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Fission of Odd-A Uranium and Plutonium Isotopes Excited by (d,p), (t,d), and (t,p) Reactions*

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Fission-fragment angular correlations and fission probabilities have been measured for a series of odd-A uranium and plutonium isotopes excited by (d,p), (t,d), and (t,p) reactions. The following fissioning nuclei have been studied: (1) ^{235}U from (d,pf), (t,pf); (2) ^{237}U from (d,pf), (t,df), (t,pf); (3) ^{239}U from (d,pf); (4) ^{241}Pu from (d,pf), (t,df), (t,pf); and (5) ^{243}Pu from (d,pf), (t,df). Fission probabilities and angular-correlation coefficients for each case are compared with previously reported (n,f) results. The direct-reaction experiments show fission thresholds at excitation energies equal to or less than threshold energies observed in (n,f) experiments. Some qualitative characteristics of the transition-state spectrum for the nuclei studied are determined from comparisons of the direct-reaction and (n,f) results.

I. INTRODUCTION

Since the concept of transition states was first introduced by Bohr,[1] there have been several attempts to try to deduce the spectra of low-lying transition states for various Th, U, and Pu isotopes from an analysis of cross sections and angular distributions for (n,f) reactions on even-even targets. Some aspects of the transition state spectra for several nuclei were determined by Lamphere[2,3] from a qualitative analysis of the structure apparent in (n,f) cross sections and angular anisotropies. More recently, attempts have been made to obtain information on the low-lying transition states for several nuclei from a simultaneous quantitative fit to (n,f) cross sections and fragment angular distributions.[4-7]

In addition to the (n,f) reaction, it is also possible to excite these same nuclei to energies above the fission barrier using various direct reactions. Results have previously been reported[8] for the angular distributions of fission fragments from the ^{234}U(d,pf) reaction. A quantitative interpretation of direct-reaction results is difficult because of the uncertainties in the characteristics of the direct-reaction process. However, for excitation energies near the neutron binding energy, the direct-reaction and neutron-capture processes excite a different distribution of angular momentum values. This can create qualitative differences in the fission probabilities and the angular distributions of the fragments for nuclei excited by neutron capture or direct reactions. For example, a low-lying transition-state band of high spin may be observed in the direct-reaction fission experiment, but may not be apparent in neutron-fission results because the neutron-capture reaction is unable to excite states of the appropriate angular momenta.

In this paper results are reported on the fission probabilities and angular correlations for (d,pf),

(t, df), and (t, pf) reactions leading to the following odd-A nuclei: ^{235}U, ^{237}U, ^{239}U, ^{241}Pu, and ^{243}Pu. Comparisons are made with similar results from (n, f) reactions.

II. EXPERIMENTAL PROCEDURE

The experimental techniques were similar to those of previous experiments[9,10] and only a few of the more important aspects will be repeated here. The experiments were performed using 18.0-MeV triton and deuteron beams from the Los Alamos Scientific Laboratory Van de Graaff accelerator facility. A multipurpose scattering chamber was used which could accomodate a ΔE-E charged-particle telescope and up to eight independent fission detectors. Outgoing proton or deuteron energy spectra were obtained in coincidence with each of the fission detectors yielding a full (up to eight angles) angular correlation of fission fragments in a single run. In some cases, data were obtained for more than one fission-detector configuration, yielding angular correlations with more than eight angles.

The ΔE detector was a 310-μ Au surface barrier and the E detector was a lithium-drifted detector of 3 mm thickness. The over-all resolution of the proton or deuteron detection systems was approximately 120 keV. The detector was collimated with a circular aperture and subtended an angle $\Delta\theta \sim 15°$. The fission detectors were phosphorus-diffused semiconductor detectors of ~400-Ω-cm silicon which were operated at reverse biases of 100–200 V. Detectors of two sizes were used: 8 mm×8 mm square and 8 mm×20 mm rectangular. For angles near the recoil angle, the square detectors were used, and rectangular detectors were used for fission detectors that were nearly perpendicular to the recoil direction. In the reaction plane, the fission detectors subtended an angle $\Delta\theta = 13°$ in the (d, pf) and (t, pf) experiments and $\Delta\theta = 9°$ in the (t, df) measurements.

The targets were prepared by vacuum evaporation on 40–80-μg/cm^2 carbon backings. The heavy elements were in the form of oxides with deposit thicknesses ranging from 100–300 μg/cm^2. The targets had the following enrichment in the isotope of interest: ^{233}U – 97.96%, ^{234}U – 99.7%, ^{235}U – 93.25%, ^{236}U – 99.88%, ^{238}U – 99.97%, ^{239}Pu – 94.41%, ^{240}Pu – 98.0%, and ^{242}Pu – 99.88%.

The reactions studied in these experiments are listed in Table I.

III. DATA REDUCTION AND ANALYSIS

The data were obtained utilizing an SDS-930 online computer with final data reduction in a larger CDC-6600 computer as described previously.[9,10] From the data analysis, coincidence proton or deuteron spectra corrected for accidental contributions are obtained for each of the fission detectors. The corrected coincidence spectra are normalized to account for differences in the solid angles of the fission detectors. Each spectrum is then converted to a new spectrum of counts versus excitation energy of the residual nucleus with standard channel widths of 50 keV. The conversion to an excitation-energy spectrum involves an

TABLE I. Characteristics of the reactions studied and the experimental setup used in obtaining the reported results. $\Delta\theta_f$ is the solid angle subtended by the fission detectors.

Reaction	Number of fission angles	Ground-state Q value (MeV)	Angle of proton or deuteron detector (deg)	$\Delta\theta_f$ (deg)
$^{233}U(t,p)^{235}U^*$	7	+3.65[a]	130	13
$^{234}U(d,p)^{235}U^*$	7	+3.08[a]	150	13
$^{235}U(t,p)^{237}U^*$	7	+3.18[b]	150	13
$^{236}U(d,p)^{237}U^*$	7	+2.90[a]	150	13
$^{236}U(t,d)^{237}U^*$	24	−1.14[a]	140	9
$^{238}U(d,p)^{239}U^*$	7	+2.59[a]	150	13
$^{239}Pu(t,p)^{241}Pu^*$	8	+3.26[b]	150	9
$^{240}Pu(d,p)^{241}Pu^*$	7	+3.02[a]	150	13
$^{240}Pu(t,d)^{241}Pu^*$	24	−1.02[a]	140	9
$^{242}Pu(d,p)^{243}Pu^*$	7	+2.81[a]	150	13
$^{242}Pu(t,d)^{243}Pu^*$	24	−1.23[a]	140	9

[a] J. R. Erskine, A. M. Friedman, T. H. Braid, and R. R. Chasman, in *Proceedings of the Third International Conference on Atomic Masses and Related Constants, Winnepeg, Canada, 1967*, edited by R. C. Barber (University of Manitoba Press, Winnepeg, Canada, 1968), p. 622.

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energy calibration of the ΔE-E system, the Q values of the reaction, and a c.m. transformation to the rest system of the fissioning nucleus. For each interval of excitation energy, each detector angle is calculated in the rest system of the fissioning nucleus (i.e., the angle relative to the kinetic recoil angle with c.m. correction). The statistical error for each point is also calculated.

For the above data reduction, the relative solid angles of the fission detectors are determined by comparing the relative singles fission rates with measured angular distributions for the corresponding (d, f) or (t, f) reaction. The energy calibration of the ΔE-E system is determined from known energy groups for reactions on ^{12}C and ^{16}O. The reaction Q values are listed in Table I. Estimated uncertainties in the calibration and the reaction Q values lead to a ±50-keV uncertainty in the calculated excitation energies.

For a given reaction, the data from all runs are combined into a single matrix of excitation energy and fission angle. Then at each excitation energy interval a least-squares fit is performed to the function

$$W(\theta) = A_0 [1 + \sum_{L=2,4,6} g_L P_L(\cos\theta)] , \qquad (1)$$

where A_0 and g_2, g_4, g_6 are adjustable parameters, and the angles are measured in the rest system of the fissioning nucleus (i.e., relative to the kinematic recoil angle).

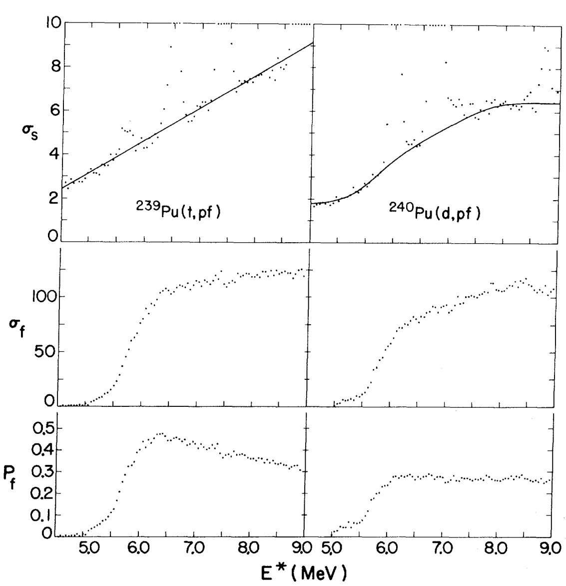

FIG. 1. Singles proton spectra (σ_s) and coincident fission cross sections (σ_f) in arbitrary units. Resultant fission probability (P_f) for the ^{239}Pu(t, pf) and ^{240}Pu(d, pf) reactions. Solid curves in σ_s represent an extrapolation underneath carbon and oxygen contaminant peaks which was used to determine P_f.

Fission probabilities are calculated at each energy interval from

$$P_f = \sigma_f / \sigma_s , \qquad (2)$$

where a relative fission cross section σ_f is determined from A_0 using measured absolute solid angles for the fission detector, and σ_s is the "singles" cross section for producing an excited residual nucleus in the appropriate energy interval. For the (t, p) and (d, p) reactions the singles cross sections are extrapolated smoothly underneath the sharp peaks from reactions with ^{16}O and ^{12}C contaminants. An example of the procedure for calculating P_f is shown in Fig. 1 for the ^{239}Pu(t, pf) and ^{240}Pu(d, pf) reactions. Because of the larger kinematic shifts in the (t, d) reaction, there were no carbon or oxygen contaminant peaks in the region of interest, and the above extrapolation procedure was not necessary.

In discussing the direct-reaction results in comparison with (n, f) cross-section data, it is necessary to obtain fission probabilities which are defined in an equivalent manner for the (n, f) and various types of direct-reaction experiments. It is most convenient to define the fission probability as the fraction of all the nuclei, in a particular excitation energy interval, which decay by fission. Thus, for neutron experiments, fission probabilities can be obtained from the ratio of the fission cross section to the total compound-nucleus cross section. However, for the direct-reaction results, the expression in Eq. (2) does not necessarily give an equivalent fission probability because not all of the outgoing particles contained in the cross section, σ_s, correspond to events which produce a residual nucleus excited to the appropriate energy. In particular, for the (d, p) reaction leaving residual nuclei excited above the neutron binding energy, the singles proton cross section can be written as the sum of two components

$$\sigma_s = \sigma_{\text{compound}} + \sigma_{\text{breakup}} ,$$

where σ_{compound} is the (d, p) stripping cross section leaving excited residual nuclei at a particular energy and σ_{breakup} is the cross section for the direct (d, pn) reaction involving the breakup of the deuteron in the field of the heavy nucleus. This direct breakup process differs from the compound process where a residual nucleus is excited and then later decays either by neutron emission or fission. Thus, the true fission probability is more correctly given by

$$P_f = \sigma_f / \sigma_{\text{compound}}$$

rather than by Eq. (2). However, in the present experiment, it is only possible to measure σ_s, and there are no available experimental or theoretical results which can be used to reliably obtain σ_{compound} from the measured values of σ_s.

In some cases a qualitative estimate of the ratio $\sigma_{\text{breakup}}/\sigma_{\text{compound}}$ for the (d,p) reaction can be obtained by comparing (t,pf) and (d,pf) results leading to the same residual nucleus at the same excitation energy. For the (t,p) reaction at excitation energies near the neutron binding energy, protons from a breakup reaction can be obtained only from a (t,d) reaction to a low-lying state followed by the breakup of the outgoing deuteron. At the backward angles used in these experiments cross sections for (t,d) reactions are much less than (t,p) cross sections, and this breakup process would be expected to have a negligible cross section. Thus the fission probabilities calculated for the (t,p) reactions should be approximately correct. Then, if the (t,p) fission probabilities are correct and the (d,p) and (t,p) process excite the same distribution of angular momentum states, the following relationship would be approximately correct:

$$\frac{P_f(t,pf)}{P_f(d,pf)} \approx 1 + \frac{\sigma_{\text{breakup}}}{\sigma_{\text{compound}}}, \tag{3}$$

where σ_{breakup} and σ_{compound} are the relative cross sections going into deuteron breakup and excitation of the residual nucleus for the (d,p) process. The total (d,p) cross section, σ_s, is proportional to $\sigma_{\text{breakup}} + \sigma_{\text{compound}}$. In general, because the (d,p) and (t,p) reactions to the same residual nucleus involve the transfer to different particles (neutron or dineutron) to targets with different spins, it is not a very good assumption that the angular momentum and parity distributions of the residual nuclei following the two types of reaction are exactly the same. However, at energies above the fission threshold, where fission is proceeding through many different transition states, one might expect Eq. (3) to become approximately correct even if there are quantitative differences between the angular momentum distributions excited in the two types of reaction.

Figure 2 shows experimental results for the ratio $P_f(t,pf)/P_f(d,pf)$ for reactions leading to residual nuclei ^{235}U, ^{236}U, ^{237}U, and ^{241}Pu as a function of an equivalent neutron energy, $E_n = E^* - B_n$. In this form, one might expect the variation of the fraction of the (d,p) cross section going to direct breakup to be the same for all targets. The results shown here are for a deuteron bombarding energy of 18 MeV. The dependence of the breakup cross section might be different for a different deuteron energy. Figure 2 shows that the ratios obtained above $E_n \approx 1$ MeV are approximately the same for residual nuclei ^{235}U, ^{236}U, and ^{241}Pu. Below $E_n \approx 1$ MeV the different nuclei tend to give somewhat different results for a variety of reasons.

In the ^{236}U case, where the residual nucleus is even-even, there are no open fission channels for states with angular momenta 1^+ or 0^- in the region below the pairing gap, $E_{2\Delta}$. The (d,pf) reaction can excite these and other states that do not have natural spin parity, while the (t,p) reaction is limited to the excitation of natural spin-parity combinations. This difference between the two reactions leads to the relatively large values for the $P_f(t,pf)/P_f(d,pf)$ ratio below the pairing energy.

For the odd-A residual nuclei, the values obtained for the ratio in the region below the half-rise point for the fission probability, $E_{1/2}$, are sensitive to the accuracy of the energy calibration and to the details of the transition-state spectrum. The deviations observed in this region for ^{235}U and ^{241}Pu are probably not significant.

The ^{237}U results tend to give ratios that are ≈50% greater than those obtained from the other reactions. The reason for this difference is not clear.

The solid line shown in Fig. 2 is meant to be a qualitative representation of all the results, with the additional constraint that the ratio should go to 1.0 at energies below the neutron binding energy. This solid line is used for a qualitative correction of all of the (d,pf) results for the effect of deuteron breakup on the fission probability. This correction is certainly not exact, but it is hoped that it is good enough to allow qualitative comparisons

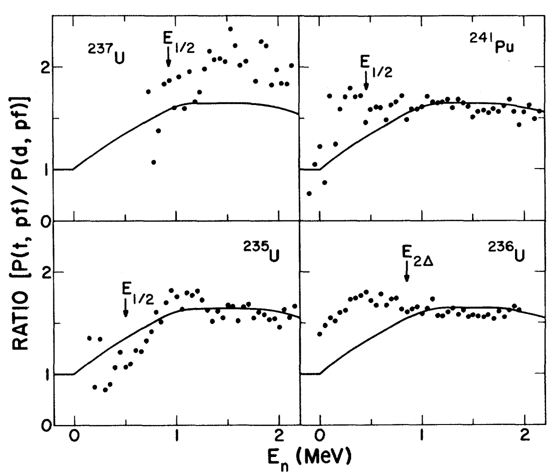

FIG. 2. Ratio of fission probabilities for the (t,pf) and (d,pf) reactions going to the same fissioning nuclei at the same excitation energy. Excitation energies have been converted to equivalent neutron bombarding energies on the appropriate even-even target. The solid curve is the same in all cases and represents the best characterization of all the results within the limitations described in the text.

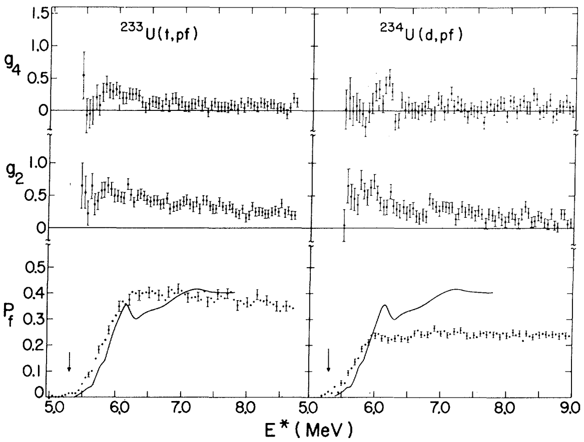

FIG. 3. Fission probability and angular-correlation coefficients from the fission of ^{235}U induced by the ^{233}U-(t,pf) and $^{236}U(d,pf)$ reactions. Arrows indicate the binding energy of the last neutron.

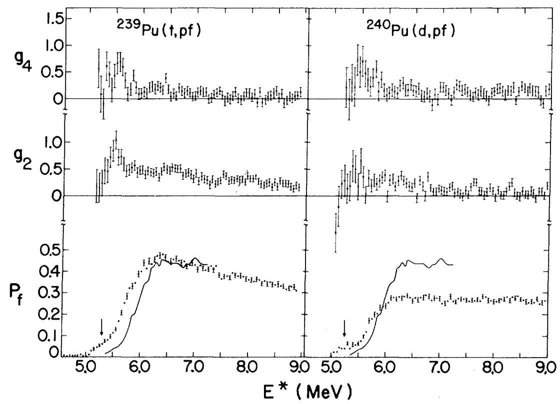

FIG. 5. Fission probability and angular-correlation coefficients from the fission of ^{241}Pu induced by the ^{239}Pu-(t,pf) and $^{240}Pu(d,pf)$ reactions. Arrows indicate the binding energy of the last neutron.

between the fission probabilities obtained from (n,f), (d,pf), and (t,pf) reactions.

Fission probabilities for the (n,f) reaction were obtained by dividing published (n,f) cross sections by calculated compound-nucleus-formation cross sections from an optical-model calculation[11] using parameters determined by Auerbach and Moore[12] from a best fit to a variety of $n+{}^{238}U$ experimental data. The neutron-fission cross sections used were taken from Lamphere[2] for targets of ^{234}U, ^{236}U, and ^{238}U; from Nesterov and Smirenkin[13] for ^{240}Pu; and from Butler[14] for ^{242}Pu.

IV. RESULTS

Fission probabilities and angular-correlation coefficients, g_2 and g_4 for the (t,pf) and (d,pf) reactions are shown in Figs. 3–6 as a function of excitation energy. In these figures the fission probabilities from the (d,pf) reactions have not been corrected for the proton cross section arising from the direct deuteron breakup reaction described in the previous section. Similar results for the (t,df) reactions are shown in Fig. 7. In all cases the solid curves show fission probabilities for the appropriate (n,f) reaction, which are obtained as described in the previous section. The error bars indicate statistical errors only and there is an additional ±10% uncertainty in the absolute fission probabilities for the direct-reaction results. The excitation energies have an estimated uncertainty of ±50 keV.

For the (d,pf) results, the fitted g_2, g_4, and g_6 coefficients were used to calculate angular aniso-

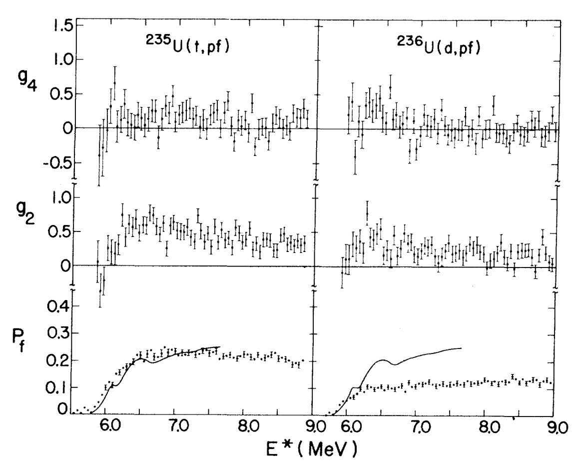

FIG. 4. Fission probability and angular-correlation coefficients from the fission of ^{237}U induced by the ^{235}U-(t,pf) and $^{236}U(d,pf)$ reactions.

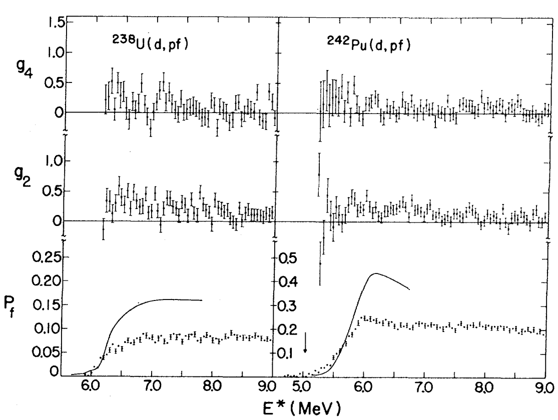

FIG. 6. Fission probability and angular-correlation coefficients from the fission of ^{239}U and ^{243}Pu induced by the $^{238}U(d,pf)$ and $^{242}Pu(d,pf)$ reactions, respectively. The arrow indicates the binding energy of the last neutron for ^{243}Pu.

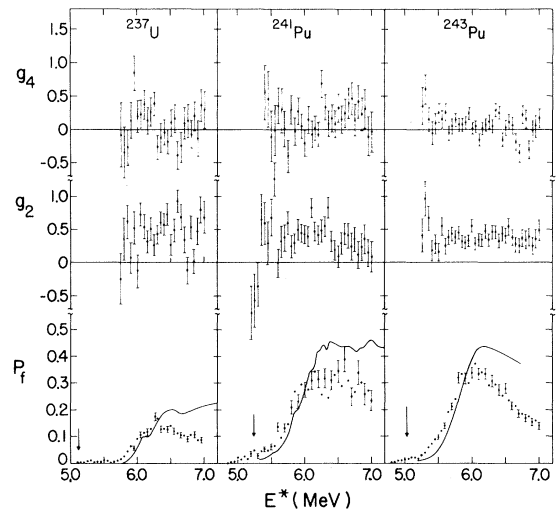

FIG. 7. Fission probability and angular-correlation coefficients from fission of ^{237}U, ^{241}Pu, and ^{243}Pu induced by the ^{236}U(t,df), ^{240}Pu(t,df), and ^{242}Pu(t,df) reactions, respectively. Arrows indicate the binding energy of the last neutron.

tropies, $W(0°)/W(90°)$. These anisotropies are shown in Fig. 8 compared with the results of Lamphere[3] for (n,f) reactions. In general, the statistical uncertainties on the (d,pf) results are rather poor, so that it is difficult to make a detailed comparison with the more precise (n,f) results. The (d,pf) anisotropies tend to be somewhat larger

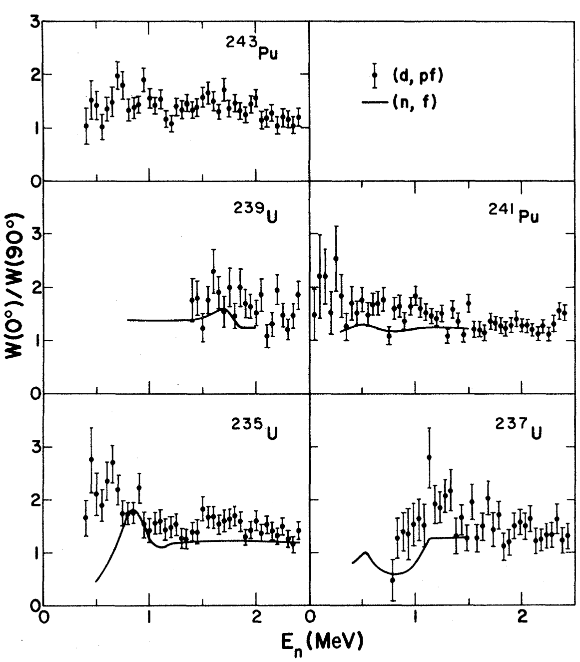

FIG. 8. Measured anisotropies for the (d,pf) reactions compared with results from (n,f) experiments (Ref. 3).

than (n,f) anisotropies but the statistical uncertainties do not allow a significant determination of the dependence of the anisotropy on excitation energy except in a very qualitative way. A previous ^{234}U(d,pf) experiment[8] yielded anisotropies which were less than those observed in the present experiment and similar to the (n,f) results. The previous experiment[8] was performed with 13-MeV deuterons and a proton detection angle of 90°. Studies[9,15] of the ^{239}Pu(d,pf) reaction have shown that at a proton angle of 90° the fragment anisotropies are about a factor of 2 less than observed at $\theta_p > 120°$ because of Coulomb-distortion effects on the (d,p) stripping process. The difference between the anisotropies observed in the present experiment with $\theta_p = 150°$ and previous results[8] at $\theta_p = 90°$ is consistent with this Coulomb-distortion effect.

Figure 9 shows fission probabilities for the (n,f), (t,pf), and (d,pf) reactions for all the cases that were studied. In this case an approximate correction (see previous section) has been applied to the (d,pf) results to account for the cross section going into deuteron breakup reactions. The apparent energy shifts between (d,pf) and (t,pf) results are consistent with the ±50-keV uncertainty in the energy calibration and reaction Q values.

The (t,df) results are generally similar to (d,pf) and (t,pf) results. Because of the limited quality of the (t,df) data no attempt was made to correct these results for the effect of breakup reactions, and these results were not included in the summary presented in Fig. 9.

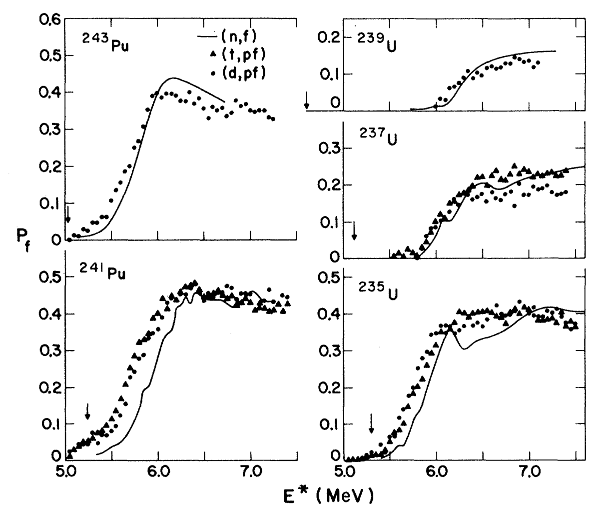

FIG. 9. Fission probabilities for the various reactions studied compared with results for (n,f) measurements. The (d,pf) results have been corrected for the effects of deuteron breakup reactions as described in the text. Arrows indicate the binding energy of the last neutron.

V. DISCUSSION

A. Transition State Spectra

Recently attempts[4-7] have been made to quantitatively fit (n,f) cross-section and angular-distribution measurements with a detailed model of the (n,f) process. In this model the neutron capture and decay widths are calculated and the characteristics of the transition-state spectrum and the fission barrier are used as adjustable parameters in an attempt to fit observed experimental results. This model used a parabolic-shape fission barrier and in order to quantitatively fit the experimental results, the curvature of the parabolic barrier was allowed to vary for different transition states. However, recent discoveries[16] that the fission barrier is double peaked instead of having the previously assumed parabolic shape add considerable complexity to the problem. In particular, calculations of penetrabilities through double-peaked barriers,[10,17,18] which appear appropriate for nuclei in the U–Pu region, show complex penetrability functions that are capable of producing, as a function of excitation energy, both sharp changes in the fragment angular distributions, and relatively smooth changes in the fission cross sections. The rapid energy variations in the angular distributions, coupled with relatively slowly varying cross sections for some (n,f) reactions, required the use of different barrier shapes for different transition states in the model fits[4-7] with parabolic barriers. If a more realistic double-peaked barrier were used to fit the (n,f) results, it is not clear that the data would still require more than one barrier shape for a given nucleus. Fits to the (n,f) data with a more realistic barrier shape would probably indicate that the important low-lying transition states are those identified in the fits[4-7] with parabolic barriers but the detailed energy spacing and ordering of these states could be quite different. Until the complexities of fission through a double-peaked barrier and the relevant barrier shapes are better known, it must be assumed that both the "quantitative"[4-7] and qualitative[2,3] interpretation of (n,f) experimental results give only an indication of the particular transition states which lie low in a particular nucleus, and the details of the spacing and ordering of the states may change when data are analyzed with a more realistic model.

Similarly, because of the complexity of both the fission and direct-reaction processes, it is not possible to determine details of the fission barrier shapes or the transition-state spectra from the present direct-reaction fission results. However, it is possible to determine a few general characteristics of the transition-state spectrum and the fission decay process from a qualitative comparison of the direct-reaction results with (n,f) data.

The fundamental difference between the (n,f) and the direct-reaction fission processes is in the distribution of angular momentum values which can be excited in each case. Figure 10 shows calculated angular momentum transfers for the (n,f) reaction at various energies compared with the calculated distribution for the (t,p) reaction. The parameters[12] used for the neutron calculations pre-

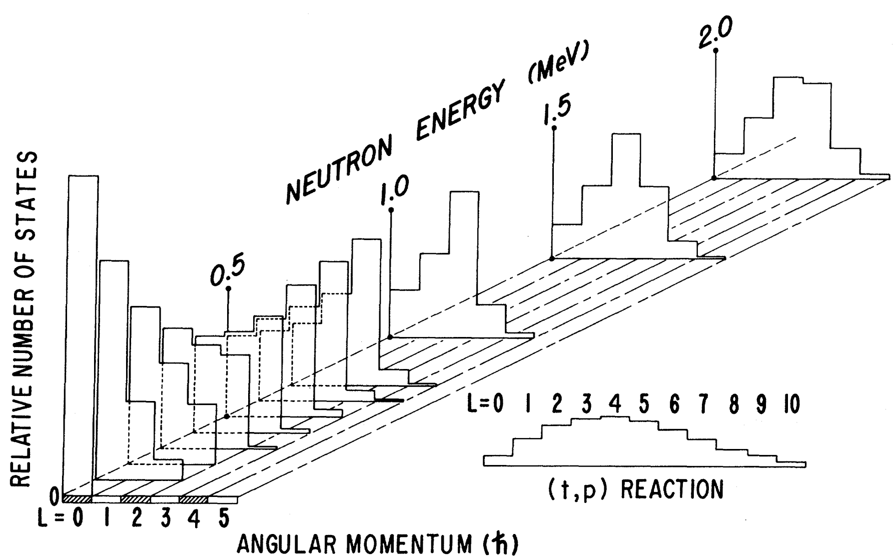

FIG. 10. Calculations of the relative probability of various orbital angular momentum transfers for the (n,f) reaction at various energies and for the 18-MeV (t,p) reaction.

dict a strong enhancement for odd-l transfers. This is evident in the dominance of $l=1$ for the range of $0.1 \leq E_n \leq 1.0$ MeV. However, this effect may be sensitive to the parameter set used in these calculations and, thus, the results presented in Fig. 10 should be used for a qualitative comparison of the results. Nevertheless, certain features of the angular momentum transfers are relatively invariant to reasonable parameter change. In particular, the calculated average angular momentum transfers in the (n,f) reaction ($l=0.6$, 1.2, 1.7, and 2.3 at $E_n=0.1$, 0.5, 1.0, and 2.0 MeV, respectively) can be compared with similar calculations for the direct reaction [$l=3.1$ and 4.3 for the (d,pf) and (t,pf) reactions, respectively].

From the neutron angular momentum transfers shown in Fig. 10 and similar calculations for the (d,p) reaction it is possible to qualitatively determine both the fraction of the fission cross section which excites states that are appropriate for fission through a particular transition-state band with a given K projection and parity, and the average angular coefficient $\bar{g}_2$ for fission through these transition states. Table II shows results of these calculations for the (d,pf) reaction and for (n,f) reactions at neutron energies of 0.1 to 1.0 MeV. From Table II the following general characteristics should be noted: (1) For $E_n \leq 0.3$ MeV the (n,f) reaction can strongly excite only states appropriate for fission through $K=\frac{1}{2}^+$, $\frac{1}{2}^-$, and $\frac{3}{2}^-$ bands, whereas the (d,pf) process can also strongly excite states in $K=\frac{3}{2}^+$ and $\frac{5}{2}^+$ bands; (2) for $E_n \leq 0.3$-MeV fission through states from $K=\frac{5}{2}^-$, $\frac{7}{2}^-$, and $\frac{7}{2}^+$ bands should not be seen at all in (n,f) reactions; (3) for $E_n > 0.5$ MeV both the relative strengths of excitation and the average coefficients $\bar{g}_2$ should be similar for (n,f) and (d,pf) reactions; (4) for the (d,pf) reactions the $\bar{g}_2$ coefficients should be strongly positive for $K=\frac{1}{2}^+$ or $\frac{1}{2}^-$ bands and negative for all other bands.

In the following discussion we will try to use the above characteristics to make some qualitative conclusions about the low-lying transition-state spectra for some of the nuclei studied. In some cases, it is possible to identify the approximate excitation energies at which particular transition-state bands start to make noticeable contributions to the experimental distributions. These "threshold" energies should not be interpreted as actual level positions for the transition-state bands, because of the complexities introduced by fission through a double-peaked fission barrier, which may or may not have the same shape for fission through all transition states.

1. ^{235}U (Figs. 3 and 9)

The (d,pf) and (t,pf) results both show a weak threshold ($P_f \sim 0.02$, $P_f/P_{f\,\max} \sim 0.05$) at $E^* \sim 5.2$ MeV. For the (n,f) reaction there is no significant fission for very low-energy neutrons suggesting that the (t,pf) and (d,pf) results correspond to fission of states other than $\frac{1}{2}^+$. There are two possible explanations for this weak threshold. First, it could correspond to fission through a high-spin transition-state band ($K=\frac{5}{2}^-$, $\frac{7}{2}^\pm$ would give $P_f/P_{f\,\max}$ 0.05–0.09, see Table II). Alternatively, fission in this region could be due to a subbarrier resonance for a lower-spin transition state (other than $\frac{1}{2}^+$). These two possibilities could be separated by observing the angular correlation of the fragments in this region. Unfortunately, the present results are not of sufficient accuracy to allow this.

TABLE II. Calculations of the relative cross sections and angular correlation coefficients $\bar{g}_2$ for exciting states appropriate for fission through various single-particle transition-state bands for the 18-MeV (d,pf) reaction and (n,f) reactions at various energies.

Transition-state band K^π	18-MeV (d,pf) $\sum\sigma(j^\pi)/\sigma_T$	$\bar{g}_2$	0.1-MeV (n,f) $\sum\sigma(j^\pi)/\sigma_T$	$\bar{g}_2$	0.3-MeV (n,f) $\sum\sigma(j^\pi)/\sigma_T$	$\bar{g}_2$	0.5-MeV (n,f) $\sum\sigma(j^\pi)/\sigma_T$	$\bar{g}_2$	1.0-MeV (n,f) $\sum\sigma(j^\pi)/\sigma_T$	$\bar{g}_2$
$\frac{1}{2}^+$	0.52	+0.7	0.45	+0.0	0.39	+0.1	0.41	+0.6	0.42	+0.8
$\frac{3}{2}^+$	0.32	−0.1	0.04	−0.3	0.16	−0.4	0.24	−0.3	0.31	−0.2
$\frac{5}{2}^+$	0.21	−1.1	0.02	−1.4	0.08	−1.4	0.13	−1.4	0.18	−1.3
$\frac{7}{2}^+$	0.05	−0.8	...	...	...	...	...	...	0.02	−1.1
$\frac{1}{2}^-$	0.48	+0.8	0.55	+0.7	0.61	+0.7	0.59	+0.7	0.58	+0.8
$\frac{3}{2}^-$	0.40	−0.6	0.38	−1.0	0.43	−0.9	0.42	−0.8	0.46	−0.3
$\frac{5}{2}^-$	0.09	−0.8	...	...	0.02	−0.6	0.06	−0.6	0.20	−0.7
$\frac{7}{2}^-$	0.05	−1.7	...	...	0.01	−1.7	0.04	−1.7	0.13	−1.7

The (n, f) results show an apparent threshold at $E^* \sim 5.5$ MeV which is more strongly excited in the (d, pf) and (t, pf) experiments than in the (n, f) reaction. The relative fission probability ($P_f/P_{f\,\max} \sim 0.1$) for the (n, f) reaction is consistent with fission through a $K = \frac{3}{2}^+$ or $\frac{5}{2}^+$ band, and the predicted probability, $P_f/P_{f\,\max} \sim 0.2-0.3$ (Table II), for the (d, pf) reaction is consistent with the (d, pf) and (t, pf) experimental results. The value of $g_2 \sim 0.5$ in the energy region $E^* = 5.5-6.0$ MeV indicates a strong contribution from fission through a $K = \frac{1}{2}^\pm$ band, and the apparent threshold at $E^* \sim 5.7$ MeV in the (n, f) data has a relative fission probability consistent with a $K = \frac{1}{2}$ band.

In summary, these results indicate the presence of low-lying states, $K = (\frac{5}{2}^-$ or $\frac{7}{2}^\pm)$, $K = (\frac{3}{2}^+$ or $\frac{5}{2}^+)$, and $K = \frac{1}{2}^\pm$ in the transition-state spectrum for ^{235}U.

2. ^{237}U (Figs. 4 and 9)

In this case the (t, pf) and (d, pf) fission probabilities are very similar to the results obtained from the (n, f) reaction. This similarity is consistent with the fact that the first threshold is at $E_n \sim 0.8$ MeV. The first threshold at $E^* \sim 6.0$ MeV corresponds to $g_2 \sim 0$ for the (t, pf) and (d, pf) measurements and an anisotropy of ~1 for (n, f) measurements, and it is excited with large probability in all experiments. These results are most consistent with a $K = \frac{3}{2}^\pm$ band. The second rise at $E^* \sim 6.3$ MeV corresponds to a significant rise in g_2 for all experiments and is most consistent with a band $K = \frac{1}{2}^\pm$.

3. ^{239}U (Figs. 6 and 9)

In this case the (d, pf) results are of rather poor quality because of the small fission probability for ^{239}U. The fission threshold is at a neutron energy $E_n \sim 1.5$ MeV and, therefore, the P_f distributions are approximately the same for (d, pf) and (n, f) results. The character and accuracy of the results do not allow any serious conclusions regarding the low-lying transition states for this nucleus.

4. ^{241}Pu (Figs. 5 and 9)

The major characteristic of these results is that in the region 5.3–5.8 MeV the fission probabilities observed in the (d, pf) and (t, pf) experiments are much larger than those observed in (n, f) experiments. This indicates strong contributions in this energy region from one or more transition state bands with $K = \frac{3}{2}^+$ or $\frac{5}{2}^+$ since these are the only bands which can give strong contributions in the (d, pf) and (t, pf) results and be weakly seen in (n, f) experiments. The weak apparent threshold ($P_f/P_{f\,\max} \sim 0.1$) at 5.2 MeV in the (d, pf) and (t, pf) experiments is most consistent with fission through a $K = \frac{5}{2}^-$ band and the negative value for g_2 in this region is consistent with this assignment (see Table II). The peak in g_2 for both (t, pf) and (d, pf) results in the region $E^* = 5.3-5.5$ MeV indicates significant contributions from fission through a $K = \frac{1}{2}^\pm$ band in this energy region.

5. ^{243}Pu (Figs. 6 and 9)

In this case both the (d, pf) and (n, f) results show a broad threshold near $E^* \sim 5.7$ MeV with relatively small values for g_2 in this region. The data are not good enough to make any serious conclusions but the small g_2 values in the region of large fission probability suggest a low-lying $K = \frac{3}{2}^\pm$ band.

B. $\langle \Gamma_n/\Gamma_f \rangle$ from (t, pf) Results

In Fig. 9 a comparison of the fission probabilities from the direct-reaction and neutron experiments shows that for neutron energies above about 1 MeV and above the fission threshold, the fission probabilities are the same to within ±10% for all the different experiments. In this energy region the differences between the (d, pf), (t, pf), and (n, f) fission probabilities are well within the uncertainties in the experiments and in the optical-model calculations used to determine P_f from the (n, f) cross sections. In the region $E_n = 1-2$ MeV the angular-momentum-transfer calculations described above indicate that the average angular momentum transfer for the (t, pf) reaction was 2–3 times greater than for the (n, f) reaction. The fact that the fission probabilities for the two cases are essentially the same indicates that the fission probability (and, therefore, $\langle \Gamma_n/\Gamma_f \rangle$) is not a strong function of angular momentum when there are a reasonable number of open fission channels. Values for $\langle \Gamma_n/\Gamma_f \rangle$ deduced from fission probabilities obtained in (t, pf) experiments are given in Table III and compared with values obtained in other experiments. These results indicate that under appropriate circumstances it should be possible to estimate (n, f) cross sections to the order of ±10% from measured fission probabilities.[19]

VI. SUMMARY

The results of the present experiment show that in cases where fission thresholds exist at excitation energies less than about 0.5 MeV above the neutron binding energy, a qualitative comparison of (d, pf) [or (t, pf)] results with (n, f) data can give information on the low-lying transition states, which

TABLE III. Comparison of $\langle\Gamma_n/\Gamma_f\rangle$ values obtained from analysis of fission probabilities for (t,pf) experiments with the compilation for $\langle\Gamma_n/\Gamma_f\rangle$ values obtained by Vandenbosch and Huizenga (see Ref. b) from other types of experiments.

Compound nucleus	(t,pf) experiments (±15%)	Vandenbosch and Huizenga Systematics	12-MeV photofission	3-MeV neutron fission
^{232}Th	16[a]	15	12	
^{234}Th	30[a]	40		
^{235}U	1.5	1.4	1.6	1.17
^{236}U	1.7[a]	1.8	2.1	1.64
^{237}U	3.3	3.0		3.01
^{238}U	4.6[a]	4.0	4.0	
^{240}U	6.1[a]	7.5		
^{241}Pu	1.4	1.0		0.74
^{242}Pu	1.3[a]	1.2		
^{244}Pu	1.8[a]	2.5		

[a]See Ref. 19.

[b]R. Vandenbosch and J. R. Huizenga, in *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1968* (United Nations, Geneva, Switzerland, 1968), Vol. 15, p. 284.

are important in the fission decay. This condition is met and some information is obtained for the transition-state spectra of ^{235}U and ^{241}Pu. For heavier uranium and plutonium isotopes the lowest fission thresholds are at higher neutron energies and there are enough possible channels open in the neutron experiments that very few conclusions can be drawn from a comparison of (d,pf) with (n,f) results. The results do not give unambiguous information on the transition-state spectra for any nucleus, but they may be useful in future attempts to quantitatively analyze the fission of these nuclei with a detailed model.

At neutron energies above ~1 MeV, in cases where there are a relatively large number of fission channels open, the fission probabilities obtained in (n,f), (t,pf), and (d,pf) experiments are remarkably similar even though the average angular momentum transfers for the (t,pf) reaction are 2–3 times greater than for the (n,f) reaction. These results suggest that the average ratio of fission to neutron widths $(\langle\Gamma_n/\Gamma_f\rangle)$ is not a strong function of angular momentum.

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Self-Consistent Calculations of Shell Effects Including the Proposed Island of Stability*

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A study is made of the characteristics of magic nuclei displayed in a Hartree-Fock calculation. It is seen that the known doubly-closed-shell nuclei are clearly distinguished by the behavior of the energy as a function of neutron and proton number. The existence of these characteristics for the superheavy nucleus with $Z=120$ and $N=178$ indicates that this may also be a magic nucleus. Single-particle-model calculations have indicated $Z=114$ as the magic nucleus. Possible reasons for this difference are discussed.

I. EXISTENCE OF SUPERHEAVY NUCLEI

The possibility of accelerating heavy ions (e.g., Ar^{40}) has resulted in much experimental and theoretical research on an island of stability with $Z>100$. Recent experimental progress has been reviewed by Flerov.[1] Though nuclei with $Z>105$ have not as yet been formed in these experiments, the existence of such stable nuclei might be determined when it becomes possible to accelerate the heavier ions.

Theoretical calculations have been performed[2-4] which indicated that an island of stability might exist in the region of $Z=114$, $N=184$. In general, the theoretical techniques applied consist of single-particle calculations of the Nilsson type[2] combined with certain features of the liquid-drop model. Though such calculations may well provide a device for extrapolating from the known nuclei to heavier nuclei, because of their uncertainties it would also be desirable to investigate less phenomenological methods for calculating properties of superheavy nuclei.

A step in this direction has been made in the approach taken by Meldner,[5] where a degree of self-consistency has been added to the single-particle-Hamiltonian method. The technique is, essentially, to solve the single-particle equation

$$\left(\frac{\hbar^2}{2m}\frac{\partial^2}{\partial r^2}-\epsilon_\mu\right)\varphi_{\mu,\tau_t}(\vec{r})=\int d^3r' K_{\tau_z}(\vec{r},\vec{r}\,')\varphi_{\mu,\tau_z}(\vec{r}\,')\,,\qquad(1)$$

where K, rather than being a single-particle potential directly derived from a two-body interaction, is assumed to be a nonlocal potential with a specific density dependence of the form

$$K_{\tau_z}(\vec{r},\vec{r}\,')=v(|\vec{r}-\vec{r}\,'|)\left\{1-\left[\frac{\rho_{\tau_z}(x)}{\rho_1}\right]^{2/3}\right\}\rho(x)\,.\qquad(2)$$

Here

$$x=\tfrac{1}{2}(|\vec{r}|+|\vec{r}\,'|)\,,$$

$$\rho_{\tau_z}=\sum_{\mu=1}^{N}|\varphi_{\mu,\tau_z}|^2\,,$$

and the nonlocality is contained in the factor v