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POSSIBLE EVIDENCE FOR SHAPE ISOMERIC GAMMA-DECAY IN μ^- ATOMS OF ^{238}U

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1. INTRODUCTION

The phenomenon of the spontaneously fissioning isomers[1] can be interpreted as fission from a shape isomeric state, e.g., a state characterized by much greater nuclear deformation than the ground state. Theoretical calculations[2] on the basis of the liquid drop model with shell and pairing corrections included are capable of describing the shape isomerism qualitatively. In the framework of this model isomeric fission occurs from a state at the second minimum of the potential energy curve at large quadrupole deformation (fig. 1).

A systematic study of all hitherto known fission isomers[3] shows that their formation cross sections are decreasing in going from $Z=94$ to lower Z-values, and for $Z<92$ no isomers are known at all. The calculations of the double-humped fission barrier[4] predict a higher outer than inner barrier for $Z<94$, which has been experimentally supported[5]. Whereas for shape isomers with $Z\geq 94$ fission is the dominating de-excitation channel for nuclei with $Z<94$ and lowered inner barrier γ- decay back into the first minimum should compete with fission. A strong γ-branch would allow one to regulate the observed formation cross sections and to establish precise excitation energies for

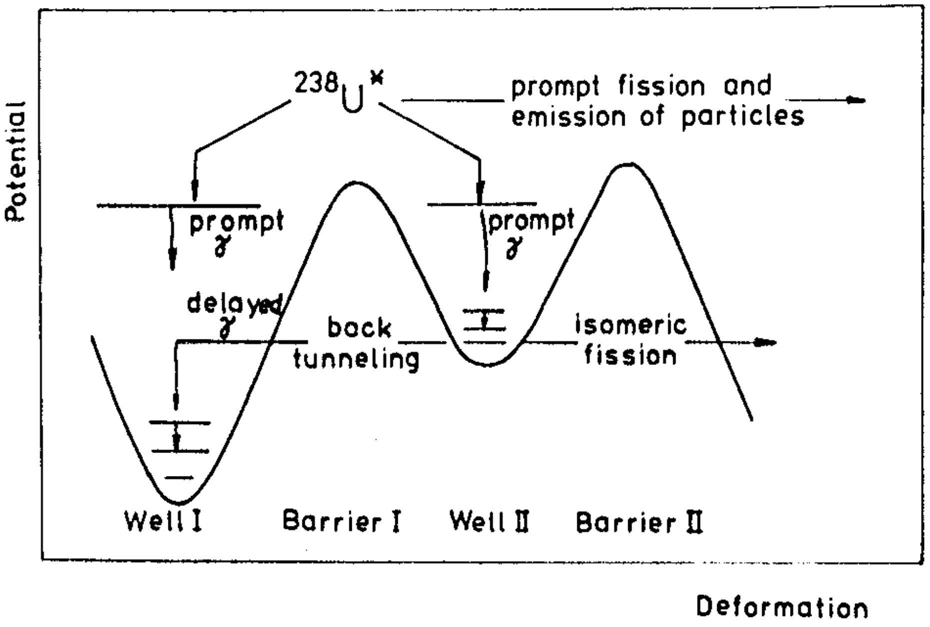

Fig. 1. Schematic representation of the fission barrier potential versus deformation and the decay modes of the isomeric state.

the shape isomeric states. Search experiments for the observation of the shape isomeric γ-decay have been performed by various groups employing the reactions $^{235}U(d,p)^{236m}U$ ref.[6], $^{238}U(p,n)^{238m}Np$ ref.[7], $^{238}U(d,pn)^{238m}U$ ref.[8]. Only the authors of ref.[8] have succeeded to observe γ-transitions from the shape-isomeric decay and precisely determined the excitation energy of this state to 2.559 MeV. One of the possibilities of obtaining physical information about the presence of large deformations is the observation of rotational

states in the second minimum through which the shape isomeric state is populated. In ref.[9] conversion electrons from such highly converted γ-transitions precurring the isomeric fission of ^{240}Pu have been observed. From the energy spacing of these states a moment of inertia twice as high as that of the ground state band was determined, which strongly supports the connection of fission isomerism with large deformations.

Another independent method would be to use the sensitivity of the muon binding energy to the nuclear charge distribution as proposed in ref.[10]. According to the calculations from refs.[11,12] the high quadrupole moment of the shape isomeric state should lower the muonic binding energy for the 1s -state by several hundred keV. The barrier heights would be changed, allowing the isomeric nucleus more easily to back-tunnel and depressing the fission channel[33].The observation of an energetically shifted shape isomeric state in the presence of the muon would give another proof for the validity of the double-humped barrier description. As another consequence of the augmentation of the barrier the lifetime of the shape isomeric state in the muonic atom would be changed, whereas one should take into account the capture of the muon resting in the state by the nucleus as a competing process.

Presumption for a successful search for the γ-branch of the decay of the shape isomeric state in muonic atoms is a sufficiently strong excitation of such states by the muonic cascade. From the different lifetimes observed for delayed fission refs.[13, 14] and the electronic decay of the muon ref.[15] in muonic ^{238}U a strong population of the shape isomer due to radiationless transitions of the muon has been postulated[16].

The aim of the present work is to search for delayed γ-radiation after the formation of muonic atoms of ^{238}U.

2. EXPERIMENTAL ARRANGEMENT

The measurements were performed at the separated μ^--beam of the Dubna synchrocyclotron. A special extension of the muon channel into the heavy shielded cave of the low background laboratory was used. The effect-to-background ratio was much improved compared with an earlier run[17] with the μ-stop telescope in the main experimental hall.

The compact geometrical arrangement of the counters in the small free space at the end of the beam line is shown in fig. 2, a simplified block-diagram of the electronic apparatus is given in fig. 3. The μ-stop signal was registered by means of a scintillation counter telescope fulfilling the coincidence relation $12\bar{3}$. With a typical intensity of the μ^--beam of 8000 s^{-1} the number of μ-stops in the 115 g uranium target was 650 s^{-1}. The γ-radiation was recorded by means of a 60 cm^3 Ge(Li) -diode with 2.8 keV FWHM at 1 MeV resolution in-beam and two NaI(Tl) crystals of 8 cm diameter and 8 cm length. The timing system[18] used has given an integral time resolution of 7 ns FWHM in the energy range from E_γ= 0.1 to 4.7 MeV in

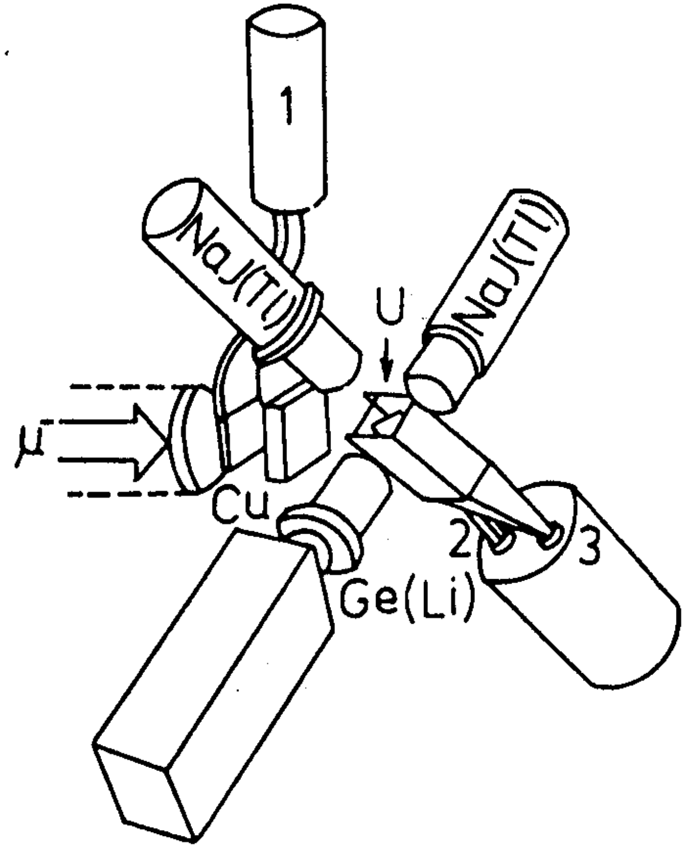

Fig. 2. Experimental arrangement. The counter telescope is given by No. 1, No. 2 and No. 3. Cu is the copper moderator. The arrow indicates the direction of the μ^--beam.

the Ge(Li) -branch. The experiment was operated on-line with the HP 2116C computer.

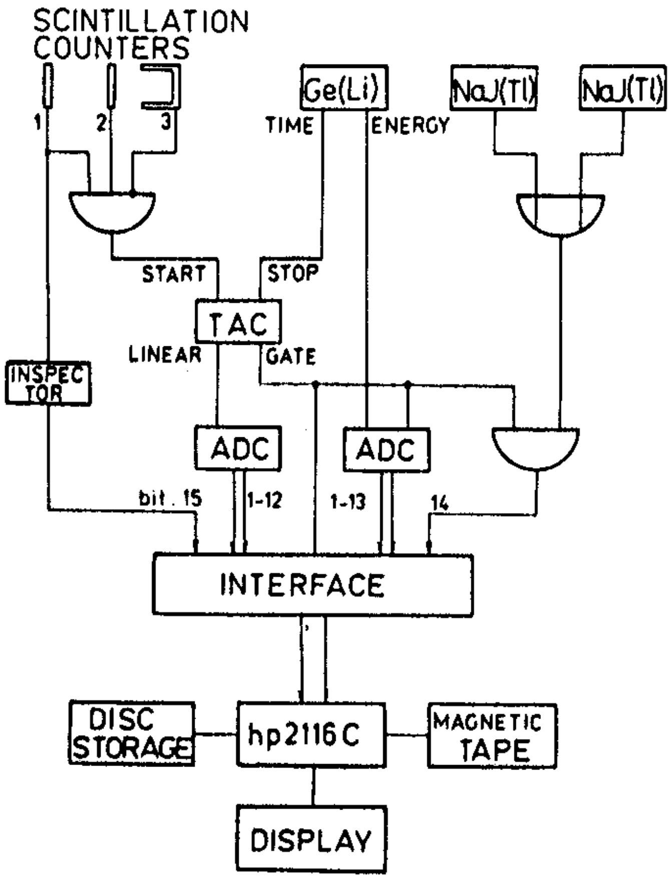

Fig. 3. Schematic Block diagram of the electronics.

The following information was combined and transferred to the computer:

i) the energy of the γ-quanta registered by the Ge(Li)-detector (up to 4.7 MeV in 8192 channels; this resolution was necessary for muonic X-ray investigations, in the present work, however, channel pairs were summed),

ii) the time between the μ-stop event and the Ge(Li)- signal ($2.5\,\mu s$ range in 4096 channels),

iii) the signal from the inspection circuit/34/. if $3\,\mu s$ before and after the μ-stop moment no other charged particle passes through the first counter (threshold for muons 100 keV),

iv) the signal of the NaI(Tl)-detectors, if at least one of them had registered a γ-ray in the time interval from 20 to 120 ns after the Ge(Li)-signal (threshold 200 keV).

The data were stored in the event-by-event mode on magnetic tape. Integral energy and time spectra were sampled and displayed simultaneously. After the 45 hour data-taking run the events were distributed into 4 groups of 8 energy spectra each. The groups were determined by the presence of the signals indicating single muon and/or delayed NaI - coincidence events. The spectra were derived according to eight time intervals producing one prompt, six successive delayed and one calibration spectra. The internal calibration lines were provided by radioactive sources registered as chance coincidences in the course of the experiment. The time scale was calibrated with a 10 MHz frequency generator.

In the delayed spectra the main part of the background activity is formed by γ-radiation following the nuclear muon capture. As far as we are mainly interested in the detection of weak delayed lines as they were expected from the decay of the shape isomeric state in muonic ^{238}U, the delayed background should be suppressed as much as possible. Except for measures as good spectroscopic resolution the delayed coincidence circuit between the Ge(Li)- and the NaI(Tl) -detectors was introduced. The coincidence condition is fulfilled only in the case where a γ-ray registered by the Ge(Li) -detector was followed by other γ-rays in a time interval typical for nuclear muon capture. Therefore, in the spectra sorted from events with such coincidences delayed γ -rays emitted before the nuclear muon capture should be registered with an efficiency equal to the response probability of the delayed coincidence setup to nuclear muon capture. This value was measured to be 10% from the intensity ratio of prompt muonic X - rays in spectra with and without the coincidence signal. The inspection circuit for single muon events was used as a further condition in order to suppress chance coincidences. In the energy spectra, separated by more than 25 ns from the maximum of the prompt peak, the background was reduced by a factor of about 220 whereas the effect should be diminished only tenfold. Therefore, taking into account the $\sqrt{N}$ -law, a 1.5-fold sensitivity improvement follows for the detection of delayed activities in spectra treated in the above mentioned manner. In the time range less than 25 ns after the maximum of the prompt peak, however, the background reduction becomes worse due to the tail of the prompt distribution, which is usually present in time spectra from large-volume Ge(Li) -detectors. Then the sum spectra over all four groups give the best detection sensitivity. The detection limit was estimated in the energy range of 0.4 to 3.2 MeV for spectra with the delayed coincidence condition to be 0.8% for 10 ns halflife and 0.3% for 30 ns halflife. The corresponding values for the summed spectra are 0.3% and 0.5% per stopped muon. This value shows a considerable improvement over the earlier experiment of Kaplan et al. /20/ in which at the sensitivity level of 1% per μ -stop no effect could be found.

3. RESULTS AND DISCUSSION

3.1. Gamma-Ray Time Distribution

The time distribution of all γ-ray events recorded from the ^{238}U target is shown in fig. 4. The decay time of the delayed γ-rays

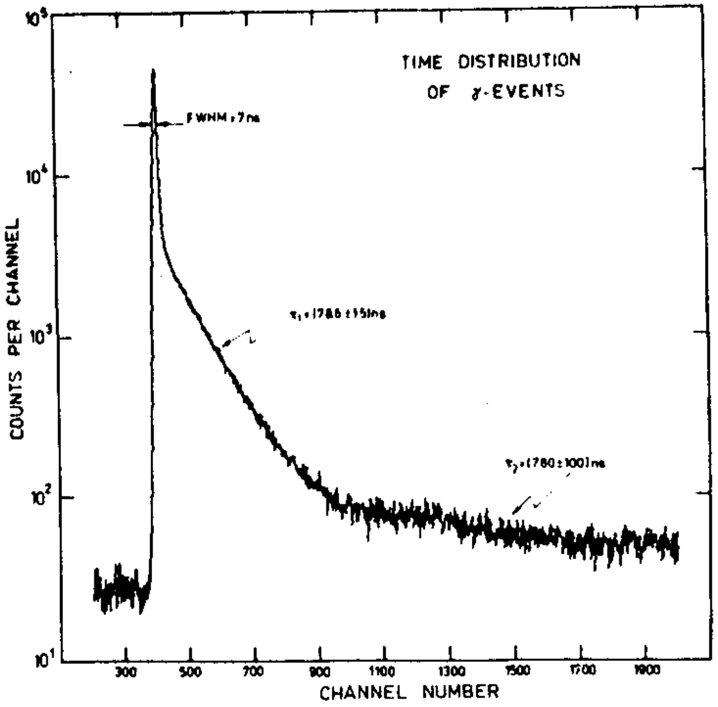

Fig. 4. Experimental time distribution of γ -quanta from μ^{-} ^{238}U.

was determined by the least squares method for two exponential decays, the longer one should account for the muon capture in components of the telescope and was found to be (780 ± 100) ns. The lifetime of the component belonging to the muonic^{238}U τ_μ=(78.6 ± 1.5) ns remains unchanged within error limits if the second decay is omitted. In Table 1 the lifetimes for muonic ^{238}U measured via fission, γ-rays and electronic decay are compared. For the assumption of a strong population of the shape isomeric state by the muon a smaller value for the lifetime measured by fission products or γ-rays with respect to muonic decay electrons is essential.

The prompt spectrum (fig. 5) exhibits all the features of the interaction of the muon

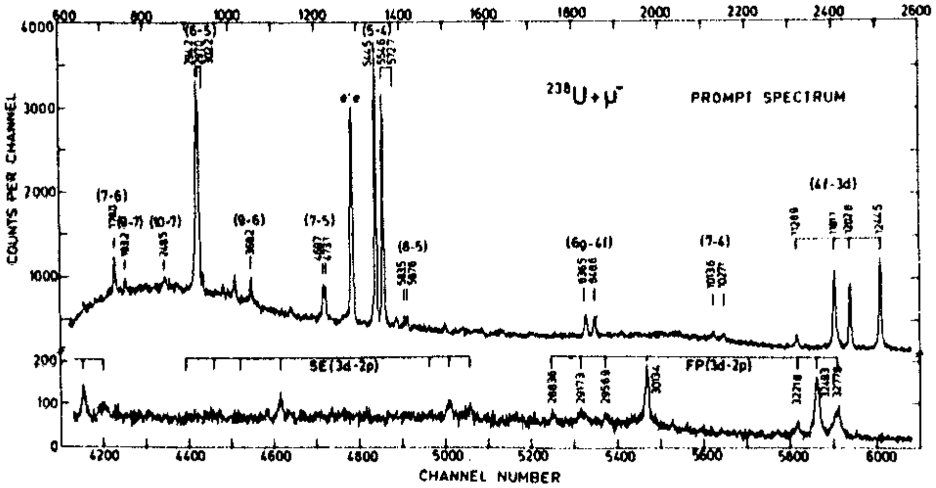

Fig. 5. The prompt spectrum of μ^- ^{238}U in the region of 500-3360 MeV.

Table 1

Experimental lifetimes in muonic atoms ^{238}U for fission, electron decay and gamma-rays

	Lifetime (ns)	References
Electron decay	τ_e = 88 ± 4	1959 Sens
	τ_e = 81.5 ± 3.0	1975 Hashimoto et al.
Fission	τ_f = 75.6 ± 2.9	1963 Diaz et al.
	τ_f = 74.1 ± 2.8	1970 Budick et al.
	τ_f = 76.1 ± 1.0	1974 Chultem et al.
Gamma-ray	τ_γ = 78.6 ± 1.5	1975 this work

and the heavy deformed nucleus. The results of the investigation of the prompt spectrum will be given in a forthcoming paper. The calibration spectrum developed from γ-events separated by more than 200 ns from the μ-stop moment shows in addition to the γ-transitions from the decay of ^{56}Co and ^{152}Eu γ-rays known from the radioactive decay of ^{238}U.

3.2. Delayed Activities

The main interest in this investigation was devoted to delayed γ-ray activities which can be produced in different ways. Except for the γ-ray decay of shape isomeric states excited by radiationless transitions in the muonic cascade which was searched for, delayed γ-lines are emitted in the nuclear μ-capture process, by the capture of neutrons emitted in connection with μ-capture and by inelastic scattering of neutrons, by the de-excitation of spin isomers excited in fission fragments as well as in the target nucleus.

On an average 100 counts per channel were recorded in the delayed spectra in the energy range above 500 keV. An essential enhancement of the statistics for delayed activities at the present facility is impossible because of intensity and beam-time limitations. An essential improvement of such a type of experiment would be possible at one of the high intensity beams of the existing meson factorities.

A careful search for delayed activities revealed two groups of γ-rays distinguished by their halflives. The first group of transitions has a lifetime of about 70 ns, which is typical for the μ-capture process in uranium nuclei (see Table 2). The energies and intensities of these lines are in good agreement with the transitions observed in the ^{238}U(n,γ) -reaction[21,22]. That means that neutrons emitted from the nucleus after μ-capture produce secondary reactions in the thick uranium target. Transitions from the ^{238}U(n,n'γ) -reaction, and from the excited residual nuclei after the ^{238}U(μ,xn)$^{238-x}$Pa -reaction could not be detected. Nuclear muon capture can be acoompanied by fission too, but for ^{238}U the delayed fission branch has a probability of 3% per captured muon only[14]. The complex γ-ray spectrum produced by the de-excitation of isomeric states in the variety of fission fragments was studied in detail using spontaneous fissioning ^{252}Cf [23]. Every isomeric transition from a fission fragment produced by muon capture induced fission is populated by a delayed process with respect to the μ-stop moment. Lines from fission fragments should therefore appear in our spectra with en-

Table 2
Intensity of Delayed gamma-lines per μ^- stop in μ^{-} ^{238}U τ_1 = (69 ± 20) ns

E_γ (keV)	552	580-612 a)	834-856 a)
I/μ^--stop (%)	6.5±1.4	39±8	22±5

a) composite peak.

larged lifetimes. Caused by the small probability of the fission process and the wide spread-out of its intensity no γ-transitions from fission fragment isomers could be detected with an intensity exceeding 0.3% per stopped muon.

The second group comprises delayed γ-transitions with a shorter mean lifetime of about 12 ns. In general the existence of a γ-line was admitted if in a series of successive spectra the counting rate of neighbouring channels was higher than the background. Unfortunately, for this short lifetime region the additional delayed coincidence with the NaI(Tl)-counters was not applicable. Totally seven transitions were found and their appearance in different time windows is shown in Fig. 6. The decay of their intensities is presented in Fig. 7 together with a least-squares-fit to a common lifetime. The intensities of the transitions are summarized in Table 3. The normalization of the delayed intensities to the number of μ-stops was performed with the intensity of the muonic 4f-3d transition in the prompt spectrum and the percentage of this transition determined by a cascade calculation/24/ starting with statistical distribution at $n = 20$. The reduction of the intensity of the detected main components of the muonic 4f-3d transition group due to dynamic quadrupole interaction, ref. /25/ was taken into account using the charge distribution parameters of ref. /26/ in the calculation for ^{238}U. Because of the short lifetime of the γ-transitions under consideration any explanation of their existence with processes connected with the capture of

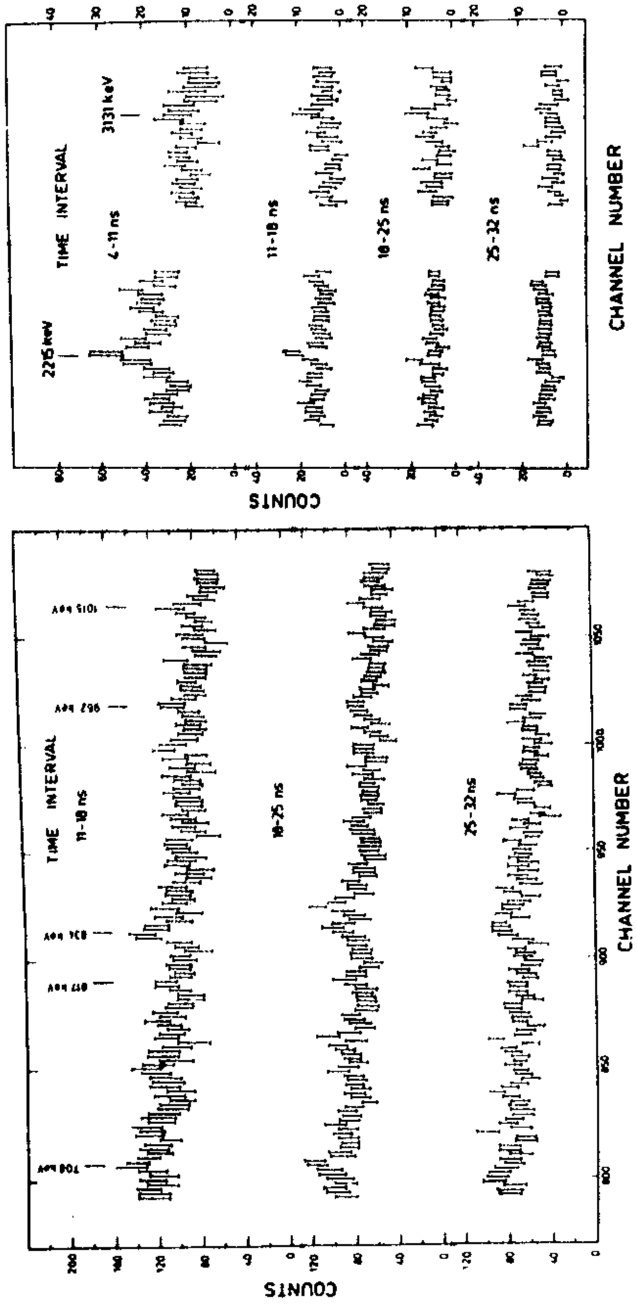

Fig. 6. Parts of the delayed spectra of μ^- ^{238}U. The energy region of 700-1100 keV is shown for 3 time intervals, the regions around 2215 and 3130 keV, for 4 time intervals.

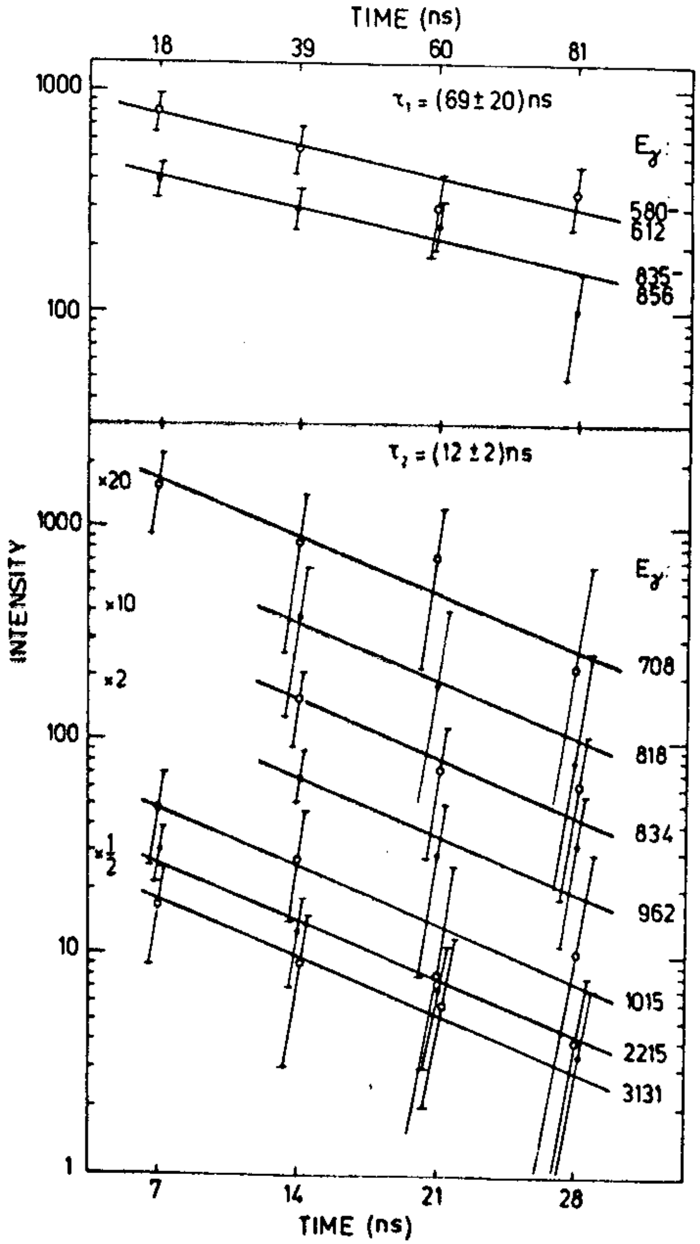

Fig. 7. Decay curves for delayed gamma-rays in $\mu^- \, {}^{238}$U.

Table 3

Intensity of delayed gamma-lines per μ^- stop in $\mu^- \; {}^{238}U$
τ_2 = (12± 2) ns

E_γ (keV)	708±1	817±1	834±1	964±1	1015±1	2215±2	3131±2
I/μ-stop (%)	0.6± ±0.2	0.4± ±0.2	0.5± ±0.3	0.4± ±0.2	0.65± ±0.25	0.5± ±0.2	0.5± ±0.2

the muon from the 1s-state is not consessive. In principle, prompt fission after radiationless excitation of the nucleus by the muon could excite isomers in the fission products which then would be recorded with their real lifetimes in the delayed spectra. Prompt fission, however, occurs in only 0.2% of μ-stops in the ^{238}U target/14/ which is beyond our detection limits.

3.3. Comparison with the ^{238}U Level Scheme

Further one has to investigate, whether or not the short-lived transitions observed could be explained by an isomeric state in ^{238}U. The level structure of ^{238}U has been studied from the decay of ^{238}Pa /27/ and by means of Coulomb excitation /28/ as well as the (n,n'γ) -reactions/29/. The ground state band was followed up to the 14^+ member at about 1.5 MeV. No short-lived spin isomeric transitions were reported so far. Also in the investigation of the ^{238}U(d,pn) reaction by Russo (ref. /8/) no spin isomeric states have been found. The existence of such isomers, however, cannot be excluded in this mass region. For example, the study of the ^{236}U(d,pn) reaction/6/ dedicated to the detection of the γ-branch from the shape-isomeric state yielded a new isomer in the first minimum with K, $j^{\pi} = 4.4^-$ at 1.052 MeV excitation in ^{236}U. A second isomer with K, $j^{\pi} = 2.2^-$ at 0.687 MeV and a halflife of 4.4 ns is also known in ^{236}U /30/. Because of the high excitation energy of the state observed in the experiment (> 3 MeV) and the limited possibilities of the excitation of states with significant spin difference to the ground state by the muon, the identification of the observed delayed γ-rays with the de-excitation of a yet unknown spin- or K-isomeric state is very improbable.

Two of the seven transitions from Table 3 fit in the level scheme of ^{238}U (Fig. 8). The 1015 keV transition represents the de-excitation of the lowest member of the γ-vibrational band (K = 2) to the 2^+ state of the ground state band, the 817 keV transition deexcites the member of the second rotational band to the 4^+ state of gsb. It should be mentioned that the $4^+ \to 2^+$ (E_γ = = 103 keV) and $2^+ \to 0^+$ (E_γ = 45 keV) transitions in the experimental arrangement could not be recorded due to their low energy and high internal conversion. Furthermore, all lines with intensities lower than 0.3% are undetectable, thus, in the case of branching at one level the de-excitation path may be lost. In addition, all transitions recorded in the presence of the muon in the 1s -state are splitted and shifted due to the interaction of nucleus and muon. The value of the magnetic hyperfine splitting of nuclear levels is of the order of 1 keV, and in most cases larger than the isomeric shift. The intensities of the delayed transitions and the limited energy accuracy are not sufficient to extract this refinements from the data. Therefore, in Fig. 8 all the levels are shown like unsplitted ones. The isomeric shift has a minor influence on the level energies, if the nuclear charge distribution (of deformation) for the levels under consideration is about equal. For large differences in the shape of the nuclear charge distribution the isomeric shift

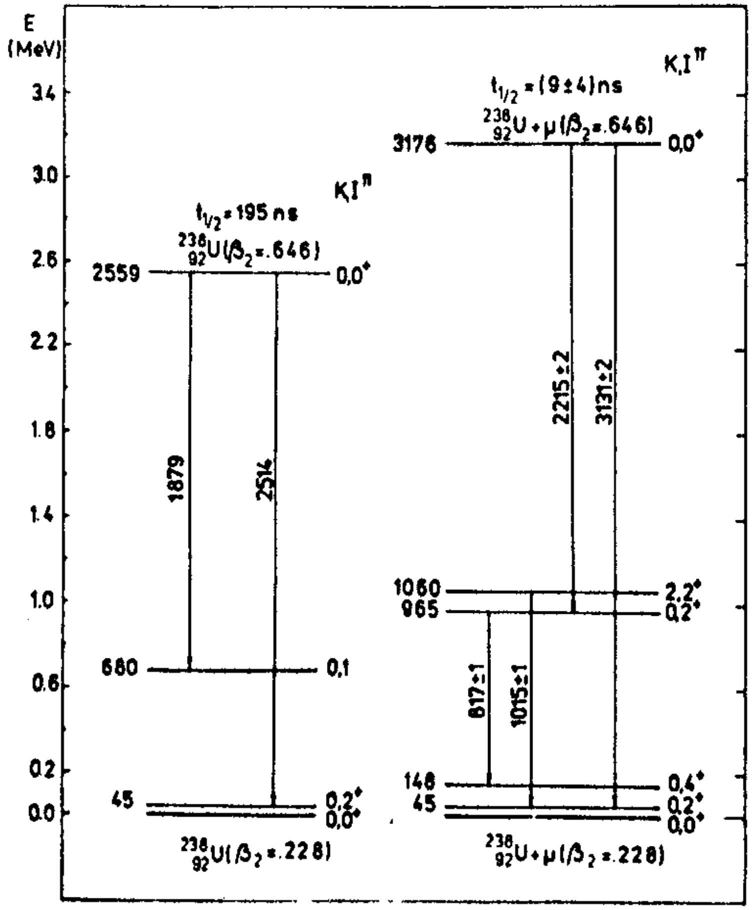

Fig. 8. Decay scheme for the ^{238}U gamma-branch from the shape isomer in ^{238}U without the muon (ref. /8/) and with the muon on the 1s -orbit. All transition energies in keV. β is the deformation in the isomeric and the ground state.

becomes the dominating part of the muonic-nuclear interaction. Hence, a large shift of the shape isomeric levels relative to states from the first minimum has to be expected. In ruling out other possibilities for short-lived high-energy transitions the two transitions with E_γ= 2215 keV and E_γ=3131 keV can be ascribed to the decay of the shape isomeric state in muonic ^{238}U to the 2^+ levels of the γ-vibrational and ground state band. The level scheme for the γ-ray decay of the shape isomer in ^{238}U, ref. /8/ and muonic ^{238}U are shown together in Fig. 8. Three of the observed delayed transitions could not be placed in the decay scheme. It is possible that intermediate states with low spin at energies over the γ-vibrational band are connected by these transitions with the shape isomeric decay. There is no experimental information of states with low spin in the excitation band higher than 1.5 MeV in ^{238}U available at present.

3.4. Influence of the Muon on the Gamma-Branch

The most striking features comparing the two level schemes presented in Fig. 8 are the large isomer shift for the shape isomeric state in the presence of the muon of ΔE = = 617 keV and its shortened lifetime. In the early work of Zaretski/11/an estimation of ΔE = 500 keV for the energy shift of the fission barrier in muonic actinide nuclei was given. The augmentation of the fission barrier can be estimated from the change of the binding energy of the muon in the 1s-state.

The binding energy of the muon was calculated by solving the Dirac equation of a deformed Fermi type charge distribution

$$\rho(r) = \rho_0 (1 + \exp(4 \ln 3 \frac{r - c\,(1 + \beta_2 Y_{20})}{t}))^{-1} .$$

In Fig. 9 the potential energy curve for ^{238}U and muonic ^{238}U are shown together. The value of the isomer shift of states in the second minimum $\Delta E = 640$ keV is in good agreement with the experimental value. The results of a recent paper by Leander and Möller /12/, where the non-relativistic Schrödinger equation for a charged particle in the Coulomb potential of a deformed charge distribution was solved, are in close connection with the values for the change of the muonic binding energy given in Fig. 9. The experimental lifetime $\tau_{exp} = 12$ ns of the delayed γ-rays observed combines the lifetime τ_i of the shape isomeric states in the presence of the muon and the lifetime τ_μ of the muonic atom itself:

$$\frac{1}{\tau_{tot}} = \frac{1}{\tau_i} + \frac{1}{\tau_\mu} ,$$

The value for τ_i can be compared with the lifetime of the shape isomer estimated on the basis of parabolic fission barriers /8/. In this framework the barrier penetrability is given as

$$P_{A,B} = \exp(-2\pi \frac{E_{A,B} - E_{II}}{\hbar\omega_{A,B}})$$

with E_{II} as the excitation energy of the shape isomeric state and the curvature parameters $\hbar\omega_{A,B}$ for the inner barrier A and the outer barrier B respectively. For equal penetrabilities fission will dominate over γ-decay by a factor of about 10^6 (ref. /4/).

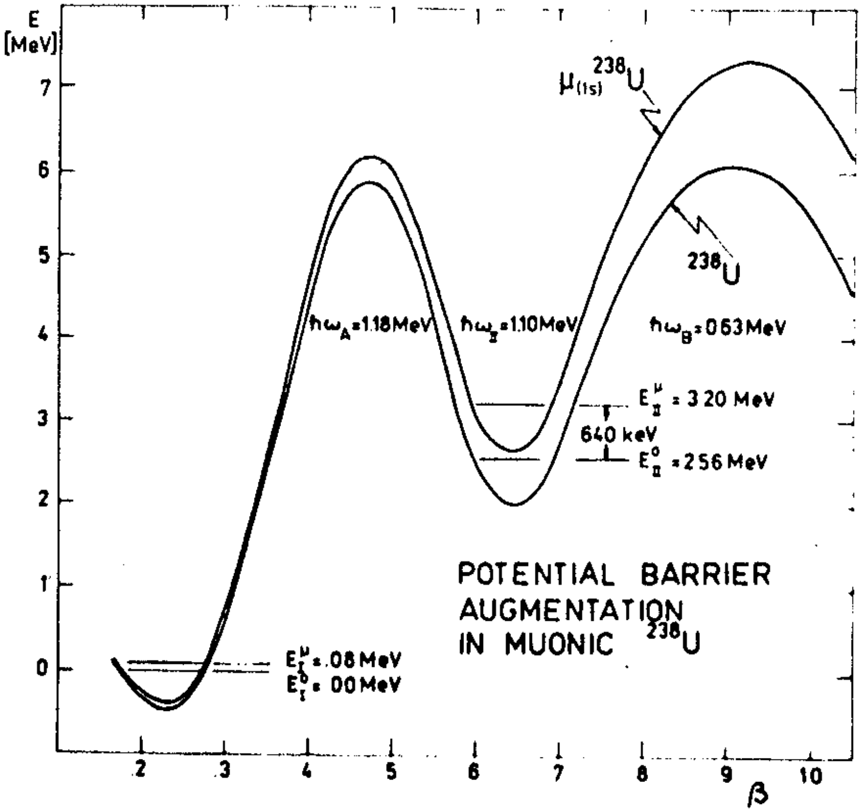

Fig. 9. Isomer shift of the potential energy curve for μ^- ^{238}U together with the calculated potential energy curve (parameters from ref. /8/) without muon.

Using for the partial lifetime for fission decay of the isomer the expression

$$\tau_{i,f} = \frac{1}{n \cdot P_B} = \frac{6 \cdot 10^{-21}}{P_B} ,$$

where n is the number of barrier assaults per second for the vibrational frequency in the second minimum of $\hbar\omega_{II} = 1$ MeV, the partial lifetime for γ-decay of the isomer becomes

$$\tau_{i,\gamma} = \frac{6\cdot 10^{-15}}{P_A} .$$

In Table 4 the estimated partial and total lifetimes for ^{238}U and muonic ^{238}U are compared. The barrier parameters were taken from ref. /5/. The augmentation of the fission barrier due to the muon was calculated as described above. The shortening of the isomeric decay with the muon in the nuclear system is reproduced. However, there are ways lef left open to enhance the agreement between the experimental and the theoretical values. For example, the curvature parameters of the fission barrier were taken not modified by the barrier height changes caused by the muon. Furthermore, damping of nuclear state according to the theory of Lynn /31/ was not included since it was felt that the experimental material did not permit to give such refinements any physical significance. The most astonishing fact is the large population probability of the second minimum by the muonic cascade. Radiationless transitions which are responsible for the excitation of the isomeric state establish about 20% of the muonic cascade. The total intensity of the γ-rays de-exciting the shape isomeric states was found to be 1.6% in this experiment. Therefore the feeding probability of the shape isomeric state by radiationless transitions is of the order of 10^{-1}, which is surprisingly large compared with the corresponding values of 10^{-4} and 10^{-5} for neutron induced excitation or charged par-

Table 4

Barrier Parameters and Lifetimes of ^{238m}U - and μ^{-} ^{238m}U

	E_A (MeV)	E_{II} (MeV)	E_B (MeV)	$\hbar\omega_A$ (MeV)	$\hbar\omega_{II}$ (MeV)	$\hbar\omega_B$ (MeV)	$\tau_{i,\gamma}$ (ns)	$\tau_{i,f}$ (ns)	τ_{tot} (ns)
^{238}U ref.$^{5)}$	5.90		6.12	1.00		0.62	5.2×10^{3}	18.64×10^{3}	4.07×10^{3}
^{238}U ref.$^{8)}$	5.90	2.56	6.12	1.18	1.10	0.63	212	10.51×10^{3}	207
Energy shift of μ^{-238}U	0.28	0.56	1.06						
μ^{-238}U	6.18	3.12.	7.18	1.18	1.10	0.63	48	1.56×10^{6}	$29^{1)}$ $(12\pm4)^{2)}$

1) The lifetime of the muon on the 1s-orbit $\tau_\mu = 75$ ns

$$1/\tau_{tot} = 1/\tau_{i,\gamma} + 1/\tau_\mu .$$

2) The experimental lifetime.

ticle reactions, respectively. The physical reason for this phenomenon could be resonance excitation of states in the second minimum via vibrational states in the first well. Such resonances have been observed in a subthreshold excitation study/32/. At present more experimental material should be accumulated to yield in a better understanding of the excitation process governing the discussed effect.

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