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TRI-PP-85-63
Aug 1985

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Muon induced fission in ^{235}U and ^{238}U

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(Submitted to Canadian Journal of Physics)

Index classification: 25.85

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ABSTRACT

Muon induced prompt and delayed fission yields in ^{235}U and ^{238}U have been measured. A coincidence with the muonic uranium K_{α} X-rays was used to identify the muon stop in the target. The experimental absolute fission yields per muon stop were 0.142 ± 0.023 for ^{235}U and 0.068 ± 0.013 for ^{238}U . The disappearance rate of muons from the 1s state of muonic uranium has also been measured in the fission mode. Muon induced fission lifetimes were 71.6 ± 0.6 ns for ^{235}U and 77.2 ± 0.4 ns for ^{238}U . No evidence for a short lifetime fission-isomer component was found. Comparison of lifetime results with previously measured values in the electron, gamma and neutron decay modes indicates that the systematic discrepancies can be explained by muon capture on fission fragments produced from prompt fission.

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1. Introduction

Nuclear fission is induced by stopping μ^- in heavy nuclei through two different mechanisms. In the predominant mode, fissions occur from excited nuclear states produced by μ^- capture. This process exhibits the time dependence of the μ^- capture process (1) and gives rise to a symmetric mass distribution (2) of the fission fragments characteristic of the mean excitation energy of 15 to 20 MeV. In heavy nuclei, fission is also induced when the nucleus is nonradiatively (3,4) excited by the muon cascading through its atomic levels. It has been suggested that both quadrupole and dipole transitions can contribute to nonradiative excitation (4,5). The time delay of the fission induced by this process is on the order of atomic cascade times ($\sim 10^{-14}$ s), and the fission fragment mass distribution is asymmetric, due to the lower excitation energies (6-10 MeV) involved. It has been speculated that a nonradiative process might also excite shape isomeric levels (6-8) which could be detected by the time dependence of subsequent fissions from the isomeric state.

At TRIUMF, we have measured the absolute fission yields (delayed and prompt) and time distributions of fission induced by μ^- stopping in ^{235}U and ^{238}U . A coincidence with muonic uranium K_α x-rays was required to tag muons captured on uranium nuclei in the fission chambers. An abbreviated report of this work (based on the first experimental run) has been published (9); the present paper includes additional data from subsequent measurements in which a number of technical improvements were implemented.

2. Experimental Method

The experiment was performed using the cloud muon beam from the TRIUMF M9 channel (10). A schematic of the experimental setup is shown in Fig. 1. Absorbers D1 and D2 were used to slow the muons and to eliminate contaminant pions from the beam. D2 was placed very near to the chamber to reduce the multiple scattering of muons before they reached the target. The thickness of absorbers D1 and D2 was adjusted to maximize muon stops in the uranium target. The muon stop signal was obtained from a conventional [1.2-3.4] scintillator telescope. In addition, a large pulse (signifying a slow muon) was required in the thin scintillator S_3 immediately upstream of the fission chamber target. Contaminant pions and electrons in the beam (96 MeV/c in the first run and 77 MeV/c in the second run) were also electronically vetoed in the stop signal by a cut on the beam time of flight in the channel. In addition, in the second run, an electrostatic crossed-field particle separator was used to deflect contaminant pions and electrons from the muon beam. Events for which two muon stops were registered within 1 μs were rejected.

Multiplate avalanche chambers were used to detect fissions; X rays from the muon cascade in uranium were detected by a large volume, high resolution Ge(Li) detector in the first run, and by three NaI(Tl) detectors (15.2 cm $\times$ 12.7 cm diameter) in the second run. All events in which a muon stop was accompanied by an X-ray or a fission were recorded on magnetic tape with a computer-based data acquisition system. For each event, the pulse amplitudes and times relative to the muon stop were recorded for the fission

chamber and Ge(Li) or NaI(Tl) detectors. Only 10% of the NaI(Tl) pulses unaccompanied by a fission were recorded.

The multiplate fission chamber (11), contained in a propane filled vessel, is shown in Fig. 2. It consisted of a stacked sandwich of parallel-plate avalanche chambers and uranium target material deposited on thin substrate foils. The uranium targets and detector plates were sufficiently thin so that both fission fragments could reach a sensitive detector region if emitted almost normally to the target plane. Technical details of the three chambers are shown in Table 1. The efficiencies of chambers "A" and "B" used in the first run, calibrated by measuring the ^{235}U fission rates in a known thermal neutron flux at the Berkeley Research Reactor (12,13), were found to be $78 \pm 12\%$ and $47 \pm 9\%$ for ^{235}U and ^{238}U respectively. A higher efficiency ^{238}U chamber, "C", was constructed for the second run. Its efficiency was calibrated by making a relative measurement of photofissions in the chamber and in a standard proportional counter containing an extremely thin $0.0867 \pm 0.0026 \text{ mg/cm}^2$ natural UF_6 foil, fission fragments from which were detected with 100% efficiency. The efficiency of this ^{238}U chamber was found to be $79 \pm 5\%$.

3. Data analysis and results

Energies and time distributions of both fissions and X-rays following a muon stop were deduced from data recorded on magnetic tape. The fission energy histograms (Figs. 3 and 4) contain, in addition to pulses from fission fragments, low energy alpha pulses. Alphas and fissions were well separated in energy in the ^{238}U chamber. In the ^{235}U chamber, because of the higher alpha

activity, there was an overlap between the low energy fissions and the alphas. In order to optimize the sensitivity of the timing spectra shown in Fig. 5 to the fission lifetimes, an energy threshold was set to reduce accidental alpha background. To determine the effect of this threshold on the detection efficiency, the energy spectrum of the fission fragments is required. This was obtained by subtracting from the total energy spectrum a spectrum taken at negative time which contained only the alpha background. The resultant spectrum for ^{235}U is shown in Fig. 6.

The absolute capture fission yield per muon stop was deduced using the X ray-fission coincidence technique. This yield is given by

$$Y_f = \frac{N_{KX}/\epsilon_f}{N_X}$$

where N_{KX} is the number of fissions that occurred in coincidence with N_X K_α X-rays, N_X is the number of K_α X-rays and ϵ_f is the fission detection efficiency. Since the NaI X-ray data have a considerable amount of neutron capture gamma background at 6 MeV, the X-ray data obtained using Ge detectors were used for absolute yield determination.

A. FISSION CHAMBER TIME RESOLUTION

The fission chamber time resolution was obtained by studying the time distribution of pion induced prompt fission events. For this measurement, the d.c. separator and the channel flight time cut were adjusted to select pions. The time distributions were measured with a time-to-digital converter (TDC), which was then calibrated with an ORTEC 462 time calibrator. The time

calibration data were fitted to a quadratic function of channel number. The time resolution of the chambers was about 4 ns (FWHM).

B. MUON INDUCED FISSION LIFETIMES

Muon induced fission lifetimes were obtained from the fission time distributions using least squares fitting methods. The fitting function consisted of a prompt component, one or two exponentially decaying components, and a time independent background, all convoluted with the Gaussian fission chamber resolution function (12). Fitting was accomplished by a modified iterative least squares procedure.

The fission time distribution for ^{235}U was fitted well over the full time range with a mean lifetime of $\tau = 71.6 \pm 0.6$ ns. The ^{238}U spectrum obtained with the fission chamber design identical to that of the ^{235}U chamber was well fitted with a single component having mean life of 77.2 ± 0.4 ns.¹ The lifetime was found not to be sensitive to the choice of fission energy threshold and the effect of nonlinearity in the time calibration of the TDC on the lifetime were found to be less than 0.1%. Typical time distributions of fission relative to muon stop in ^{235}U and ^{238}U are shown in Fig. 5. The solid curves in the figure show the least squares fit to the data. Results obtained from both runs are summarized in Table 2. Taking into account the different conditions of the two runs, preferred values were also selected. Our preferred

¹No evidence was found for a second component with $\tau = 18 \pm 5$ ns (intensity $8 \pm 2\%$) as was required in fitting our previous data (9).

values along with results from previous measurements (2,8,14-24) are summarized in Table 3.

C. MUON CAPTURE RATE

The muon disappearance rates ($1/\tau$) in ^{235}U and ^{238}U in the fission mode were obtained from the time distributions of fission fragments after the muon stop. Muon capture rates, γ_c , were deduced from the disappearance rates using the relation:

$$\gamma_c = \frac{1}{\tau} - R\gamma_d$$

where γ_d is the free muon decay rate and $R(\sim 0.85)$ accounts for the reduction of the decay rate due to the effect of atomic binding (25). Using $\tau = 77.2 \pm 0.4$ ns for ^{238}U and 71.6 ± 0.6 ns for ^{235}U , muon capture rates of $(125.7 \pm 0.7) \times 10^5$ per second and $(135.8 \pm 1.2) \times 10^5$ per second, respectively, were obtained.

The difference in the muon capture rates in ^{235}U and ^{238}U may be explained as a consequence of the isotope effect pointed out by Primakoff (26). The relative capture rates are in good agreement with Primakoff's original calculation but not in as good agreement with the later calculation of Goulard and Primakoff (27).

D. PROMPT-TO-DELAYED FISSION YIELD RATIO

The prompt-to-delayed fission yield ratios were calculated using the parameters obtained from the least squares fit to the fission time distributions (12). The contribution from the π^- induced fission to the muon induced prompt fission in the first run was < 0.002 per μ^- stop. This value was estimated from the probable pion contamination in the muon beam and the value of the

fission yield per pion stop (14). In the second run, the pion contamination was further suppressed by the electrostatic separator and no correction was needed. Present results of prompt-to-delayed fission yield ratios along with other results are summarized in Table 4.

E. ABSOLUTE FISSION YIELD

The absolute yields of prompt and delayed fission induced by negative muons in ^{235}U and ^{238}U were obtained using the X-ray fission coincidence data. Muon stops in the uranium target material were identified by the muonic uranium K_α and L_α X rays in coincidence with the muon stop. Absolute delayed fission yields were obtained by identifying muon capture in uranium with a muonic uranium K_α X-ray preceding the fission event. The delayed fission yields per K_α X-ray were 0.125 ± 0.023 for ^{235}U and 0.062 ± 0.013 for ^{238}U . Using the preferred prompt-to-delayed fission yield ratios shown in Table 2, the absolute prompt fission yields, (0.017 ± 0.003) for ^{235}U and (0.006 ± 0.001) for ^{238}U per muon stop were calculated. The absolute total yield per muon stop which is the sum of delayed and prompt yield was 0.142 ± 0.023 for ^{235}U and 0.068 ± 0.013 for ^{238}U .

This is the first direct absolute fission yield measurement in ^{235}U . Previously absolute fission yields were inferred (14,15) from relative measurements (14,31) with respect to ^{238}U . Thus to some extent the disagreement among ^{235}U results reflects the variations in the ^{238}U yields used. Large variations in the absolute fission yield of ^{238}U indicated a systematic experimental uncertainty which might be due to inaccurate estimation of the

fraction of muons captured by uranium. The present absolute fission yields per muon stop in ^{235}U and ^{238}U using X-ray fission coincidence technique are summarized along with previous results (14,15,29-31) in Table 4.

F. LIFETIME DISCREPANCY

Muon capture lifetimes measured in the fission mode are consistently slightly shorter than the lifetimes determined from other decay modes as shown in Table 3. The possible reason for this apparent difference has been theoretically explained (6,7). Using Hadermann's approach (7) the mean lifetime measured by detecting the decay electron (τ_e) can be expressed as (12)

$$\tau_e = \tau + \beta_e \gamma_p (\tau' - \tau)$$

where τ is the muon induced fission lifetime of uranium, $\tau' \approx 130$ ns is the average of the muon capture lifetime of the fission fragment, $\beta_e = \tau'/\tau$ is the muon decay electron yield on the fragment relative to that on uranium, and γ_p is the measured prompt fission yield per muon stop. Using our prompt fission yields and fission lifetimes, $\tau_e = \tau(1.9 \pm 0.4)$ ns in ^{235}U and (0.6 ± 0.1) ns in ^{238}U . Lifetime differences measured in the neutron of gamma modes may also be calculated by using for $\beta_{n,\gamma}$ the ratios of the neutron or gamma ray multiplicities from fragment muon capture to those from uranium capture. The values ($\tau_{n,\gamma} - \tau$) will be considerably smaller than ($\tau_e - \tau$) if $\beta_{n,\gamma} \lesssim 1$ is assumed. However, the lifetime differences estimated above are smaller than some of the measurements would suggest.

In the present measurement, evidence for a short fission lifetime component (9,23) in ^{238}U to be associated with a possible isomer excitation has been searched for (12). At the 95% confidence level, no candidates for a short lifetime fission isomer component have been found with yields greater than 0.05% per stopping muon. This does not eliminate the possibility of isomer excitation (6) during the muonic Coulomb cascade. In the presence of a 1s muon, since the inner barrier becomes more transparent compared to the outer barrier, the fission mode of decay would be suppressed, allowing the excited nucleus in the shape isomeric state to decay back to the ground state with the emission of a gamma. However a back decay gamma component (8), suggesting possible isomeric excitations, has not been found with yields greater than 0.2% per stopping muon (32,33).

4. Acknowledgements

The authors are indebted to J.L. Gallant for his preparation of the uranium foils, and to R.M. Pearce, M.S. Dixit and A. Miresghi for their help during the data taking phase of the experiment. One of the authors (S.A.) is grateful to the Canadian Commonwealth Fellowship Committee and to the Government of Bangladesh for kind support. This experiment was supported by the Natural Science and Engineering Research Council of Canada under grant IEP-11, and by the U.S. Department of Energy under contract W-7905ENG48.

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TABLE 1. Technical Details of the Fission Chambers

	²³⁵ U chamber "A"	²³⁸ U chamber "B"	Redesigned ²³⁸ U chamber "C"
Number of target plate	17	10	19
Chemical form of target	UF ₄	UO ₂ (NO ₃) ₂ , 6H ₂ O	UF ₄
Total mass of ²³⁵ U	270 mg	0.13 mg	3.8 mg
Total mass of ²³⁸ U	14 mg	470 mg	528 mg
Enrichment	95%	99.97% 0.028% ²³⁵ U	99.3% 0.7% ²³⁵ U
Total Uranium/cm ²	10.2 mg/cm ²	19.6 mg/cm ²	13.9 mg/cm ²
Thickness per plate:			
Uranium	0.60 mg/cm ²	1.96 mg/cm ²	0.73 mg/cm ²
F, O, N, etc.	0.20 mg/cm ²	2.18 mg/cm ²	0.23 mg/cm ²
Target support	0.70 mg/cm ² (aluminum)	0.50 mg/cm ² (wyilar)	0.30 mg/cm ² (aluminum)
Thickness of one avalanche counter foil	0.25 mg/cm ²	0.50 mg/cm ²	0.25 mg/cm ²

TABLE 2. Present results for muonic ^{235}U , ^{238}U

Target Used	Mean Lifetime τ_μ (ns)	Fission Probability (Prompt Delayed)	Absolute delayed fission yield per muon stop
^{235}U First run (9)	71.5 ± 0.9	0.138 ± 0.009	0.125 ± 0.023
Second run	71.6 ± 0.7	0.133 ± 0.007	-
Preferred values	$71.6 \pm 0.6^*$	$0.133 \pm 0.007^{**}$	0.125 ± 0.023
^{238}U First run (9)	76 ± 1.3 18 ± 5	0.089 ± 0.017	0.062 ± 0.013
Second run	77.2 ± 0.4	0.099 ± 0.005	-
Preferred values	$77.2 \pm 0.4^{***}$	$0.099 \pm 0.005^{**}$	0.062 ± 0.013

*Weighted average value.
 **Values obtained using electrostatically separated μ^- beam.
 ***Values obtained using the redesigned chamber identical to ^{235}U chamber.

TABLE 3. Experimental mean lifetimes for muonic ^{235}U , ^{238}U

Mode of registration	Mean lifetimes τ_μ (ns)		Reference
	^{235}U	^{238}U	
electron	-	88.0 ± 4.0	(16)
	78.0 ± 4.0	81.5 ± 2.0	(17)
	75.4 ± 1.9	73.5 ± 2.0	(18)
gamma	-	79.1 ± 0.5	(19)
	-	78.6 ± 1.5	(8)
neutron	75.0 ± 0.7	78.3 ± 1.0	(20)
fission	66.5 ± 4.2	75.6 ± 2.9	(21)
	65.3 ± 2.8	74.1 ± 2.8	(22)
	75.6 ± 2.3	76.0 ± 1.0	(14)
	-	77.1 ± 0.2	(23)
	72.8 ± 0.6	77.7 ± 0.6	(15)
	72.9 ± 0.9	77.9 ± 0.5	(24)
	-	78.0 ± 2.0	(2)
	71.6 ± 0.6	77.2 ± 0.4	present

TABLE 4. Fission yields per muon stop and prompt-to-delayed fission yield ratios

Nucleus	Fission probability			Reference
	prompt/delayed	per muon stop		
^{235}U	0.111 ± 0.021	-		(21)
	$0.094 \pm 0.027^*$	-		(28)
	0.170 ± 0.010	0.037 ± 0.009		(14)
	0.115 ± 0.005	0.310 ± 0.060		(15)
	0.133 ± 0.006	0.142 ± 0.023		(present)
^{238}U	-	0.070 ± 0.030		(29)
	-	0.150 ± 0.060		
	-	0.070 ± 0.008		(30)
	0.072 ± 0.014	-		(21)
	$0.080 \pm 0.024^*$	-		(28)
	0.071 ± 0.003	0.031 ± 0.007		(14)
	-	0.150 ± 0.030		(31)
	0.079 ± 0.006	-		(14)
	0.088 ± 0.003	-		(2)
	0.093 ± 0.005	0.068 ± 0.013		(present)

*The published results (20) have been corrected in Rushon's Ph.D. thesis (26).

FIGURE CAPTIONS

1. Experimental setup: (a) First run (b) Second run.
2. A general view of a fission chamber.
3. Typical ^{238}U fission chamber energy spectrum.
4. Typical ^{235}U fission chamber energy spectrum.
5. Typical time distributions of fission events relative to time of muon stop. The solid curve is the least squares fit to the data (2.926 ns/channel).
6. Typical background-subtracted fission amplitude spectrum in ^{235}U .

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6