Relative Excitations of the ^{237}Pu Shape Isomers*

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(Received 14 May 1973)

The excitation energy of the 1120-nsec spontaneous-fission isomer of ^{237}Pu has been shown to be 0.30 ± 0.15 MeV above that of the 82-nsec isomer. The half-life of the longer-lived isomer may therefore be determined by its radiation width rather than by its direct fission width.

I. INTRODUCTION

^{237}Pu has been shown[1-7] to exhibit two isomeric states, both of which decay, directly or indirectly, by spontaneous fission. These two isomeric states are attributed to different single-particle states at the deformation of the second or shape-isomeric well. A study of the dependence of the relative isomer population on the angular momentum deposition in the reactions producing them has shown[5] that the shorter-lived isomeric state has the lower spin. The purpose of the present experiment is to determine which isomer has the lower excitation energy. This is of particular interest since the decay mode of the longer-lived species is ambiguous if this isomer lies higher in energy than the short-lived species. The observed lifetime in such a situation may be determined by γ-decay of the long-lived state to the short-lived state rather than by direct fission from the long-lived excited state. Although the latter case would preclude interpretation of the lifetime of the long-lived isomer as a fission lifetime, it would open the possibility for observing deexcitation through the rotational band built on the lower state.

II. EXPERIMENTAL METHOD

The half-lives of the two shape isomers are approximately 80 and 1100 nsec.[5] The only suitable reactions for ^{237}Pu^m threshold determinations are the ^{235}U$(\alpha, 2n)$ and ^{237}Np$(d, 2n)$ reactions. The former was chosen since in this reaction the Coulomb barrier for the incident projectile is lower relative to the expected threshold for this reaction. The delayed-fission yields associated with the shape-isomer decay are only about 5×10^{-6} of the prompt-fission yield at the peak of the excitation function. In order to measure the thresholds one must measure relative yields which are at least an order of magnitude smaller. Thus it is critical to select a detection scheme with high efficiency. The method[1] of observing the fragments from fission of recoils in flight has been used. The fragments are observed in plastic-foil track detectors. The collimated recoiling compound nuclei from a thin target travel down an evacuated path surrounded for the first 12 inches in near-2π axial geometry by plastic foils. The effective cone half-angle of the collimator-detector system is approximately 12°. The path length along which detection plates may be usefully placed is limited by this cone angle, because the area of detector material along a unit length of path will increase until the background due to imperfections in the foils becomes excessive. To increase the efficiency of the detector system a 10-μg/cm^2 carbon foil was placed just downstream from the target. This slows the recoils to permit a larger fraction of the longer-lived isomer to decay within the sensitive region of the detection system. Prompt-fission fragments, screened from the plastic foils by collimation, are detected by a solid-state counter sitting at 50° above the beam axis.

The ^{235}U target (isotopic enrichment of 93%) was 48 μg/cm^2 thick and was prepared by vacuum volatization onto a nickel and copper backing. The helium-ion beam from the University of Washington two-stage tandem Van de Graaff passed through the backing losing 0.5 MeV before reaching the ^{235}U target. The target and detector plates were located in a large 60-in.-diam scattering chamber.

The plastic foils were 1.1-mg/cm^2-thick Kimfoil.[8] They were chemically etched using a variant,[9] devised by Henderson, of the usual procedure. The foils were floated onto 6 N NaOH in such a way that the top surface remained dry. They were etched in this position for 60 min at 60°C. They were then washed on both sides and dried. This developing procedure resulted in much lower and more reproducible backgrounds (0.023 tracks/cm^2) than were achieved in earlier attempts in this laboratory. The tracks were identified by the holes produced on an aluminized Mylar electrode[10] when sparked at 1000 V.

The distribution of tracks along the recoil path was converted into a time-decay curve using the calculated velocity of the recoils after passing through the carbon foil. The decay curves were resolved by a least-squares analysis into 82- and

1120-nsec half-life components.

III. RESULTS

The ratios of delayed-to-prompt fission yields for each of the two components are given in Table I. Only relative values of the ratios are given as the absolute delayed-fission detection efficiency, common to both components, is not well known.

The excitation functions of the delayed-to-prompt ratios are plotted in Fig. 1. They have been fitted with a modified Jackson neutron-evaporation model[11-13] to extract the thresholds. As employed here the model has three parameters in addition to an arbitrary normalization constant. These parameters are the nuclear temperature T, the isomer excitation energy E_i, and the height of the outer barrier E_B. The latter is important if the height of the outer barrier is lower than the neutron binding energy of the final nucleus. A search routine was used to vary E_i and E_B for selected values of the nuclear temperature. The best fits were obtained with temperatures between 0.4 and 0.6 MeV. The isomer excitation energy is most sensitive to the leading edge of the excitation function, and decreases slightly as the temperature is increased. The outer barrier height is determined by the falloff on the high-energy side of the excitation function. The final values obtained at $T = 0.5$ are $E_i(1120\text{ nsec}) = 3.45 \pm 0.10$, $E_i(82\text{ nsec}) = 3.10 \pm 0.15$, and $E_B = 4.8 \pm 0.4$. The uncertainties encompass values obtained with $T = 0.4$ and 0.6 and also values for which the χ^2 value for the fit is a factor of 2 worse. If a common nuclear temperature is assumed for both fits the differenence in isomer excitation energies E_i is 0.35 ± 0.12 MeV. In the above analysis we have neglected the perturbation of the relative population of the low- and high-spin isomers due to the increasing angular momentum in the compound nucleus as the bombarding energy increases. A statistical-model calculation similar to that described in our previous work[5] has been performed and a correction for spin fractionation introduced. This reduces the difference in isomer excitation energies to 0.30 ± 0.15 MeV.

The above E_i values can be compared with a value of 3.4 MeV obtained in a similar analysis by Burnett *et al.*[4] in an experiment which did not distinguish between the short- and long-lived isomers. This measurement has been extended by Britt *et al.*[14] and the results have been analyzed using a more sophisticated model. This type of analysis results in a lower isomer excitation energy, giving a value of 2.9 ± 0.2 MeV for the unresolved ^{237}Pu^m isomer data. A still later analysis gives an even lower value.[15] The assumptions in the newer analyses should not appreciably affect the difference in the isomer excitation energy for

TABLE I. Ratios of isomeric-to-prompt fissions. The ratios for the two isomers are given in arbitrary units and have to be multiplied by a factor $\sim 10^{-7}$ to give peak ratios consistent with measured absolute values (see Refs. 5 and 14). The ratios for the 82-nsec isomer relative to those for the 1120-nsec isomer, however, are absolute. The errors are based on the standard deviations of the least-squares fit and do not include possible systematic errors.

E_α(lab) (MeV)	Yield ratio 82-nsec isomer	Yield ratio 1120-nsec isomer
22.0	3.6 ± 0.6	3.9 ± 1.4
22.1	9.7 ± 1.5	4.0 ± 2
22.2	15 ± 3	3.2 ± 3.2
22.3	10 ± 3	8.6 ± 4
22.5	20 ± 4	36 ± 8
22.7	14 ± 4	21 ± 6
23.0	44 ± 8	121 ± 18
23.5	73 ± 9	142 ± 53
24.0	60 ± 9	154 ± 21
25.0	41 ± 4	71 ± 8
25.5	37 ± 3	49 ± 5

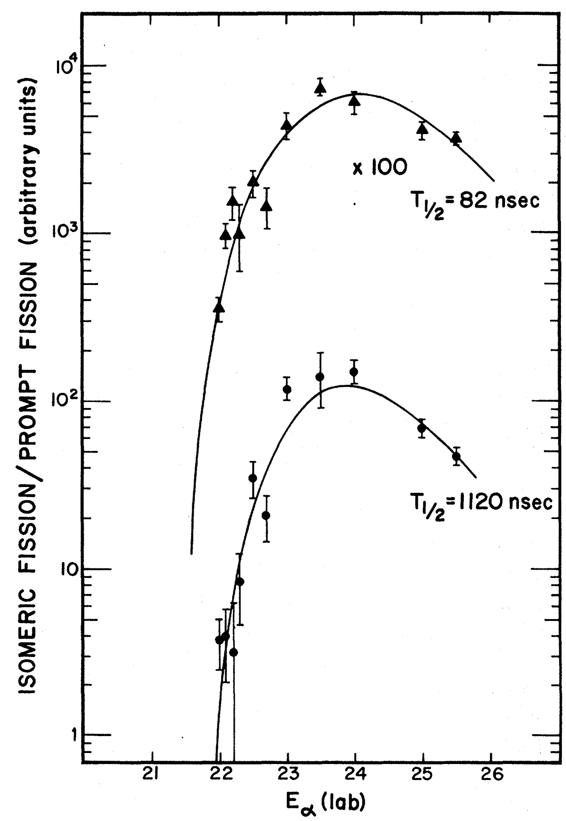

FIG. 1. Excitation functions for the delayed-to-prompt-fission yield ratios.

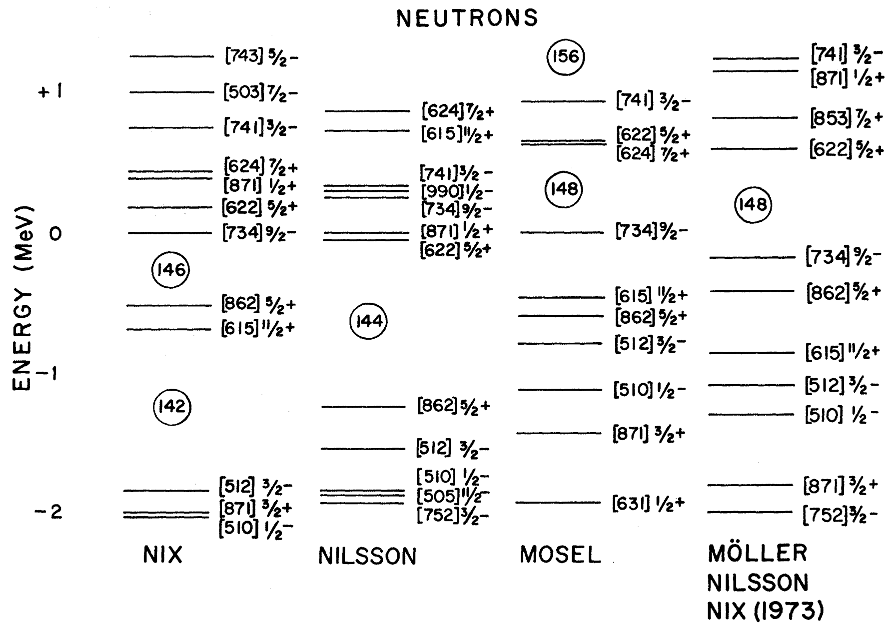

FIG. 2. Single-particle energy levels for neutrons at a deformation corresponding to the second minimum in the potential energy surface. Adapted from a figure constructed by J. Pedersen from results of calculations described in Refs. 16–19.

the two isomers. We have reanalyzed the extended data set of Britt *et al.* using the model employed here and obtained a value of $E_i = 2.8$ MeV, in fair agreement with the present results.

IV. DISCUSSION

The possible spin assignments for the ^{237}Pu isomers consistent with the isomer-ratio studies were discussed in a previous publication.[5] A likely spin for the high-spin long-lived isomer was $\frac{11}{2}$, and it was suggested on the basis of the Nilsson diagram[16] then available that this could be the $[505]\frac{11}{2}^-$ orbital. Since that publication additional single-particle diagrams have become available.[17–19] The results of three single-particle calculations at the approximate deformation expected for the second minimum are shown in Fig. 2. Pedersen[20] has noted that the newer calculations suggest that an alternative assignment for the high-spin state is the $\frac{11}{2}^+[615]$ orbital. This orbital is at the Fermi surface ($N = 143$) in the calculation of Bolsterli, Fiset, Nix, and Norton,[17] and just above the Fermi surface in the calculation of Mosel and Schmitt.[18] One should not expect the calculations to give the exact ordering, but it appears that an $\frac{11}{2}$ spin state is to be expected very close if not at the Fermi surface.

It is interesting to note that even if the 143rd neutron were expected on the basis of the single-particle level diagram to occupy the $\frac{11}{2}^+$ single-particle state, the influence of the pairing correlation is likely to make it appear as an excited shape-isomeric state rather than the lowest shape-isomeric state. This is a consequence of the stronger pairing interactions for a pair of nucleons in a high-spin orbital than in a low-spin orbital. Thus the promotion energy for moving a nucleon from a lower filled orbital into the otherwise half-filled high-spin orbital may be more than compensated for by the gain in pairing energy. This effect occurs for nuclei between the $Z = 50$ and $Z = 82$ shells, where ground states with $\frac{11}{2}$ and $\frac{13}{2}$ spins are never observed even though such orbitals become occupied before the $Z = 82$, $N = 126$ shell closure. Thus our observation that the higher-spin state lies higher in energy is consistent with expectations even if the high-spin orbital were to correspond to the lowest single-particle orbital for the 143rd neutron.

Based on the isomer-ratio results and the theoretical single-particle level ordering, the most likely spin assignments are $I = \frac{5}{2}$ and $I = \frac{11}{2}$. Although these spin assignments are far from being established, one might question whether the lifetime of the long-lived, high-spin isomer is consistent with the lifetime expected for a γ transition between the $\Omega = \frac{11}{2}$ state and one of the rotational states built on the $\Omega = \frac{5}{2}$ single-particle state. This transition would correspond to a change of 3 units in K, the projection of I on the nuclear symmetry axis. An examination of the limited decay data available for single-particle energy levels at the equilibrium deformation indicates that a lifetime of 1100 nsec would not be inconsistent with that expected for such a γ decay. Whether the 1120-nsec isomer also decays by spontaneous fission directly is not obvious from the experimental data. There is, however, reason to believe that the fission barrier would be large for an $I=\frac{11}{2}$ level because of specialization energy effects. This would favor γ decay to the 82-nsec isomer followed by fission from that level. It may be possible to determine the branching ratio between spontaneous fission and γ decay, however, by comparing the yield of delayed fission with the absolute yield of conversion electrons associated with transitions between the high-spin isomer and the rotational band built on the low-spin isomer.

ACKNOWLEDGMENTS

We wish to thank Robert Wilson and Rick Methot for their assistance in target preparation, foil etching and sparking, and data reduction. We thank Jørgen Pedersen for permission to use Fig. 2, and J. Rayford Nix and Ulrich Mosel for supplying the level energies used in constructing this figure. We also thank W. Jacobs for providing the least-squares program used in resolving the decay curves.

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†Work supported in part by the U.S. Atomic Energy Commission.
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