ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

ДУБНА

E15 - 8134

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1974

ЛАБОРАТОРИЯ ЯДЕРНЫХ ПРОБЛЕМ

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Submitted to *Nuclear Physics*

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E15 - 8134

Деление ^{232}Th, ^{238}U и ^{235}U при взаимодействии с отрицательными мюонами

Получены экспериментальные данные о вероятности мгновенного и запаздывающего деления тория-232, урана-235 и урана-238 при взаимодействии с отрицательными мюонами.

Проводится сопоставление полученных данных о фотоделении.

Препринт Объединенного института ядерных исследований.
Дубна, 1974

Chultem D., Cojocaru V., Ganzorig Dz., Kim Si Chwan, Krogulski T., Kuzniecow W.D., Ortlepp H.G., Polikanov S.M., Sabirov B.M., Schmidt U., Wagner W.

E15 - 8134

Fission of ^{232}Th, ^{238}U and ^{235}U Induced by Negative Muons

The absolute yields of prompt and delayed fission induced by negative muons in ^{232}Th, ^{238}U and ^{235}U have been measured. The yields of delayed fission are much lower than could be predicted from Γ_n/Γ_f systematics for 15-20 MeV nuclear excitation. The systematics of prompt fission yields is compared with photofission data recently obtained. It is suggested that prompt fission can be used for investigating the channel structure of the fission barrier.

Preprint. Joint Institute for Nuclear Research.
Dubna, 1974

1. Introduction

The muon can interact with the nucleus on both stages of the capture: in atomic processes, cascading down its orbits, and in the absorption process: $(Z,N) + \mu^- \to (Z-1,N+1)$.

In heavy nuclei the latter proceeds with a characteristic mean-life time about 80 nsec and the mean excitation energy of the nucleus is estimated to be 15-20 MeV /1/. This is sufficient to open the fission channel in the de-excitation modes of the nucleus.

As has been pointed out by Wheeler /2/, the interaction between the muon and the heavy nucleus in the muonic atom could lead to the excitation of the nucleus also sufficient to involve the fission channel. But that kind of fission differs clearly from the other one mentioned. It proceeds in time interval characteristics for muon cascading, e.g., 10^{-14} - 10^{-15} sec from the moment of μ-atomic capture. Therefore, it is referred to as the prompt fission process. There are some of its features which seem to be quite interesting from the point of view of fission barrier investigations.

2. Experimental Set-Up

In the present experiment we used the fast many-plate methane-filled ionization chamber. In the control measurement of the $f-\gamma$ prompt coincidences the resolution time was 2.5 nsec. The chamber was loaded with about 300 mg of ^{235}U and the fragments were detected with efficiency not poorer than 90%.

All our measurements were performed simultaneously for two isotopes: the investigated one and ^{238}U, as a reference. The targets were deposited by sedimentation on the aluminium foils about 13 mg/cm^2 thick. The natural uranium and ^{235}U (95% of isotopic purity) were deposited in the form of U_3O_8 oxides. A thorium target (monoisotope ^{232}Th) was prepared in the form of ThO_2 oxide.

The targets were prepared in the form of discs with a diameter of 47 mm and 2.6 mg/cm^2, 0.7 mg/cm^2 and 3.1 mg/cm^2 thick for ^{238}U, ^{235}U, and ^{232}Th, respectively. In the measurements the chamber contained: 857 mg of ^{238}U, 237 mg of ^{235}U, and 1020 mg of ^{232}Th.

The measurements were performed with the μ^- and π^- separated beams of the Dubna 680 MeV synchrocyclotron. The μ^- beam of 98% purity with a mean energy of 85 MeV and dispersion $\Delta E = 9$ MeV was slowed down in the moderator of 30 g/cm^2 equivalent thickness.

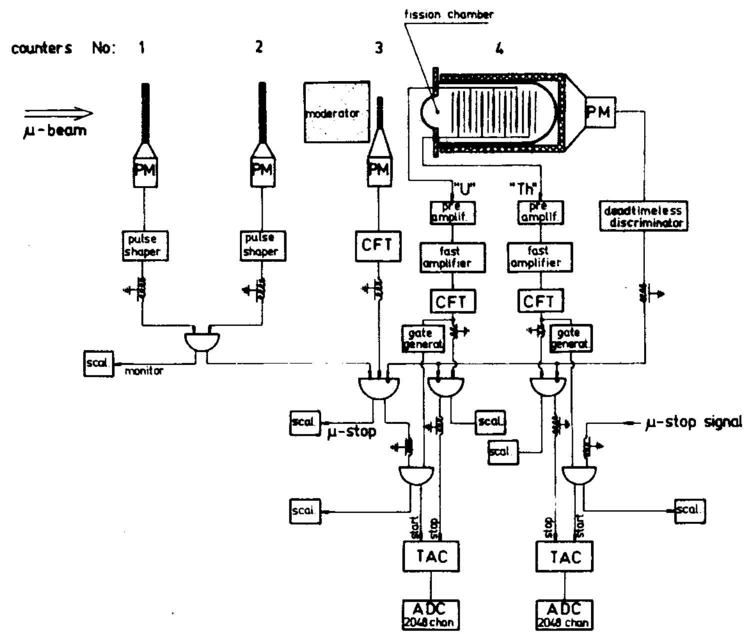

Fig. 1. A block-diagram of the electronics used in conjunction with the fission chamber and telescope counters.

Two identical fast systems working in the constant fraction regime were used for developing the signals from both halves of the fission chamber. The used system is described in details in ref./3/ and only the preamplifiers have been adapted specially to operate with our chamber.

3. Measurements and the Analysis

In every run typically 30-35 hours long both μ^- and π^- beams were used. Due to the measurements with π^- we could control the stability of the timing system and analyse the response curve of prompt coincidences for every run independently.

The monitoring system included the control of:

a) the beam intensity - (1,2) coincidence rate;

b) the rate of μ -stops - (1,2,3,$\bar{4}$) coincidences;

c) the rates of the counts in the chamber;

d) the rates of (1,2,3,$\bar{4}$,f) coincidences.

All these quantities had fluctuated but normalized to the beam intensity or μ -stop rate were quite stable. The ratio of the coincidence rates (1,2,3,$\bar{4}$,f) from the two halves of the chamber was very stable in all measurements.

All these measurements were analysed in the standard procedure order. Firstly, the primary spectrum was summed over every 20 channels and fitted by an exponential curve on the whole interval except the prompt peak region and the region, where the background was comparable with the measured effect. The random coincidence level was measured in the negative part of the time spectrum (to the left of the prompt peak). The sensitivity of the results to the choosen fitting interval and the estimated background level was checked.

In the second step, the time distribution of fission induced by μ^- were fitted by the curve composed of: 1) a prompt coincidence response curve defined in the measurements with π^-, 2) its convolution with the exponential curve of the mean life-time defined in the first step of the analysis, 3) the constant background.

The final results of the described procedure have given us the ratios of prompt to delayed fission and the yields of fission induced in ^{232}Th and ^{235}U relative to the yield of fission in ^{238}U.

The yield of fission per μ-capture, Y_{μ}^{f}, has been measured for ^{238}U. We detected simultaneously the number of the fission events and the mesic X-ray spectrum of uranium, aluminium and other materials being inside the chamber. For X-ray spectrum detection a 27 ccm, coaxial Ge(Li) spectrometer was used.

The X-ray spectrum was measured in coincidence with μ-stop events with the time resolution of $2\tau = 10$ nsec. The chamber contained about 1.6 g of U_3O_8. It has occured to be too small an amount to observe the muonic transitions in uranium with sufficient statistics. At the same time the known $2p-1s$ transition of aluminium has been observed with good statistics. As in our chamber the aluminium was present only in the form of the electrodes, the geometrical conditions relatively to the μ-beam have been practically identical to that for the uranium targets. It made possible to find the total μ-capture rate in uranium having this quantity for aluminium and using the range-energy relations for muons in Aℓ and U.

This quantity for aluminium is

$$N_{\mu}^{A\ell} = \frac{N_{\gamma}^{A\ell}}{\epsilon_{\gamma} I_{2p-1s}^{A\ell}},$$

where $N_{\mu}^{A\ell}$ is the number of muons stopped in aluminium, $N_{\gamma}^{A\ell}$ is the observed intensity of the $K_{\alpha}(A\ell)$ transition, ϵ_{γ} is the efficiency of the Ge(Li) detector for the energy of this transition and $I_{2p-1s}^{A\ell}$ is the intensity of K_{α} transition per μ-capture in Aℓ taken from ref. /4/.

As soon as the number of μ-captures in uranium targets N_{stop} is established, the fission yield per μ-capture, Y_{μ}^{f}, is:

$$Y_{\mu}^{f} = \frac{N^{f}}{N_{stop} \times \epsilon_{f}},$$

where N_f is the number of fission events measured simultaneously with the X-spectrum, and ϵ_f means the efficiency of the fission chamber.

We found the efficiency of the chamber in the experimental conditions to be (58 ± 5)%.

4. Results

4.1. π^- Induced Fission

The examples of the time distribution of π^- induced fission and the results of the Gaussian curve fitting are shown in fig. 2.

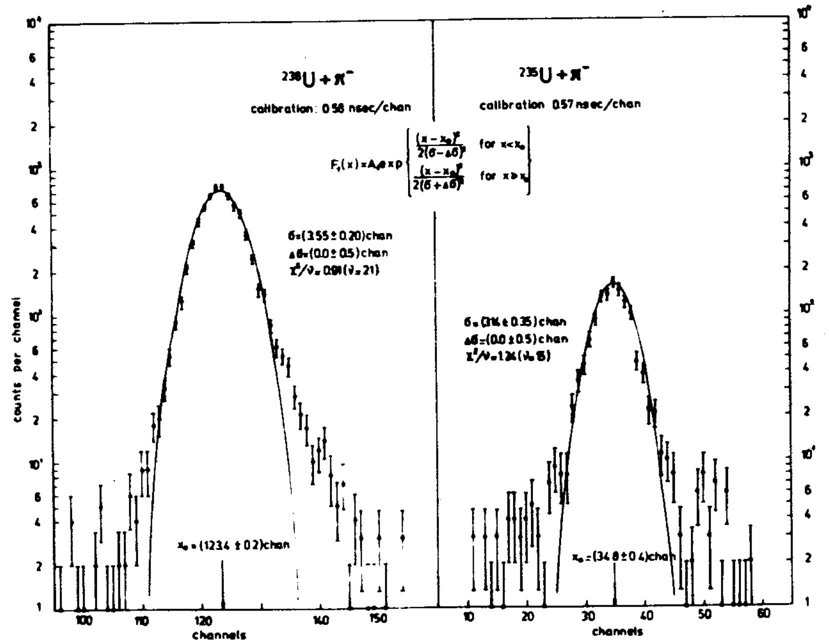

Fig. 2. Two examples of the time distribution of fission induced by π^-.

We have obtained the relative probability of fission induced by π^- in ^{238}Th, ^{235}U, , and ^{238}U. They are (43±3)%, (120±5)% and 100%, respectively. Many papers have been published (for instance see refs. /5,6/) in which the fission probability per π^--capture in ^{238}U has been measured. If one takes the mean value 0.45 ± ± 0.10, then the values 0.22 ± 0.05 for ^{232}Th and 0.54 ± ± 0.12 for ^{235}U are obtained from our results.

4.2. μ^--Capture Mean-Lives

The time distributions of fission events induced by μ^- in three investigated isotopes are shown in fig. 3. To

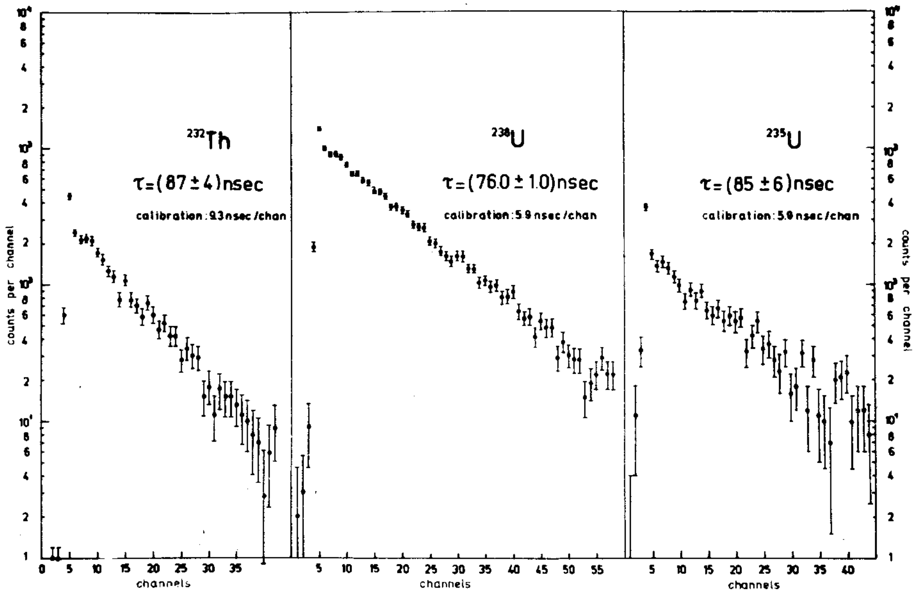

Fig. 3. Time distribution of the fission events in ^{232}Th, ^{238}U , and ^{235}U indiced by μ^- (Background has been subtracted).

obtain them, every 20 channels in the primary spectra have been summed. The mean lives have been found by the least squares method, with the confidence level of 0.05. Our results are compared with the known data in Table 1. Satisfactory good agreement for the ^{238}U isoto-

pe (except Sens' result) and significant disagreement for ^{232}Th and ^{235}U are observed.

Table 1

	mean lives τ_μ(nsec)		
	^{232}Th	^{238}U	^{235}U
J.C.Sens [8)]		88 ± 4	
J.A.Diaz et al [9)]	74.2 ± 5.6	75.6 ± 2.9	66.5 ± 4.2
B.Budick et al [10)]		74.1 ± 2.8	65.3 ± 2.8
present work	87 ± 4	76.0 ± 1.0	84 ± 6

4.3 Ratio of the Prompt-to-Delayed Fission Yields

The final analysis of the time distribution is shown in fig. 4. The weighted yield ratios of the prompt-to-delayed fission for the three measured isotopes are compared with the other results in table 2. In connection with some evident disagreements in table 2 it should be noted that treating simply the prompt component as a deviation from the exponential law, a systematic error of 50% is possible if the time resolution of the used system is about 3÷5 nsec.

4.4. The Fission Yield per μ^--Capture

The compilation of all available data on the fission yield per μ^--capture is presented in table 3. Using the data from tables 2 and 3 we obtain our final experimental

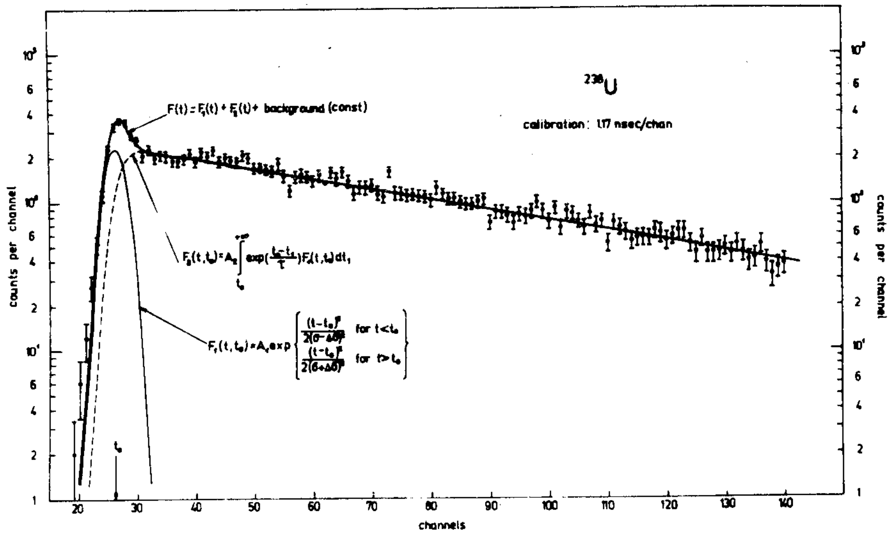

Fig. 4. Illustration of the time distribution analysis procedure.

Table 2

	prompt to delayed fission yields ratio		
	^{232}Th	^{238}U	^{235}U
J.A.Diaz et al.[9]	0.064 ± 0.022	0.072 ± 0.014	0.111 ± 0.021
B.Budick et al.[10]		0.048 ± 0.025	0.063 ± 0.025
present work	0.130 ± 0.012	0.071 ± 0.003	0.17 ± 0.01

Table 3

	fission yield per μ-capture		
	^{232}Th	^{238}U	^{235}U
W.Galbraith and W.J.Whitehouse [14]		<0.25	
W.John and W.F.Fry [13]		0.15 ± 0.06 0.07 ± 0.03	
M.G.Petrascu, A.K.Mihul [11]	0.018 ± 0.012		
G.E.Belovitskii et al [12]		0.070 ± 0.008	
present work *)	0.0043 ± 0.0010	0.031 ± 0.007	0.037 ± 0.009

x) the errors of the relative fission yield $\frac{Y^f(^{232}Th)}{Y^f(^{238}U)}$ and $\frac{Y^f(^{235}U)}{Y^f(^{238}U)}$ are about one order lower

Table 4

	fission yield per μ-capture		
	^{232}Th	^{238}U	^{235}U
prompt	$(5.0 \pm 1.2) \times 10^{-4}$	$(2.03 \pm 0.45) \times 10^{-3}$	$(5.1 \pm 1.2) \times 10^{-3}$
delayed	$(3.8 \pm 0.9) \times 10^{-3}$	$(2.90 \pm 0.65) \times 10^{-2}$	$(3.2 \pm 0.8) \times 10^{-2}$

results: the absolute probability of the prompt and delayed fission yields. They are presented in table 4 and make the basis of the discussion in the next section.

5. Discussion

Figure 5 shows our results as a function of the fissility parameter x , defined accordingly to Meyers and Swiatecki /7/. The quoted errors represent uncertainty in the relative yields only and do not include the 25% uncertainty in the absolute yield values (see, table 4).

5.1. Delayed Fission

If the nuclear μ^--capture reaction is well described in the equilibrium process terms, i.e., as going via the compound system from the very beginning, then our results on the fission yields of ^{232}Ac , ^{238}Pa , and ^{235}Pa should be describable in the Γ_n/Γ_f terms. As for these isotopes we have no data we inter- and extrapolated the known systematics of Γ_n/Γ_f values /15/. The results of the analysis are presented in table 5.

It is quite evident from this table that the assumption on the nucleus attaining the equilibrium state at the full excitation energy 15÷20 MeV characteristics for μ^--capture is unacceptable in any case even taking into account the uncertainties of the extrapolation procedure (estimated to be mostly the factor two).

5.2. Prompt Fission

Figure 5 shows the photofission probability for photon energy corresponding to K_α mesic transitions in the nuclei under consideration. The results published in refs./16–18/ and Huizenga's estimation of the photoabsorption cross section in the actinides /19/ were used. Figure 6 shows the measurements of Khan and Knowles /16/ , Yester et al. /17/ and Anderl et al. /18/ dealing with the near threshold photofission cross section of ^{232}Th , ^{238}U ,

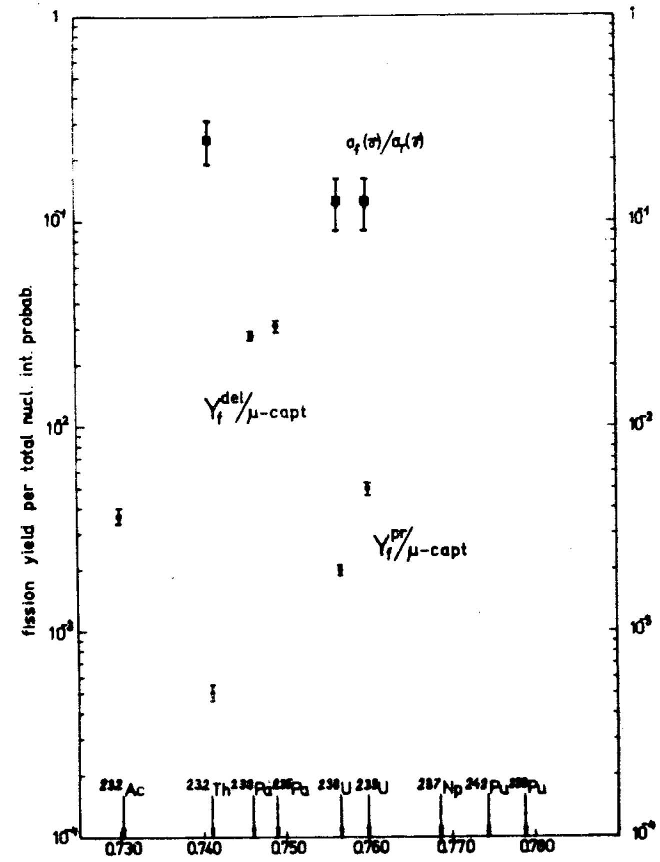

Fig. 5. The absolute yield of prompt and delayed fission induced by μ^- capture as a function of the fissility parameter x /7/. The probability of the photofission for the photon energy corresponding to K_α transitions is also quoted (cross sections $\sigma_f(\gamma)$ from ref. /16/ and $\sigma_T(\gamma)$ from ref./19/ are taken).

Table 5

delayed fission yield per μ-capture

nucleus	estimation: E^x (MeV)	estimation: first chance fission	estimation: first+second chance fission	exp. results (present work)
^{232}Ac	$15 \div 20$	8×10^{-3}	24×10^{-3}	$(3.8 \pm 0.9) \times 10^{-3}$
^{231}Ac	$8 \div 12$	16×10^{-3}		
^{238}Pa	$15 \div 20$	5×10^{-2}	13×10^{-2}	$(2.90 \pm 0.65) \times 10^{-2}$
^{237}Pa	$8 \div 12$	8×10^{-2}		
^{235}Pa	$15 \div 20$	12×10^{-2}	32×10^{-2}	$(32 \pm 0.8) \times 10^{-2}$
^{234}Pa	$8 \div 12$	20×10^{-2}		

and ^{235}U. The vertical bars indicate the energies of K_a mesic transitions in these nuclei. The prominent structure in the excitation curves is discussed by the authors as revealance of the sub- and near-barrier channel structure. The neutron competition can be practically excluded as responsible for such a structure, but a fluctuation in the photoabsorption cross section cannot be a priori rejected.

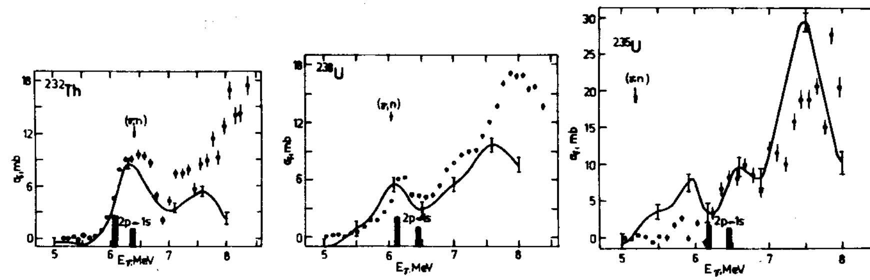

Fig. 6. The compilation of photofission excitation functions for ^{232}Th, ^{238}U, and ^{235}U as measured by Khan and Knowles /16/ Yester et al./17/ and Anderl et al. /18/ The energy and relative intensities of the spin-orbit muonic doublets $2p_{3/2} - 1s_{1/2}$ and $2p_{1/2} - 1s_{1/2}$ are also indicated.

Below we shall discuss to what extent our results on the radiationless transition fission are coherent with the photofission data and their interpretation, and what additional information about the fission barrier could be obtained dut to this analysis.

Firstly, we should analyse if K_β (and higher Lyman's series transitions) and quadrupole transitions $3d - 1s$ could compete with K_a transition in the observed prompt fission yields.

One general assumption, as it has been pointed out in section 1, is unavoidable: The nuclear excitation in the radiationless transitions of the muon in its atomic orbits is governed by the same interaction like photoabsorption. We understand this assumption in the sense presented by

Zaretsky and Novikov /20/. Thus, we mean that photoabsorption cross sections for thorium and uranium isotopes are in the same relation like the radiationless transition probabilities

$$\frac{\Gamma_{r\ell}}{\Gamma_{r\ell}+\Gamma_x} \propto \sigma_T(\gamma) ,$$

where $\Gamma_{r\ell}$ and Γ_x are the widths of radiationless and muonic transitions. Secondly, we state that the nonradiative fission is not affected by the moun bound in the 1s orbit, if the excitation energy is $\gtrsim$ 9 MeV (the region of K_β, K_γ and 3d-1s transitions).

In view of that we can reject the supposition that the observed fission yields are conditioned up for all three isotopes by higher than K_α transitions.

In fact, the photofission cross sections for the investigated nuclei are comparable in the region of 8.5 - 9.0 MeV (see, fig. 6). Thus, if the $\sigma_T(\gamma)$ values are also comparable, then

$$P_0^f(\mathrm{Th}) \approx P_0^f(\mathrm{U})$$

but:

$$P_\mu^f(\mathrm{Th}) << P_\mu^f(\mathrm{U})$$

as follows from our results (P_μ^f , P_μ^f are the photofission probabilities for the nucleus with and without the moun in its 1s orbit).

It is easily seen that any other relation between the $\sigma_T(\gamma)$ values for thorium and uranium isotopes preserves the same strong difference between P_0 and P_μ . But it is hardly understable as far as the excitation of 9 MeV is considered.

Resuming, we state:

a) it is impossible to explain all the fission yields observed in our measurement as conditioned up by higher than K_α transitions;

b) higher transitions as well as K_α transitions can be responsible for the fission yield in ^{232}Th case.

We present our results once more in fig. 7. It was assumed here that only K_α radiationless transition contribute to the measured fission yields. Consequently these yields were normalized to the probability of the K_α nonradiative transitions. It was taken as equal to 0.2, on average, for all three nuclei in view of relatively large errors quoted in ref. /21/.

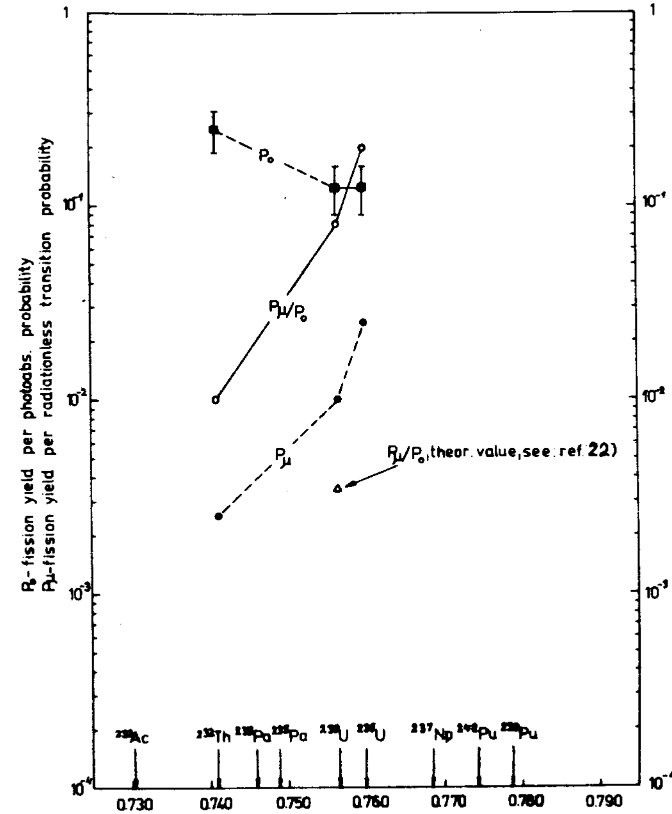

Fig. 7. Comparison of the photofission and radiationless transition fission probabilities for ^{232}Th , ^{238}U , and ^{235}U.

Figure 7 shows the photofission probability also. The ratio P_μ/P_0, as the comparison of the photofission yields with and without the muon in the 1s muonic orbit is presented along with its theoretical value for ^{238}U taken from ref. /22/.

Two features are quite clear. Firstly, the photofission probability of the investigated nuclei with the muon in the 1s orbit P^f_μ is much lower than P^f_0 (without the muon in the 1s orbit). Secondly, quite steep variation of the ratio P_μ/P_0 from one nucleus to the other is observed when practically no variation is observed for P_0 alone.

One can try to explain these facts by the effect of the muon binding energy on the fission barrier height. In the region of uranium according to Zaretsky and Novikov's result /20/ the heights of the first and second barriers increase by 150 MeV and 600 keV, respectively /22/ But these calculations have been performed only for the reflection symmetric shapes of the nucleus. It seems to be a generally accepted point of view that the second saddle appears at the stabilized reflection-asymmetric shape for which the correction of its height due to the muon presence in the 1s orbit should be smaller than quoted 600 keV. On other hand, the height of the second saddle should be comparable or some hundreds of kiloelectronvolts lower than the $(1,0^-)$ channel of the first barrier. It follows from the fact that the first and second barrier for thorium and uranium are comparable according to the experimental systematics /23/. For thorium, they are: S_A=(5.9±0.2)MeV and S_B=(6.1±0.2)MeV and for uranium: (5.7±0.2) MeV and (5.8±0.2) MeV, respectively. This relation can be changed in the presence of the muon and excited energy of the $(1,0^-)$ channel becomes practically comparable or even slightly lower than the height of the second saddle. So, the net result is as follows:

In the region of the first barrier we are about 150 keV out off the $(1,0^-)$ resonance and phenomenologically it corresponds to the shift of the energy scale in fig. 6 by 150 keV on the left.

In the region of the second barrier the nucleus ^{232}Th excited by the $2p_{3/2}-1s_{1/2}$ component of K_a muonic transition is maximally 700 keV below the barrier and excited by a $2p_{1/2}-1s_{1/2}$ component not more than 400 keV. In the case of ^{238}U the first component excites the nucleus to 300 keV below the barrier while the second excites it to the energy of the second saddle height.

We can estimate the decrease of the fission probability as the product of the decreases in the region of the first and second barrier. The first factor is maximally 0.7 for ^{232}Th and unity for ^{238}U in accordance with Yester's et al. /17/ and Anderl's et al. /18/ results. The second factor, the ratio of penetrabilities for the nucleus with and without the muon we estimate as 0.25 and 0.7 taking the data from ref. /22/

Finally, the total decrease of the fission probability caused only by the change of the barrier heights in the presence of the muon in the orbit is maximally six times for ^{232}Th and 30% for ^{238}U. We see that the observed difference for these two isotopes is well reproduced. But the observed absolute values of the fission probabilities P^f_μ cannot be reproduced being hundred, ten and five times lower than P^f_μ, for ^{232}Th, ^{238}U, and ^{235}U, respectively.

We can explain such deep depression of the fission probability if the resonances observed in refs. /16-18/ in the region of 6.3 MeV for ^{232}Th and ^{238}U and interpreted there as the $(1,0^-)$ fission channel are in fact composed of the $(2,2^+)$ channel, i.e., a gamma-vibration nature one, probably lying in the region of 6 MeV /25/ and the $(1,0^-)$ channel of octupole vibration nature. It would mean that the widths of the dipole resonances are considerably smaller than those observed in refs. /16-18/. So, in radiationless transition fission the excitation energy of the nucleus is much more out off the dipole resonance than it can be judged from fig. 6.

This conclusion is even more evident for ^{232}Th, if in the observed fission yield we have some contributions from higher than K_a transitions.

In other words our result could be interpreted as the indication of the complex channel structure in the region of the 6 MeV excitation.

It does open up a new possibility in the barrier structure investigations: non-radiative transition fission as induced by the pure dipole transition of the energy near the top of the fission barrier can help in revealing the nature of fission channels.

In view of that it seems very interesting to spread the systematics described in this paper on such nuclei like ^{240}Pu and ^{242}Pu. As it is claimed in Vandenbosch's analysis /24/, the first dipole channel $(1,0^-)$ in ^{242}Pu should lie in the region of 6.7 MeV. In this nucleus the upper edge of the K_α transitions is about 6.5 MeV. It means that the ^{242}Pu nucleus excited in the radiationless K_α transition will be about 0.5 MeV off the $(1,0^-)$ resonance and the relatively low yield of the prompt fission should be observed.

Acknowledgements

We are grateful to Professors V.P.Dzhelepov and L.I.Lapidus and K.Ya.Gromov for help and interest in this work.

It is a pleasure to acknowledge Drs. K.Andert, A.I.Kalinin and V.G.Zinov's very helpful electronic assistance.

We are indebted to Dr. A.Zglinski who kindly rendered at our disposal the computer codes.

We thank N.G.Zaitseva and Mrs. L.Gumnerova for their aid in the target preparation.

Dr. A.V.Strelkov's kind help in the ionization chamber preparation is appreciated.

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Received by Publishing Department
on July 25, 1974.