Ratio of Asymmetric to Symmetric Fission of Pu^{239} and Pu^{241} as a Function of Neutron Energy

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Further radiochemical experiments have been carried out to test the theoretical suggestion that in low-energy neutron-induced fission, the asymmetric to symmetric fission ratio should depend on the spin of the fissioning nucleus. In the present work two kinds of experiments were done. In the first, with both Pu^{239} and Pu^{241}, comparisons were made of the asymmetric/symmetric yield ratios for thermal fission with the corresponding ratios for gross resonance fission with epi-Sm neutrons. In the second kind, monoenergetic neutrons were employed to study variations of Pu^{239} fission yields with neutron energy over the 0.297-ev resonance. If the assumption is made that spin difference is the principal cause of change in the asymmetric/symmetric ratio in this energy region, the results indicate that this ratio differs by a factor of at least 5.3 between the two spin states of Pu^{239}. This is the largest effect of this kind observed to date.

INTRODUCTION

In previous work,[1] an experimental study was made of the theoretical suggestion[2,3] that the ratio of asymmetric to symmetric fission should be different for the two possible spin states of the compound system formed upon the addition of a neutron to the nucleus. In that work, radiochemical determinations of the ratio of asymmetric to symmetric fission yields were carried out for fission of U^{233} both with thermal neutrons and with monoenergetic neutrons of energies corresponding to the first several prominent fission resonances. This approach was taken because a multilevel analysis[4] of the U^{233} cross section had suggested that a large fraction of the cross section at thermal is associated with one spin state in the compound nucleus, while the first two prominent resonances are associated with the other possible spin state. Comparison of the asymmetric/symmetric ratios for thermal and for resonance fission thus was expected to provide a suitable test of the existence of possible differences between spin states. This work has now been extended to Pu^{239} and to Pu^{241}. In each case thermal and resonance fission have been compared.

EXPERIMENTAL PROCEDURES

In all thermal neutron measurements, sample irradiations were made in the VG-23 facility in the graphite zone of the Materials Testing Reactor where the experimental Cd ratio for In is ~ 800 and where the thermal neutron flux is $\sim 2\times 10^{10}$ n/cm^2 sec. For resonance fission measurements, two kinds of experiments were done. In the first kind, the Pu samples were enclosed in small hollow Sm metal cylinders for irradiation in the VG-7 facility of the reactor where the thermal flux is $\sim 1.0 \times 10^{13}$ n/cm^2 sec and where the experimental Cd ratio for a 70-μg/cm^2 Au detector is ~ 3. The Sm metal cylinders were 0.165 in. o.d., $\sim$1.125-in. long, and had wall thicknesses of 0.020 in. These filters served to accentuate resonance fission while minimizing thermal fission. In these experiments it was calculated that $<1\%$ of the fissions were attributable to thermal neutrons. Sm coverings were chosen instead of the usual Cd since the effective "Sm cutoff" is $\sim$0.1-ev lower than the "Cd cutoff", which lies close to the middle of the first large fission resonance in both Pu^{239} and Pu^{241}. By the use of Sm filters more neutrons were available for fission occurring in the first resonance. Actually, because it was not possible to fabricate from Sm metal neutron-tight covers for the irradiation cylinders, a small Cd cover had to be substituted at the one open end of each cylinder. As a result, a small fraction of the neutrons reaching the samples during these irradiations were really epi-Cd neutrons, although the large majority (calculated as $>98\%$) were epi-Sm neutrons. In a few early experiments with epi-Sm neutrons, the use of pressed Sm oxide coverings was attempted. The results were quite erratic presumably due to "windows" in the oxide filters arising from nonuniform packing. With the use of Sm metal this difficulty was corrected.

In the second and more refined kind of resonance fission experiments, monoenergetic neutrons were employed. These were provided by the MTR crystal spectrometer[5] fitted with a beryllium crystal. The fissionable samples were irradiated in the Bragg beam resulting from diffraction from the $10\bar{1}1$ planes. In the regions in which experiments were run, the energy resolution was $\sim 7.5\%$ with the collimation employed. The monoenergetic neutron intensities were quite low. Samples of Pu^{239} which were about one-third "black" at the peak of the 0.297-ev resonance and which intercepted the entire Bragg beam had fission rates of $\sim 1\times 10^5$ fissions/sec. Background neutrons accounted for 2.5% of the

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1 R. B. Regier, W. H. Burgus, and R. L. Tromp, Phys. Rev. **113**, 1589 (1959).
2 J. A. Wheeler, Physica **22**, 1103 (1956).
3 J. A. Wheeler, *Proceedings of the Conference on Neutron Physics by Time-of-Flight*, Gatlinburg, Tennessee, November, 1956 [Oak Ridge National Laboratory Report ORNL-2309 (unpublished)].
4 M. S. Moore and C. W. Reich, Phys. Rev. **118**, 718 (1960).
5 J. E. Evans, Atomic Energy Commission Report IDO-16120, 1953 (unpublished).

total fissions occurring during spectrometer irradiations. As a result of the low-neutron intensities and resolutions presently available, only irradiations in the vicinity of the large 0.297-ev resonance of Pu^{239} were possible, and experiments at the higher resonances (7.9 ev and above) could not be carried out. In order to test for possible interference effects in fission, which sometimes result in fairly major changes in measurements made across a resonance (e.g., in measurements of eta,[6] the number of neutrons produced per neutron absorbed in fissile material) asymmetric/symmetric ratios at several energies across the 0.297 resonance were obtained. These were at 0.06, 0.22, 0.297 (peak), and 0.36 ev.

The Pu^{239} samples used throughout this work had the following isotopic composition: Pu^{239}, 95.40%; Pu^{240}, 4.31%; Pu^{241}, 0.28%; Pu^{242}, 0.01%. For thermal and epi-Sm irradiations carried on inside the reactor, 0.4 and 0.2 mg samples, respectively, were used. These samples were prepared by dissolving Pu metal in HCl and evaporating aliquots of the solution to dryness on Al foils. For spectrometer irradiations, samples were strips of Pu-Al alloy of dimensions $\frac{1}{2}\times 2\frac{1}{4}\times\frac{1}{16}$ in., each containing about 0.17 g of Pu.

The Pu^{241} used in this work had the following isotopic composition: Pu^{239}, 1.10%; Pu^{240}, 2.18%; Pu^{241}, 96.60%; Pu^{242}, 0.12%. Because of the limited amount of Pu^{241} available, spectrometer irradiations were not possible, and only thermal and epi-Sm resonance irradiations (inside the reactor) could be carried out. Samples each consisted of 22 μg of Pu^{241} in nitrate solution evaporated on Al foils.

After irradiations of Pu samples with neutrons of various energies, the asymmetric to symmetric fission yield ratios were obtained by radiochemical isolation and measurement of the following fission products: 66-hr Mo^{99} (characteristic of asymmetric fission); and 53-hr Cd^{115}, 27-hr Sn^{121} and 10-day Sn^{125} (characteristic of symmetric fission). Similar chemical procedures were employed in processing all samples. Each of the irradiated samples (including Al backing or Al alloy) was dissolved in 6*M* HCl and diluted to volume in a volumetric flask. Duplicate small aliquots (4% of the solution) were removed and Mo carrier added prior to isolation of the Mo fraction. Sn and Cd carriers were then added to the remaining bulk of the solution. After treatment to assure radiochemical exchange of Sn and Cd, Pu was reduced to Pu(III) with HI and was then removed by passing the entire solution through an anion exchange column loaded with Dowex-1 anion exchange resin in Cl^- form. The resin was washed with 1*M* HCl to thoroughly remove Pu, which passes through the column while Sn and Cd are retained on the resin. The resin was then "wet ashed" with hot $HNO_3-H_2SO_4$ mixture and the oxidation was completed with a few drops of $HClO_4$. This radical treatment was necessary to assure maximum recovery of the Cd and Sn fractions which had very low counting rates after the spectrometer irradiations. After separation of the Sn and Cd by an ammonium polysulfide procedure, the Mo, Sn, and Cd fractions were purified radiochemically by the procedures described by Kleinberg.[7] Because of their relatively low counting rates, fission product samples isolated from spectrometer-irradiated Pu^{239} were all counted on a low-level beta-counting system having a background of ~2 counts/min. For purposes of comparison, samples from in-pile irradiated Pu^{239} were also counted on this system. The fission product samples isolated from in-pile irradiated Pu^{241} had sufficiently high counting rates to be counted on an end-window flowing methane proportional counter and the low-level system was not employed in the Pu^{241} experiments.

Because only the ratios of mass yields of asymmetric to symmetric fission products for thermal and for resonance fission were required, determinations of absolute disintegration rates and absolute fission yields were not necessary. It was sufficient to compare saturated activities of the various fission products (which are directly proportional to their absolute fission yields), each product being counted in an arbitrary, but standard, reproducible manner. The counting rates of all samples were corrected for chemical recovery and decay, and all samples were normalized to an arbitrary standard sample thickness by means of self-absorption—self-scattering correction curves empirically constructed for this purpose. Decay corrections and resolutions of complex decay curves were based on weighted least squares analyses of the counting data, carried out on an IBM-650 data processing machine. Irradiation periods ran from 15 min in the highest flux in-pile irradiations to 120 hr in the longest spectrometer irradiations.

EXPERIMENTAL RESULTS

Comparisons of the asymmetric to symmetric fission ratio at thermal and at resonance energies are made in terms of a quantity R defined as follows:

$$R=\frac{A_{asy}'/A_{sym}'}{A_{asy}/A_{sym}},$$

TABLE I. Comparison of thermal and resonance fission of Pu^{239} and Pu^{241}.

Pu isotope	Neutron energy, ev	R, indicated by Mo^{99}/Cd^{115}	Mo^{99}/Sn^{121}	Mo^{99}/Sn^{125}
239	0.06	1.33±0.06	1.26±0.05	1.16 ±0.04
	0.22	2.60±0.15	2.82±0.32	1.85 ±0.18
	0.297	3.00±0.28	3.28±0.31	2.05 ±0.07
	0.36	3.24±0.14	3.22±0.25	1.78 ±0.16
	All epi-Sm	2.41±0.15	2.34±0.06	1.79 ±0.06
241	All epi-Sm	1.02±0.03	1.01±0.08	0.995±0.15

[6] E. H. Magleby, J. R. Smith, J. E. Evans, and M. S. Moore, Atomic Energy Commission Report IDO-16120, 1956 (unpublished).

[7] J. Kleinberg, Los Alamos Scientific Laboratory Report LA-1721 (unpublished).

Table II. Comparison of calculated and measured R values for Pu^{239}.

Energy	Thermal	0.06 ev	0.22 ev	0.297 ev	0.36 ev
Contribution to $\sigma_{n,f}$ from 0.297-ev level	358 b (48%)	303 b (61%)	1100 b (97.4%)	3265 b (99%)	1120 b (97.4%)
Contribution to $\sigma_{n,f}$ from other spin state	388 b (52%)	192 b (39%)	~30 b (2.6%)	~30 b (~1%)	~30 b (2.6%)
Calculated R value	1.00 (normalized)	1.21	2.97	3.21 (assumed)	2.97
Observed R value					
Mo^{99}/Cd^{115}	1.00 (normalized)	1.33±0.06	2.60±0.15	3.00±0.28	3.24±0.14
Mo^{99}/Sn^{121}	1.00 (normalized)	1.26±0.05	2.82±0.32	3.28±0.31	3.22±0.25

where A is the saturated counting rate for a nuclide, subscript "asy" refers to an asymmetric fission product (Mo^{99} in this case), and subscript "sym" refers to a symmetric fission product (Cd^{115}, Sn^{121}, or Sn^{125}). Primed activities refer to resonance fission and unprimed activities to thermal fission.

In Table I are summarized the values of R computed from measurements described above. These values are based on the averages of at least three independent irradiations at each energy, and the errors expressed in the table are the standard error of the mean.

DISCUSSION

For each experiment where neutrons of a given discrete energy were employed, and for each where gross epi-Sm neutrons were used, the values of R based on Cd^{115} and Sn^{121} listed in Table I are indistinguishable within the errors quoted. The R values based on Sn^{125}, however, are significantly lower than the corresponding values based on Cd^{115} and Sn^{121}. This is not unexpected inasmuch as the thermal fission yields of Cd^{115} and Sn^{121} are nearly identical and both nuclides are produced by fission events which may be described as close-to-completely-symmetric fission. On the other hand, Sn^{125} is several mass numbers removed from completely symmetric fission and therefore might be expected to be a less sensitive indicator of change in the symmetric/asymmetric yield ratio.

If it is assumed, as the theory[2,3] suggests, that difference of spin results in a difference in the ratio of asymmetric to symmetric fission, and further if it is assumed that that portion of the thermal cross section not attributable to the 0.297-ev resonance is associated with the spin state differing from that of the 0.297-ev level, then the measured R values of Table I may be used to calculate the ratio of the asymmetric/symmetric ratios typical of each spin state. A recent multilevel analysis of the Pu^{239} cross section carried out by Vogt[8] is not in disagreement with the above assumption concerning spin states. Vogt's analysis indicates the presence of a bound level which is responsible for a large part of the thermal cross section, and which may differ in spin from (does not interfere with) the 0.297-ev level. The single-level Breit-Wigner formula is thus adequate to describe this level, and on this basis, calculations have been made of the contribution of the 0.297-ev level to the fission cross section at the five different energies at which R values were measured. The parameters given in the BNL "Neutron Cross Sections" compilation[9] have been used in these computations. With the respective contributions to fission cross sections as weighting factors, with the asymmetric/symmetric ratio normalized to 1.00 for fission in the spin state associated with the bound level, and with an assumed R value of 3.2 obtained from Mo^{99}, Sn^{121}, and Cd^{115} data at the peak of the 0.297-ev resonance, it is calculated that the asymmetric/symmetric ratio characteristic of the spin state predominating at the 0.297-ev level is 5.3 times the ratio typical of the other spin state. Actually if at negative energies there were an additional level or levels of the same spin as the 0.297-ev level, and if these were to contribute significantly to the thermal cross section, then a value even larger than 5.3 would result. Comparisons of experimentally measured R values with calculated values are listed in Table II.

The self-consistency of the interpretation assumed above is shown by comparison of the calculated R value of 1.21 for 0.06-ev neutrons with the experimentally measured values of 1.33±0.06 and 1.26±0.05. All three values are in agreement within the errors of the assumptions and experiments. The R values obtained at 0.22, 0.297, and 0.36 ev indicate that, as expected, no important interference effects operate to change the asymmetric/symmetric ratio across the resonance studied.

Since the ground-state spin and parity[10] of Pu^{239} are $\frac{1}{2}+$, the Pu^{240} compound nuclei formed upon neutron addition may have spin and parity of either $0+$, or $1+$. The present results, of themselves, do not permit assignment of spins to each of the two states for which there is an apparent characteristic asymmetric/symmetric fission ratio. It is interesting to note that Mostovaya[11] has compared the mass distribution for thermal neutron-induced fission of Pu^{239} with that for spontaneous fission of Pu^{240} in its ground state and has found that the "peak-

[8] E. Vogt, Phys. Rev. **118**, 724 (1960).

[9] *Neutron Cross Sections*, compiled by D. J. Hughes and R. Schwartz, Bookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

[10] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 822 (1958).

[11] T. A. Mostovaya, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), Vol. 15, p. 433.

to-valley" ratio is much larger for the latter case. However it does not appear possible to make definite spin-state assignments in the present experiments on the basis of Mostovaya's work, since in the latter work the observed effects are doubtless mainly due to large excitation energy differences, while in the present work the effects are presumably due to a different cause (spin difference) at the same excitation level. It is well known that differences in excitation energy produce marked changes in asymmetric/symmetric fission ratios.[12]

The recent work of Fraser and Schwartz,[13] coupled with the multilevel analysis of Vogt[8] does, however, permit indirect assignment of spins to the 0.297 ev and to the bound level. By means of scattering experiments Fraser and Schwartz were able to assign directly spin $J=1$ to the 7.8, 10.9, and 11.9-ev resonances. Vogt's analysis indicates that the 0.297-ev level does not involve the same spin as the 7.8-ev level or the bound level. Since the 7.8-ev level has spin $J=1$, and the analysis indicates that the spin of the bound level is the same, then the spin of the 0.297-ev level may be deduced as $J=0$. However, this is in contradiction to the deductions of Bollinger, Coté, and Thomas,[14] who assigned spin $J=1$ to the 0.297-ev level.

Finally the data of Table I for fission with epi-Sm neutrons indicate that, in contrast to Pu^{239}, Pu^{241} gross resonance fission is similar to thermal fission. This is rather interesting since the Pu^{239} and Pu^{241} fission cross section curves are quite similar; both have a single strong resonance at $\sim$0.3 ev followed by an absence of strong resonances up to $\sim$6 ev, and both have high thermal fission cross sections with bound levels indicated. If spin is indeed a factor in determining the asymmetric/symmetric fission ratio at the same excitation energy, then the R values of Table I may be taken to indicate that for Pu^{241}, thermal fission involves the same weighting of spin states as that involved in gross resonance fission with epi-Sm neutrons; for Pu^{239} a considerably different weighting must be involved.

As higher intensity monoenergetic neutron sources become available, further study of Pu^{239} offers the following advantages over other fissile nuclei: the asymmetric to symmetric ratios can undergo large changes from resonance to resonance; the resonances are well separated and do not have large interference effects; direct assignments of spins to several resonances have already been made; and Pu^{240} spontaneous fission provides a possible important comparison point for the same fissioning nucleus at a different excitation level but with a known spin.

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[12] R. B. Leachman, reference 11, Vol. 15, p. 229.

[13] J. S. Fraser and R. B. Schwartz, Bull. Am. Phys. Soc. **5**, 294 (1960).

[14] L. M. Bollinger, R. E. Coté, and G. E. Thomas, *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), Vol. 15, p. 127.