Fission Mass Yield Dependence on Angular Momentum*

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(Received 6 July 1964; revised manuscript received 12 October 1964)

Fission mass yields from the compound nucleus U^{236} have been measured for 19–29-MeV alpha-particle-induced fission of Th^{232} and 9–18-MeV neutron-induced fission of U^{235} with particular attention to the yields of symmetric fission. Decreases in the symmetric fission yield are seen at the 25-MeV onset of $(\alpha,2nf)$ fission and the corresponding 14-MeV onset of $(n,2n'f)$ fission. Above these energies, the symmetric fission yields are approximately 13% greater for alpha-particle-induced fission than for neutron-induced fission at the same excitation energies. This increase is partially explained by the fission-chance ratio of higher energy fissions relative to fissions of the most de-excited nuclei being greater for alpha particles than for neutrons. Angular-momentum-dependent cross sections for these fissions were calculated, and the use of reasonable values of level densities with a rigid moment of inertia resulted in a calculated effect of about one-half this observed effect. The difference remaining is possibly explained by fissions following direct interactions. Mass yields of 9.1–18.1-MeV neutron-induced fission of Th^{232} and 4.7–18.1-MeV neutron-induced fission of U^{238} were also measured. Some indication of a small central peak in the mass distribution was observed in the yields from U^{236} compound-nucleus fission, but not from Th^{233} compound-nucleus fission.

I. INTRODUCTION

AMONG the many complexities of the fission process, the predominantly asymmetric division observed in heavy nucleus fission plays a central role in attempts to understand fission. Several nuclear models have been used. The intuitively satisfying liquid-drop model of fission indicates that the saddle-point configuration, the low point of the potential surface in the configuration space of nuclear deformation, is symmetric.[1] Thus, if the mass distribution of the fission products is established at the saddle-point configuration, then the liquid-drop model suggests symmetric fission to be dominant. The statistical model[2] attempts to explain asymmetric fission by the density of states of the nascent fission fragments, and thus emphasizes the scission configuration of the fissioning nuclei. Measured excitation energies of fragments[3] from various asymmetries of fission are inconsistent with this explanation of fission asymmetry. The shell-model explanation[4] involves arguments about the deformability of nascent fragments and the effect upon the division of fission energy into Coulomb energy, deformation energy, and excitation energy. The energy required for deformation is particularly large near closed-shell nuclei, which for heavy element fission are on the shoulders of mass yield curves. Thus, shells are probably associated with fission asymmetry, but in a somewhat complicated manner. The unified model has been applied to fission.[5] For low-energy fission channels, the nuclear shapes of the saddle-point collective levels are identified by the spin and parity. Thus, in this energy region below the approximately 2.6 MeV required to break a nucleon pair,[6] fission through odd parity states is expected to result in smaller symmetric fission yield than through even parity states.[5]

In this paper we principally consider the effects of angular momentum on fission asymmetry for the compound nucleus U^{236} formed by $n+U^{235}$ and $\alpha+Th^{232}$ at the same excitation energies between 16 and 25 MeV. We find only a small (about 13%) increase of symmetric fission yield with angular momentum. In contrast, increases apparently attributable to angular momentum have been found to be many times this large for $\leqq 0.2$-MeV neutron-induced fission.[7,8] For the experimental conditions of these present and previous measurements, changes in the symmetric fission yields result from one or more of the following causes: (1) changes in parity, (2) differences in moments of inertia, (3) fission-chance split, and (4) fraction of direct-interaction fissions. The first two involve the shape of the nuclei undergoing fission, and the last two involve the distributions of excitation energies at which fissions occur.

The large increase in symmetric relative to asymmetric fission for neutron energy increase from 65 to 200 keV observed by Cuninghame *et al.*[7] could be a result of the increased number of even parity fissions induced by *p*-wave neutrons rather than the small in-

* Work performed under the auspices of the U. S. Atomic Energy Commission.
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[2] P. Fong, Phys. Rev. **102**, 434 (1956).
[3] W. E. Stein and S. L. Whetstone, Jr., Phys. Rev. **110**, 476 (1958); and V. F. Apalin, Yu. N. Gritsyuk, I. E. Kutikov, V. I. Lebedev, and L. A. Mikaélyan, Zh. Eksperim. i Teor. Fiz. **43**, 329 (1962) [English transl.: Soviet Phys.—JETP **16**, 235 (1963)].
[4] L. Meitner, Arkiv Fysik **4**, 383 (1951); W. Brunner and H. Paul, Ann. Phys. (Paris) **8**, 146 (1961); J. Terrell, Phys. Rev. **127**, 880 (1962); R. Vandenbosch, Nucl. Phys. **46**, 129 (1963); and J. C. D. Milton, Institute for Atomic Energy, Kjeller, Report KR-64, 1963 (unpublished).
[5] Aa. Bohr, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 151; and L. Wilets, in *Proceedings of the Rehovoth Conference on Nuclear Structure, 1957* (North-Holland Publishing Company, Amsterdam, 1958), p. 122.
[6] H. C. Britt, R. H. Stokes, W. R. Gibbs, and J. J. Griffin, Phys. Rev. Letters **11**, 343 (1963).
[7] J. G. Cuninghame, G. P. Kitt, and E. R. Rae, Nucl. Phys. **27**, 154 (1961).
[8] G. A. Cowan, B. P. Bayhurst, and R. J. Prestwood, Phys. Rev. **130**, 2380 (1963); and K. T. Faler and R. L. Tromp, Phys. Rev. **131**, 1746 (1963).

creases in energy and average angular momentum. This parity effect, listed as (1) above, is expected from the unified model.[5] For electron-volt neutrons, experimental energy resolutions and the separations of fission levels have enabled studies of isolated levels of fission.[8] Here, changes in the symmetric fission yields are again large, but are difficult to coordinate with theory. For our large bombarding energies, parity effects are not expected because the fission channels are at energies allowing intrinsic, as well as collective, excitations and because populations of the two parities are nearly equal for the large number of orbital angular momenta l values of both alpha-particle and neutron irradiations.

The Pik-Pichak[9] explanation of the angular momentum effect on fission probability can be extended to treat the dependence of fission asymmetry on the angular momentum causes (2) and (3) above. This Pik-Pichak explanation for fission probabilities is based on statistical-model densities for fission channels at the saddle point and for levels of the final nucleus resulting from the competing process of neutron emission. The width $\Gamma_{x,f}$ for fission relative to the width $\Gamma_{x,n}$ for neutron emission is, in the simplest form,

$$\frac{\Gamma_{x,f}}{\Gamma_{x,n}} \approx p \exp\left[\frac{\hbar^2 J_x(J_x+1)}{2T}\left(\frac{1}{\mathcal{I}_n}-\frac{1}{\mathcal{I}_\perp}\right)\right], \qquad (1)$$

where p is the ratio of fission to neutron width for zero angular momentum, T is a nuclear temperature assumed in Eq. (1) to be the same for both statistical distributions, and $\mathcal{I}_\perp$ and $\mathcal{I}_n$ are, respectively, the moment of inertia of the saddle-point nucleus with respect to an axis perpendicular to the symmetry axis and the moment of inertia of the nucleus resulting from neutron emission. Since the probability for other de-excitations of the compound nuclei by gamma rays and charged particles is generally neglected, the partial fission cross sections are related to the fission probability $\gamma_x^{(1)} \equiv \Gamma_{x,f}^{(1)}/[\Gamma_{x,f}^{(1)}+\Gamma_{x,n}^{(1)}]$ [analogous definitions apply to $\gamma_x^{(2)}$ and $\gamma_x^{(3)}$] and to the compound-nucleus cross section $\sigma_{x,c}$ for particle x by

$$\sigma_{x,f}^{(1)} = \gamma_x^{(1)} \sigma_{x,c}, \qquad (2a)$$

$$\sigma_{x,f}^{(2)} = \gamma_x^{(2)} [\sigma_{x,c} - \sigma_{x,f}^{(1)}], \qquad (2b)$$

$$\sigma_{x,f}^{(3)} = \gamma_x^{(3)} [\sigma_{x,c} - \sigma_{x,f}^{(1)} - \sigma_{x,f}^{(2)}], \qquad (2c)$$

where each γ_x is evaluated for the nucleus, excitation energy, and angular momentum applicable at that stage of de-excitation by neutron emission. Throughout the paper, the following identifications of indices apply: The first of a subscript pair is the input channel of α, n, or x representing alpha particles, neutrons, or either, respectively; the second of the pair is c, n, f, a, or s, which represent, respectively, compound-nucleus formation, neutron emission, fission, asymmetric fission product, or symmetric fission product. (Subscript pairs on moments of inertia $\mathcal{I}$ are an exception in representing first the axis and then the fission product.) Superscripts (1), (2), and (3) represent, respectively, first-chance (x,f) fissions, second-chance (x,nf) fissions, and third-chance $(x,2nf)$ fissions for the three stages of de-excitation considered. No superscript denotes the combination of these available stages.

An increase of fission cross section $\sigma_f^{(1)}$ with angular momentum J predicted by Eqs. (1) and (2a) has been observed by Gilmore *et al.*[10] for cases of heavy-ion induced fission with large differences in orbital angular momenta.

The possibility of different moments of inertia $\mathcal{I}_{\perp,a}$ and $\mathcal{I}_{\perp,s}$ from different saddle-point configurations leading to asymmetric and symmetric fission, respectively, listed as (2) above, gives

$$\frac{Y_{x,s}^{(1)}}{Y_{x,a}^{(1)}} \approx \left[\frac{Y_{x,s}^{(1)}}{Y_{x,a}^{(1)}}\right]_{J_x=0} \times \exp\left[\frac{\hbar^2 J_x(J_x+1)}{2T}\left(\frac{1}{\mathcal{I}_{\perp,a}}-\frac{1}{\mathcal{I}_{\perp,s}}\right)\right], \qquad (3)$$

from Eq. (1), where the yields Y are taken as the percentage of fissions resulting in the given nucleus as a fission product.[11] Corresponding equations apply for $Y_{x,s}^{(2)}/Y_{x,a}^{(2)}$ of (x,nf) fissions and $Y_{x,s}^{(3)}/Y_{x,a}^{(3)}$ of $(x,2nf)$ fissions.

The meager evidence existing about the relative magnitudes of $\mathcal{I}_{\perp,a}$ and $\mathcal{I}_{\perp,s}$ predicts very little change of symmetric fission yield with angular momentum from this different moment-of-inertia cause (2), as is shown by the following qualitative arguments. The kinetic energies of fragments from fission measured[12] in the energy range of the present measurements were about 5% lower for symmetric fission than for the high-yield masses of asymmetric fission. Moments of inertia increase roughly as the square of the saddle-point elongation, and the kinetic energy of fragments generally varies inversely as scission elongation.[13] Thus, the moment of inertia should vary roughly as the inverse of the square of the kinetic energies of fragments for various fission modes, provided the scission-point elongations, which are assumed to determine the kinetic energies, are proportional to the saddle-point elongations involved in Eq. (3). Thus, we use $\mathcal{I}_{\perp,s}/\mathcal{I}_{\perp,a} \approx 1.1$. We also use $T \approx 0.8$ MeV, $\mathcal{I}_n \approx 0.4\mathcal{I}_\perp$,[1] a rigid moment of inertia value of

[9] G. A. Pik-Pichak, Zh. Eksperim. i Teor. Fiz. **36**, 961 (1959) [English transl.: Soviet Phys.—JETP **9**, 679 (1959)].

[10] J. Gilmore, S. G. Thompson, and I. Perlman, Phys. Rev. **128**, 2276 (1962).

[11] G. P. Ford and R. B. Leachman, Bull. Am. Phys. Soc. **6**, 376 (1961).

[12] H. C. Britt and S. L. Whetstone, Jr., Phys. Rev. **133**, B603 (1964); and S. L. Whetstone, Jr., Phys. Rev. **133**, B613 (1964).

[13] J. Terrell [Phys. Rev. **113**, 527 (1959)] has shown this general property for the average kinetic energy of fission as a function of the mass number A of the compound nucleus. However, such a variation with the mass number of fragments from symmetric and asymmetric fission of a given compound nucleus has not been established.

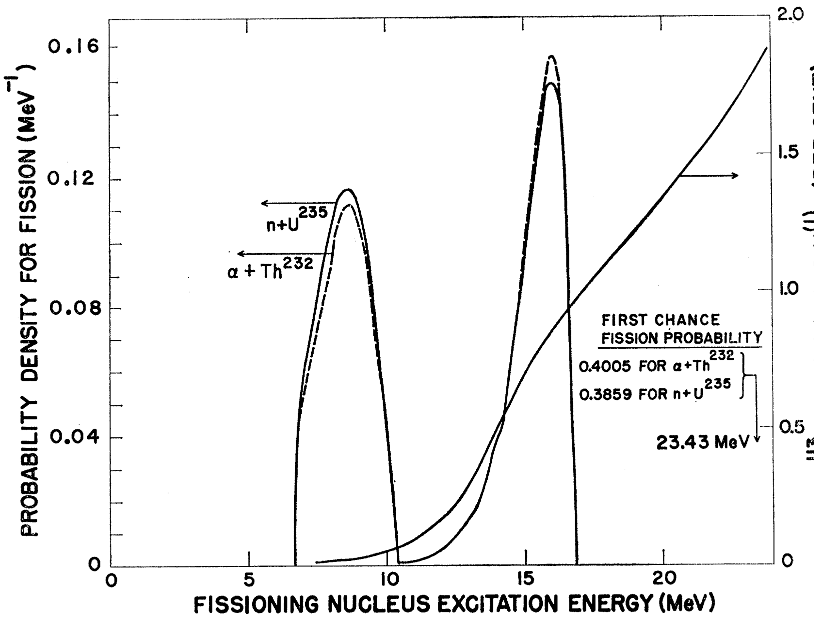

FIG. 1. Calculations for $Th^{232}(\alpha,f)$ and $U^{235}(n,f)$ showing the probabilities of first-chance fission and also the probability densities of second- and third-chance fissions as a function of the excitation energy of the nucleus undergoing fission. The initial excitation energy of the compound nucleus U^{236} is 23.43 MeV in this example. Second-chance fissions occur at about 16-MeV excitation energy and third-chance fissions at about 8-MeV excitation energy. To determine the yield of a symmetric fission product, it is necessary to weight the yield as a function of excitation energy with the probabilities of fission at these excitation energies. An example of such a symmetric fission yield is illustrated by the yield of Ag^{113} in the figure. This yield was derived from the unfolding process of Sec. IIIB, and the fission probabilities and probability densities were calculated by the Markov chain method of Sec. IIIB with the parameters $N=3$, $a_n=23.4$ MeV^{-1} of Table II.

$\hbar^2/2\mathcal{I}_n \approx 3$ keV, $\langle l_\alpha(l_\alpha+1)\rangle_{av}=83$ for 28.5-MeV alpha particles[14] and $\langle l_n(l_n+1)\rangle_{av}=43$ for the equivalent 17-MeV neutrons.[15] [We use $l_x(l_x+1)\approx J_x(J_x+1)$ by neglecting the particle and target spins compared to the orbital angular momentum l_x.] From these values, $Y_{\alpha,s}^{(1)}/Y_{\alpha,a}^{(1)}$ for alpha-particle-induced fission is 1% greater than $Y_{n,s}^{(1)}/Y_{n,a}^{(1)}$ for neutron-induced fission. Since $l(l+1)$ does not change significantly with prefission neutron emission, this result is expected to apply also for second- and third-chance fissions and thus for the observed ratios $Y_{\alpha,s}/Y_{\alpha,a}$ and $Y_{n,s}/Y_{n,a}$ at all energies inducing fission if the relative values of $\mathcal{I}_{\perp,s}$ and $\mathcal{I}_{\perp,a}$ do not change. This small effect from possible differences in moments of inertia is neglected in the following considerations.

The third cause[16] for a change of fission asymmetry with angular momentum is termed "fission-chance split" in (3) of the listing above and is illustrated in Fig. 1. When third-chance fission is energetically possible, the cross-section ratio $[\sigma_{x,f}^{(1)}+\sigma_{x,f}^{(2)}]/\sigma_{x,f}^{(3)}$ is expected from Eqs. (1) and (2) to be greater for alpha-particle-induced fission than for neutron-induced fission. The symmetric fission yield $Y_{x,s}^{(1)}$, illustrated by the Ag^{113} yield in Fig. 1, is relatively large for fissions produced at the 23.43-MeV and approximately 16-MeV excitation energies of first- and second-chance fissions, respectively. However, because of the exponentially decreasing yields[17–19] below about 15-MeV excitation energy, the symmetric fission yield of third-chance fission induced at approximately 8 MeV is relatively very low. The combination of partial cross sections and yields into the combined yields

$$Y_{\alpha,s}=[Y_{\alpha,s}^{(1)}\sigma_{\alpha,f}^{(1)}+\langle Y_{\alpha,s}^{(2)}\rangle_{av}\sigma_{\alpha,f}^{(2)}+\langle Y_{\alpha,s}^{(3)}\rangle_{av}\sigma_{\alpha,f}^{(3)}]/\sigma_{\alpha,f}, \quad (4a)$$

$$Y_{n,s}=[Y_{n,s}^{(1)}\sigma_{n,f}^{(1)}+\langle Y_{n,s}^{(2)}\rangle_{av}\sigma_{n,f}^{(2)}+\langle Y_{n,s}^{(3)}\rangle_{av}\sigma_{n,f}^{(3)}]/\sigma_{n,f}, \quad (4b)$$

thus causes $Y_{\alpha,s}/Y_{n,s}$ to be greater than unity. This is seen by approximating the yields $Y_{x,s}^{(2)}$ and $Y_{x,s}^{(3)}$ by $Y_{x,s}^{(1)}$ so that all yields in Eqs. (4) are replaced by the energy-dependent $Y_{n,s}^{(1)}$. In Eqs. (4), averages are over the range of excitation energies at which the indicated type of fissions occur after neutron emission.

Relative values of the partial cross sections $\sigma_{x,f}^{(1)}$, etc., in Eqs. (2) and (4) have previously been considered[20] without angular momentum effects, but in this paper we consider these partial cross sections with angular momentum effects[21] in considerable detail to

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[15] J. R. Beyster, R. G. Schrandt, M. Walt, and E. W. Salmi, Los Alamos Scientific Laboratory Report, LA-2099, 1956 (unpublished). Unpublished calculations with extrapolations of these optical-model parameters have been made by L. Blumberg and J. Matthews.

[16] We are grateful to R. Vandenbosch for this suggestion (private communication).

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[18] T. T. Sugihara, P. J. Drevinsky, E. J. Troianello, and J. M. Alexander, Phys. Rev. **108**, 1264 (1957); and R. Gunnink and J. W. Cobble, Phys. Rev. **115**, 1247 (1959).

[19] H. G. Hicks, H. B. Levy, W. E. Nervik, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., Phys. Rev. **128**, 700 (1962).

[20] R. B. Leachman, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), Vol. 15, p. 229; G. Rudstam and A. C. Pappas, Nucl. Phys. **22**, 468 (1961); V. M. Pankratov and V. M. Strutinskii, At. Energ. (USSR) **14**, 171 (1963) [English transl.: Soviet J. At. Energy **14**, 161 (1963)].

[21] J. R. Huizenga and R. Vandenbosch, in *Nuclear Reactions*, edited by P. M. Endt and P. B. Smith (North-Holland Publishing Company, Amsterdam, 1962), Vol. II, p. 42.

calculate relative yields from Eqs. (4). The increase of the symmetric fission yield in our energy range for alpha-particle-induced fission relative to neutron-induced fission is calculated in Sec. III to be less than 10%, and so the considerations of Eqs. (1), (2), and (3) all indicate the effect from fission-chance splits is not significantly more than experimental accuracies in fission-yield measurements.

The cause (4) above, the fraction of direct-interaction fissions, results from the possibility that more direct interactions followed by low-energy induced fission (having low symmetric fission yields), occur for neutron irradiations than for alpha-particle irradiations.[22] For 14-MeV neutron irradiations of bismuth, angular correlation measurements[23] of emitted neutrons indicated that at most 80% of the $(n,2n)$ reactions are compound nuclei, and angular distributions[24] indicated that direct interactions with 4–8-MeV neutrons emitted are roughly 3% of the reaction cross section. Such direct (n,n') reactions on U^{235} with $\leqq$8-MeV neutrons emitted could be followed by fissions, which would be at low excitation energy. If the cross section for (n,α) direct interactions followed by fission is negligible, then a few percent lower symmetric fission yield for neutron irradiation can be expected from this cause.

In the course of the present measurements, some mass yields from fission of Th^{232} by 9.1–18.1-MeV neutrons and from fission of U^{238} by 4.7–18.1-MeV neutrons were also measured.

II. EXPERIMENT

To provide the desired mass resolution in the symmetric-fission yield measurements, radiochemical analyses were used. Yields of several nuclei, including both symmetric and asymmetric fission products, were measured for both $Th^{232}(\alpha,f)$ and $U^{235}(n,f)$.

A. Irradiations

Alpha-particle irradiations were made with 1.3-cm diam beams from the variable energy cyclotron typically upon 5-mg/cm² evaporated deposits of Th^{232} on 19-mg/cm² gold backings with a similar cover foil of gold. At some of the highest energies, 0.2-mg/cm² Th^{232} deposits were used without a cover foil before the deposit (to avoid degrading the alpha-particle energy); corrections were made for center-of-mass effects that caused 0.8% more symmetric-fission products than heavy fragments to be caught in the $2\pi-$sr catcher. Beam currents up to 6 μA were used. In duplicate runs with greatly different currents, no significant yield changes attributable to target heating were detected. Irradiations were typically 4 h in duration. For the lowest energy 18.9-MeV alpha particles, a large quantity of Th^{232} external to the beam was analyzed to estimate the number of fissions in the target from neutrons produced by (α,n) reactions; less than 0.07% were from this cause. This background is less at higher energies. Measurement of the alpha-particle energies was by the stopping-foil technique in Sec. IIIA of the previous paper[25]; corrections were made for the energy loss in the first gold catching foil and in Th^{232}. For all runs, ranges in emulsions were measured to confirm the energy determination and to estimate the spread in energy. Including the energy loss in Th^{232}, the energy spreads were $\pm$0.17 MeV from the median energy.

Irradiations providing neutrons with the following energies and energy spreads were: the $H^2(d,n)$ reaction with a Van de Graaff neutron source of 2 cm length at 3 atm pressure behind cooled double 1.1-mg/cm² nickel windows for 4.7$\pm$0.14-MeV neutrons; the $H^2(d,n)$ reaction with a cyclotron neutron source of 2 cm length at 15 atm pressure behind cooled 10- and 23-mg/cm² molybdenum windows and a Van de Graaff neutron source of 3 cm length at 12 atm pressure behind a single 10-mg/cm² molybdenum window, both for 9.1$\pm$0.3-MeV neutrons; the $T(d,n)$ reaction from a tritium-impregnated-metal neutron source on a Cockcroft-Walton accelerator for 13.4$\pm$0.17-, 14.1$\pm$0.17-, and 14.9$\pm$0.25-MeV neutrons; the $T(d,n)$ reaction with a Van de Graaff neutron source of 1 cm length at 5 atm pressure behind a single 10-mg/cm² molydenum window for 16.3- and 17.2-MeV neutrons; and the last, except for 3 cm length, for 18.1-MeV neutrons. Irradiations were nominally 4 h; for gaseous sources the beam currents were between 3 and 7 μA. Neutron energies and their spreads were calculated by the method in Sec. IIIA of the preceding paper.[25] For 16.3- and 17.2-MeV neutron conditions the energy losses in the window were important, so the energies were confirmed by emulsion ranges of proton recoils for both energies and by ranges of alpha particles from $C^{12}(n,n'3\alpha)$ for 16.3-MeV. The angles subtended by the fissionable materials with respect to the beam spot established the energy and most of its spread for the Cockcroft-Walton irradiations.

For these neutron irradiations, the fissionable targets were disks of metal plated with 0.003-cm thick nickel. The U^{235} was 93.4% U^{235}, 1.1% U^{234}, and 5.5% U^{238}; the U^{238} was 99.9% U^{238} and 0.1% U^{235}. No corrections were made for fissions of minority isotopes. The thicknesses of the disks were nominally 0.14 cm for Th^{232}, 0.08 cm for U^{235}, and 0.05 cm for U^{238}. The diameters were all 1.2 cm.

Some fissions were induced by neutrons lower in energy than the nominal energy. Corrections were made in fission yields for the following: (1) For U^{235} with the 9.1-MeV conditions, measurements with evacuated neutron sources showed 19% of the asymmetric fissions and

[22] We are grateful to J. S. Gilmore for this suggestion (private communication).

[23] A. Ádám, P. Hraskó, G. Pálla, and P. Quittner, Nucl. Phys. **49**, 489 (1963).

[24] L. Rosen and L. Stewart, Phys. Rev. **107**, 824 (1957) and private communication of further details of these data.

[25] R. B. Leachman and L. Blumberg, Phys. Rev. **137**, B814 (1965).

3% of the symmetric fissions for the cyclotron conditions and 11 and 3%, respectively, for Van de Graaff conditions to result from neutrons produced by deuteron irradiation of the windows and gold stoppers of the neutron sources; for cyclotron conditions these respective results were 3.6 and 0.8% for Th^{232} and 7.1 and 4.5% for U^{238}. (2) For the 9.1-MeV condition, a calculated 6% of the $U^{235}(n,f)$ fissions was induced by low-energy neutrons from the $D(d,np)$ reaction[26]; for $Th^{232}(n,f)$ and $U^{238}(n,f)$, thresholds reduced this to an estimated 3%. (3) Depending upon the number of disks stacked in each irradiation, the percentages of fissions induced by low-energy neutrons emitted by other fissions within the fissionable disks were calculated[27] for U^{235} to be between 3 and 7% and for both Th^{232} and U^{238} to be between 1 and 3%. (A repeat of the 13.4-, 14.1-, 14.9-, and 18.1-MeV irradiations but with a 0.6-cm thick U^{235} disk having a 0.8-cm diam hole, for which geometry this calculated effect is only 2%, confirmed the magnitude of this type of correction.) (4) For neutrons from the Cockcroft-Walton, a calculated 3% of the fissions was induced by neutrons degraded in energy by structural materials in the neutron source.

At the other energies used, fissions from effect (1) were measured to be less than 1%, and neutrons from effect (2) were not energetically possible. Since the fission yields from effects (2) through (4) were not readily measurable, for simplicity these fissions were assumed all to be induced by neutrons degraded to 1 MeV, which is roughly the most probable energy measured[26] for (2) and calculated[27] for (3). Estimated yields for 1-MeV neutron-induced fission were used for these corrections. No uncertainties in these corrections were included in the data, but only for the cases of 9.1-MeV neutrons would these additional uncertainties be large compared to the quoted uncertainties. Measurements with and without cadmium wrappings around the U^{235} disks confirmed that the number of thermal-neutron-induced fissions was always negligible.

B. Chemistry and Counting

For the U^{235} irradiations, the nickel-coated disks of metal were dissolved in concentrated hydrochloric acid with a little nitric acid. Foils of thorium-oxide on gold were dissolved in the same way except that a little ammonium fluorosilicate was added. In every case, the nickel or gold was completely dissolved and the nitric acid was boiled away.

Suitable size aliquots of the solution were taken for the various elements. Usually silver, palladium, europium, and cesium were done on the same aliquot. Carriers for each were added and the solution boiled for several minutes to encourage exchange. The separation of these elements was then based on the insolubilities of Ag_2S, PdS, AgCl, and $Eu(OH)_3$. The other elements were usually done on separate aliquots. Duplicates were generally analyzed for each element for each irradiation. Standard radiochemical procedures[28] were used for decontamination. The final products were mounted for counting as described by Bayhurst and Prestwood[29] except that, to increase the counting rate, a piece of 0.03-cm platinum was used to back all samples except molybdenum and cadmium. The effect of the platinum was determined for each mass number in a separate experiment.

Corrections for the platinum backing were applied to counting efficiencies estimated by the method of Bayhurst and Prestwood.[29] Published decay schemes[30] were used. It should be noted that the estimates of counting efficiencies depend strongly on the decay scheme. (For example, the ratio of the estimates for Ba^{140} to La^{140} counting efficiencies was 1.29, but the actual ratio of efficiencies was measured to be 1.42.) The results of the counting efficiency estimates are: Zr^{97}, 0.470; 1-min Nb^{97}, 0.148; 74-min Nb^{97}, 0.438; Pd^{109}, 0.446; Ag^{111}, 0.404; Pd^{112}, 0.089; Ag^{112}, 0.540; Ag^{113}, 0.496; 54-h Cd^{115} with 4.4-h I^{115}, 0.572; Cs^{136}, 0.148; Ba^{139}, 0.495; Eu^{156}, 0.392. For Mo^{99}, 0.388 was used.[31] Each sample was counted many times over an appropriate time interval and the decay curve resolved by the method of least squares with the techniques of Moore *et al.*[32] In the least-squares calculation, each count rate and each background was weighted by the reciprocal of the estimate of its variance. Corrections were made for decays during the sometimes variable irradiations.

The number of fissions were based on either Zr^{97} assays or the average of Zr^{97} and Ba^{139} assays when Ba^{139} results were available. It was assumed for all compound nuclei that these two fission products have the same energy-dependent percentage yields as given by Hicks *et al.*[19] for Mo^{99}. Yields of Zr^{97} and Ba^{139} from our measurements generally differed by less than 4%, twice by 9%, and once (22.3-MeV alpha-particle irradiation of Th^{232}) by 20%.

During the course of the experiment, occasional unirradiated samples were analyzed for most elements to obtain the activity blanks. The errors in these activities were assumed to be absolute rather than percentage errors and were considered to be those of the population except for cesium and barium, which were

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[27] Kindly calculated by W. J. Worlton with the Los Alamos TDC reactor code.

[28] Los Alamos Scientific Laboratory Report LA-1721, edited by J. Kleinberg, 2nd ed., 1958 (unpublished).

[29] B. P. Bayhurst and R. J. Prestwood, Nucleonics **17**, 82 (1959).

[30] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences-National Research Council, Washington 25, D. C.), NRC 58-9-42, NRC 58-1-57, NRC 58-3-62, NRC 58-5-60, NRC 58-10-62, NRC 58-8-50, NRC 58-10-66, NRC 59-1-83, NRC 59-1-85, NRC 59-5-59.

[31] Measured by J. S. Gilmore by $4\pi-$sr beta counting (private communication).

[32] R. H. Moore and R. K. Zeigler, Los Alamos Scientific Laboratory Report, LA-2367, 1959 (unpublished) and Paul McWilliams, LA-2367 *addenda*, 1962.

surprisingly small. For cesium and barium the standard deviation was taken to be 1.5 counts per minute. The gross activity observed for an element was assigned to each isotope of that element. These activity blanks and their standard deviations in counts per minute were: Zr, 1.47±1.68; Pd, 0.66±0.49; Ag, 2.61±1.86; Cs, 0±1.5; Ba, 2.38±1.5; and Eu, 2.96±0.93. Compared to activities from irradiated samples, these activities were almost always less than 2% of the observed activities, except for the isotopes Ag^{111} for a few irradiations and Eu^{156} for all irradiations. The following errors arising from nonreproducibility of the chemical procedures were determined from six irradiations of U^{235} with thermal neutrons: Zr^{97}, 0.87%; Pd^{109}, 0.47%; Ag^{111}, 2.12%; Pd^{112}, negligible; Ag^{113}, 1.21%; Cs^{136}, 4.09%; Ba^{139}, 0.43%; and Eu^{156}, 1.35%. The error due to the Poisson variance of counting rates (counting statistics) was a by-product of the least-squares calculation. If the weighted sum of squares of the least-squares fit was less than its expectation value, the error was taken as given; otherwise, the error due to the Poisson variance was multiplied by the square root of the weighted sum of squares divided by the expectation of the weighted sum of squares.[33]

The above errors were combined by the usual rules for the propagation of error. In calculations of averages, the error was taken to be the larger of the propagated error or the error based on the variance of the numbers being averaged.

C. Results

Shown in Fig. 2 is the excitation function for the fission cross section $\sigma_{\alpha,f}$ measured for $Th^{232}(\alpha,f)$. Compound-nucleus cross sections of the compound-nucleus U^{236} are less than twice the fission cross sections from neutron irradiations in this energy range[20]; therefore, the disagreements of more than a factor of 2 between measurements and calculations in Fig. 2 indicate inadequacies in both the optical model calculations[14] and square-well calculations.[34] (In the preceding paper[25] this matter is considered in Sec. IVA and its Ref. 17.)

Yields are given in Table I for all the individual fission products measured. Some disagreements exist with earlier measurements of the U^{236} compound nucleus yields[7,19,35–37] and of $Th^{232}(n,f)$ yields[38] and $U^{238}(n,f)$ yields[36,39] from 15-MeV neutron irradiations. These disagreements can possibly be attributed to corrections (Sec. IIA), normalizations, and estimates of counting efficiencies. Enhanced yields for symmetric fission have been observed in triple peaks of mass yield from fission of compound nuclei with smaller mass number.[40] Our smallest mass number case of $Th^{232}(n,f)$ has symmetry at mass 114 after neutron emission, but no enhanced yield of symmetric fission is indicated by the neighboring Ag^{113} yields in Table I. For the next heavier case, mass distributions from $Th^{232}(\alpha,f)$ derived from some measurements of fragment energies[12] also gave no indication of an additional peak for symmetric fission. However, for the compound nucleus U^{236}, our yields of Cd^{115}, which after neutron emission lie near the mass 116 of symmetric fission, give indication in Table I of a slightly enhanced symmetric fission yield just as was indicated by the fragment energies observed by Seki *et al.*[41]

The increases of the measured symmetric fission yields $Y_{\alpha,s}$ from $Th^{232}(\alpha,f)$ and $Y_{n,s}$ from $U^{235}(n,f)$ with

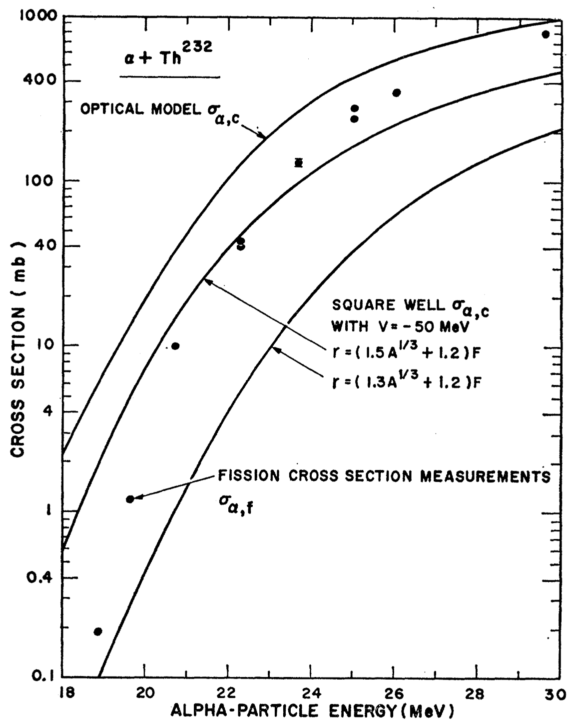

FIG. 2. The measured fission cross section $\sigma_{\alpha,f}$ for $Th^{232}(\alpha,f)$. Uncertainties are smaller than the size of the points unless otherwise indicated. The optical-model calculation of the compound-nucleus cross section $\sigma_{\alpha,c}$ is from Ref. 14; the square-well calculations of $\sigma_{\alpha,c}$ are from Ref. 34.

[33] W. E. Deming, *Statistical Adjustment of Data* (John Wiley & Sons, Inc., New York, 1938), 1st ed., p. 37.

[34] Kindly calculated by M. L. Gursky by the method of J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 319.

[35] A. S. Newton, Phys. Rev. **75**, 17 (1949) and B. M. Foreman, Jr., W. M. Gibson, R. A. Glass, and G. T. Seaborg, Phys. Rev. **116**, 382 (1959).

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[37] S. Katcoff, Nucleonics **18**, 201 (1960) and H. B. Levy, H. G. Hicks, W. E. Nervik, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., Phys. Rev. **124**, 544 (1961).

[38] A. N. Protopopov, G. M. Tolmachev, V. N. Ushatskii, R. V. Venediktova, I. S. Krisiuk, L. P. Rodionova, and G. V. Iakovleva, At. Energ. (USSR) **5**, 130 (1958) [English transl.: Soviet J. At. Energy **5**, 963 (1958)]; V. A. Vlasov, Y. A. Zysin, I. S. Kirin, A. A. Lbov, L. I. Osyaeva, and L. I. Sel'chenkov, in *Soviet Progress in Neutron Physics* (Consultants Bureau, New York, 1963), p. 172.

[39] J. G. Cunninghame, J. Inorg. Nucl. Chem. **5**, 1 (1957); and K. M. Broom, Phys. Rev. **126**, 627 (1962).

[40] H. C. Britt, H. E. Wegner, and J. C. Gursky, Phys. Rev. **129**, 2239 (1963).

[41] M. Seki, A. Katase, M. Sonoda, A. Yoshimura, Ts. Akiyoshi, and S. Yamawaki, Phys. Letters **8**, 263 (1964).

TABLE I. Measured fission yields. Yields are in percent of fissions resulting in the indicated yields. The spreads of energies for the irradiations are indicated.

Reaction	Incident particle energy (MeV)	Excitation energy, E (MeV)	Number of irradiations	Measured fission yields (percent) Pd^{109}	Ag^{111}	Pd^{112}	Ag^{113}	Cd^{115}	Cs^{136}	Eu^{156}
$U^{235}+n$	4.7 ±0.14	11.18	1	0.222 ±0.061	0.129 ±0.019	0.15 ±0.02	0.105 ±0.006			
$Th^{232}+\alpha$	18.9 ±0.17	14.00	1		0.83 ±0.06	0.37 ±0.08	0.25 ±0.09			
$Th^{232}+\alpha$	19.6 ±0.17	14.71	2		0.43 ±0.02[a]		0.37 ±0.03			
$U^{235}+n$	9.1 ±0.3	15.56	4	0.622 ±0.010	0.517 ±0.006	0.494 ±0.007	0.364 ±0.009	0.572 ±0.003[a,c]		0.070 ±0.013[b]
$Th^{232}+\alpha$	20.6 ±0.17	15.68	1	0.60 ±0.03	0.525 ±0.018	0.453 ±0.013	0.369 ±0.012	0.529 ±0.009[c]		
$Th^{232}+\alpha$	20.7 ±0.17	15.75	1	0.652 ±0.019		0.478 ±0.007			0.25 ±0.15[c]	
$Th^{232}+\alpha$	22.3 ±0.17	17.35	2	1.04 ±0.05	0.72 ±0.03	0.76 ±0.03	0.59 ±0.02		0.150 ±0.017	0.056 ±0.004
$Th^{232}+\alpha$	23.3 ±0.17	18.32	1	1.26 ±0.05	1.11 ±0.02	1.004 ±0.010	0.814 ±0.013			0.0627 ±0.0019
$Th^{232}+\alpha$	23.7 ±0.17	18.68	1	1.09 ±0.03	0.91 ±0.03	0.94 ±0.02	0.73 ±0.02			0.135 ±0.04
$U^{235}+n$	13.4 ±0.17	19.84	3	1.09 ±0.04	0.992 ±0.016	0.96 ±0.02	0.720 ±0.017	1.020 ±0.013[a,c]		
$Th^{232}+\alpha$	25.0 ±0.17	20.01	2	1.44 ±0.04	1.08 ±0.3	1.07 ±0.04	0.91 ±0.04		0.184 ±0.008	0.0554 ±0.0009
$U^{235}+n$	14.1 ±0.16	20.53	3	1.22 ±0.06	1.014 ±0.015	0.954 ±0.019	0.748 ±0.009	1.07 ±0.03[a,c]		0.050 ±0.012[a]
$Th^{232}+\alpha$	26.0 ±0.17	20.94	2	1.271 ±0.013	1.05 ±0.02	1.087 ±0.014	0.87 ±0.03			
$U^{235}+n$	14.9 ±0.25	21.34	3	1.06 ±0.03	1.039 ±0.016	0.991 ±0.014	0.773 ±0.017	1.113 ±0.009[a,c]		0.055 ±0.010[b]
$Th^{232}+\alpha$	27.7 ±0.17	22.65	2	1.38 ±0.03	1.25 ±0.02	1.279 ±0.016	0.968 ±0.014	1.419 ±0.013[a,c]	0.260 ±0.018	0.0371 ±0.0014
$U^{235}+n$	16.3 ±0.2	22.73	1	0.93 ±0.14	1.16 ±0.04	1.25 ±0.02	0.902 ±0.017	1.18 ±0.02[c]		
$U^{235}+n$	17.3 ±0.2	23.73	1	1.5 ±0.3	1.34 ±0.05	1.29 ±0.15	1.00 ±0.04		0.22 ±0.17	
$Th^{232}+\alpha$	29.0 ±0.17	23.92	1	1.43 ±0.11	1.34 ±0.03	1.42 ±0.03	1.098 ±0.019		0.40 ±0.09	0.058 ±0.014
$Th^{232}+\alpha$	29.6 ±0.17	24.51	1	1.47 ±0.14	1.55 ±0.07	1.57 ±0.05	1.23 ±0.06			
$U^{235}+n$	18.1 ±0.43	24.52	2	1.36 ±0.12	1.35 ±0.04	1.36 ±0.04	1.03 ±0.03	1.544 ±0.018[a,c]		
$U^{238}+n$	4.7 ±0.14	9.51	1	0.169 ±0.18	0.08 ±0.05	0.20 ±0.08	0.058 ±0.011			
$U^{238}+n$	9.1 ±0.3	13.89	1	0.55 ±0.08	0.338 ±0.011	0.28 ±0.03	0.187 ±0.005			
$U^{238}+n$	13.4 ±0.17	18.17	1	1.14 ±0.12	1.07 ±0.11	1.09 ±0.11	0.64 ±0.07			0.109 ±0.013
$U^{238}+n$	14.1 ±0.16	18.81	1	1.20 ±0.06	1.08 ±0.05	1.12 ±0.05	0.66 ±0.03			0.109 ±0.008
$U^{238}+n$	14.9 ±0.25	19.67	1	1.08 ±0.03	1.07 ±0.03	1.09 ±0.04	0.666 ±0.018			0.109 ±0.005
$U^{238}+n$	17.3 ±0.2	22.06	1	1.52 ±0.12	1.12 ±0.07	0.93 ±0.04	0.73 ±0.03			
$U^{238}+n$	18.1 ±0.43	22.85	1		1.11 ±0.05		0.86 ±0.04			
$Th^{232}+n$	9.1 ±0.3	14.12	1	0.7 ±0.3	0.54 ±0.03	0.54 ±0.10	0.436 ±0.014			
$Th^{232}+n$	13.4 ±0.17	18.40	1	1.80 ±0.09	2.06 ±0.05	1.88 ±0.04	1.44 ±0.02			0.032 ±0.004
$Th^{232}+n$	14.1 ±0.16	19.04	1	1.61 ±0.10	1.85 ±0.04	1.77 ±0.02	1.34 ±0.02			0.056 ±0.007
$Th^{232}+n$	14.9 ±0.25	19.90	1	1.53 ±0.07	1.85 ±0.06	1.71 ±0.05	1.28 ±0.04			0.044 ±0.008
$Th^{232}+n$	18.1 ±0.43	23.08	1	2.2 ±0.3	2.52 ±0.12	2.70 ±0.12	1.92 ±0.10			

[a] Result of one irradiation.
[b] Result of two irradiations.
[c] Number of fissions from average of Zr^{97}, Mo^{99}, and Ba^{139}.

increasing energy are compared in Fig. 3 for common excitation energy $E^{(1)}$ of the compound nucleus U^{236}. The symmetric fission yield has been observed previously to increase exponentially[17–19] with energy to 15-MeV excitation and then linearly[18,19] with energy. There is evidence of a slight dip in the symmetric fission yield at the 20-MeV excitation energy onset of $(x,2nf)$ fissions. Analogous effects in the anisotropy of fragments have been observed[25]; for proton-induced fission, distinct decreases in symmetric yields have been observed.[42] At 16-MeV excitation energy, data in Fig. 3 indicate no difference between the neutron-induced $Y_{n,s}$ and alpha-particle-induced $Y_{\alpha,s}$ yields of symmetric fission, but for 20-MeV excitation and above, the data indicate $Y_{\alpha,s}$ to be about 13% greater than $Y_{n,s}$. We believe this difference to be clearly beyond experimental uncertainties.

[42] B. J. Bowles, F. Brown, and J. P. Butler, Phys. Rev. **107**, 751 (1957); J. P. Butler, B. J. Bowles, and F. Brown, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), Vol. 15, p. 156; and G. R. Choppin, J. R. Meriwether, and J. D. Fox, Phys. Rev. **131**, 2149 (1963).

III. CALCULATIONS

We wish to estimate the dependence of fission asymmetry on the angular momentum effect caused by the fission-chance split. Present fission data and computational facilities are adequate to enable rather detailed calculations. For both neutron- and alpha-particle-induced fission and for each of the compound nucleus stages U^{236}, U^{235}, and U^{234}, the partial fission cross sections $\sigma_{x,f}^{(1)}$, $\sigma_{x,f}^{(2)}$, and $\sigma_{x,f}^{(3)}$, which are functions of the respective excitation energies $E^{(1)}$, $E^{(2)}$, and $E^{(3)}$, are to be combined in Eqs. (4a) and (4b) with the respective energy-dependent yields $Y_{x,s}^{(1)}$, $Y_{x,s}^{(2)}$, and $Y_{x,s}^{(3)}$ (each, however, approximated by $Y_{n,s}^{(1)}$) of symmetric fission.

A. Statistical Considerations

Statistical considerations of level density and of exits from compound states are expected to apply for the excitation energies involved in the present study. For fission of the U^{236} compound nucleus at an excitation energy $E^{(1)}$, the fission width is then

$$\Gamma_{x,f}^{(1)}(E^{(1)}) = K[2\pi\rho(E^{(1)})]^{-1} \times\int_0^{E^{(1)}-B_f-\mathcal{E}_{x,f}} \epsilon^\nu \rho(E^{(1)}-B_f^{(1)}-\mathcal{E}_{x,f}-\epsilon)d\epsilon, \quad (5)$$

with corresponding widths for second- and third-chance fission, where the power ν of the exit energy ϵ for fission is 0 for $N=1$ space coordinate in fission[43] and 1 for $N=3$ effective space coordinates.[44] Also, $\rho(E^{(1)})$ is the level density before fission, $\rho(E^{(1)}-B_f^{(1)}-\mathcal{E}_{x,f}-\epsilon)$ is the level (fission channel) density of the nucleus at the saddle point, $B_f^{(1)}$ is the fission-barrier threshold energy, $\mathcal{E}_{x,f}=\hbar^2 J_x(J_x+1)/2\mathcal{I}_\perp$ is the rotational energy, and K is a parameter discussed below.

Similarly, the neutron width $\Gamma_{x,n}^{(1)}(E^{(1)},E^{(2)})$ for de-excitation to $E^{(2)}$ is approximated[21] by

$$\Gamma_{x,n}^{(1)}(E^{(1)},E^{(2)}) = 2A^{2/3}r_0^2 m[\pi^2\hbar^2\rho(E^{(1)})]^{-1} \times\int_{E^{(2)}}^{E^{(2)}+dE^{(2)}} \epsilon\rho(E^{(1)}-B_n^{(1)}-\mathcal{E}_{x,n}-\epsilon)d\epsilon \quad (6)$$

and corresponding equations for $\Gamma_{x,n}^{(2)}(E^{(2)},E^{(3)})$ and $\Gamma_{x,n}^{(3)}(E^{(3)},E^{(4)})$, where A is the mass number, $r_0=1.5F$ is the nuclear radius parameter used, m is the mass of the neutron, $B_n^{(1)}$ is the neutron-binding energy, and $\mathcal{E}_{x,n}=\hbar^2 J_x(J_x+1)/2\mathcal{I}_n$ is the rotational energy. Limits 0 and $E^{(1)}-B_n^{(1)}-\mathcal{E}_{x,f}$ for the integral in Eq. (6) give the width $\Gamma_{x,n}^{(1)}$ for all neutron exits.

Calculations were made with both the Fermi-gas level density

$$\rho_x(E)=C_a \exp[2(a_x E)^{1/2}] \quad (7)$$

and the constant-temperature level density

$$\rho_x(E)=C_T \exp(E/T_x), \quad (8)$$

where C_a and C_T are constants. Necessary integrations for Eqs. (5) and (6) are conveniently given by Vandenbosch and Huizenga.[21] Note that the Pik-Pichak ratio of widths[9] in Eq. (1) is simply the exponential term from the width ratio of $\Gamma_{x,f}^{(1)}$, obtained by substituting Eq. (8) in Eq. (5), relative to $\Gamma_{x,n}^{(1)}$, obtained from substituting Eq. (8) in Eq. (6) with 0 and $E-B_n^{(1)}-\mathcal{E}_{x,n}$ integration limits.

To determine the partial fission cross sections, $\sigma_{x,f}^{(1)}$, $\sigma_{x,f}^{(2)}$, and $\sigma_{x,f}^{(3)}$, the fission probabilities, $\gamma^{(1)}$, $\gamma^{(2)}$, and $\gamma^{(3)}$ in Eqs. (2a), (2b), and (2c) are needed. In the calculation of these fission probabilities, all available experimental quantities were used, and fits to data of neutron-induced fission cross sections were used to estimate unavailable quantities. Measured fission thresholds[45] [$B_f^{(1)}=5.45$ MeV for the compound nucleus U^{236} and $B_f^{(3)}=5.27$ MeV for U^{234}] were increased by a 1.3-MeV pairing energy[6] for even-even compound nuclei for use in the level densities Eqs. (7) and (8); similarly, reported[46] neutron-binding energies to even-odd com-

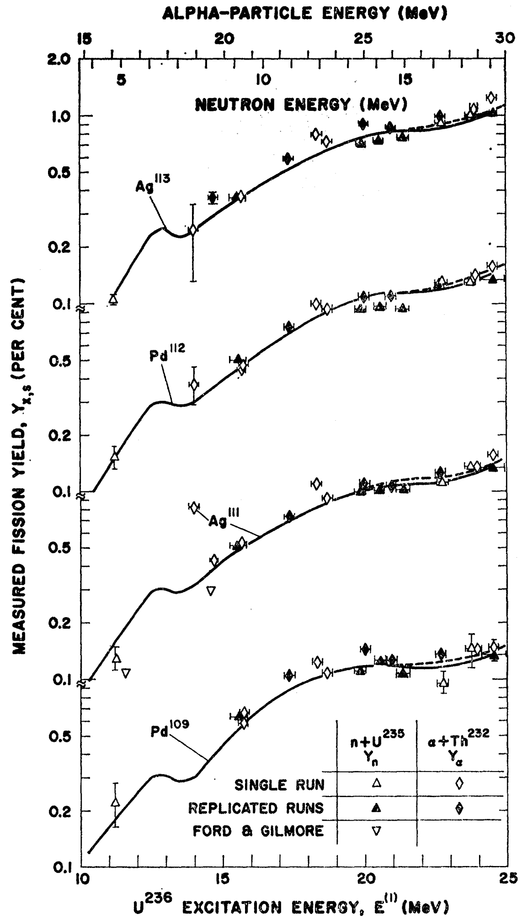

FIG. 3. Measured fission yields $Y_{x,s}$ of symmetric fission products from $Th^{232}(\alpha,f)$ and $U^{235}(n,f)$. Spreads in the irradiation energies are indicated. Yield uncertainties are less than the size of the points unless otherwise indicated. The solid lines are yields calculated from fission-chance splits by the Markov chain method of Sec. IIIB for the neutron-induced fission of U^{235}. The dotted lines are similarly calculated for alpha-particle-induced fission of Th^{232} and are not distinguishable from the neutron results below about 20-MeV excitation energy. Parameters were $a_n=23.4$ MeV^{-1}, $N=3$, and $\mathcal{I}_n=0.4\mathcal{I}_\perp$. The Ford and Gilmore results are from Ref. 36.

[43] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[44] I. Halpern [Ann. Rev. Nucl. Sci. **9**, 245 (1959)] has raised the question of "essential dimensionality" of the fission process. For our case of fission induced by energies considerably above the fission threshold, we consider the possibility that $\nu>0$ might be appropriate. Since the exact value of ν is not known, $\nu=1$ is used in our exploratory calculations. Actually, including the spin-dependent rotational energy $\mathcal{E}$ in the level densities ρ affects the function of ϵ in Eqs. (5) and (6), but we use the standard formalism of Ref. 21 for our calculations.

[45] J. A. Northrop, R. H. Stokes, and K. Boyer, Phys. Rev. **115**, 1277 (1959).

[46] Binding energies of neutrons to U^{236} from G. B. Holm, J. R. Burwell, and D. W. Miller [Phys. Rev. **122**, 1260 (1961)] and to U^{235} and U^{234} from R. A. Glass, S. G. Thompson, and G. T. Seaborg [J. Inorg. Nucl. Chem. **1**, 3 (1955)].

TABLE II. Calculations of fission probabilities. The average excitation energies for $U^{235}(n,n'f)$ and $U^{235}(n,2n'f)$ induced by 17-MeV neutrons $\bar{E}^{(2)}$ and $\bar{E}^{(3)}$, respectively. The standard deviations in the distribution in $E^{(2)}$ and $E^{(3)}$ are denoted by std. dev.

Space coordinates assumed N	Level parameter assumed[a]	Parameters from fits: Level parameter[a]	Parameters from fits: U^{235} fission threshold $B_f^{(2)}$ (MeV)	Parameters from fits: Level density coefficient K	17-MeV neutron calculation results: $\sigma_{n,f}^{(1)}$ b	$\sigma_{n,f}^{(2)}$ b	$\bar{E}^{(2)}$ (MeV)	std. dev. $E^{(2)}$ (MeV)	$\sigma_{n,f}^{(1)}$ b	$\bar{E}^{(3)}$ (MeV)	std. dev. $E^{(3)}$ (MeV)
1	$a_n=23.4$	$a_f=27.0$	6.10	0.747	1.732	0.582	15.2	1.03	0.371	8.3	0.88
3	$a_n=23.4$	$a_f=22.6$	6.05	10.5	1.000	0.818	15.2	1.07	0.772	8.3	0.87
1	$T_n=0.827$	$T_f=0.80$	6.25	5.33	1.550	0.584	15.1	1.14	0.356	8.1	0.89
1	$T_n=0.36$	$T_f=0.36$	6.15	2.85	1.152	0.729	16.0	0.49	0.711	9.3	0.70
3	$T_n=0.827$	$T_f=0.80$	6.05	6.93	1.577	0.657	15.1	1.14	0.274	8.1	0.91
3	$T_n=0.36$	$T_f=0.35$	6.05	6.05	1.822	0.605	16.0	0.48	0.274	9.3	0.70

[a] a_n and a_f are in units of MeV^{-1}; T_n and T_f are in MeV.

pound nuclei were increased by a 0.7-MeV pairing energy[47] to account for the low-energy gap in the level densities of residual even-even nuclei from neutron emission. The rotational-energy constants $\hbar^2/2\mathscr{I}_n$ used were 3 keV for excitations above a 3.4-MeV critical energy and 7-keV below.[21] The values of $\langle J_x(J_x+1)\rangle_{av}$ used were from optical-model calculations[14,15] for each irradiation energy and were held fixed through all stages of the calculations. The measured level-density constants used for neutron emission were $a_n=23.4$ for the Fermi-gas density[48] of Eq. (7) and $T_n=0.827$ MeV[48] or 0.36 MeV[49] for the constant temperature density of Eq. (8). Compound-nucleus cross sections $\sigma_{n,c}$ used were calculated with optical-model parameters extrapolated from those of Beyster *et al.*[15]

Fits to the data of both $U^{235}(n,f)$ cross sections[50] and $U^{233}(n,f)$ cross sections[51] in the 1- to 6-MeV neutron energy region of first-chance fission determined the magnitudes of the parameter pair a_f of Eq. (7) and the coefficient K of Eq. (5). (This coefficient K was introduced to give the analysis a semi-empirical quality.) The fission threshold $B_{n,f}^{(2)}$ for the compound-nucleus U^{235} is not known experimentally with the same standards as with U^{236} and U^{234} and thus it was the free parameter in the 1- to 6-MeV fit to the $U^{234}(n,f)$ cross section; the fission thresholds for U^{235} obtained from the various fits are consistent with measurement.[51,52] These results are shown in Table II.

The effect of pairing[53] on the density of fission channels is to favor the constant-temperature density of Eq. (8) rather than the Fermi gas density of Eq. (7). For this reason cross-section fits for both $N=1$ and $N=3$ space coordinates were attempted with constant-temperature density for fission and Fermi-gas density for neutron emission. These fits were much poorer than for the parameter combinations of Table II, and so this was not pursued further.

B. Markov Chain Calculations

Fission probabilities were calculated as a function of excitation energy along the stages of de-excitation that are energetically possible. Since the fission width $\Gamma_{x,f}$ and neutron width $\Gamma_{x,n}$ at any state in the de-excitation are independent of the previous decay history, a Markov chain treatment[54] is applicable.

In our calculations the populations of 101 states of the system are given by a 101-dimensional vector. States 1 through 99 are nuclei in one of 99 excitation-energy intervals. The system is in state 100 if a nucleus in a previous stage has fissioned, and the system is in state 101 if both neutron emission and fission were energetically impossible for a nucleus in a previous stage. For the compound nucleus U^{236} at our fixed irradiation energies, this population vector $\mathbf{q}^{(1)}=(q_1^{(1)},\cdots,q_{101}^{(1)})$ is simply $\mathbf{q}^{(1)}=(1,0,\cdots,0)$, but the corresponding population vectors $\mathbf{q}^{(2)}$ for U^{235}, $\mathbf{q}^{(3)}$ for U^{234}, and $\mathbf{q}^{(4)}$ for U^{233} are to be calculated. The probability of a nucleus being in state i after the first neutron emission is $q_i^{(2)}$, and the conditional probability $p_{ij}^{(2\to3)}$ is the probability of such a nucleus contributing to the probability $q_i^{(3)}$ of state j by means of neutron emission (or fission, or neither). These conditional probabilities are calculated from Eq. (6) [or Eq. (5) for fission] and are matrix elements of the transition matrix $P^{(2\to3)}$. The transition matrices $P^{(1\to2)}$ and $P^{(3\to4)}$ have corresponding meanings. Then

$$\mathbf{q}^{(2)}=\mathbf{q}^{(1)}P^{(1\to2)}\,, \tag{9a}$$

$$\mathbf{q}^{(3)}=\mathbf{q}^{(2)}P^{(2\to3)}=\mathbf{q}^{(1)}P^{(1\to2)}P^{(2\to3)} \tag{9b}$$

$$\mathbf{q}^{(4)}=\mathbf{q}^{(3)}P^{(3\to4)}=\mathbf{q}^{(1)}P^{(1\to2)}P^{(2\to3)}P^{(3\to4)}\,. \tag{9c}$$

The last equations result from the Chapman-Kolmogorov equation $P^{(u\to w)}=P^{(u\to v)}P^{(v\to w)}$ for $u<v<w$. (Only the fission state 100 was needed for $q^{(4)}$.) This detailed calculation of the decay chain was made for about 20 initial energies $E^{(1)}$ over the span of energies measured. For each incident energy of the calculations, the elements $q_i^{(2)}p_{i,100}^{(2\to3)}$ give the probability of second-

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chance fission for nuclei in the energy interval i. A normalized example is shown in Fig. 1 along with similar results for $(1 \to 2)$ and $(3 \to 4)$.

We now wish to test the various parameters in Table II by comparing the results from this Markov chain calculation with measured cross sections for U^{235} fission as a function of incident neutron energy. The calculated cross sections are $\sigma_{n,f} = \sum_k \sigma_{n,c} q_{100}^{(k)}$, and are compared with measured values in Fig. 4. However, these do not provide an adequate basis for deciding the number of effective space coordinates in fission nor a preference between level densities of Eqs. (7) and (8). This is because the fits are of similar quality and because three free parameters (Table II) are used in the calculations. These parameters were used in fitting only 1–6-MeV data, and so result in only moderately good fits at higher energy. Also shown in Fig. 4 are the partial fission cross sections $\sigma_{n,f}^{(1)}$, $\sigma_{n,f}^{(2)}$, and $\sigma_{n,f}^{(3)}$ obtained with the parameters $a_n = 23.4$ MeV^{-1}, $N = 3$ chosen for further analyses.

The energy-dependent yield $Y_{n,s}^{(1)}$ replacing all yields in Eq. (4) can now be determined by the Markov chain calculations from the measured yield $Y_{n,s}$ of symmetric fission in Table I and Fig. 3. This was done by the following unfolding process: (1) In the 0–6-MeV neutron energy region of exclusively $U^{235}(n,f)$, the yield $Y_{n,s}^{(1)}$ is simply the measured yield $Y_{n,s}$. (2) In the 6–13-MeV region of both $U^{235}(n,f)$ and $U^{235}(n,n'f)$, $Y_{n,s}^{(1)} = (Y_{n,s}\sigma_{n,f} - \langle Y_{n,s}^{(2)} \rangle_{av} \sigma_{n,f}^{(2)}) / \sigma_{n,f}^{(1)}$, where $Y_{n,s}^{(2)}$ of the U^{235} compound nucleus was approximated by $Y_{n,s}^{(1)}$ of U^{236} at each residual excitation energy. Cross sec-

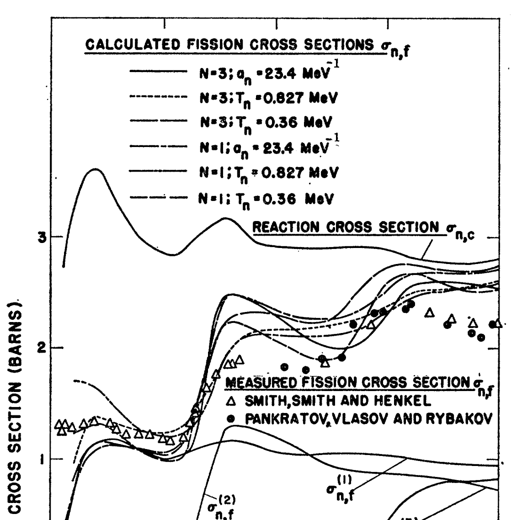

FIG. 4. Fits of $U^{235}(n,f)$ fission cross sections $\sigma_{n,f}$ calculated from fission-chance splits with the Markov chain method of Sec. IIIB and the parameters of Table II and in Sec. IIIA. The number of space coordinates for fission in these calculations is N; a_n is the level density parameter for neutron emission in Eq. (7), and T_n is the similar parameter for Eq. (8). The calculated partial cross sections $\sigma_{n,f}^{(1)}$, $\sigma_{n,f}^{(2)}$, and $\sigma_{n,f}^{(3)}$ for the $a_n = 23.4$ MeV^{-1}, $N = 3$ conditions of Table II are also shown. The calculated compound-nucleus cross section $\sigma_{n,c}$ is from Ref. 15; the measured fission cross sections are from Ref. 50.

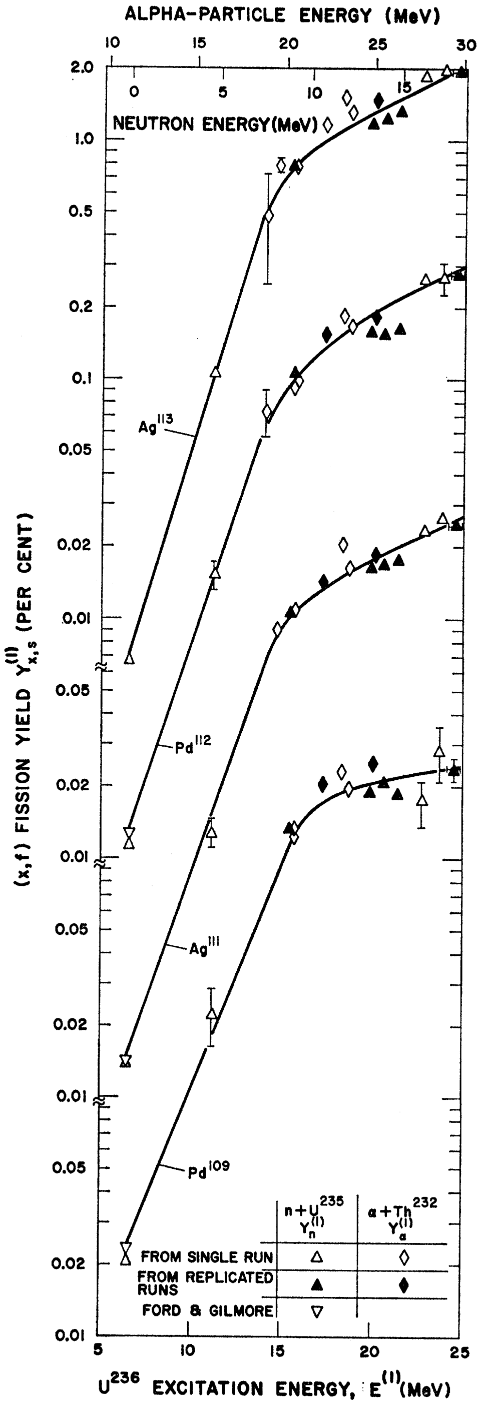

FIG. 5. First-chance fission yields $Y_{x,s}^{(1)}$ from fission of the compound nucleus U^{236}. These fission yields were calculated from the data of Table I and of Ref. 36 by the unfolding process of Sec. IIIB.

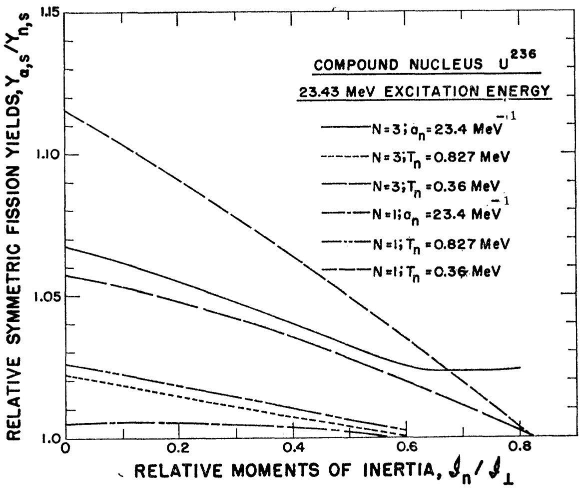

FIG. 6. Calculated ratio of symmetric fission yields $Y_{\alpha,s}$ from alpha-particle-induced fission of Th^{232} relative to $Y_{n,s}$ from neutron-induced fission of U^{235}. Calculations were from fission-chance splits by the Markov chain method of Sec. IIIB with the parameters of Table II and in Sec. IIIA.

tions and energy probabilities were calculated from the Markov chain. (3) Above this energy, Eq. (4b) was similarly solved for $Y_{n,s}^{(1)}$ by substituting the results of (1) and (2). The results of this process (1), (2), and (3) for obtaining the symmetric products Ag^{111}, Ag^{113}, Pd^{109}, and Pd^{112} are shown in Fig. 5.

From these yields, $Y_{n,s}^{(1)}$ in Fig. 5 and the Markov chain calculation of the energy-dependent partial cross sections, the yields $Y_{n,s}$ and $Y_{\alpha,s}$ were calculated for the predicted[1] $\mathcal{I}_n=0.4\mathcal{I}_\perp$; Eqs. (4a) and (4b), respectively, and the simplification and approximation following these equations were used. The results in Fig. 3 show no difference below the 20-MeV excitation energy onset of $(x,2nf)$ but show a calculated 4% difference above this energy. The calculated ratios $Y_{\alpha,s}/Y_{n,s}$ for other moment-of-inertia ratios and for the other parameters are shown in Fig. 6. The Fig. 6 calculations involved an approximate yield expression

$$Y_{n,s}^{(1)}=\frac{8.79(10^{-4})\exp(0.449E)}{1+8.79(10^{-4})C_Y\exp(0.449E)}, \qquad (10)$$

where the constant C_Y was 0.6 for all parameter sets in Table II except for the $a_n=23.4$, $N=3$, and $T_n=0.36$, $N=1$ cases, for which $C_Y=0.4$. The result shown in Fig. 3 for the calculation with unfolded yields $Y_{n,s}^{(1)}$ agrees with the result shown in Fig. 6 for $\mathcal{I}_n=0.4\mathcal{I}_\perp$ and the parameter set $a_n=23.4$, $N=3$.

IV. DISCUSSION

The experimental results in Table I and Fig. 3 qualitatively confirm the theoretical expectations that in the excitation-energy region of overlapping levels the effects of angular momentum on the asymmetry of fission are small. The calculation results in Fig. 6 for reasonably large ratios $\mathcal{I}_n/\mathcal{I}_\perp$ of moments of inertia account for generally half or less of the approximately 13% effect observed for yields in Fig. 3. Possibly this difference can be attributed to improper parameters or inadequacies of Eqs. (5) through (8) used in the calculations.

The results in Fig. 3 calculated from the fission-chance split indicate no dependence of the symmetric yields upon angular momentum below the approximately 20-MeV excitation energy onset of $(x,2nf)$. Anisotropy[25] and cross section[50] measurements show that the number of $(x,2nf)$ fissions are insignificant at 20 MeV. On this basis, data of 20 MeV and below give a measure of the importance of other causes. One cause is the moment of inertia difference ($\mathcal{I}_{\perp,\alpha}\neq\mathcal{I}_{\perp,s}$). Comparison at 20-MeV excitation energy shows the symmetric fission yields in Table I and Fig. 3 for $Th^{232}(\alpha,f)$ to be generally more than 9% greater than for $U^{235}(n,f)$; however, the symmetric fission yields at about 15.7-MeV excitation energy are about the same. This is consistent with the fraction of direct-interaction fissions increasing with neutron energy. Thus, these fission yield comparisons at 20 MeV and below indicate the possibility that above this energy at least several percent of direct-interaction fissions might explain the difference between the observed yield ratios and yield ratios calculated from fission-chance splits (discussed in Sec. I).

The agreement of yield ratios at about 15.7-MeV excitation energy argues against the possibility of an anomalously large moment-of-inertia difference producing symmetric fission yield differences. However, the unusually large corrections for neutrons degraded in energy for these 9.1-MeV neutron irradiations (see Sec. IIA) somewhat reduce the significance of this comparison.

ACKNOWLEDGMENTS

The authors appreciate the chemistry and data-handling assistance of Mrs. D. L. Copley and Mrs. T. K. Kelley, Van de Graaff irradiations by R. K. Smith, and Cockcroft-Walton irradiations by R. W. Davis. We thank D. D. Armstrong and J. E. Midgley for preparing targets and holders for irradiations.