PHYSICAL REVIEW C VOLUME 10, NUMBER 4 OCTOBER 1974

Fission-neutron multiplicity and total prompt gamma-ray energy for resonances in ^{239}Pu†

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(Received 1 July 1974)

The fission-neutron multiplicity $\bar{\nu}$ and the total prompt γ-ray energy have been studied for the resolved resonances in ^{239}Pu between 10 and 170 eV. There was a weak correlation found between $\bar{\nu}$ and the spin of the resonances. There was a significant correlation between the total γ-ray energy and the spin of the resonance. These correlations can be explained by competition of the $(n,\gamma f)$ reaction with the (n,f) reaction in the $J=1$ resonances which have a narrow fission width.

[NUCLEAR REACTIONS neutrons per fission and total prompt γ energy from ^{239}Pu resonance fission, $E_n=10$–170 eV. Correlation with Γ_f due to ^{239}Pu$(n,\gamma f)$.]

INTRODUCTION

Experimental determinations of the fission-neutron multiplicity $\bar{\nu}$ for the resonances of ^{239}Pu have been very confusing historically. Quite strong correlations of $\bar{\nu}$ with the resonances of the two spin states have been reported with opposite correlations.[1,2] Preliminary results of this experiment which indicated no significant correlations have been reported.[3] More recent reports by other experimenters report weak correlations.[4,5]

In ^{239}Pu there is the possibility that in some cases the $(n,\gamma f)$ process can be comparable in probability with the (n,f) process. The effect would be correlated with the spin of the resonances since the $J=0$ resonances tend to have large fission widths which would mask such an effect, whereas part of the $J=1$ resonances have fission widths comparable to the expected width for the $(n,\gamma f)$ reaction (about 3 meV for $J=1$ resonances).[6] Experimental evidence of the competing $(n,\gamma f)$ process is the sparsity of resonances in ^{239}Pu with very narrow fission widths.[6] In the cases where the total fission width is comparable with the $(n,\gamma f)$ width of the resonance, there may be other experimentally observable effects.

The $(n,\gamma f)$ reaction could conceivably affect both the total prompt γ-ray energy following fission as well as $\bar{\nu}$, as has been proposed by Shackleton *et al.*[4] There is a recently reported experiment with evidence that the total γ-ray energy following fission is higher for the resonances of ^{239}Pu with very narrow fission widths.[4] This can be explained by the fact that a γ ray is given off first in the $(n,\gamma f)$ reaction before the statistical processes toward fission progress very far. Because of this initial γ ray, one might expect the total prompt γ-ray energy to be higher.

There is also the possibility that the $(n,\gamma f)$ reaction could affect the neutron multiplicity $\bar{\nu}$ for the resonances of narrow fission widths. This experimental effect has been suggested in a recent report.[4] The reason for this effect could be that the initial γ ray of the $(n,\gamma f)$ reaction carries off part of the available excitation energy. With less total excitation energy available, fewer neutrons on the average might be emitted. Alternative means of energy balance such as kinetic energy of fission fragments and neutrons also exist.

Both the average neutron multiplicity and the total γ-ray energy following fission in resonances of ^{239}Pu were studied in this experiment.

EXPERIMENTAL PROCEDURE

For these studies a fission chamber was used to detect the occurrence of a fission event, fast neutron detectors were used to detect the average number of fission neutrons per fission, and γ-ray detectors were used to detect the prompt γ rays given off by the fission event. The Oak Ridge electron linear accelerator was used as a source of neutrons. The energies of the neutrons causing the detected fission events were measured by their time of flight from the neutron producing target to the fission chamber along a flight path of 20 m.

The ^{239}Pu fission chamber was of multiple, parallel plate design[7] and contained 1.4 g of Pu with an average areal density of 0.8 mg/cm^2. The fission fragment pulses were amplified by ten separate, current amplifiers (one for each independent section of the fission chamber) because of the high natural α-decay rate of the Pu. As biased, the chamber's efficiency was ~30% with a gross α pileup count rate of 300 counts/sec. The background due to the α pileup events could be accurately determined between Linac bursts after the neutrons had passed.

The fast neutron detectors were liquid scintillators (NE-213) which were 10 cm in diameter and 5 cm thick. The two scintillators were mounted on 12.7-cm photomultipliers (58 AVP). Pulse-shape discrimination was done on the pulses from the scintillators in order to discriminate between fast neutrons and γ rays. The pulse-shape discrimination was of a type developed by Forte.[8] The liquid scintillators were mounted opposing each other, each at a distance of 11 cm from the center of the fission chamber. Prompt fission γ rays given off simultaneously with the fission neutrons as well as γ rays delayed by <1 μsec with respect to the fission neutrons could have been sufficiently intense to confuse the γ-neutron discrimination circuit. To reduce such effects 1.3 cm of Pb was placed between the fission chamber and each scintillator. In order to check that capture γ rays were not detected by the neutron counters, an experimental test was made under operating conditions except with an additional Au sample in the neutron beam. The compound efficiency for detecting the capture γ rays from Au and the discrimination circuit indicating neutrons was about 1 part in 600.

The efficiency for fission-neutron detection was about $\frac{1}{2}\%$ per detector, per fission neutron. This efficiency was low enough to suppress effects to multiple neutron detection even though there is a correlation between the fragment axis and the direction of emission of fission neutrons. Evidence of multiple neutron events in the scintillator being a small effect is the fact that the ratio of coincidences to singles between the two liquid scintillators was about 1%. Further tests were made with Cf and ^{235}U which have appreciably different values of $\bar{\nu}$. These tests also indicated that fission neutrons were being reliably detected.

The total γ-ray energy detectors were liquid scintillators (NE-226) of the same size (10 cm diam and 5 cm thick) as the neutron detectors. The γ-ray detectors were of a nonhydrogenous fluorocarbon scintillator to minimize the sensitivity to the fast fission neutrons. In order to obtain the average total γ-ray energy following fission, the detectors were used in a mode described by Mack-

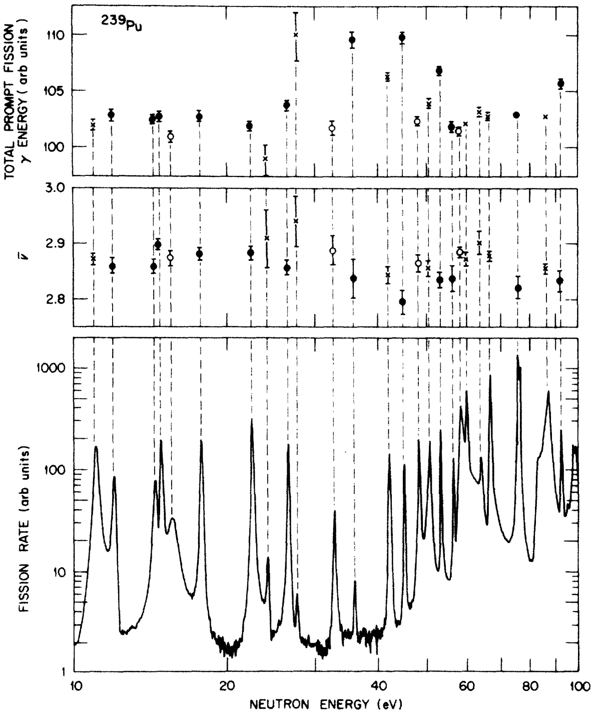

FIG. 1. The average neutron multiplicity $\bar{\nu}$ and the average total prompt γ-ray energy following fission for the resonances of ^{239}Pu in the neutron energy range of 10 to 100 eV. Open circles are $J=0$ resonances. Closed circles are $J=1$. Crosses are resonances of uncertain spin or unresolved groups of different J values.

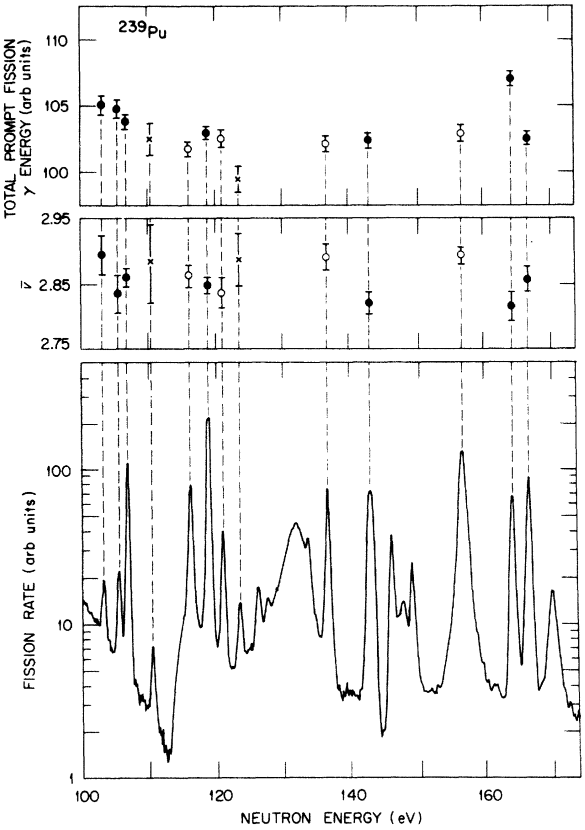

FIG. 2. The average neutron multiplicity $\bar{\nu}$ and the average total prompt γ-ray energy following fission for the resonances of ^{239}Pu in the neutron energy range of 100 to 170 eV.

lin and Gibbons.[9] The pulse height of each γ-ray event in coincidence with fission was recorded. These were individual γ-ray events since the probability of pileup was small (~1%). By weighting the event according to its pulse height, the detectors could be given an effective efficiency which is directly proportional to γ-ray energy. With such an efficiency which is linear with γ-ray energy, the net efficiency for the γ-ray cascade following fission would be proportional to the total γ-ray energy. This is the same principle on which the well-known Moxon-Rae detector[10] works except in this case a higher effective efficiency can be obtained. These γ-ray detectors were run in coincidence with the fission chamber simultaneously with the fast neutron detectors.

EXPERIMENTAL RESULTS

The results of the experiment are presented in Figs. 1 and 2. Figure 1 is for the neutron energy range 10 to 100 eV and Fig. 2 is for the neutron energy range 100 to 200 eV. At the bottom of the two figures is plotted the detected fission rate vs the neutron energy. The plot of the fission rate simply indicates the resonances for which the values of $\bar{\nu}$ and $\bar{E}_\gamma$ are given in the top part of the figure. The values of $\bar{\nu}$ and $\bar{E}_\gamma$, shown on an expanded scale in the top part of the figures, represent an average over a resonance. The errors shown are those due to counting statistics. There are no known systematic effects that would cause errors comparable or as large as the statistical errors. The normalization of the data is arbitrary since accurate absolute efficiencies were not determined. The normalization used was $\langle\bar{\nu}\rangle = 2.868$ over the energy range 10 to 100 eV. This normalization is consistent with the number of prompt neutrons per fission for thermal neutron induced fission.[11] The data are presented in tabular form in Table I.

Figures 1 and 2 show an obvious correlation between the total fission γ-ray energy $\bar{E}_\gamma$ and the spin of the resonances. The very narrow resonances which tend to be $J=1$ resonances tend to have higher $\bar{E}_\gamma$ than the broad resonances which tend to be $J=0$. There are no obvious indications in the case of $\bar{\nu}$ that there are correlations with the spin of the resonance. However, a weighted mean of $\bar{\nu}$ for the resonances which are clearly $J=1$ is almost two standard deviations away from the weighted mean of $\bar{\nu}$ for the resonances which are clearly $J=0$. This difference of $\frac{1}{2}$% between the weighted means with the $J=0$ resonances having the higher $\bar{\nu}$ is small but possibly significant. Since the narrow resonances have the largest statistical errors, it may also be significant that the unweighted means have a larger difference of 0.8%. The difference in the means of $\bar{E}_\gamma$ (2.5%) and $\bar{\nu}$ is very close to the value which would be expected if the two quantities were directly related. That is, the reported experimental measurements[12] of the rate of change of $\bar{\nu}$ with excitation energy is just about right to explain the change in the mean $\bar{\nu}$ by the change in mean total γ-ray energy $\bar{E}_\gamma$.

TABLE I. Values of the average neutron multiplicity and total γ-ray energy for the resonances of ^{239}Pu.

E_{res} [a] (eV)	J [a]	Γ_f [a] (meV)	$\bar{\nu}$	Error	Total E_γ	Error
10.9, 11.5	1, 0	143, 10	2.873	0.007	101.8	0.3
11.89	1	24	2.861	0.012	102.9	0.5
14.31	1	67	2.860	0.011	102.5	0.3
14.68	1	30	2.900	0.007	102.8	0.3
15.46	0	656	2.875	0.012	101.0	0.5
17.66	1	34	2.884	0.010	102.8	0.3
22.29	1	62	2.886	0.008	102.1	0.3
23.94		38	2.910	0.051	99.0	
26.24	1	44	2.859	0.012	103.8	0.4
27.24		5	2.943	0.047	110.1	2.2
32.31	0	110	2.888	0.027	101.8	0.6
35.5	1	4	2.840	0.036	109.8	0.5
41.42, 41.66	1, ?	4, 46	2.845	0.015	106.5	0.3
44.48	1	5	2.797	0.020	109.9	0.5
47.6	0	248	2.868	0.013	102.5	0.3
49.7, 50.1	0, 1	746	2.858	0.012	104.2	0.3
52.6	1	9	2.836	0.013	106.8	0.3
55.6	1	21	2.840	0.022	102.0	0.5
58.8, 57.4	0, 0	445	2.886	0.007	101.6	0.2
59.2, 60.9	1, 0	123, 6736	2.875	0.009	102.3	0.2
63.1		111	2.903	0.020	103.3	0.4
65.4, 65.7	?, 1	50, 71	2.880	0.007	103.0	0.2
74.1, 75.0	1, 1	32, 84	2.822	0.020	103.4	0.1
90.8	1	9	2.837	0.015	105.9	0.4
103.0	1	10	2.895	0.031	105.2	0.7
105.3	1	5.4	2.834	0.031	104.8	0.7
106.7	1	26	2.860	0.014	103.9	0.4
110.4		13	2.887	0.060	102.5	1.2
116.0	0	218	2.863	0.014	101.7	0.4
118.8	1	42	2.846	0.010	103.1	0.3
121.1	0	39	2.838	0.024	102.6	0.6
123.4		39	2.891	0.018	102.2	0.5
136.8	0	84	2.891	0.018	102.2	0.5
142.9, 143.5	1	82, 31	2.822	0.015	102.3	0.4
157.1	0	540	2.892	0.011	102.9	0.3
164.5	1	8	2.813	0.021	107.1	0.5
167.1	1	69	2.855	0.018	102.6	0.3

[a] See Ref. 13.

Figure 3 is a plot of $\bar{E}_\gamma$ and $\bar{\nu}$ vs the reciprocal of the fission width,[13] Γ_f, of the resonance for the resonances which are clearly $J=1$. This type plot has been used by Frehaut and Shackleton[4] to accentuate the effect of the variation of $\bar{E}_\gamma$ and $\bar{\nu}$ with resonances of fission width comparable to the width for the $(n, \gamma f)$ reaction. The competition of the $(n, \gamma f)$ process with the (n, f) process is strongest for the resonances of small fission width.

Figure 3 shows a striking, apparently linear relationship between $\bar{E}_\gamma$ and $1/\Gamma_f$. The intercept for zero $1/\Gamma_f$ should be 8.0 MeV[14] which is the total prompt fission γ-ray energy. This strong correlation can be interpreted as evidence that the $(n, \gamma f)$ process is competing in the resonances with narrow fission widths.

In Fig. 3 the plot of $\bar{\nu}$ vs $1/\Gamma_f$ does not indicate such clear results as in the case of $\bar{E}_\gamma$. The line marked "energy balance" would be the result if it were assumed that all the increase in $\bar{E}_\gamma$ with $1/\Gamma_f$ appeared as a decrease in $\bar{\nu}$ through the experimentally determined relationship: $(d\nu/dE_n) = 0.128$ neutron per MeV.[12] This $d\nu/dE$ was derived by adding excitation energy via kinetic energy of the incoming neutron and the applicability to the present case is an assumption. The least squares fit to the data does not show as steep a slope as the "energy balance" line. However, a weighted least squares fit shows a somewhat steeper slope. The slopes of these lines must not be taken too seriously because of the large uncertainties in the data as compared to the variation of $\bar{\nu}$ from its average value. There seem to be variations in the data which are not explained by correlation with the fission width of the resonance or with the statistical uncertainties. This is indicated by the poor fit in Fig. 3 and examination of Fig. 1 indicates there may be variations in $\bar{\nu}$ with neutron energy over ranges of energy which are large compared to the resonance spacing.

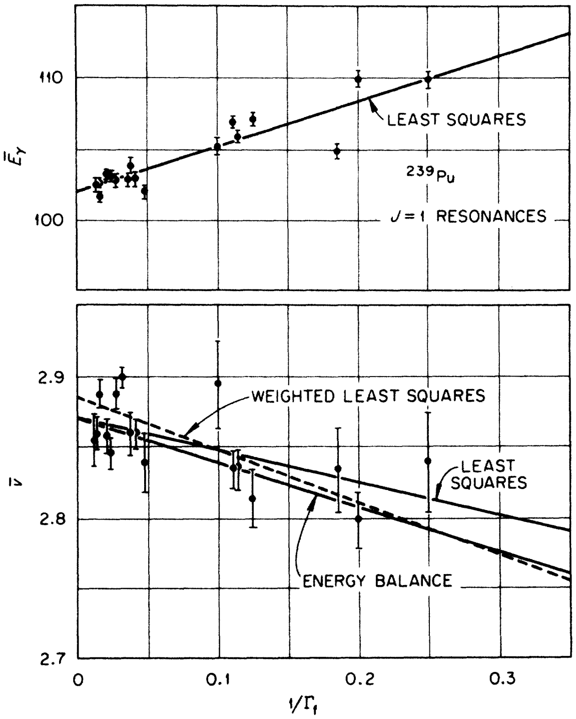

FIG. 3. Total fission γ-ray energy $\bar{E}_\gamma$ and fission neutron multiplicity $\bar{\nu}$ as a function of the reciprocal of the fission width of the resonance.

Such a weak correlation between $\bar{\nu}$ and the spin of the resonance is in contradiction with the data of others.[1,2,4,5] This difference between the present experiment and the others is not understood. Most of the other experiments used a quite different technique. That is, they used a large liquid scintillator tank loaded with gadolinium or cadmium to detect fission neutrons with high efficiency. After a coincidence between the fission chamber and the tank signaled that a fission had occurred, the pulses caused by neutron absorptions in the tank were counted. Since the other experimental results which used the scintillator tank method are poorly correlated with each other, it is difficult to speculate that there are systematic differences between the two types of experiments. One shortcoming of the present experiment is that the neutron detectors are sensitive to the kinetic energy of the neutrons. If there were changes in the energy spectrum of the neutrons from resonance to resonance, this could cause uncertainties in the measurement of $\bar{\nu}$. Such variations in the energy spectrum of the prompt fission neutrons are not expected because a large part of their kinetic energy is derived from the motion of the fission fragments from which they are emitted. An advantage of the present method is the very low background in neutron detection as compared to the other experiments.

CONCLUSIONS

The present experiments shows a strong correlation between the total prompt γ-ray energy following fission and the fission width of the resonance. This also correlates with resonance spin since the $J = 1$ resonances have narrower fission widths. The total γ-ray energy is higher for the resonances of very small fission width where the $(n, \gamma f)$ reaction can compete with the (n, f) reaction. These results are in agreement with those of Shackleton *et al.*[4]

The present data show a possible correlation between resonance spin and $\bar{\nu}$. The correlation is not as strong or significant as with other experiments.[1,2,4,5] The present data give an average weighted difference between the value of $\bar{\nu}$ for the two spin states of $\sim\frac{1}{2}\%$. Theoretical predictions from nuclear models do not yield a unique reinforcement of any particular set of data.

In the resonance region of neutron energies the data indicate that the variation of $\bar{\nu}$ from resonance to resonance is small enough and random enough that this effect could be ignored in reactor calculations. The variation in $\bar{\nu}$ can also be ignored in fission cross section measurements involving the detection of fission neutrons, since the uncertainties in these measurements are generally larger than the variations in $\bar{\nu}$. The fission cross section measurements are usually for reactor applications where the resonances of very narrow fission widths which have the greatest variation of $\bar{\nu}$ from the average are of small consequence.

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†Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

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