the Seventeenth Conference on Magnetism and Magnetic Materials, Chicago, Illinois, November, 1971 (American Institute of Physics, New York, to be published); R. J. Birgeneau, R. Dingle, M. T. Hutchings, G. Shirane, and S. L. Holt, Phys. Rev. Lett. 26, 718 (1971); R. E. Dietz, F. R. Merritt, R. Dingle, D. Hone, B. G. Silbernagel, and P. Richards, Phys. Rev. Lett. 26, 1186 (1971).

[2]M. T. Hutchings, G. Shirane, R. Birgeneau, R. Dingle, and S. Holt, J. Appl. Phys 42, 1265 (1971), and Phys. Rev. B (to be published).

[3]M. E. Fisher, Amer. J. Phys. 32, 343 (1964).

[4]F. B. McClean and M. Blume, J. Appl. Phys. 42, 1380 (1971).

[5]P. Resibois and M. DeLeener, Phys. Rev. 152, 305 (1966); K. Kawasaki, Progr. Theor. Phys. 39, 285 (1968); M. Blume and J. Hubbard, Phys. Rev. B 1, 3815 (1970); J. Hubbard, J. Phys. C: Proc. Phys. Soc., London 4, 53 (1971), and J. Appl. Phys. 42, 1390 (1971).

[6]F. Carboni and P. Richards, Phys. Rev. 177, 889 (1969); P. Richards and F. Carboni, Phys. Rev. B (to be published).

[7]M. Blume, R. E. Watson, and G. H. Vineyard, Bull. Amer. Phys. Soc. 16, 629 (1971).

[8]P. M. Richards, Phys. Rev. Lett. 27, 1800 (1971).

[9]H. Mori and K. Kawasaki, Progr. Theor. Phys. 27, 529 (1962).

[10]H. Mori, Progr. Theor. Phys. 33, 423 (1965), and 34, 399 (1965).

[11]M. F. Collins and W. Marshall, Proc. Phys. Soc., London 92, 380 (1967).

[12]$\delta_3 = [\overline{\langle\omega^6\rangle}/\delta_1 - (\delta_1+\delta_2)^2]\,\delta_2^{-1}$ and $\langle\omega^6\rangle$ was evaluated from the result given by D. G. McFadden and R. A. Tahir-Kheli, Phys. Rev. B 1, 3671 (1970).

[13]S. W. Lovesey, J. Phys. C: Proc. Phys. Soc., London 4, 3057 (1971).

[14]C. G. Windsor, *Inelastic Scattering of Neutrons* (International Atomic Energy Agency, Vienna, 1968), Vol. 2, p. 83.

[15]A. Tucciarone, L. M. Corliss, and J. M. Hastings, J. Appl. Phys. 42, 1378 (1971).

Investigation of γ-Ray Emission Preceding Isomeric Fission of ^{236}U†

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Measurements were made to detect γ rays preceding isomeric fission in ^{236}U induced by eV-range neutrons captured in ^{235}U. A limit of $\leq 6\times10^{-5}$ was placed on the ratio of the rate of isomeric fission events with prefission γ rays to the rate for prompt fission events. This experiment provides direct evidence that the penetration of the outer barrier is much greater than that for the inner barrier for 3^- and 4^- states in ^{236}U.

About thirty fission isomers have been identified in transuranium nuclei to date. These isomers can be considered as shape isomers associated by Strutinsky[1] with a double-humped fission barrier. This model has provided a satisfactory explanation for many aspects of fission isomerism such as the isomeric half-life, excitation energy of the isomer, and the height of the fission barrier. With the hypothesis of a double-humped fission barrier, the formation of the isomeric state is assumed to proceed in many reactions by penetration of the first barrier from an excited state near the static equilibrium deformation followed by γ decay to the isomeric state in the second minimum. The detection of the prefission γ rays is a vital test of the Strutinsky model. However, these γ rays have not yet been observed. The purpose of this experiment, therefore, was to detect the γ decay to an isomeric state followed by the subsequent fission of this state.

Several experiments[2-5] have identified a fission isomer in ^{236}U with a half-life approximately 100 nsec. Elwyn and Ferguson[4] populated this fission isomer by bombarding ^{235}U with 0.5- and 2.2-MeV neutrons. In the present experiment, a 90-mg ^{235}U sample was bombarded with neutrons from the pulsed neutron source of the Livermore linac in an attempt to detect γ-ray emission prior to fission events corresponding to the 100-nsec ^{236m}U isomer. The neutron energy was determined by the time of flight of the neutrons to the ^{235}U sample located at 14.5 m from the neutron source. Measurements were made for the neutron energy range between 1 and 100 eV. Fission fragments were detected using an ionization chamber. The γ rays were detected by a pair of deuterated-benzene (C_6D_6) scintillators which subtended a fractional solid angle ($\Omega/4\pi$) of approximately 0.8. The outputs of the C_6D_6 scintillators were summed resulting in an efficiency of approximately 15% (including solid angle) with a

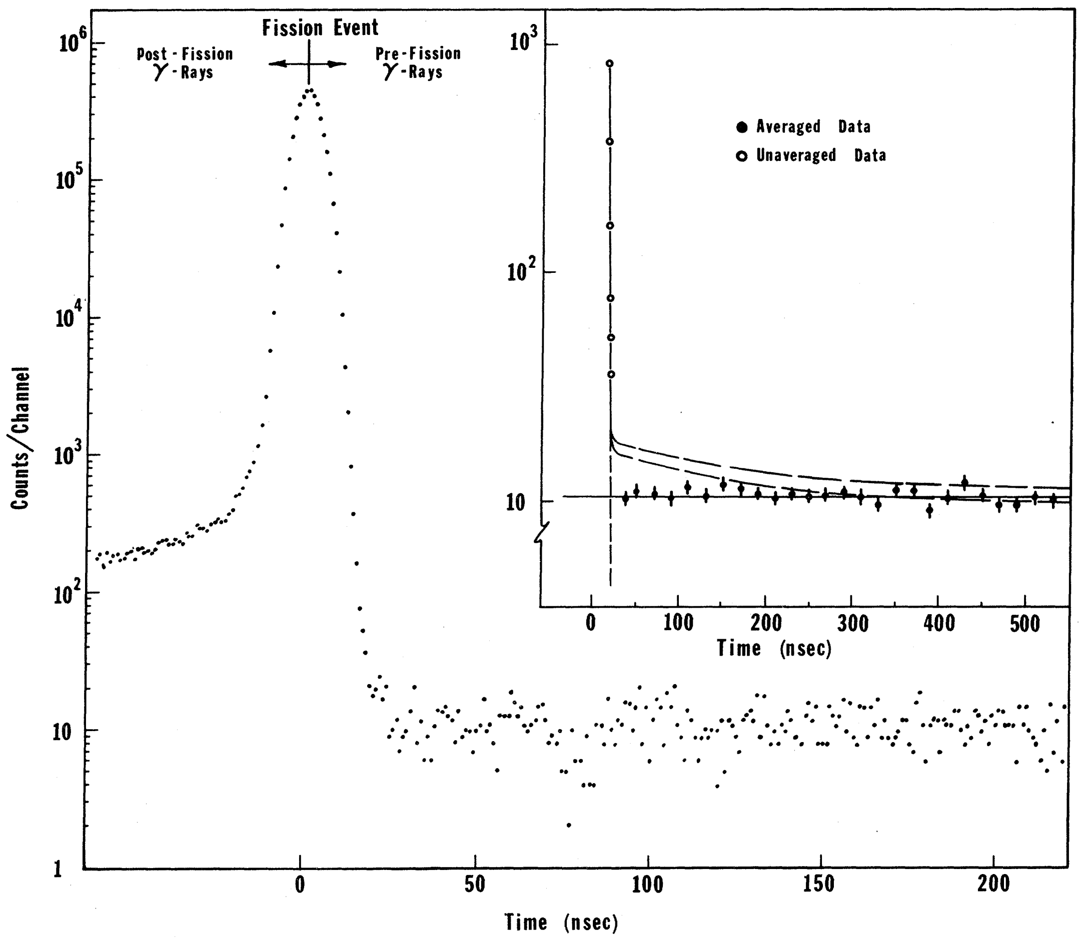

FIG. 1. Coincidence spectrum between γ rays and fission fragments for incident neutron energies between 1 and 100 eV. The inset on the upper right shows the data averaged in 20-nsec intervals. The horizontal line represents the average of the counts between $t = 25$ and 1025 nsec. The two dashed nearly parallel curves represent ± 1 standard deviation from the curve that is expected under the assumption that $\sigma_{\text{iso}}/\sigma_{\text{prompt}} = 6 \times 10^{-4}$ ($E^* = 6.5$ MeV) and $\bar{\mu}_{\text{iso}} = 2$.

flat response[6] for γ rays above the bias level of 0.5 MeV. Both the γ-ray and fission-fragment signals were processed by fast-timing logic units whose outputs were sent to a time digitizer to determine the time relationship between the signals. Precautions were taken to reduce background events which make a half-life determination more difficult. For example, the fission chamber was constructed of beryllium to reduce the effects of neutron capture in the chamber, and the γ-ray detectors contained deuterated benzene rather than ordinary benzene to reduce background from neutron capture on hydrogen.

Figure 1 presents the time relationship of the γ rays and fission fragments for incident neutron energies between 1 and 100 eV. The main peak consists of coincidences between fission fragments and prompt fission γ rays. The slope on the left of the peak corresponds to delayed γ rays from the fission fragments. Prefission γ rays would occur to the right of the peak. The full width at half-maximum of the peak is 9 nsec. Although only about 0.5 μsec of data are shown in the plot, the data extend out to approximately 2 μsec to the right of the prompt peak. To improve the statistical accuracy, the data beyond the peak were averaged in 20-nsec intervals; the inset in Fig. 1. shows the results of this averaging. A simple measure of the sensitivity of the experiment was obtained by calculating the average number of counts/channel in the time range 25 to 525 nsec (corresponding approximately to the first five half-lives) and comparing it with the average in the time range 525 to 1025 nsec. The average number of counts-channel are 10.79 ± 0.15 and 10.52 ± 0.15, respectively. This simple analysis, therefore, shows no evidence of the 100-nsec fission isomer half-life.

The efficiency for detecting a coincidence for a prompt fission event and for an isomeric fission event depends on the average number of γ rays that are emitted in each process. Several experiments[7-9] have shown that there are approximately an average of five γ rays per prompt fission ($\overline{\mu}_{\mathrm{prompt}} = 5$) whose energies are above 0.5 MeV. Since the corresponding value for isomeric fission is not known, we have assumed an average of two γ rays per isomeric fission ($\overline{\mu}_{\mathrm{iso}} = 2$) above 0.5 MeV. This is a reasonable assumption if the isomeric level is the ground state of the second minimum (excitation energy[10] $E^* \approx 2-3$ MeV) since there would be at least 3.5 MeV of energy to be emitted in reaching this level. Although the γ decay in the second minimum will probably have a somewhat lower-energy γ-ray spectrum than the usual (n,γ) process, one might still expect an average of two γ rays above 0.5 MeV because the difference in level density between the two wells will result in more widely spaced transitions in the second minimum. Then on the basis of a Poisson distribution we calculate that the efficiency per prompt fission for detecting prompt γ rays is twice that for the prefission γ rays.

The ratio of the number of isomeric fission events to the number of prompt fission events ($\sigma_{\mathrm{iso}}/\sigma_{\mathrm{prompt}}$) was determined by Elwyn and Ferguson[4] to be 3.1×10^{-4} for 2.2-MeV neutrons [$E^*(^{236}\mathrm{U}) \approx 8.7$ MeV]. Britt and Erkkila[5] have shown in a recent (d,pf) study on ^{235}U that this ratio increases by approximately a factor of 2 as the excitation energy in ^{236}U is decreased from 7.5 to 6.5 MeV (neutron binding energy). Therefore, if we assume a ratio $\sigma_{\mathrm{iso}}/\sigma_{\mathrm{prompt}} = 6 \times 10^{-4}$ for our excitation energy (≈ 6.5 MeV), and use the number of prompt coincidences (4.2×10^6), the relative γ-ray efficiency discussed above and a half-life of 100 nsec, we obtain an exponential curve which represents the expected decay of the ^{236m}U isomer. The two dashed nearly parallel curves in the inset of Fig. 1 represent the standard deviation ($N \pm \sqrt{N}$) of the expected decay curve added to the average background. From this plot it is apparent that the isomer half-life should be visible under the above assumptions.

To estimate the sensitivity of our experiment we concentrated on the region of the first half-life ($t = 25$ to 125 nsec). The average number of background counts in this interval is 1066 ± 32. We will assume that 2 standard deviations is the lower limit for the number of counts in this interval that we require to see the effect of an isomer added to the average background. In a normal distribution, 2.3% of the area lies above 2 standard deviations from the mean. From this we calculate a limit on the isomer ratio of $\sigma_{\mathrm{iso}}/\sigma_{\mathrm{prompt}} \leq 6 \times 10^{-5}$ for γ rays followed by delayed fissions.[11] This value is approximately an order of magnitude lower than the value deduced from Elwyn and Ferguson's neutron capture experiment.[4]

The experiment also can be compared with (d,pf) experiments. Britt and Erkkila[5] in a $^{235}\mathrm{U}(d,pf)$ measurement obtain an isomer ratio of $\sigma_{\mathrm{iso}}/\sigma_{\mathrm{prompt}} = 2 \times 10^{-4}$ which is an integral value for all excitation energies less than 6.5 MeV. Pedersen and Rasmussen,[12] also using the $^{235}\mathrm{U}(d,pf)$ reaction, measured the proton spectra associated with both the prompt fission of ^{236}U and the delayed fission of the 100-nsec isomer in ^{236}U. Their measurements show that the isomer-production cross section peaks for excitation energies near the fission threshold (≈ 5.3 MeV) while the prompt-fission cross section peaks near the neutron binding energy (6.5 MeV). Combining the integral measurements of Britt and Erkkila[5] with the relative data of Pedersen and Rasmussen,[12] we estimate that the isomer ratio for the 100-nsec ^{236}U fission isomer is approximately 3×10^{-5} for production in the (d,pf) reaction at our excitation energy of 6.5 MeV.

However, spin plays a significant role in the comparison of experiments. The present experiment was conducted with low-energy neutrons so that the only initial states formed were 3^- and 4^-. These spin states and others would be excited in a (d,p) reaction as a result of the larger amount of orbital angular momentum which may be transferred. If the total width for γ decay in the second minimum is essentially constant and independent of spin as it is for γ decay in the first minimum, then the spin of the initial state should not affect the width for γ decay to the isomeric state. However, if the width for prompt fission through the outer barrier is not the same for all spins, then our experiment and the (d,pf) experiment are not strictly comparable.

Our limit on the isomer ratio allows us to perform a simple calculation of the relative penetrability of the inner and outer barriers for the 3^-

and 4^- states of ^{236}U. The ratio of isomeric-to-prompt fission is proportional to $\Gamma_{\gamma 2}/\Gamma_{B2}$, where $\Gamma_{\gamma 2}$ is the total γ-decay width for the class-II state in the second minimum and Γ_{B2} is the decay width for fission of the excited state through the outer barrier. (We assume that there is no tunneling of the isomer back to the ground-state deformation.) Our results show that $\Gamma_{\gamma 2}/\Gamma_{B2} \leq 6 \times 10^{-5}$. Following Lynn's prescription,[13] we calculate a value of $\Gamma_{\gamma 2}$ equal to about 8 meV assuming the isomer excitation energy is approximately 2.5 MeV. This value of $\Gamma_{\gamma 2}$ is about 20% of the average γ-decay width of class-I states in the first minimum. With this value of $\Gamma_{\gamma 2}$ we find that $\Gamma_{B2} \geq 130$ eV. Since this value of Γ_{B2} is several orders of magnitude larger than the observed average fission width of the 3^- and 4^- class-I states (≈ 40 meV), it is clear that the fission width of the 3^- and 4^- class-I states is determined solely by the width for penetration of the inner barrier.

Since the above analysis indicates that the width of the 3^- and 4^- class-II states are of the order of 100 eV or greater, from the magnitude of Γ_{B2} above, it follows that any intermediate structure in the low-energy neutron-induced ^{235}U fission cross section will be characterized by fluctuations with an average width at least as large. For the intermediate structure to be evident in the fission cross section, the level spacings of the 3^- and 4^- class-II states, D_{II}, must be equal to or greater than the width of the class-II states (i.e., $D_{II} \geq 100$ eV). Since the level spacing of the 3^- and 4^- class-I states (D_I) is approximately 0.8 eV, the ratio D_{II}/D_I must therefore be on the order of 100 if there is to be any possibility of the existence of intermediate structure in the neutron-induced fission of ^{235}U. The factor of 100 is close to the experimental results of this ratio for the Pu isotopes,[13,14] so that intermediate structure is not definitely ruled out.

In summary, the existence of γ rays preceding isomeric fission events has not been confirmed experimentally. However, this experiment does show that the penetrability of the outer barrier is much larger than the inner barrier for the 3^- and 4^- states of ^{236}U, which yields the result that any intermediate structure in the low-energy neutron-induced ^{235}U fission cross section will be characterized by widths on the order of 100 eV or greater. However, the detection of the prefission γ rays still remains as an important test for confirming and extending the present model for isomeric fission.

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[1]V. M. Strutinsky, Nucl. Phys. A95, 1 (1967).

[2]N. L. Lark, G. Sletten, J. Pedersen, and S. Bjørnholm, Nucl. Phys. A139, 481 (1969).

[3]K. L. Wolf, R. Vandenbosch, P. A. Russo, M. K. Mehta, and C. R. Rudy, Phys. Rev. C 1, 2096 (1970).

[4]A. J. Elwyn and A. T. G. Ferguson, Nucl. Phys. A148, 337 (1970).

[5]H. C. Britt and B. H. Erkkila, Phys. Rev. C 4, 1441 (1971).

[6]J. B. Czirr, Nucl. Instrum. Method 72, 23 (1969).

[7]F. Maienschein *et al.*, Oak Ridge National Laboratory Report No. 1142 (unpublished).

[8]V. Verbinski and R. E. Sund, DASA Report No. 2234 (unpublished), and Gulf General Atomic Report No. 9148 (unpublished).

[9]F. Pleasonton and H. Schmitt, Bull. Amer. Phys. Soc. 16, 1149 (1971), and private communication.

[10]S. Bjørnholm and V. M. Strutinsky, Nucl. Phys. A136, 1 (1969).

[11]A measurement was also made for neutron energies less than 4 eV. A similar analysis yielded a limit on the isomer ratio of $\leq 8 \times 10^{-5}$.

[12]J. Pedersen and B. Rasmussen, to be published.

[13]J. E. Lynn, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1969), p. 249.

[14]G. F. Auchampaugh, J. A. Farrell, and D. W. Bergen, Nucl. Phys. A171, 31 (1971).