo β-delayed fission. Proven by the present experiments this really widespread occurrence of β-delayed fission opens up new prospects for studying the probability and mechanism of low-energy fission at considerable variations of Z, N and N/Z of the fissioning nucleus.

* * *

We take pleasure in thanking Academician G. N. FLEROV for his encouraging attitude towards this work. Our debt concerning this work is owed to the late A. A. PLEVE, who made a valuable contribution to the development of the highly stable, isotopically enriched samarium targets. We wish to thank also B. N. GIKAL, V. B. KUTNER and the U-400 cyclotron staff for providing these experiments with the intense beams of calcium and argon ions.

REFERENCES

[1] OGANESSIAN YU. TS. and LAZAREV YU. A., in *Treatise on Heavy Ion Science* edited by, BROMLEY D. A., vol. 4, (Plenum Press, New York, N. Y.) 1985, pp. 1-251.

[2] LAZAREV YU. A., OGANESSIAN YU. TS. and KUZNETSOV V. I., *Paper presented at the International IUPAC Symposium on the Synthesis and Properties of New Elements, Dubna, September 23-27, 1980*; JINR preprint E7-80-719, Dubna (1980).

[3] KUZNETSOV V. I., SKOBELEV N. K. and FLEROV G. N., *Yad. Fiz.*, 4 (99) (1966) 279; 5 (1967) 271.

[4] MYERS W. D., *Droplet Model of Atomic Nuclei*, (IFI/Plenum, N. Y.) 1977.

[5] MYERS W. D. and SWIATECKI W. J., *Nucl. Phys.*, 81 (1966) 1; Report UCRL-11980, Berkeley (1965).

[6] SIERK A. J., *Phys. Rev. C*, 33 (1986) 2039.

[7] MÖLLER P. and NIX J. R., *Nucl. Phys. A*, **361** (1981) 117; *At. Data Nucl. Data Tables*, **26** (1981) 165.

[8] IGNATYUK A .V., SMIRENKIN G. N., ITKIS M. G., MUL'GIN S. I. and OKOLOVICH V. N., *Fiz. Elem. Chastits At. Yadra*, 16 (1985) 709.

[9] LIRAN S. and ZELDES N., *At. Data Nucl. Data Tables*, 17 (1976) 411.

[10] OGANESSIAN YU. TS., in *Proceedings of the International School-Seminar on Heavy Ion Physics, Alushta, USSR, April 14-23, 1983* JINR Report D7-83-644, p. 55, Dubna (1983).

[11] OGANESSIAN YU. TS., HUSSONOIS M., DEMIN A. G., KHARITONOV YU. P., BRUCHERTSEIFER H., CONSTANTINESCU O., KOROTKIN YU. S., TRETYAKOVA S. P., UTYONKOV V. K., SHIROKOVSKY I. V. and ESTEVEZ J., *Radiochim. Acta*, 37 (1984) 113.

[12] SCHNEIDER J. R. H., HOFMANN S., HESSBERGER F. P., MÜNZENBERG G., REISDORF W. and ARMBRUSTER P., *Z. Phys. A*, **312** (1983) 21.

[13] TAKAHASHI K., YAMADA M. and KONDOH T., *At. Data Nucl. Data Tables*, **12** (1973) 101.

[14] KLAPDOR H.-V., *Prog. Part. Nucl. Phys.*, **10** (1983) 131.