Measurement of total muon-capture rates in ^{232}Th, ^{235,238}U, and ^{239}Pu†

M. W. Johnson, W. U. Schröder, and J. R. Huizenga
*Nuclear Structure Research Laboratory**
and Departments of Chemistry and Physics, University of Rochester, Rochester, New York 14627

W. K. Hensley and D. G. Perry
Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544

J. C. Browne
Lawrence Livermore Laboratory, Livermore, California 94450
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The disappearance rate of muons from the 1*s* state of muonic actinide atoms has been measured by detecting muon-decay electrons. The experimental muon lifetimes are (79.2 ± 2.0), (75.4 ± 1.9), (73.5 ± 2.0), and (73.4 ± 2.8) nsec for muonic ^{232}Th, ^{235,238}U, and ^{239}Pu, respectively. The muon-capture rates deduced from the data are compared with the predictions of various theories. The experimental values are consistent with the systematics of muon-capture rates in the heavy elements but are generally higher than theoretical predictions, which take deformation of the nuclear charge distribution into account. The results provide no evidence for a muon-induced isomeric fission process.

[RADIOACTIVITY Muonic atoms ^{232}Th, ^{235,238}U, ^{239}Pu; measured muon-disappearance rates, deduced total muon-capture rates, upper limit for population of shape-isomeric states.]

I. INTRODUCTION

Investigating the muon-capture process in the actinide region is of considerable interest from several points of view: Very little experimental data[1-3] on total muon-capture rates are available for this region of nuclei, where the simple Primakoff theory,[4] which successfully predicts experimental capture rates for a wide range of other target nuclei,[5] is expected to fail. Hence, accurate measurements of muon-capture rates in actinides provide a sensitive test of recently developed, improved theories[6-9] on muon capture. Another interesting aspect of such studies has been pointed out by Błocki[10] and Bloom,[11] who suggested that an actinide nucleus may be excited to states in the second well of the double-humped fission barrier by a radiationless muonic cascade transition. Bloom[11] predicts that, due to isomeric fission of the nucleus in the second well, the rate of muon-induced fission should be higher than the muon-capture rate. However, available data on muon-induced fission[12-16] and muon-capture[1-3] in actinides are not conclusive in this respect.

The present study aims at an accurate determination of muon-capture rates by a measurement of of electrons from the decay of muons in the 1*s* orbit of muonic ^{232}Th, ^{235,238}U, and ^{239}Pu. Some details on the experiments and data reduction, as well as the results, are presented in Sec. II. In Sec. III the total muon-capture rates are compared with theoretical predictions and the implications of the present results on the possibility of muonic excitation of second-well states are discussed.

II. EXPERIMENTS AND RESULTS

The experiments were performed at the LAMPF stopped-muon channel of the Los Alamos Scientific Laboratory. A schematic diagram of the experimental setup is shown in Fig. 1. Muons and electrons were measured with separate conventional plastic counter telescopes. Targets were metallic sheets of 5–10 g/cm^2 thickness. A time-to-amplitude converter was started with a fast $123\bar{4}$ coincidence signal indicating a muon stop in the tar-

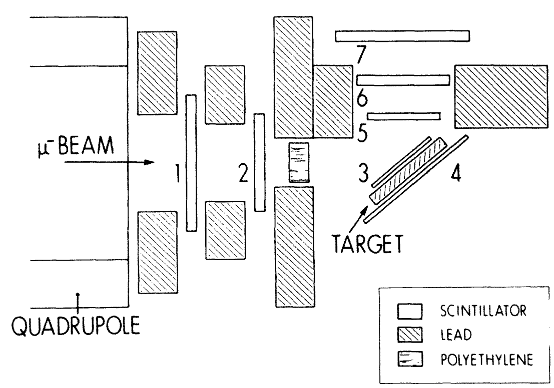

FIG. 1. Experimental arrangement of the muon telescope (1, 2, 3, 4) and the electron telescope (5, 6, 7).

get and stopped by a 567 coincidence signal signifying an electron. In order to reduce background, events were rejected either where more than one muon was following or preceding a muon stop within 5 μsec or where an electron signal appeared in prompt coincidence with a signal from either of the thin (1.59 mm) counters 3 or 4, except when the former belonged to a muon stop coincidence.

Decay electrons were accepted during a time interval from −200 nsec to 3.8 μsec with respect to a muon stop. In Fig. 2 a part of such an electron-time distribution is shown, together with a least-squares fit indicated by the full curve. The total background resulted mainly from events random in time but had a 20% component decreasing exponentially in time with a decay constant τ_B between 1 and 2 μsec, characteristic of muon lifetimes in light materials near the target. Therefore, fits to the ^{232}Th and ^{238}U data were performed using a sum of two exponentials and a constant. Since the targets of ^{235}U and ^{239}Pu were clad in 0.1 g/cm^2 copper foil, another exponential was added to the fit function, in these cases, using the known muon lifetime of $\tau_{\mathrm{Cu}} = 163.5 \pm 2.4$ nsec (Ref. 5) in copper. The results of the fits thus obtained are compared in Table I with those of other authors. In view of the possibility that there might be an excitation mechanism leading to the population of states in the second well,[11] lifetimes deduced from measurements of fragments from muon-induced fission and of nuclear γ rays emitted following muon capture are also included in Table I. The uncertainties quoted for the present results include statistical errors and allow for different assumptions in the decay time of the background and the time interval chosen for the fit. Uncertainties due to a nonlinearity of the system and those of the time calibration were negligible.

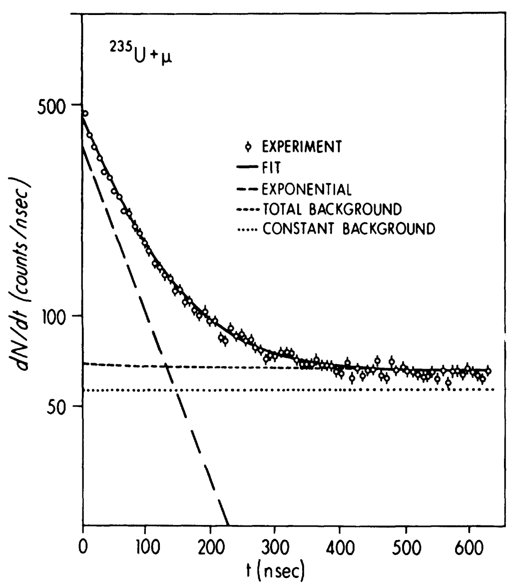

FIG. 2. Part of the experimental electron time distribution of muonic ^{235}U. The solid line represents the fit to the data point. The total background (dashed) is a sum of a random background (dotted) and two exponentials corresponding to muon lifetimes in copper and light materials, respectively.

TABLE I. Lifetimes of muons in muonic actinides, deduced from measurements of decay electrons (e), muon-induced fission fragments (f), and nuclear γ rays (γ) emitted following muon capture.

Isotope	Mode	τ (nsec)	Ref.
^{232}Th	f	74.2 ± 5.6	12
	f	87 ± 4	14
	e	80.4 ± 2.0	2
	e	79.2 ± 2.0	Current work
^{235}U	f	65.3 ± 2.8	13
	f	66.5 ± 4.2	12
	f	84 ± 6	14
	e	78 ± 4	2
	e	75.4 ± 1.9	Current work
^{238}U	f	75.6 ± 2.9	12
	f	74.1 ± 2.8	13
	f	76.0 ± 1.0	14
	e	88 ± 4	1
	e	81.5 ± 2.0	2
	γ	79.5 ± 0.5	3
	e	73.5 ± 2.0	Current work
^{239}Pu	f	74 ± 14	15
	f	70 ± 3	16
	e	77.5 ± 2.0	2
	e	73.4 ± 2.8	Current work

III. DISCUSSION OF RESULTS

The values for the muon lifetimes found in this work are systematically lower than the results of the most recent experiment[2]; however, the agreement between the two sets of electron data is good, except for the case of ^{238}U. Here the two results differ by four standard deviations. An explanation for this discrepancy is presently not available. As can be seen from Table I, the muon lifetimes of this work agree rather well with those deduced from fission measurements, although the latter data show considerable scatter.

From the experimental muon lifetimes τ_e, the total muon-capture rates λ_c^{exp} can be evaluated according to $\lambda_c^{\mathrm{exp}} = 1/\tau_e - R\lambda_0$, where λ_0 is the decay rate of the free muon and R is the Huff factor.[17,18] The latter factor describes the reduction of the muon-decay rate due to the effects of atomic binding ($R \approx 0.85$ for heavy elements). The resulting experimental capture rates are summarized in

Table II and compared with two theoretical predictions. Eckhause *et al.*[5] were able to fit experimental capture rates for a wide range of nuclei with the formula

$$\lambda_c^{\mathrm{th}\,1}(A,Z)=168.44Z_{\mathrm{eff}}^4\left(1-3.14\,\frac{A-Z}{2A}\right)\mathrm{sec}^{-1}, \quad (1)$$

the structure of which was predicted by the Primakoff theory.[4] Here, Z_{eff}^4 represents the overlap integral of the $1s$ muonic wave function with the nuclear charge distribution. Goulard and Primakoff[6] have determined unknown matrix elements of their improved model by a fit to experimental data yielding

$$\lambda_c^{\mathrm{th}\,2}(A,Z)=218.41Z_{\mathrm{eff}}^4\left(1-\frac{\epsilon_\mu}{m_\mu}\right)^2\left[1-2\left(\frac{m_\mu-\epsilon_\mu}{2m_N}\right)\right] \times\left[1-0.03\,\frac{A}{2Z}+0.25\,\frac{A-2Z}{2Z}-3.24\left(\frac{A-Z}{2A}+\frac{|A-2Z|}{8ZA}\right)\right], \quad (2)$$

where ϵ_μ is the $1s$ muon binding energy of approximately 12 MeV, and m_μ and m_N are the muon and nucleon masses, respectively. The values Z_{eff}^4 used to generate the theoretical rates of Table II were calculated by solving numerically the Dirac equation for a deformed nuclear charge distribution $\rho(\vec{r})$ of the Fermi type[19]:

$$\rho(\vec{r})=N[1+\exp(4\ln 3\{r[1-\beta Y_0{}^2(\theta)]-c\}/t)]^{-1}, \quad (3)$$

where N is a normalization factor, c and t are the half-density radius and the surface thickness, respectively, and β is a deformation parameter. All charge-distribution parameters were taken from de Wit *et al.*[19] The ratios $Z_{\mathrm{eff}}^4(\beta)/Z_{\mathrm{eff}}^4(0)$ of the overlap integrals evaluated with a deformed and a spherical nuclear charge distribution have been included in Table II. They show that the decrease of Z_{eff}^4 by ~5% with respect to the spherical value caused by nuclear deformation is of the same order as the experimentally observed variation of the capture rates. Hence, it is clearly not legitimate to neglect this effect for actinide muonic atoms.

Comparing the experimental muon-capture rates with the theoretical predictions as listed in Table II, one notices that the general trend of increasing experimental capture rate with increasing Z is reproduced by theory. However, the theoretical values are generally too low, and the theoretical dependence of the capture rate on A and Z appears to be somewhat stronger than experimentally observed. This applies, in particular, to the simpler Primakoff theory. Whereas in the present experiment no significant isotopic effect of the capture rate in ^{235,238}U was found, theories predict an effect of 10–20%. A similar overestimation of the isotopic effect by theory has been observed also in the cases of muon capture in ^{203,205}Tl (Ref. 20) and ^{206,207,208}Pb (Ref. 21). It appears, however, that the Goulard-Primakoff formula Eq. (2) represents a definite improvement over the older Primakoff formula Eq. (1).

The present results suggest that, within the limits of experimental accuracy, there is no difference $\Delta\lambda=\lambda_f-\lambda_e$ between the disappearance rate $\lambda_e=1/\tau_e$ and the rate λ_f of muon-induced fission except possibly in the case of ^{235}U. Bloom[11] derives a relation

$$\Delta\lambda=\frac{\epsilon\lambda_{\mathrm{if}}}{\alpha\lambda_c}(\lambda_{\mathrm{if}}+\lambda_{\mathrm{bt}}), \quad (4)$$

where ϵ is the relative population of states in the second well as compared with those of the first well, α is the branching ratio of fission following muon capture, λ_{if} is the isomeric-fission rate, and λ_{bt} is the rate for the system in the second well to tunnel back into the first well. As has been noticed by Bloom himself, the quantities entering the calculation are subject to serious uncertainty. Taking the present value for the muon-capture lifetime of muonic ^{238}U and a weighted average of the corresponding experimental fission lifetimes, one arrives at $\Delta\lambda\leqslant 4\times10^4$ sec^{-1}. With $\alpha=0.03\pm0.007$,[22] $\lambda_{\mathrm{bt}}=5\times10^6$ sec^{-1},[23] and $\lambda_{\mathrm{if}}=1.2\times10^5$ sec^{-1},[23] Eq. (4) leads to a relative population of second-well states of $\epsilon\leqslant3\times10^{-2}$ during the muon cascade. However, Eq. (4) was derived under the assumption that λ_c and α are the same for both

TABLE II. Experimental muon-capture rates λ_c^{exp} compared with the predictions of the Primakoff formula ($\lambda_c^{\mathrm{th}\,1}$) and of the Goulard-Primakoff theory ($\lambda_c^{\mathrm{th}\,2}$). Capture rates are given in units 10^7 sec^{-1}. $Z_{\mathrm{eff}}^4(\beta)/Z_{\mathrm{eff}}^4(0)$ is the ratio of capture rates evaluated with a deformed and spherical nuclear charge distribution.

Target	λ_c^{exp}	$Z_{\mathrm{eff}}^4(\beta)/Z_{\mathrm{eff}}^4(0)$	$\lambda_c^{\mathrm{th}\,1}$	$\lambda_c^{\mathrm{th}\,2}$
^{232}Th	1.22 ± 0.03	0.966	0.94	1.11
^{235}U	1.29 ± 0.03	0.959	1.08	1.20
^{238}U	1.32 ± 0.04	0.955	0.89	1.08
^{239}Pu	1.33 ± 0.04	0.952	1.18	1.28

wells, and λ_{if} was taken to be that of the bare nucleus. These assumptions are not justified. The fission barrier is raised due to the muon-nuclear Coulomb interaction,[10] and λ_{if} may be reduced by a factor of 10. Furthermore, because of the larger deformation, the capture rate in the second well may be lower by ~5% and α may be considerably higher than the corresponding values for the nucleus in the first well.

It is straightforward to modify Bloom's relations to take these effects into account. Denoting the relevant quantities for the first and second well with the superscripts I and II, respectively, one defines $\Delta\lambda_0 = \lambda_0^{\mathrm{I}} - \lambda_0^{\mathrm{II}}$, $\Delta\lambda_c = \lambda_c^{\mathrm{I}} - \lambda_c^{\mathrm{II}}$, and $\Delta\alpha = \alpha^{\mathrm{I}} - \alpha^{\mathrm{II}}$. Following Bloom's procedure, one arrives at

$$\Delta\lambda = \frac{\epsilon\lambda_{if}}{\alpha^{\mathrm{I}}\lambda_c^{\mathrm{I}}}(\lambda_{if} + \lambda_{bt} - \Delta\lambda_c) \times\left(1 - \frac{\lambda_c^{\mathrm{I}}}{\lambda_{if}}\Delta\alpha - \frac{\alpha^{\mathrm{I}}}{\lambda_{if}}\Delta\lambda_c + \frac{\alpha^{\mathrm{I}}\lambda_c^{\mathrm{I}}}{\lambda_{if}\lambda_0^{\mathrm{I}}}\Delta\lambda_0\right) \qquad (5)$$

instead of Eq. (4).

Of the three correction terms, the one including $\Delta\lambda_0$ can be neglected, since the Huff factor is expected to be nearly independent of the nuclear deformation. However, both of the other terms have to be taken into account. Calculations suggest that $\Delta\lambda_c$ is of order 10^6 sec^{-1} for actinide muonic atoms. Even larger corrections may result from the $\Delta\alpha$ term. Muon capture by a nucleus in the second well will lead to an increase of the mean excitation energy with respect to the value for the first well equal to the energy difference between the ground states in the first and second well. Consequently, the probability for second- and third-chance fission will be enhanced. Although estimation of the importance of this effect is difficult, it is likely that $|\Delta\alpha/\alpha|$ may be as high as 0.5 for lighter actinides.

Since $\Delta\alpha$ is expected to be negative and $|\lambda_c^{\mathrm{I}}\Delta\alpha| > |\alpha^{\mathrm{I}}\Delta\lambda_c|$ for most cases, the $\Delta\lambda$ values obtained from Eq. (5) should be larger than those resulting from Eq. (4). This implies that the present results set an even lower limit on ϵ than discussed above. This result is consistent with known systematics[23] of cross sections for the population of shape-isomeric states by nuclear reactions. Although radiationless muonic excitation is a mechanism considerably different from particle-induced excitation, there is no reason for expecting the former to lead to excitation of isomeric states with a probability two orders of magnitude higher than the latter process, as would be necessary in order to produce a measurable difference $\Delta\lambda$.

It should be emphasized that the experiments performed up to now on muon-capture rates in the actinides have led to results which cannot be unambiguously interpreted. A variety of reactions may be induced by radiationless muonic cascade transitions, such as prompt neutron emission or fission, and the muon may then decay or be captured in a reaction product. Consequently, the observed time distribution of decay electrons or capture products would be a superposition of different components, which will be weighted in different ways, depending on the particular reaction channels studied in the experiment. Recently, Hadermann[24] arrived at a similar conclusion. He included the process of prompt fission followed by muon capture on one of the fission fragments and predicted a nonexponential behavior of the time distributions of decay electrons or fission fragments. As can be seen in Fig. 2, the present results indicate a pure exponential decay of the electron time distribution and do not confirm this effect. However, the situation is certainly more complex than considered by Hadermann, e.g., prompt-neutron emission preceding muon capture as well as the finite width of the prompt-fission mass distribution should be accounted for. On the other hand, more refined experiments, e.g., measurements where decay or capture products are studied in coincidence with high-energy muonic x rays or prompt neutrons are needed to study these effects in detail.

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