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POPULATION OF THE ^{236}U SHAPE ISOMER IN A PHOTONUCLEAR REACTION †

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Abstract: A fission isomer with a half-life of 115 ± 5 ns and a yield ratio $Y_{iso}/Y_{prompt} = (2.02 \pm 0.16) \times 10^{-5}$ was observed in bremsstrahlung-induced fission of natural uranium. The isomer is ascribed to ^{236}U populated via a ^{238}U(γ, 2n) reaction. The integrated cross section for isomeric fission is determined to be $\sigma_{int} = 32 \pm 6$ μb · MeV. Comparing this value with a calculated total isomer production cross section, a branching ratio of the isomer decay of $\Gamma^{\gamma}_{II}/\Gamma^{f}_{II} \approx 6$ can be deduced.

E | NUCLEAR REACTIONS ^{238}U(γ, 2n), $E = 45$ MeV bremsstrahlung; measured isomer/prompt yields; deduced σ for isomer production. ^{236m}U shape isomer deduced $T_{1/2}$, $\Gamma_{\gamma}/\Gamma_{f}$. Natural target.

1. Introduction

The superposition of deformation-dependent shell corrections to the smooth potential of the liquid-drop model leads to a second minimum in the potential energy surface of nuclei in the actinide region ($92 \leq Z \leq 97$) [1]). The observed fission isomers can be explained as shape or form isomers. Their lifetimes are determined by the heights and widths of the double-humped fission barrier. The experiments of Specht *et al.* [2]) and Habs *et al.* [3]), who measured the moment of inertia and lifetimes of excited states in the second well, respectively, showed direct evidence that fission isomers are metastable nuclei with nearly twice the deformation of the deformed ground state.

Photonuclear reactions seem to be favourable for the investigation of fission isomers. One advantage is the low angular momentum transfer (mainly electric dipole excitation), leading to excited states with low spins. Additionally, compound states of low excitation energy are easily populated in photonuclear reactions, due to the missing Coulomb barrier. Therefore, the number of open exit channels is reduced compared with particle-induced reactions. However, only two investigations of photonuclear reactions leading to fission isomers have been reported so far, possibly due to the experimental difficulties, mainly the lack of intense monochromatic photon sources and the inherent background problems at photon beams. Gangrsky *et al.* [4]) observed long-lived isomers (μs-ms region) in ^{239}Pu, ^{241}Pu, ^{240}Am using the bremsstrahlung beam of a microtron (E_{max} = 7–16 MeV). The isomeric fission

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yields compared with the prompt fission yields were of the order of 10^{-3}. Tamain *et al.* [5]) investigated sub-μs isomers in uranium and plutonium using the bremsstrahlung beam of the Giessen linac ($E_{max} = 53$ MeV). They found isomers in uranium ($T_{\frac{1}{2}} = 200 \pm 50$ ns) and plutonium ($T_{\frac{1}{2}} \approx 500$ ns) with a low delayed-to-prompt fission ratio of the order 10^{-5}. Due to the poor statistics the isomers could not be ascribed uniquely to a certain isotope [^{236}U($T_{\frac{1}{2}} = 116 \pm 7$ ns) [6]); ^{238}U($T_{\frac{1}{2}} = 195 \pm 30$ ns)[7])]. The low isomeric-to-prompt fission ratio of 10^{-5} has the same order of magnitude as the ratios observed for ^{236}U in (d, pf) and (n, f) reactions (see table 2). The small isomeric fission yield suggests a relatively low inner barrier for the uranium isotopes, as expected from barrier systematics. This may favour a possible γ-decay branch of the shape isomeric state back to the first minimum. Only for ^{238}U have Russo *et al.* [8]) observed a high energetic γ-transition (2.514 MeV) which was attributed to such a competing γ-decay of the shape isomer.

An exact measurement of the half-lives and the relative cross section for isomeric fission via a photonuclear reaction of the uranium isotopes seemed to be of interest, in particular with respect to a possible γ-decay branch.

2. Experiment

2.1. EXPERIMENTAL SET-UP

The experiments were performed at the improved bremsstrahlung facility of the 65 MeV Giessen electron linac. The electron beam is analysed in energy by a 90° achromatic deflecting system and focused on a 1 mm tungsten bremsstrahlung target. A dumping magnet together with a collimator and beam-hardener system in the 3 m concrete wall between the experimental area and the bremsstrahlung target produces a well-defined electron-free photon beam. The fission fragments were detected in a vacuum chamber by two large-area surface-barrier detectors (900 mm^2) and conventional electronics. The natural uranium targets consisted of UF_4 (≈ 1 mg/cm^2) evaporated on formvar backings and of metallic uranium (up to 2×4 mg/cm^2), respectively.

2.2. TIMING METHODS

The time distributions were measured by pulsed-beam technique, operating the linac with 100 ns pulses and a repetition rate of 800 Hz ($E = 45$–50 MeV). The fragment pulses served as start signals for the time to pulse height converter. Three delayed sources were investigated as stop signals:

(i) a fast scintillation counter detecting the γ-flash;

(ii) the gun trigger pulse of the linac;

(iii) a ferrite monitor measuring the electron beam pulse.

The time scale was absolutely calibrated with the Ortec time calibrator.

2.3. TESTS

The quality of the timing as well as the background level from fast neutron induced fission were checked by test measurements on ^{232}Th, where no delayed fission is expected. The measured prompt time distribution reproduced the time profile of the beam burst with a full half-width of 80–100 ns and a slope of 10–20 ns for a decrease by a factor of ten. The ^{232}Th measurements showed that the background is negligible.

2.4. EVALUATION

The half-life for isomeric fission was determined by a least-squares fit to the delayed time distribution after summing up an appropriate number of channels in order to improve the statistic of the channel contents. The ratio of isomeric to prompt fission yields can be evaluated from the measured prompt events and the extrapolation of the delayed time distribution. At this procedure the form of the prompt time distribution has to be taken into account.

3. Results

Six measurements were performed. Fig. 1 shows a typical time distribution. Fig. 2 gives an example for a decay curve together with the result of the least-squares fit. In table 1 we have summarized the results of all six measurements. A total sum of

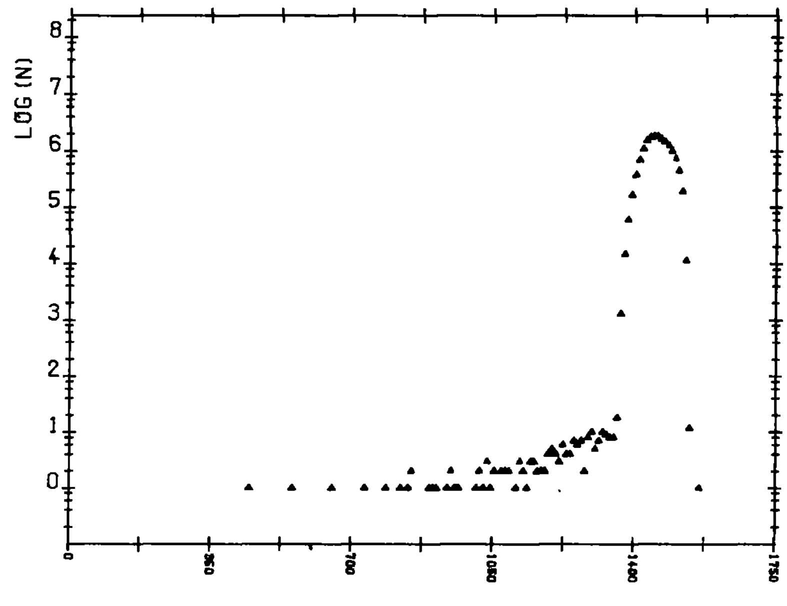

Fig. 1. Example of a measured time distribution of the fission fragments.

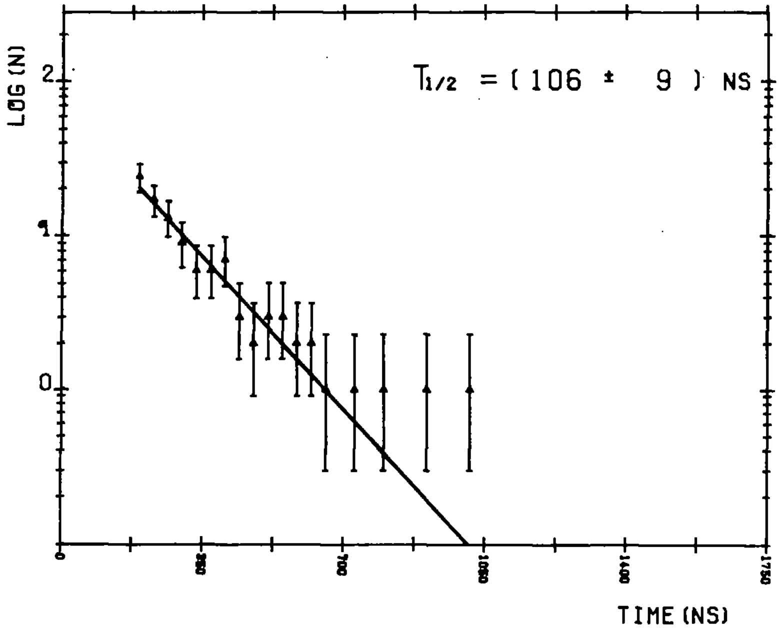

Fig. 2. Example of a measured decay curve (the straight line represents a least-squares fit to the data).

about 7.4×10^7 prompt fission fragments was detected. For all measurements the half-lives as well as the ratios of isomeric to prompt fission yields agree within the statistical errors. The weighted mean values are

$$T_{\frac{1}{2}} = 115 \pm 5 \text{ ns}, \qquad \frac{Y_{\text{iso}}}{Y_{\text{prompt}}} = (2.02 \pm 0.16) \times 10^{-5}.$$

TABLE 1

Experimental results

E_0 (MeV)	Timing	$N_{\text{prompt}} \times 10^{-6}$	$T_{1/2}$ (ns)	$(Y_{\text{iso}}/Y_{\text{prompt}}) \times 10^5$
50	scintill.	5.2	127±11	1.94±0.41
45	trigger	7.7	107±17	1.59±0.47
45	trigger	13.7	118±13	2.09±0.42
45	ferrite	16.0	106± 9	2.13±0.37
45	ferrite	14.8	119±10	2.18±0.37
45	ferrite	19.4	111±10	2.02±0.32
		total:	weighted mean:	
		73.8	115± 5	2.02±0.16

4. Discussion

4.1. COMPARISON WITH RESULTS FROM PARTICLE-INDUCED REACTIONS

In table 2 our results are compared with those from particle-induced reactions populating the shape isomer in ^{236}U. A good agreement of our measured half-life is evident, in particular with the most exact data from Christiansen *et al.* [6]). Therefore we ascribe the observed isomer to ^{236}U populated by the ^{238}U(γ, 2n) reaction. The isomeric-to-prompt fission ratio in the photonuclear reaction is of the same order of magnitude as in particle-induced reactions, although the reaction mechanisms are quite different.

TABLE 2

Comparison of our results with previous investigations of the ^{236}U form isomer

Reaction	Energy of the projectile (MeV)	$T_{1/2}$ (ns)	$(\sigma_{\text{iso}}/\sigma_{\text{prompt}})\times 10^5$	Ref.
^{235}U(d, p)	11.0	130±30	0.96±0.27	[9])
	13.0	110±50	0.31±0.05	[9])
	12.0	130±15	8.6 ±1.3	[10])
	11.0		0.9 ±0.1	[11])
	11.0	130±40	1.46±0.73	[12])
	11.0	116± 7	1.24±0.06	[6])
^{236}U(d, pn)	18.0	105±20		[7])
^{235}U(n, γ)	therm.		<5	[13])
			<4	[14])
			1.0 ±0.2	[15])
	0.5		<2.7	[16])
	2.5		<5	[16])
^{238}U(γ, 2n)	<45	115± 5	2.02±0.16	this work

4.2. THE INTEGRATED ISOMERIC FISSION CROSS SECTION

Since we worked with bremsstrahlung the yield ratio is given by

$$\frac{Y_{\text{iso, f}}}{Y_{\text{prompt}}} = \frac{\int^{E_{\max}} N(E_\gamma, E_{\max})\sigma_{\text{II, f}}(E_\gamma)\text{d}E_\gamma}{\int^{E_{\max}} N(E_\gamma, E_{\max})\sigma_{\gamma,\text{F}}(E_\gamma)\text{d}E_\gamma}, \tag{1}$$

where $N(E_\gamma, E_{\max})$ is the shape of the bremsstrahlung spectrum, $\sigma_{\text{II, f}}(E_\gamma)$ is the isomeric fission cross section, and $\sigma_{\gamma,\text{F}}(E_\gamma)$ is the total prompt photofission cross section. The spectrum shape $N(E_\gamma, E_{\max})$ can be well approximated by a theoretical Schiff spectrum for our arrangement [17]). Absolute photofission cross sections were measured [18, 19]) in the most important region of the giant resonance up to 18 MeV and can be extrapolated to higher energies in a reliable way [20]) using experimental Γ_n/Γ_f values [19]). The energy dependence of the cross section for the isomer population can be calculated within an evaporation model given by Vandenbosch [21]) for two-neutron evaporation reactions. This cross section is concentrated in a small energy interval, in our case at about 16 MeV. Using absolute prompt photofission cross sections [19]), a Schiff bremsstrahlung spectrum and the calculated shape of $\sigma_{\mathrm{II, f}}$, it is therefore possible to extract from our measured yield ratio an absolute integrated cross section for the isomeric fission in the reaction $^{238}\mathrm{U}(\gamma, 2\mathrm{n})^{236}\mathrm{U}$ of 32 ± 6 μb · MeV. The quoted error is determined by the statistical error of the measured yield ratio and the systematic uncertainty of the absolute photofission cross sections.

4.3. ESTIMATION OF THE BRANCHING RATIO $\Gamma^{\gamma}_{\mathrm{II}}/\Gamma^{\mathrm{f}}_{\mathrm{II}}$

In the following section we try to estimate the branching ratio $\Gamma^{\gamma}_{\mathrm{II}}/\Gamma^{\mathrm{f}}_{\mathrm{II}}$ for the decay of the shape isomer in ^{236}U by comparing the experimentally observed cross section for isomeric fission with a calculated cross section for the total production of the isomer. Here $\Gamma^{\gamma}_{\mathrm{II}}$ represents the width for a γ-decay of the shape isomer back to the first minimum, and $\Gamma^{\mathrm{f}}_{\mathrm{II}}$ that for isomeric fission. The meaning of all other width symbols used in this section can be seen in figs. 3 and 4.

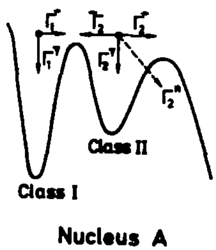

Fig. 3. Meaning of the different decay and penetration widths in the two wells.

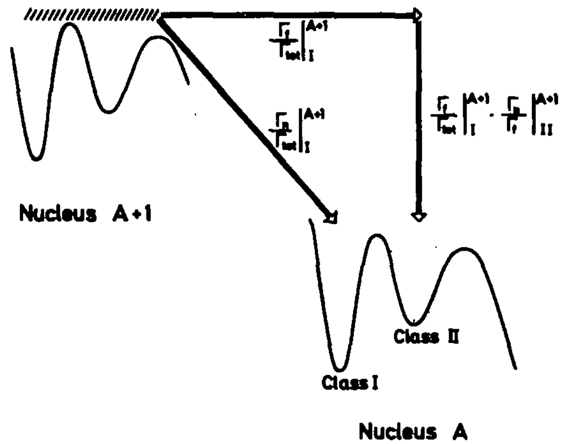

Fig. 4. Scheme for the population of the two wells of the nucleus A by neutron evaporation from compound states of the nucleus $A+1$.

Energy dependence of the isomer production. The energy dependence of the isomer production cross section σ_{II} can be calculated using Vandenbosch's model [21]) for two-neutron evaporation reactions:

$$\frac{\sigma_{\mathrm{II}}}{\sigma(\gamma, 2n)} \approx \frac{\rho_{\mathrm{II}}}{\rho_{\mathrm{I}}} \frac{I(\Delta_2^{\mathrm{i}}, 3) - I(\Delta_3, 3)}{I(\Delta_2, 1) - I(\Delta_3, 3)}. \tag{2}$$

The Pearson functions I represent integrals over level densities; the arguments contain known barrier parameters, binding energies and the mean temperature T. The $\rho_{\mathrm{I,II}}$ are the level densities in the two wells respectively. Assuming a simple level-density formula $\rho \approx e^{E/T}$ the ratio $\rho_{\mathrm{II}}/\rho_{\mathrm{I}}$ is a constant. For the calculations we used the absolute (γ, 2n) cross section of the Livermore group [19]) and the parameters $\rho_{\mathrm{I}}/\rho_{\mathrm{II}} = 250$ [ref. [22])], $E_{\mathrm{B}}^{236} = 6$ MeV, $E_{\mathrm{II}}^{236} = 2.3$ MeV and a mean temperature of $T = 0.5$ MeV (E_{B} is the height of the outer barrier, and E_{II} is the height of the second minimum).

Normalization of the calculated cross section σ_{II}. The absolute scale of σ_{II} was adjusted following a consideration of Anderson *et al.* [15]), applied to the case of ^{235}U(n, γ). In the most important energy region near the barriers of ^{236}U, the ratio $\sigma_{\mathrm{II}}/\sigma_{\mathrm{I}}$ of the population into the wells can be expressed in terms of the corresponding widths (see fig. 3):

$$\frac{\sigma_{\mathrm{II}}}{\sigma_{\mathrm{I}}} = \frac{\Gamma_2^{\gamma}\Gamma_1^{\rightarrow}}{\Gamma_1^{\gamma}\Gamma_2^{\mathrm{tot}}} \qquad (\Gamma_2^{\mathrm{tot}} = \Gamma_2^{\gamma}+\Gamma_2^{\mathrm{n}}+\Gamma_2^{\rightarrow}+\Gamma_2^{\leftarrow}). \tag{3}$$

The ratio of the γ-widths is given by

$$\frac{\Gamma_2^{\gamma}}{\Gamma_1^{\gamma}} = \frac{(E-E_{\mathrm{II}}-\Delta)^2}{(E-\Delta)^2}, \tag{4}$$

where E is the excitation energy and Δ is the pairing energy ≈ 1 MeV. With

$$\frac{\Gamma_1^{\rightarrow}}{\Gamma_2^{\mathrm{tot}}} \approx \frac{\Gamma_1^{\rightarrow}}{\Gamma_2^{\leftarrow}+\Gamma_2^{\rightarrow}}, \tag{5}$$

and introducing the number of open channels N_{A} and N_{B} at the inner and outer barrier respectively we get

$$\frac{\sigma_{\mathrm{II}}}{\sigma_{\mathrm{I}}} = \frac{(E-E_{\mathrm{II}}-\Delta)^2}{(E-\Delta)^2}\,\frac{N_{\mathrm{A}}}{N_{\mathrm{A}}+N_{\mathrm{B}}}\,\frac{\rho_{\mathrm{II}}}{\rho_{\mathrm{I}}}. \tag{6}$$

Assuming $N_{\mathrm{A}} = N_{\mathrm{B}}$ for the low spin states populated in the (γ, 2n) reaction, we adjusted the isomer production cross section calculated within the Vandenbosch model at an excitation energy of 6.4 MeV in ^{236}U (neutron binding energy, corresponding to $E_\gamma \approx 17$ MeV). The so calculated cross section $\sigma_{\mathrm{II}}(E_\gamma)$ has a maximum value of ≈ 64 μb at 16 MeV, a half-width of 3.5 MeV and amounts to an integrated value of 208 μb · MeV.

Comparison of σ_{II} *with the* $\Gamma_{\mathrm{n}}/\Gamma_{\mathrm{f}}$ *systematics.* To check our calculations the maximum value of $\sigma_{\mathrm{II}}(E_\gamma)$ can be compared with the systematics of isomer production cross sections reported by Borggreen *et al.* [23]) for two-neutron evaporation reactions.

In this systematic the quantity $[\Gamma_{\mathrm{n}}/\Gamma_{\mathrm{f}}]_{\mathrm{II}}^{A+1}$ is plotted, describing the branching ratio for neutron emission and fission of the nucleus $A+1$ at the deformation of the second minimum. The ratio $\sigma_{\mathrm{II}}/\sigma_{\mathrm{I}}$ of the cross sections for the population of the two wells can be written as (see fig. 4)

$$\frac{\sigma_{\rm II}}{\sigma_{\rm I}} = \left[\frac{\Gamma_{\rm n}}{\Gamma_{\rm f}}\right]_{\rm II}^{A+1} \bigg/ \left[\frac{\Gamma_{\rm n}}{\Gamma_{\rm f}}\right]_{\rm I}^{A+1}. \tag{7}$$

In our case $\sigma_{\rm I}$ is given by the $\sigma(\gamma, 2n)$ cross section for ^{238}U. Using the peak value of our calculated isomer population cross section ($\sigma_{\rm II\,max} = 64$ μb) and the experimental values reported by the Livermore group [19]) ($[\Gamma_{\rm n}/\Gamma_{\rm f}]_{\rm I}^{A+1} = 2.35$ for ^{237}U; $[\sigma(\gamma, 2n)]_{E_\gamma = 16\,\rm MeV} = 120$ mb) we get:

$$\left[\frac{\Gamma_{\rm n}}{\Gamma_{\rm f}}\right]_{\rm II}^{237} = 1.25 \times 10^{-3}, \tag{8}$$

which fits very well the systematics. This shows that the calculated isomer population cross section is quite reasonable.

The branching ratio $\Gamma_{\rm II}^{\gamma}/\Gamma_{\rm II}^{\rm f}$. The calculated isomer population cross section of 208 μb·MeV must be compared with the measured isomeric fission cross section of 32 ± 6 μb·MeV. The discrepancy may be attributed to a competing γ-decay mode back to the first minimum. With this assumption one can deduce a branching ratio for the isomer decay of

$$\Gamma_{\rm II}^{\gamma}/\Gamma_{\rm II}^{\rm f} \approx 6. \tag{9}$$

This value for the branching ratio is in reasonable agreement with most recent estimates [15]) and preliminary experimental results of the Heidelberg group [24]).

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