## MUON- AND PION-INDUCED FISSION OF URANIUM ISOTOPES\*

B. Budick<sup>†</sup>, S. C. Cheng, E. R. Macagno, A. M. Rushton, and C. S. Wu Department of Physics, Columbia University, New York, New York 10027 (Received 21 January 1970)

We have measured the energy distributions of fission fragments following formation of muonic and pionic  $^{233}$ U and  $^{235}$ U. We have confirmed earlier results on the mean lives for muon capture by  $^{235}$ U and  $^{238}$ U and extended the study to  $^{233}$ U. Additional data have also been obtained on the process whereby a muon induces fission by transferring its excitation energy to the nucleus in a radiationless transition to the 1s level.

The capture of a pion near the nuclear surface results in a deposition of approximately 80 MeV of excitation energy in the nucleus. Emulsion studies,<sup>1</sup> on the absorption of slow pions by <sup>238</sup>U nuclei, have shown that the subsequent fission process is predominantly a symmetric division of the nucleus. Muons can also induce fission by being captured from a 1s orbit. In this case, however, most of the  $\mu$  rest mass is carried off by a neutrino and only about 15 MeV is left in the form of nuclear excitation energy, on the average.

In 1958 Zaretsky<sup>2</sup> pointed out that fission may be an alternative mode of de-excitation of a muonic atom. For heavy nuclei (Z > 90) the energy of the 2p-1s muonic transition is roughly equal to the height of the fission barrier and to neutron binding energies in these nuclei. The muon may therefore transfer its excitation energy in a radiationless process producing an excited nucleus with a muon in a 1s orbital. Both fission and neutron emission are open channels to the excited nucleus. Following fission the muon may remain with one of the fission fragments and may either be internally converted or captured by the fission fragment nucleus. Of course muon decay is also possible. The emission of prompt neutrons without fission has recently been observed.<sup>3</sup> Zaretsky estimated that the ratio of fission induced by  $\mu$  capture to that induced by a radiationless muonic transition is about 4:1.

Recently we have investigated, with the aid of silicon surface-barrier detectors, the nuclear fission induced in <sup>233</sup>U, <sup>235</sup>U, and <sup>238</sup>U by pions and muons. Both the distribution of the kinetic energies and the time spectra of the fission fragments were separately recorded on multichannel analyzers and subsequently analyzed. (1) From the time spectra of the fission events in muonic atoms, a ratio of capture to radiationless fission of roughly 20:1 was observed which is in disagreement with the theoretical prediction of 4:1 by Zaretsky, but is in good accord with an earlier work of a Berkeley group where a gas scintillation chamber was used.<sup>4</sup> (2) The isotope capture effects in <sup>233</sup>U, <sup>235</sup>U, and <sup>238</sup>U were studied by measuring the mean lives (in nanoseconds) of the delayed fissions, and their relative ratios 61.7:65.3:74.1 are in agreement with those predicted by the well-known Primakoff formula.<sup>5</sup> (3) The analysis of the energy distribution of fission fragments from <sup>235</sup>U induced by muon capture revealed that the fission is mostly of an asymmetrical character. If there is any occurrence of symmetric fission in this process, its admixture must be very small. (4) The spectrum of fission-fragment energies following the absorption of slow pions by <sup>235</sup>U nuclei can be interpreted as a mixture of symmetric and asymmetric fission. A general discussion concerning the experimental results and the theoretical predictions will be presented.

Our apparatus consisted of two banks of 400mm<sup>2</sup> silicon surface-barrier detectors (Ortec 7904 series) between which thin targets of fissionable material could be placed. The target and detectors were placed in a brass cylinder 8 in. in diam and 8 in. long that was closed at both ends by thin plates of aluminum and evacuated. A conventional beam telescope<sup>6</sup> signaled the stopping of a pion or a muon in the chamber. The pion and muon beams were separated by inserting different thicknesses of aluminum absorber.

The pulses from the 24 detectors in both banks were fed to units that contained both charge-sensitive and voltage-sensitive preamplifiers. Fast pulses from the voltage-sensitive preamplifiers connected to one bank of detectors were added in parallel, and could be put in coincidence with a pulse from any of the detectors in the other bank. A triple coincidence could then be established with a pulse from the beam telescope. The output of the final coincidence drove a gate generator that enabled one or more multichannel analyzers. For work in which coincidence between the two fragments was not desired, the fast outputs of both banks were first connected in parallel. A second fast output from the detectors was fed to the start input of a time-to-amplitude converter (TAC). The stop signal from the TAC is a delayed (300 nsec) pulse from the beam telescope. The output of the TAC, corresponding to the time distribution of the arrival of fission fragments at the detectors, was stored in one of the 400-channel analyzers whose gating is described in the previous paragraph.

The slow pulses from the charge-sensitive preamplifiers that contain the energy information were added together and stored in another gated 400-channel analyzer. Provision was also made in the logic design for observing the energy distribution of delayed fission events on a third analyzer.

All targets were in the form of a V with a right angle between the arms. Each arm had an area of 4 in.  $\times$ 4 in. Four distinctly different types of targets were used in the experiment: (1) For the coincidence work on pion-induced fission of <sup>235</sup>U a thin target was prepared by an electrospraying technique. A relatively uniform layer, approximately 500  $\mu$ g/cm<sup>2</sup> thick, of uranyl nitrate was deposited on a nickel foil whose thickness was 100  $\mu$ g/cm<sup>2</sup>. The uranium in the nitrate was 93%  $^{235}$ U. (2) The work on the energy distribution of fragments from <sup>235</sup>U and <sup>233</sup>U required thin targets. Thick aluminum foils with <sup>235</sup>U electroplated on both sides to a thickness of 500  $\mu g/cm^2$ were used. The isotope purity of  $^{235}$ U was 99.76%. The <sup>233</sup>U target had 117  $\mu$ g/cm<sup>2</sup> electroplated on one side. Impurities in this target did not exceed 42 parts per million. (3) To obtain good statistics in the time distribution of fragments following  $\mu$  capture on <sup>235</sup>U, a target was prepared by rolling a piece of uranium to a thickness of 5 mg/cm<sup>2</sup>. The targets were fabricated from material that was 93%<sup>235</sup>U. (4) The <sup>238</sup>U target was a thick piece of natural uranium, which is 99.3% <sup>238</sup>U.

Prior to a run the detectors with their individual charge-sensitive preamplifiers were first calibrated on the  $\alpha$  particles from radioactive thorium or polonium sources. Then the resolution of the entire apparatus and associated electronics was determined by observing the neutroninduced fission spectrum from the target to be run. In addition, the energy scale of the multichannel analyzer was calibrated with the aid of a <sup>252</sup>Cf spontaneous-fission source before and after each run.

The principal difficulty encountered in the experiment, which in fact made a coincidence experiment compulsory in the case of pions, was star formation in the silicon detectors. This presented a problem for the muon work as well, since prompt fissions from scattered pions are indistinguishable from radiationless muon-induced fission. By observing the number of stars formed in our detectors as a function of aluminum absorber thickness, we were able to conclude that at a thickness corresponding to the maximum number of muons stopping in the fission chamber, the pion contamination was less than 0.1%.

The slow-neutron background in the experimental area was studied by displacing the apparatus out of the beam and observing the total number of fission events without requiring coincidence with a stop signal from the muon telescope. The fission rate was low enough to be completely negligible when the coincidence requirement was imposed. The slow-neutron content in the beam at the target position was investigated by increasing the amount of absorber slightly. A rapid disappearance of fission was noted, indicating the muon origin of the fission. Finally, the possibility that neutrons from  $\mu$  capture in the target vicinity were initiating fission was eliminated by doubling the aluminum backing on the target itself while observing no increase in fission rate.

In Fig. 1(a) we show data obtained in the coincidence experiment on  $\pi$ -induced fission. The data were analyzed in terms of a mixture of symmetric and asymmetric fission. Our statistics were insufficient to deduce accurately the strengths of the two fission modes. This is a worthy project for meson factories that will be available in the near future. The peak whose origin lies in a symmetric fission process has a width of roughly 20 MeV, and is centered at an energy of 57 MeV. The most probable energies of the heavy and light fragments from neutron-induced fission in our target are 60 and 84 MeV, respectively. These energies represent a degradation of the fission fragment energy in passing through the target material and/or the nickel backing. However, it is clear that the most probable average energy of the two fragments following the capture of slow pions is considerably lower than the average fragment energy for fission induced by slow neutrons. A similar displacement of the peak corresponding to symmetric fission of <sup>238</sup>U by protons of 156-MeV incident energy (which also deposit roughly 80 MeV of excitation energy in the nucleus) has also been observed.<sup>7</sup>



FIG. 1. (a) The energy distribution of fission fragments, observed in coincidence, following pion-induced fission of  $^{235}$ U. (b) The spectrum of fission-fragment energies measured (without a coincidence requirement) subsequent to the formation of muonic  $^{235}$ U. Note change of horizontal scale.

On the other hand, our results indicate that muon-induced fission is an asymmetric division of the nucleus. Although 15-20 MeV are typically available for nuclear excitation in the case of  $\mu$ capture, no definite indication of an increased preference for symmetric fission was observed, compared with fission induced by slow neutrons.

In Fig. 1(b) we show the muon-induced fission spectrum obtained with our  $500 - \mu g/cm^{2}$  <sup>235</sup>U target. For this target the most probable energies of the heavy and light fragments from nuetron-induced fission are 58 and 85 MeV, respectively. The analysis revealed that the data could be fitted quite well with two peaks centered at energies of 55 and 82 MeV. Thus the neutron-induced and muon-induced fission processes are quite simi-

lar. This was true for  $^{233}$ U as well. Only the time distribution of the fission events can definitely tell them apart.

Table I summarizes the information we obtained on the time distribution of fission events after muons stopped in targets of all three isotopes. Between 1200 and 4000 events were observed for each of the isotopes over a time interval of 250 nsec. The inherent time resolution of the surface-barrier detectors and of the electronics is of the order of 3 nsec. The actual resolution, however, is degraded to roughly 10 nsec due to the flight time of the fission fragments from target to detector. The decrease in mean life with decreasing mass number is easily comprehended in terms of the Primakoff theory.<sup>2</sup> In fact, if the <sup>235</sup>U result is taken as a normalization value, mean lives of 71.5 and 60.9 nsec are predicted for <sup>238</sup>U and <sup>233</sup>U, respectively.

Fission events that originate from the radiationless transfer of energy from muonic atom to nucleus occur approximately  $10^{-13}$  sec after the muon enters a high atomic orbit. These events manifest themselves as an excess of events in the first few bins of the time distribution, over and above the number expected from muon capture in the earliest time intervals. The small percentage of radiationless fission events observed in our experiment confirms the results of Diaz, Kaplan, and Pyle.<sup>4</sup> An explanation for the rarity of prompt events has been given by Zaretsky and Novikov.<sup>8</sup> The presence of the 1s muon inhibits the fission process by raising the barrier by approximately 0.1 MeV. This is sufficient to decrease the fission probability by an order of magnitude.

Our attention has been called to a paper by Cojocaru <u>et al.</u><sup>9</sup> in which the ratio of radiationless to capture fission in <sup>239</sup>Pu was measured and found to be  $0.43 \pm 0.09$ . The authors suggest that the enhanced radiationless fission probability is connected with the low fission barrier of <sup>239</sup>Pu.

Table I. Past and present results obtained from an analysis of the time distribution of muon-induced fission events in targets of three uranium isotopes. Both the mean lives against  $\mu$  capture and the ratio of the number of fission events induced by a radiationless muonic transition to the number induced by  $\mu$  capture are given.

	Mean life (nsec)		Radiationless events µ-capture events	
Isotope	This experiment	Earlier work <sup>a</sup>	This experiment	Earlier work <sup>a</sup>
${}^{238}{}^{ m U}{}^{235}{}^{ m U}{}^{233}{}^{ m U}{}^{ m U}$	$74.1 \pm 2.8 \\ 65.3 \pm 2.8 \\ 61.7 \pm 3.8$	$75.6 \pm 2.9 \\ 66.5 \pm 4.2$	$\begin{array}{c} 0.048 \pm 0.025 \\ 0.063 \pm 0.025 \\ 0.046 \pm 0.030 \end{array}$	$\begin{array}{c} 0.072 \pm 0.022 \\ 0.111 \pm 0.021 \end{array}$

<sup>a</sup>See Ref. 4.

This interpretation is not supported by our results.

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<sup>†</sup>Permanent address: Department of Physics, New York University, The Bronx, N. Y. 10453.

<sup>1</sup>H. A. Periflov, O. V. Lozhkin, and V. P. Shamov, Zh. Eksperim. i Teor. Fiz. <u>28</u>, 655 (1955) [Soviet Phys. JETP <u>1</u>, 439 (1955)].

<sup>2</sup>D. F. Zaretsky, in <u>Proceedings of the Second United</u> <u>Nations International Conference on the Peaceful Uses</u> <u>of Atomic Energy, Geneva, 1958</u> (United Nations, Geneva, Switzerland, 1958).

<sup>3</sup>C. K. Hargrove <u>et al</u>., Phys. Rev. Letters <u>23</u>, 215 (1969).

<sup>4</sup>J. A. Diaz, S. N. Kaplan, and R. V. Pyle, Nucl. Phys. <u>40</u>, 54 (1963).

<sup>5</sup>H. Primakoff, Rev. Mod. Phys. <u>31</u>, 802 (1959).

<sup>6</sup>See, for example, T. T. Bardin <u>et al</u>., Phys. Rev. 160, 1043 (1967).

<sup>7</sup>L. Kowalski, thesis, University of Paris, 1963 (unpublished).

<sup>8</sup>D. F. Zaretsky and V. M. Novikov, Nucl. Phys. <u>28</u>, 177 (1961).

<sup>9</sup>V. Cojocaru <u>et al</u>., Phys. Letters <u>20</u>, 53 (1966).

## ISOSPIN POLARIZATION IN THE NUCLEAR MANY-BODY PROBLEM

C. A. Engelbrecht

Atomic Energy Board, Pelindaba, Transvaal, South Africa

## and

## R. H. Lemmer\*†

Institut für Theoretische Physik der Universität Frankfurt am Main, Frankfurt am Main, Germany (Received 19 January 1970)

For nuclei with N > Z, isospin invariance is explicitly broken in the Hartree-Fock (HF) approximation. It is shown that isospin polarization correlations in the HF ground state restore this symmetry and lead to a natural framework for a microscopic description of analog states. We discuss the approximations under which the Lane potential model is obtained.

In this note we discuss the question of isospin invariance in connection with the shell-model description of analog states in heavy nuclei. If all electromagnetic effects (including the neutron-proton mass difference) are suppressed, the nuclear part of the many-body Hamiltonian H commutes with all components of the isospin operator. In particular,  $[T_{-},H]=0$ . If  $|\varphi_0\rangle$  is any eigenstate of H (for example, the ground state corresponding to a particular neutron number N and proton number Z), this immediately implies that the analog state  $T_{-}|\varphi_0\rangle$  is also an eigenstate with the same energy eigenvalue. This property, and with it the whole framework of isospin invariance, may be lost in any approximate treatment of the eigenstates of H. The Hartree-Fock (HF) approximation for a system with N > Z is a case in point. The HF ground state  $|0\rangle$  does not have good isospin since (even without electromagnetic interactions) equivalent neutron and proton states have different wave functions on account of the presence of the symmetry-potential terms associated with the excess neutrons. The breaking of isospin invariance has to do with the approximations introduced, and not with the forces.

We show below that this trouble develops in HF due to the neglect of <u>neutron-protron correlations</u> in  $|0\rangle$ , and that an isospin-conserving description of at least the ground state is recovered by including such correlations within the framework of the random phase approximation (RPA).<sup>1</sup>

Consider a system of N neutrons and Z protons with a general two-body interaction which we write in the form  $V = V^0 + V^T P^T$  after separating off the charge exchange part ( $P^T$  is the usual charge-exchange operator). The HF states  $a, b, \cdots$  for neutrons and  $\alpha, \beta, \cdots$  for protons may be used as basis for the definition of creation and destruction operators (e.g.,  $n_a^{\dagger}$  creates a neutron in state a, etc.).