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Slow-Neutron Cross Sections of Pu^{240}, Pu^{242}, and Am^{243}*

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Measurements of the total cross sections of Pu^{240}, Pu^{242}, and Am^{243} have been made using the Argonne "fast chopper." The results on Pu^{240} include only the resonance structure which could be studied through the Pu^{240} impurity in some samples of Pu^{239} which were used in an exhaustive study of the latter nuclide. The values of the resonance parameters of the resonances at 1.053, 20.4, and 38.2 ev comprise these results. The studies of Pu^{242} and Am^{243} were made possible by the separation of these nuclides from samples of Pu^{239} which had undergone extensive neutron irradiation. Resonances were observed at 2.65 and 53.6 ev in Pu^{242} and at 0.976, 1.353, 1.74, 3.42, 5.12, 6.54, 7.84, 10.3, 12.8, 13.1, and 15.3 ev in Am^{243}. Resonance parameters are reported for all of these resonances and in addition, the resonance capture integrals derived from these parameters are compared with the results of other measurements of these quantities.

INTRODUCTION

MEASUREMENTS of the neutron cross sections of the heavy artificial nuclides are important both to the design of nuclear reactors and to the understanding of the nucleus itself. For reactor studies, the importance[1] of these cross sections comes from their very definite role in determining long-term reactivity variations and neutron economy. Their importance to the physics of the nucleus lies mainly in the fact that they increase the range of nuclides over which theory and experiment can be compared.

In heavy nuclei, not near closed shells, the spacing of the neutron resonances is generally small, so that many resonances lie in the energy range in which the present time-of-flight spectrometers have extremely fine resolution. Consequently, studies of these nuclei make it possible to determine the resonance parameters for a large number of resonances. That large numbers of these parameters are necessary is a consequence of the fact that most of the theoretical work in this field has resulted in the prediction of certain average values of some of these parameters or in the specification of statistical distribution functions for some of them. For example, there are proposed distribution functions for the reduced neutron widths[2] and for the level spacings[3]; there are predictions of the variation of the neutron strength function with nucleon number.[4,5]

Experimentally, the only difference between measurements on these rare heavy nuclides and those of other heavy nuclides is in the availability of samples. The former are difficult to obtain at all and can be had, at best, only in extremely limited amounts. This factor severely determines both the quantities which can be measured and the methods which can be used. For those nuclides which have fission thresholds above the region of interest (roughly defined as being the energy region from zero to several hundred electron volts), the only measurements which can be made, aside from integral type measurements, are those of the total cross section. Because of the further limitation of the small size of the samples, measurements of the total cross section can best be made with a "fast chopper" such as the machine at Argonne[6] with which these measurements were made.

EXPERIMENTAL DETAILS

Since it is only the small size of the samples which made parts of the present studies different from most normal measurements of the total cross section by a transmission experiment, details relevant to this problem are the only ones which are discussed. All of the remarks concerning size of sample are directed at the Pu^{242} and Am^{243} samples, since no effort was made to procure special samples of Pu^{240}.

The exit collimator of the fast-chopper system provides space into which sample holders containing up to 2 in. of material may be placed. The design of the collimator is such that each of its seven slits, which correspond to those of the rotor, tapers (though not linearly) from a width of 0.025 in. at the near end (i.e., the end near the rotor) to 0.045 in. at the far end (i.e., the end towards the detector). Furthermore, the design is such that a full 2 in. of material must be inserted in the form of two 1 in. portions, one close to each end of the collimator. Because of the taper, samples made for the far end should be 0.065 in. wide while those for the near end can be made 0.035 in. wide. For the Pu^{242} and Am^{243} samples, only the narrower width could be used; and even so, there was material enough to allow use of

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[1] H. Hurwitz, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August, 1955* (United Nations, New York, 1956), Vol. 4, p. 261.

[2] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956); H. A. Bethe, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, p. 321; J. M. C. Scott, Phil. Mag. **45**, 1322 (1954).

[3] S. Blumberg and C. E. Porter, Phys. Rev. **110**, 786 (1958); N. Rosenzweig, Phys. Rev. Letters **1**, 24 (1958); I. I. Gurevich and M. I. Pevzner, J. Exptl. Theoret. Phys. U.S.S.R. **31**, 162 (1956) [translation: Soviet Phys. JETP 4, 278 (1957)].

[4] Feshbach, Peaslee, and Weisskopf, Phys. Rev. **71**, 145 (1947).

[5] Feshbach, Porter, and Weisskopf, Phys. Rev. **96**, 448 (1954); Chase, Willets, and Edmonds, Phys. Rev. **110**, 1080 (1958).

[6] Bollinger, Coté, and Thomas, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva,* 1958.

TABLE I. The isotopic constitution of the samples used for the analysis of the resonances in Pu^{240}.

Thickness of sample (number of Pu^{240} atoms/cm^2)	Percent abundance Pu^{239}	Pu^{240}	Pu^{241}
1.17×10^{19}	99.38±0.01	0.61±0.01	0.015±0.001
2.40×10^{19}	94.51±0.08	5.11±0.07	0.38 ±0.03
5.86×10^{19}	99.38±0.01	0.61±0.01	0.015±0.001
7.74×10^{19}	94.51±0.08	5.11±0.07	0.38 ±0.03
$17.7\ \times10^{19}$		1.47	

only a small fraction of the height of one of the seven slits. In the case of Am^{243} only 5 mm of one of the slits could be used, i.e., the cross sectional area of the sample was only 5 mm.^{2}

The primary effect of being able to use only a small fraction of the available slit area is a reduced counting rate which results in a much less favorable signal-to-background ratio than is normally obtained. In order to have a signal-to-background ratio high enough to make it possible to obtain data of suitable statistical accuracy, it is necessary to use the shortest flight path compatible with the resolution required to study the resonant structure of the nuclides under consideration. Most of the results of the present investigations were obtained from data taken with a flight path of 25 m. With a 25-m flight path, the resolution can be as good as 0.08 μsec/m, a value which corresponds to full-speed operation of the chopper. The data on the thermal cross section of Am^{243} were taken with a flight path of only 7 m, however.

SAMPLES

The resonance parameters for Pu^{240} were obtained from measurements on Pu^{239} samples in which Pu^{240} was an impurity. These samples were large enough to cover the area corresponding to all seven slits and had thicknesses equal to 1.17×10^{19}, 2.40×10^{19}, 5.86×10^{19}, 7.74×10^{19}, and 17.7×10^{19} atoms of Pu^{240}/cm^2. The isotopic abundances of the various plutonium isotopes present in these samples are shown in Table I.

Extensive neutron irradiation of Pu^{239} produced[7] the plutonium and americium used in these measurements. These were purified from other products of irradiation, separated from each other, and analyzed spectrographically. The only impurities present at this point were such as would not interfere with the measurements.

The americium and plutonium were required in an anhydrous form to avoid neutron scattering (and beam attenuation) by hydrogen. There also might have been a potential health hazard from the formation of peroxide, hydrogen, and oxygen from the dissociation of water by the intense alpha radiation.

Plutonium was precipitated as an oxalate and converted to a water-free oxide at 400°C. The oxide was pulverized in a nickel crucible and loaded into an aluminum-Lucite capsule via a quartz funnel. Work was done in a glove-box protected from the atmospheric moisture by a barrier of magnesium sulfate.

Since the volume of americium oxide used (7 mg of metal) was too small to have intercepted an appreciable portion of the beam, the oxide was dispersed (with the numbers of atoms in the ratio Am:Al = 1:9) in alumina. The Am and Al were thoroughly mixed in solution and precipitated as hydroxides by the addition of ammonia gas. The precipitate was dried and ignited to 1000°C in air to convert to the nonhygroscopic oxide. The resultant oxide was pulverized and encapsulated in the same manner as the plutonium.

The thickness of the Pu^{242} sample was obtained from an area analysis of the 1.05-ev resonance in Pu^{240}, which was observable because of the 7.5% of this isotope present in the sample. The complete isotopic constitution of the sample is shown in Table II. From the values listed in this table it is clear that an error of 1% might exist in the thickness of the Pu^{242} sample because of the uncertainty in the amount of Pu^{240} present. In addition, errors of 4.5% and 9% are possible in the determination of the thicknesses of the thick and thin samples, respectively, because of errors in the measured areas of the transmission dips and of uncertainties in the resonance parameters of Pu^{240}. The values of the parameters of the 1.05 ev resonance in Pu^{240} which were used for the determination of the thickness of the Pu^{242} samples are $\sigma_0 = 1.73\times10^5$ b and $\Gamma = 0.0345$ ev. The thicknesses of the Pu^{242} samples determined in this manner are $(3.48\pm0.05)\times10^{20}$ atoms/cm^2, $(4.02\pm0.2)\times10^{19}$ atoms/cm^2 and $(1.48\pm0.15)\times10^{19}$ atoms/cm^2.

The thicknesses of the sample of Am^{243} used in these measurements were $(3.38\pm0.06)\times10^{20}$ and $(1.89\pm0.09)\times10^{20}$ atoms/cm^2. The only impurities known to be in the sample are 0.469% Am^{241}, 0.0048% Am^{242}, and 0.02% cerium.

RESULTS

Pu^{240}

Since the measurements on Pu^{240} were made simply because the samples used in a very thorough study of Pu^{239} contained various amounts of the former, the results are rather fragmentary. They have been included in this presentation because they are results of high quality on a nuclide which is of importance to reactor physics.

TABLE II. The isotopic constitution of the sample of Pu^{242}.

Isotope	Percent abundance	Isotope	Percent abundance
238	0.391±0.007	241	3.33 ±0.03
239	0.226±0.005	242	88.51 ±0.08
240	7.51 ±0.07	244	0.033±0.002

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[7] Bently, Diamond, Fields, Friedman, Gindler, Hess, Huizenga, Inghram, Jaffey, Magnusson, Manning, Mech, Pyle, Sjoblom, Stevens, and Studier, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, August, 1955* (United Nations, New York, 1956), Vol. 7, p. 261.

The usual area techniques[8] have been used, under the assumption that the fission cross section is negligibly small, to obtain the resonance parameters. Leonard, Seppi, and Friesen[9] have shown that the resonance fission in the resonance at 1.05 ev is equal to 356 b. Comparison of this value with the peak total cross section of 1.73×10^5 b shows that the assumption about the fission cross section is well justified.

The parameters obtained for the three resonances observed are listed in Table III. A value of Γ_γ could be obtained only for the resonance at 1.05 ev, so a value close to this value was assumed for Γ_γ in the analysis of the resonances at 20.4 and 38.2 ev. The results are in excellent agreement with those of Simpson and Fluharty[10] who have made a thorough study of this nuclide. It is of interest that the value of Γ for the first resonance deduced by Westcott and Walker[11] from the resonance absorption integral and thermal capture cross section is also in excellent agreement with the value reported herein.

Pu^{242}

As complete an investigation as possible was made on this nuclide over the energy range from thermal energies to several kev, with the rather disappointing result that only two resonances were found which could positively be assigned to it. The parameters for these resonances, along with their respective errors, are listed in Table IV. The contribution of the uncertainty in the thickness of the thin sample to the uncertainty in the parameters is included in these errors. The values of the parameters reported previously[12] were based on the analysis of data obtained from measurements of a single thick sample of Pu^{242}. The difference between the present results and these earlier values can be attributed to uncertainties in the size of the background, to which these early results were very sensitive.

TABLE III. The resonance parameters for the first three resonances in Pu^{240}. An average $\Gamma_\gamma=(32\pm3)\times10^{-3}$ ev was assumed for the 20.4- and 38.2-ev resonances, for which values of Γ_γ could not be deduced.

E_0 (ev)	Γ (10^{-3} ev)	Γ_γ (10^{-3} ev)	Γ_n (10^{-3} ev)	σ_0 (b)
1.053	34.5 ± 3	32.1 ± 3	$2.4\ \pm0.05$	$(1.73\pm0.15)\times10^5$
20.4	34.3 ± 3	$32\ \ \pm3$	2.26 ± 0.26	$(8.84\pm0.15)\times10^4$
38.2	$48\ \ \pm3$	$32\ \ \pm3$	$15.7\ \pm0.9$	$(2.22\pm0.17)\times10^4$

TABLE IV. The resonance parameter of the resonance observed in Pu^{242}. A value of $\Gamma_\gamma=(25.1\pm2.6)\times10^{-3}$ ev was assumed for the resonance at 53.6 ev in order to obtain the parameters listed.

E_0 (ev)	Γ (10^{-3} ev)	Γ_γ (10^{-3} ev)	Γ_n (10^{-3} ev)	σ_0 (b)
2.65	27.0 ± 2.7	25.1 ± 2.6	1.9 ± 0.2	$(6.65\pm1.3)\times10^4$
53.6	70.0 ± 6.2	25.1 ± 2.6	44.9 ± 6.0	$(3.1\ \pm0.2)\times10^4$

Thick and thin samples were available for the study of the resonance at 2.65 ev, so a value of Γ_γ could be obtained. Unfortunately, the resolution width was comparable to the width of the resonance so that a meaningful comparison of a theoretical curve (based on the parameters obtained from the area analysis) with the observed shape requires an accurate knowledge of the shape of the resolution function. Since the shape of this function is not well known, a calculation of the expected resonance shape was made in which a simple triangular resolution function was assumed. The width of this function was determined by the flight path, channel width and rotor speed which were used for the data being compared. The agreement is good except near the peak of the resonance, where the effect of the finite resolution is of greatest importance.

On the basis of a provisional assignment of the resonance at 2.65 ev to Pu^{242}, Egelstaff, Gayther, and Nicholson[13] have reported a value of $\Gamma_n=(1.7\pm0.8)\times10^{-3}$ ev, which agrees with that reported herein. The only other measurements on Pu^{242} which can be related to the present measurement are those of Butler, Lounsbury, and Merritt[14] (hereafter referred to as BLM). They have measured the resonance capture integral and have listed the neutron capture cross section for neutrons with speeds of 2200 m/sec. No direct comparison of this latter cross section can be made because the present measurements were not carried out in the thermal energy region because of the large amount of Pu^{240} present in the sample. However, a value of the capture cross section for 2200-m/sec neutrons resulting from the epithermal resonances can be obtained from the measured resonance parameters. The value of 19.2 ± 3.3 b which is obtained is in good agreement with the 18.6 ± 0.8 b of BLM; their result is based on the assumption that Pu^{242} is a "$1/v$" absorber.

A value of the resonance capture integral can also be obtained from the measured resonance parameters. The value obtained is 1050 ± 150 b if the contribution, as estimated by a formula due to Dresner,[15] of levels above 55 ev is added to those of the two known resonances. The agreement between this value and the 1275 ± 30 b of BLM is not as good as for the thermal capture cross sections.

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[8] Melkonian, Havens, and Rainwater, Phys. Rev. **92**, 702 (1953); Seidl, Hughes, Palevsky, Levin, Kato, and Sjostrand, Phys. Rev. **95**, 476 (1954).

[9] Leonard, Seppi, and Friesen, Oak Ridge National Laboratory Report ORNL-2309 (unpublished).

[10] O. D. Simpson and R. G. Fluharty, Bull. Am. Phys. Soc., Ser. II, **2**, 219 (1957).

[11] C. H. Westcott and W. H. Walker, Atomic Energy Commission Report TID-7547, 1957 (unpublished), and private communication.

[12] Coté, Bollinger, Barnes, and Diamond, Atomic Energy Commission Report TID-7547, 1957 (unpublished).

[13] Egelstaff, Gayther, and Nicholson, J. Nuclear Energy **6**, 303 (1958).

[14] Butler, Lounsbury, and Merritt, Can. J. Phys. **35**, 147 (1957).

[15] L. Dresner, J. Nuclear Energy **2**, 118 (1955).

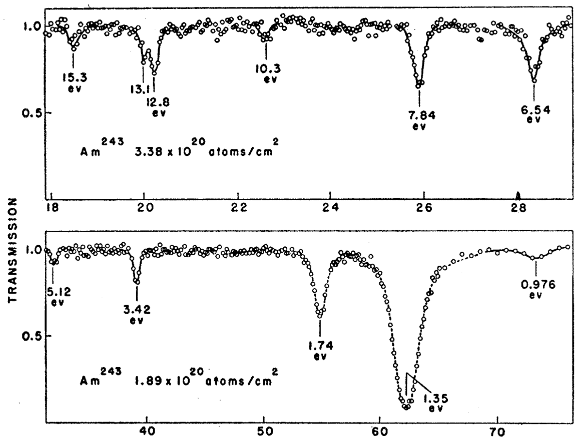

FIG. 1. The transmission *vs* time-of-flight of neutrons for samples of Am^{243}. The dashed curve drawn through the region which includes the 1.35- and 1.74-ev resonances was computed from the single-level Breit-Wigner equation using the parameters obtained from an area analysis of the data of several independent measurements made under different experimental conditions.

Am^{243}

A portion of the data obtained on Am^{243} is shown in Fig. 1, in which transmission is plotted against the time-of-flight of the neutrons. Eleven resonances were observed and, on the basis of the assumption that the statistical factor g has the value $\frac{1}{2}$, the resonance parameters listed on Table V were obtained by area analysis. It was possible to make a determination of Γ_γ for the first three resonances, although a good measurement was possible only for the resonance at 1.35 ev. A weighted average of the values obtained for Γ_γ for these three resonances was used to deduce the parameters from the areas obtained for the remaining nine resonances. Although application of "wing"[16] analysis to the data on the resonance at 1.35 ev, does not provide an ideal case for the method, it does yield a value of $\sigma_0\Gamma^2=37.4\pm4$ which is in fair agreement with the value $\sigma_0\Gamma^2=34.5\pm3.3$ based on the results of the area analysis. Since this resonance is nearly resolved, a shape analysis can be performed to obtain the resonance parameters. The value of $\sigma_0\Gamma^2$ obtained from these parameters is 35.7 ± 2. Furthermore, an examination of Fig. 1 shows that the shape computed from the resonance parameters derived from the area analysis of the resonances at 1.35 and 1.74 ev is in excellent agreement with the measured transmission.

TABLE V. The resonance parameters of the resonance observed in Am^{243}. Values of Γ_γ were obtained for the first three resonances and a weighted average of these, namely $\Gamma_\gamma=(42.0\pm3)\times10^{-3}$ ev, was used in the analysis of the remaining eight resonances.

E_0 (ev)	σ_0 (b)	Γ (10^{-3} ev)	Γ_γ (10^{-3} ev)	Γ_n (10^{-3} ev)
0.976	295± 63	78 ±29	78 ±29	0.017±0.003
1.353	18000±2700	43.8± 3.3	43 ± 3.3	0.82 ±0.08
1.74	4360± 950	30.5± 8.1	30.2± 8.1	0.18 ±0.01
3.42	1900± 90	42.2± 3	42 ± 3	0.21 ±0.01
5.12	1350± 115	42.2± 3	42 ± 3	0.22 ±0.02
6.54	3860± 190	42.8± 3	42 ± 3	0.83 ±0.04
7.84	3580± 200	42.9± 3	42 ± 3	0.93 ±0.05
10.3	755± 165	42.2± 3	42 ± 3	0.23 ±0.05
12.8	3450± 475	43.5± 3	42 ± 3	1.50 ±0.2
13.1	1860± 405	42.8± 3	42 ± 3	0.80 ±0.2
15.3	1250± 570	42.6± 3	42 ± 3	0.63 ±0.3

Since the americium sample was very pure, measurements of the total cross section in the thermal energy region was made. The variation of this cross section with neutron energy is shown in Fig. 2, from which it is evident that the cross section is not proportional to $1/v$. The total cross section for neutrons with a speed of 2200 m/sec is 190 b, of which 7.2 b can be attributed to scattering and 42.3 b to resonant capture due to the eleven epithermal resonances listed in Table V. The remainder of 140.5 b must be assigned to some other source. A resonance, which can be postulated at 0.0107 ev with $\sigma_0=340$ b and $\Gamma=0.034$ ev, will account for this difference and also satisfy the shape of the curve of capture cross section *vs* energy throughout the thermal region. The reduced neutron width $\Gamma_n{}^0$ for this resonance is only 10^{-6} ev. This is not the smallest reduced neutron width to have been observed; but on the basis of the Porter-Thomas distribution and the measured reduced neutron width for Am^{243}, only 1.6% of the resonances might be expected to be smaller. In view of this, although the resonance does not correspond in energy to any that are known to exist and in spite of the purity of the sample, the possibility exists that this resonance may be caused by some impurity.

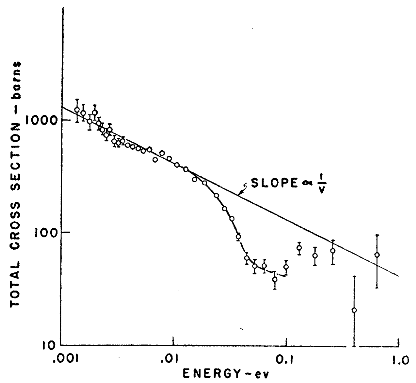

FIG. 2. The total cross section of Am^{243} as a function of neutron energy in the thermal energy region.

The present results on Am^{243} are in considerably less satisfactory agreement with those of BLM than were the Pu^{242} results. Their values of the capture cross section at 2200 m/sec and of the resonance capture integral are 73.6 ± 1.8 b and 2290 ± 50 b, respectively, while those of the present work are 183.8 ± 8 b and 1470 ± 135 b, respectively. The consideration of these radically different results brings up several facts which

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[16] R. R. Palmer and L. M. Bollinger, Phys. Rev. **102**, 228 (1956).

should be mentioned. If the total cross section of Am^{243} actually does vary as shown in Fig. 2, the assumption made by BLM that the capture cross section is proportional to $1/v$ is not correct and the value of 73.6 b given by them is not the capture cross section σ_γ, but $f\sigma_\gamma$, where f is the usual correction factor[17] which can be obtained only by means of an integration over the actual cross section. A computation of this correction factor for Am^{243} shows that it is not large enough to account for the discrepancy. Of course, if the cross-sectional variation observed in the present work is due entirely to an impurity in this sample, the true capture cross section may be proportional to $1/v$ and the value of BLM should be correct. Presumably then, our value should be made up of contributions from the epithermal resonances observed in the present experiment, and from observed levels below the binding energy of the compound nucleus. On the basis of the parameters listed in Table V, the contribution from the positive energy levels is 42.3±5 b, not nearly large enough to account for the value of BLM. The contributions of the resonance at 1.35 ev to the thermal capture cross section and to the resonance capture integral make up a major part of these quantities, about 75–80% in fact.

This points up a possible major source of error in the present work. Because of the limited range of sample thicknesses available and because of the limited time during which the measurements could be made, no data on a truly thin sample were obtained for this resonance. The consequence of this is that the results of the analysis are rather strongly dependent on a fairly accurate determination of the background. However, because of very favorable past experience with the method that was used to determine the background for these measurements, a high degree of confidence is placed in the values obtained. Nevertheless, the errors associated with the parameters of the resonance at 1.35 ev and the errors quoted for the thermal capture cross section and the resonance capture integral include an uncertainty based on an error of about 15% in the background. The other results are quite insensitive to such variations in the background.

TABLE VI. A comparison between the measured value of the radiation width Γ_γ and values obtained from the empirical relations of Cameron[a] and Stolovy and Harvey.[b]

Nuclide	Γ_γ(meas.) (10^{-3} ev)	Γ_γ(Cam.) (10^{-3} ev)	Γ_γ(S-H) (10^{-3} ev)
Pu^{240}	32.1±3	30	39
Pu^{242}	25.1±2.6	21	30
Am^{243}	43 ±3	33	31

[a] See reference 19.
[b] See reference 20.

SUMMARY

The nucleonic compositions of the nuclides under consideration are not such that strong shell effects would be expected. In fact, there is no reason to expect that the average properties of these nuclides would be much different from the other heavy nuclides with nearly as large a mass. The results of the present measurements confirm this.

Values of the strength function $\bar{\Gamma}_n^0/D$ for nuclides in this mass region are all about 10^{-4}. The values for Pu^{242} and Am^{243} are $(3.7_{-2.5}^{+3.7})\times10^{-4}$ and $(0.84_{-0.22}^{+0.28})\times10^{-4}$, respectively, neither of which indicate a large departure from the value of 10^{-4} which is predicted by both the black[4] nucleus and optical[5] models of the nucleus.

The total radiation width Γ_γ has also been the object of much investigation[18] and it might be felt that for nuclides in this mass region, far from closed shells, accurate predictions of this quantity might be inferred from the rather large amount of empirical evidence available. Cameron[19] and Stolovy and Harvey[20] have recently published semiempirical relationships between the radiation width and the nuclear binding energy and level spacing which are based on this evidence. The results of the present measurements are shown in Table VI along with the values obtained from the equations of Cameron and of Stolovy and Harvey. These results present little evidence upon which a choice between the two can be based, but they are consistent with the statement of Stolovy and Harvey that Cameron's relation is slightly better for $A>100$.

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[17] D. J. Hughes, *Pile Neutron Research* (Addison-Wesley Press, Cambridge, Massachusetts, 1953).

[18] J. S. Levin and D. J. Hughes, Phys. Rev. **101**, 1328 (1956).

[19] A. G. W. Cameron, Can. J. Phys. **35**, 666 (1957).

[20] A. Stolovy and J. A. Harvey, Phys. Rev. **108**, 353 (1957).