PHYSICAL REVIEW VOLUME 181, NUMBER 4 20 MAY 1969

Fission-Fragment Angular Correlations for ^{233}U, ^{235}U, and ^{239}Pu(*t*, *df*) Reactions*

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(Received 4 November 1968)

Measurements have been made of the angular correlations of fission fragments emitted from the (t, df) reaction on ^{233}U, ^{235}U, and ^{239}Pu targets with an 18-MeV incident triton beam for deuterons observed at 130°. The results have been fitted to the function $W(\theta)=A_0[1+\Sigma_L g_L P_L(\cos\theta)]$, and coefficients A_0, g_2, g_4, and g_6 were determined as a function of excitation energy. Fission probabilities were determined from the A_0 coefficient. The results are generally very similar to previous (d, pf) results.

I. INTRODUCTION

IN a recent paper[1] attempts have been made to identify the low-lying vibrational bands in the transition-state spectra of ^{234}U, ^{236}U, and ^{240}Pu from an analysis of (d, pf) and (t, pf) angular-correlation results. One major difficulty in a detailed analysis of these results was lack of information about the direct reaction used to excite the fissioning nucleus. From the analysis of a single set of data it is difficult to isolate effects due to the direct-reaction formation process from those occurring in the fission decay process. A possible method for obtaining a better understanding of the direct-reaction fission process is to study the same fissioning nucleus while varying the characteristics of the direct-reaction process. Studies of this type have been performed for the fissioning system ^{236}U by two methods. A comparison was made between ^{235}U(d, pf) and ^{234}U(t, pf) results,[1] and a study of the ^{235}U(d, pf) reaction at various deuteron bombarding energies has been reported.[2]

Unfortunately, the number of available targets in the actinide region and the variety of direct reactions which have sufficiently large cross sections for depositing 4–8 MeV of excitation energy are severely limited. Therefore, there are only a few cases for which the same fissioning system can be produced by different direct reactions. In this paper we report results on studies of the (t, df) reaction which can be compared directly with previous (d, pf) results.[1] Basically, the (t, d) and (d, p) reactions are very similar, since both involve the transfer of a single neutron. However, there are several important differences between the two reactions. In particular, (t, d) reactions involve somewhat higher angular momentum transfers, and (t, d) spectra are less sensitive to light-element contaminants in the target because of the larger kinematic shifts. In addition, the (d, p) cross sections tend to show a monotonic increase with excitation energy for $E^*>6$ MeV. This increase is not predicted by distorted-wave-born-approximation calculations and may indicate that a significant fraction of the protons leading to excitation energies above 6 MeV might be coming from a compound-nucleus evaporation process rather than from a direct stripping process. If this were the case, the analysis of the (d, pf) results which assumes a direct stripping process could be in error. Because of the small binding energy for the deuteron, it should be extremely unlikely for the (t, d) reaction to proceed by means of a compound-nucleus evaporation process.

In this paper, we report experimental results on angular correlations and fission probabilities for the (t, df) reaction on targets of ^{233}U, ^{235}U and ^{239}Pu. These results are compared in detail to previous (d, pf) results.[1]

II. EXPERIMENTAL PROCEDURE

The experimental techniques were essentially identical to those of the previous experiment,[1] and only a few of the more important aspects will be repeated here. The experiments were performed using an 18.0-MeV triton beam from the Los Alamos Scientific Laboratory Van de Graaff accelerator facility. A multipurpose scattering chamber was used which could accommodate a ΔE-E charged-particle telescope and up to eight independent fission detectors. Deuteron energy spectra were obtained in coincidence with each of the fission detectors yielding a full (up to eight angles) angular correlation 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-drift detector of 3-mm thickness. The over-all resolution of the deuteron detection system was approximately 120 keV. The deuteron detector was collimated with a circular aperture and subtended an angle $\Delta\theta\sim15°$. The fission detectors were phosphorus-diffused semiconductor detectors of $\sim$400-Ω cm silicon which were operated at reverse biases of 100–200 V. Detectors of two sizes were used: 8×8 mm square and 8×20 mm rectangular. For angles near the recoil angle, the square detectors were used, and for fission detectors that were nearly perpendicular to the recoil direction, rectangular detectors were used. In the reaction plane the fission detectors subtended an angle $\Delta\theta=13°$.

The targets were prepared by vacuum evaporation

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* Work performed under the auspices of the U.S. Atomic Energy Commission.

[1] H. C. Britt, F. A. Rickey, Jr., and W. S. Hall, Phys. Rev. **175**, 1525 (1968).

[2] R. Vandenbosch, K. L. Wolf, J. Unik, C. Stephan, and J. R. Huizenga, Phys. Rev. Letters **19**, 1138 (1967).

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 isotopic compositions: ^{233}U—97.96, ^{234}U—1.37, ^{235}U—0.07, and ^{238}U—0.60%; ^{235}U—93.25, ^{234}U—1.03, ^{236}U—0.28, and ^{238}U—5.44%; and ^{239}Pu—94.41, ^{240}Pu—5.23, and ^{241}Pu—0.36%.

The data were obtained utilizing an SDS-930 on-line computer with final data reduction in a larger CDC 6600 computer as was described previously.[1] From the data analysis, coincidence 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, and then each spectrum is converted to a new spectrum of number of counts versus excitation energy in the residual nucleus (fissioning nucleus) with standard channel widths of 50 keV. The conversion to an excitation energy spectrum involves a deuteron 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.[3] For each excitation energy interval, the angle of each detector is calculated in the rest system of the fissioning nucleus (i.e., angle relative to kinematic recoil angle with c.m. correction). The statistical error on 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 18-MeV ^{240}Pu(t,f) reaction. The deuteron energy calibration of the ΔE-E system is determined from known energy deuteron groups for reactions on ^{12}C and ^{16}O. The reaction Q values were determined from (d,p) and (d,t) experiments[4,5] in the U isotopes and from mass tables[6] for the ^{239}Pu(t,df) reaction. The Q values used are as follows: ^{239}Pu(t,d_0), $Q_0=0.196$; ^{235}U(t,d_0), $Q_0=0.295$; and ^{233}U(t,d_0), $Q_0=0.578$.

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

$$W(\theta)=A_0\{1+\sum_{L=2,4,6} g_L P_L[\cos(\theta-\theta_0)]\},$$

where A_0, θ_0, and g_2, g_4, and g_6 were adjustable parameters, and the angles are measured in the rest system of the fissioning nucleus (i.e., relative to the kinematic recoil angle.) From the ^{239}Pu(t,df) reaction, an average value of θ_0 was determined. Final fits were obtained for all cases with θ_0 held fixed at $-2°$. The value $\theta_0=-2°$ was within the accuracy to which the zero position for the fission detector angular scale is known and, therefore, this measurement is not considered to be evidence for a difference between the symmetry angle of the fragment correlations and the kinematic recoil angle.

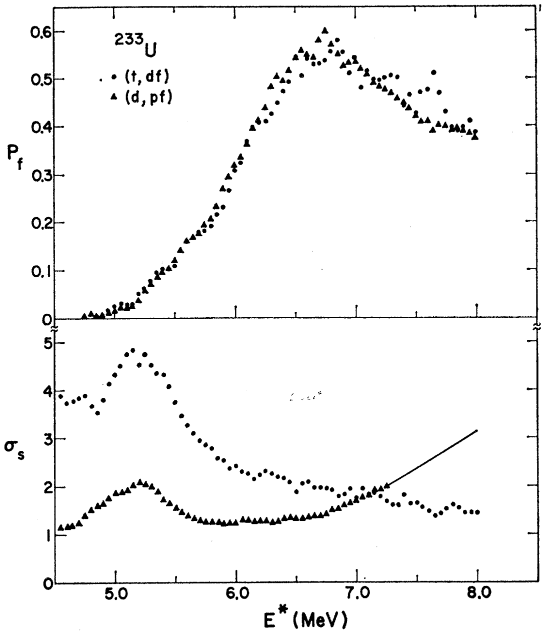

FIG. 1. Comparison of fission probability and singles distributions for the ^{233}U(t,df) with ^{233}U(d,pf) results from Ref. 1.

Fission probability distributions P_f were determined for each case by converting the coefficients A_0 to a fission cross section σ_f using measured absolute solid angles for the fission detectors and dividing by the singles-deuteron cross sections σ_s. Because of the larger kinematic shifts in the (t,d) reaction, there were no carbon or oxygen contaminant peaks in the excitation energy region of interest.

In all cases, deuterons were detected at an angle of 130°, which gives a corresponding kinematic recoil angle of $\theta_R \sim +20°$ for excitation energies near the fission threshold. Data were obtained for the ^{239}Pu(t,df) reaction at 13 fission angles from $-60°$ to $+160°$ in the laboratory system. For the ^{233}U and ^{235}U(t,df) reactions, data were obtained at seven fission angles from $+20°$ to $+160°$ in the laboratory system. The coincidence counting rates for the (t,df) reaction were approximately 10% of those obtained in (d,pf) measurements. Consequently, the (t,df) results have somewhat poorer statistical accuracy than previous (d,pf) results, except for ^{233}U, where the statistical accuracies are comparable for the two measurements.

III. RESULTS AND DISCUSSION

The singles cross sections and fission probabilities are shown in Figs. 1–3 for the present (t,df) results and the previous[1] (d,pf) results on ^{233}U, ^{235}U, and

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[3] H. C. Britt and F. Plasil, Phys. Rev. **144**, 1046 (1966).

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

[5] F. A. Rickey, Jr., and H. C. Britt (to be published).

[6] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **61**, 1 (1965).

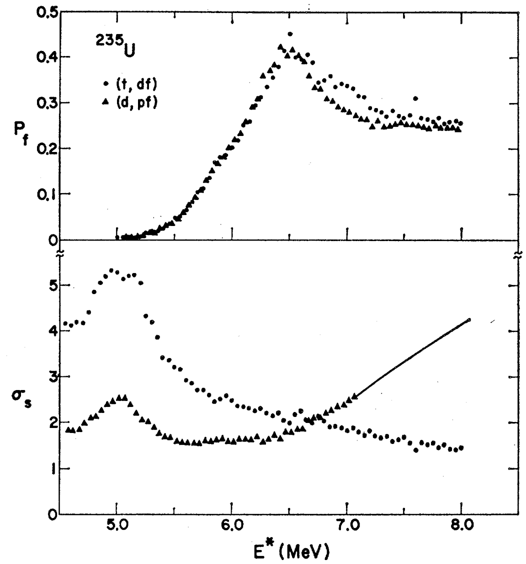

FIG. 2. Comparison of fission probability and singles distributions for the ^{235}U(*t, df*) with ^{235}U(*d, pf*) results from Ref. 1.

^{239}Pu. The (*t, d*) singles spectra show gross structure peaks at $E^*\approx 5$ MeV which are very similar to (*d, p*) spectra reflecting the common single-neutron transfer mechanism in the two reactions. However, the (*t, d*) results show a monotonic decrease in cross section with increasing excitation energy above $E^*\approx 6$ MeV as compared to an increasing (*d, p*) cross section in this region. This may be evidence that there is some compound-nucleus contribution to the (*d, p*) process at these higher excitation energies, but this conclusion is not very firm because of the differences expected in the stripping reactions due to the very different Q values involved in (*t, d*) and (*d, p*) reactions to the same excitation energy. One result of the different shape for the (*t, d*) singles spectra and the absence of carbon and oxygen contaminant peaks is that the structure near threshold in the fission probability distributions appears somewhat more well defined in the (*t, df*) results than in the (*d, pf*) measurements. In general, the fission probability distributions agree quite well with previous (*d, pf*) results, and the values are well within the estimated ±10% uncertainty on the absolute scale.

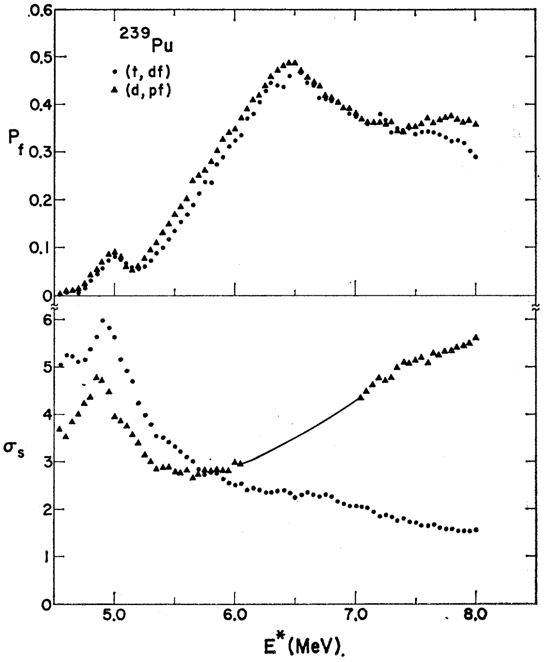

FIG. 3. Comparison of fission probability and singles distributions for the ^{239}Pu(*t, df*) with ^{239}Pu(*d, pf*) results from Ref. 1.

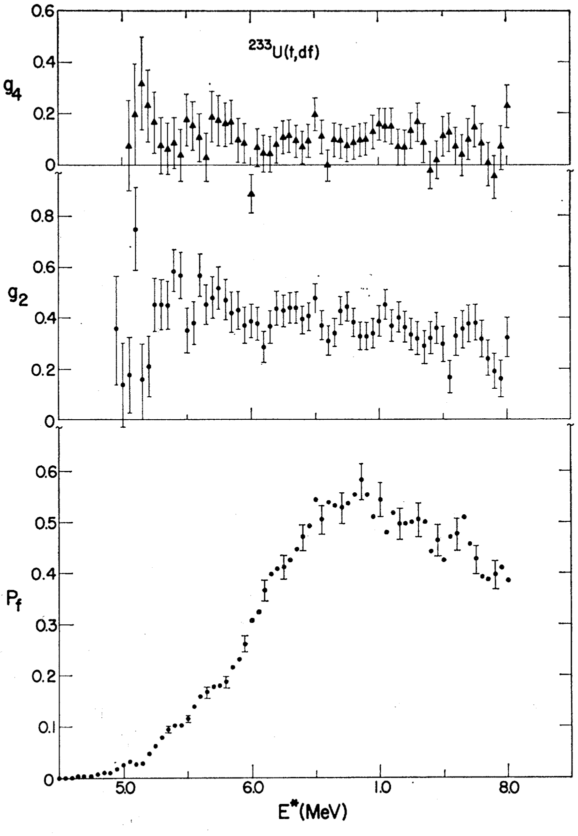

FIG. 4. Fission probability and angular-correlation coefficients (g_2 and g_4) distributions for the ^{233}U(*t, df*) reaction. Error bars represent statistical standard deviations.

The fission probabilities and angular-correlation coefficients g_2 and g_4 are shown in Figs. 4–6 for the three reactions. Figure 7 shows a comparison for (*t, df*) and (*d, pf*) results of the g_2 distributions for the three reactions studied. In general, it is seen that P_f and g_2 distributions obtained in the (*t, df*) reactions are very similar to previous (*d, pf*) results. In particular, the general structure observed in the P_f and g_2 distributions from (*d, pf*) measurements is faithfully reproduced in the (*t, df*) results. Small differences in the distributions will be discussed in detail below.

In order to facilitate discussion of the differences between the (*d*, *pf*) and (*t*, *df*) results, a comparison of calculated distributions of orbital angular momentum transfers for the (*t*, *d*) and (*d*, *p*) reactions is shown in Fig. 8. The orbital angular momentum transfers were calculated with the code JULIE[7] using measured parameters[1,5] for an excitation energy of 5 MeV. The previous (*d*, *pf*) results were obtained at deuteron bombarding energies of 15 MeV for ^{239}Pu and 18 MeV for ^{233}U and ^{235}U. It can be seen from Fig. 8 that the differences between the distributions of angular momentum transfers in the various reactions are not very great. The calculated distribution for the (*t*, *d*) reaction at 18 MeV is approximately equivalent to that which would be obtained for a (*d*, *p*) reaction at ~21 MeV. In the following, comparisons between (*t*, *df*) and (*d*, *pf*) results for the specific targets are discussed.

A. ^{233}U (Figures 1 and 4)

In the (*t*, *df*) reaction, the structure in the fission probability distribution in the region 5–6 MeV appears more pronounced than in (*d*, *pf*) results. The structure between 5 and 5.5 MeV may be accentuated in the (*t*, *df*) reaction because of the more prominent gross structure observed in the (*t*, *d*) singles spectrum.

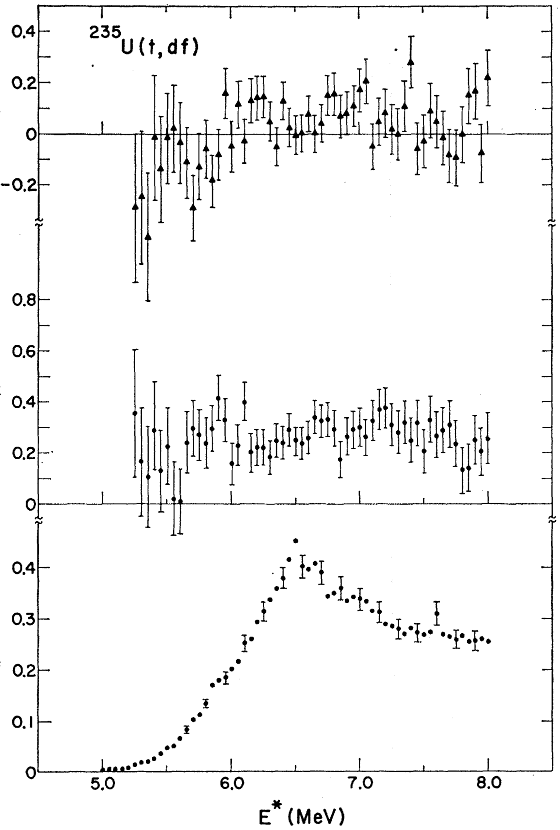

FIG. 5. Fission probability and angular-correlation coefficient (g_2 and g_4) distributions for the ^{235}U(*t*, *df*) reaction. Error bars represent statistical standard deviations.

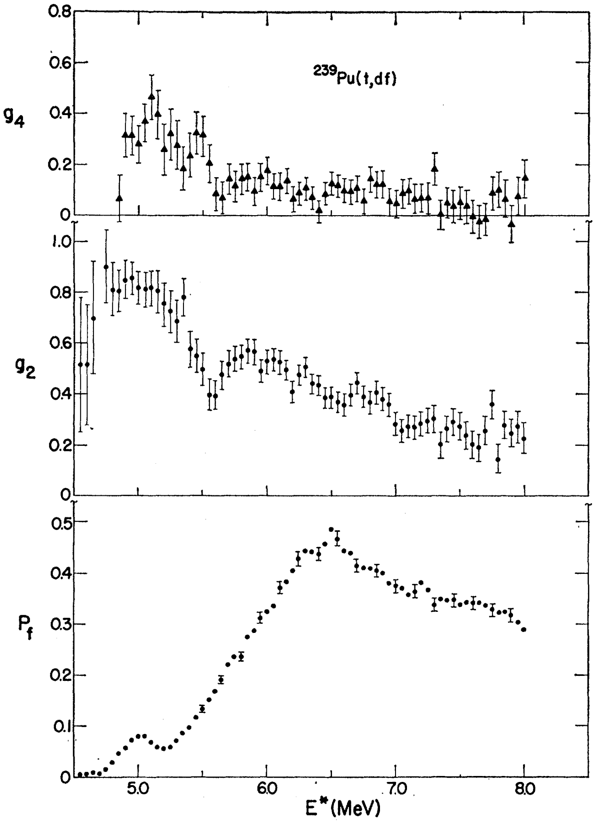

FIG. 6. Fission probability and angular-correlation coefficient (g_2 and g_4) distributions for the ^{239}Pu(*t*, *df*) reaction. Error bars represent statistical deviations.

In the (*t*, *df*) results, the g_2 distribution shows a definite decrease in g_2 as the excitation energy is decreased below threshold. This effect is predicted because of the separation in energy of the states in the $K=0^+$ band of the transition-state spectrum and was observed in the (*d*, *pf*) results on ^{239}Pu and ^{235}U. However, a decrease was not observed in the ^{233}U(*d*, *pf*) results. The reason for not observing the effect in the ^{233}U(*d*, *pf*) results is still not clear but it may be due to the poorer statistical accuracy of the ^{233}U(*d*, *pf*) data in the threshold region. At present, there does not appear to be any apparent physical reason for a difference of this type between the (*t*, *df*) and (*d*, *pf*) results. Above threshold the g_2 distributions are identical for the two reactions to within the statistical accuracies, as is seen in Fig. 7.

B. ^{235}U (Figures 2 and 5)

In this case the P_f distribution for the (*t*, *df*) results show a definite indication of two thresholds which were present in the fit to the ^{235}U(*d*, *pf*) results, but

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[7] We are indebted to R. M. Drisko and R. H. Bassel (Oak Ridge National Laboratory) for supplying us with a copy of this code.

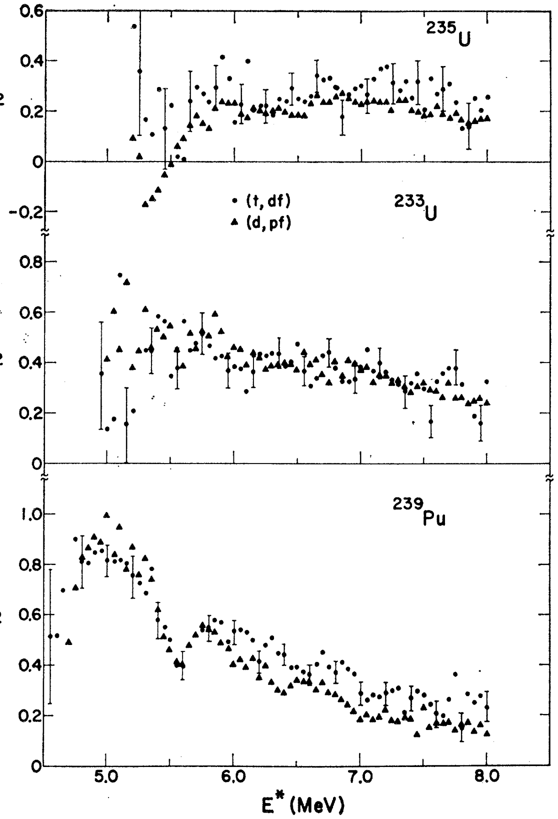

FIG. 7. Comparison of g_2 angular-correlation coefficients from (t, df) reactions with (d, pf) results from Ref. 1. Representative error bars are shown for (t, df) results. For ^{235}U and ^{239}Pu, the error bars for the (d, pf) measurement are similar or smaller. For ^{233}U, the (d, pf) results have larger error bars near threshold (5–6 MeV) than do the present (t, df) measurement (see Ref. 1).

not so apparent in the experimental distribution. The statistical accuracy in the g_2 distribution is much poorer in the (t, df) case, but the results do indicate that the values of g_2 do not drop as significantly at energies below threshold as they do in previous (d, pf) results[1,2] at $E_d = 12$–18 MeV. The (t, df) results appear most similar to previous (d, pf) measurements at 21 MeV,[2] which is consistent with the similarity between the average angular momentum transfers in the two reactions. Qualitatively, these results are consistent with the explanation proposed[2] for the negative values of g_2 observed below threshold in (d, pf) measurements at deuteron energies 12–18 MeV. Again at excitation energies above threshold the (t, df) and (d, pf) results[1] agree to within the statistical accuracies.

C. ^{239}Pu (Figures 3 and 6)

In this case the structure observed in both the P_f and g_2 distribution for the (d, pf) reaction is confirmed by the (t, df) reaction. The only significant differences are in the g_2 distributions where the (t, df) results give a slightly lower value for g_2 at $\sim$5 MeV and a greater value of g_2 for $E^* > 6$ MeV. The larger values of g_2 are consistent with the higher average angular momentum transfers (see Fig. 8) in the (t, df) reaction as compared to the 15-MeV (d, pf) reaction [previous ^{233}U and ^{235}U(d, pf) data were obtained at 18 MeV, so that smaller differences would be expected in the g_2 distributions as is observed in Fig. 7]. The reason for the smaller values of g_2 near threshold is not clear but it could be an indication of a small breakdown in the plane-wave approximation[1,8] for the (t, df) reaction at 18 MeV and $\theta_d = 130°$. A deviation from plane-wave predictions would be most apparent near threshold, where predominantly $K = 0$ transition states are involved.[1]

IV. CONCLUSIONS

In general, the results from (t, df) and previous (d, pf) measurements are found to be very similar. Experimentally, there are significant differences between the (t, df) and (d, pf) experiments (counting rates, energies of contaminant peaks in the singles spectra, etc.), and thus the agreement between the experiments tends to increase confidence in the complex experimental techniques used for these measurements. The difference in the shapes of the singles spectra from (d, p) and (t, d) reactions suggests that there may be some compound-nucleus processes contributing to the (d, p) reaction at excitation energies above 6 MeV, but the general agreement in the coincidence data in this region indicates that the assumption of a direct-reaction mechanism for the (d, p) process should not lead to erroneous conclusions.

If the simplified model used to fit previous (d, pf) measurements[1] is applied to the present (t, df) results, all of the conclusions regarding positions and identifica-

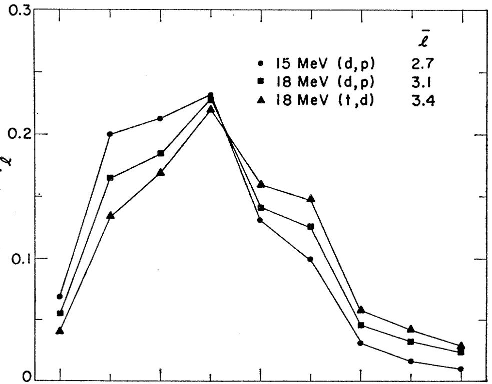

FIG. 8. Relative cross sections $(2l+1)\phi_l$ for various orbital angular momentum transfers calculated with the code JULIE for (d, p) and (t, p) reactions on uranium targets. Cross sections have been normalized to give $\Sigma_l(2l+1)\phi_l = 1$.

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[8] K. L. Wolf, R. Vandenbosch, and W. D. Loveland, Phys. Rev. **170**, 1059 (1968).

tions of the vibrational bands in the transition-state spectra of ^{234}U, ^{236}U, and ^{240}Pu remain unchanged. Somewhat different statistical parameters do result from detailed fits to the (t, df) data. However, at present, the simplified model is too qualitative to exploit the small differences between the (d, pf) and (t, df) results. At the present time, refinements to the earlier simplified model are limited by the lack of precise information on the direct-reaction process and the fission decay through a barrier that is most likely much more complex than was previously believed.

ACKNOWLEDGMENTS

We would like to thank Frank A. Rickey, Jr., for suggestions and assistance in obtaining the results presented in this paper. We gratefully acknowledge the help of W. S. Hall in developing and running the codes used for final analysis of the results, and Mrs. Judith C. Gursky and J. G. Povelites for preparing the targets used in the experiments. We are grateful to J. S. Levin and M. P. Kellogg for developing the computer programs used in the on-line data reductions.

PHYSICAL REVIEW VOLUME 181, NUMBER 4 20 MAY 1969

^{208}Pb(n, γ) Cross Sections by Activation between 10 and 200 keV*

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(Received 9 January 1969)

The ^{208}Pb(n, γ)^{209}Pb activation cross section has been measured in the energy range relevant to stellar nucleosynthesis. A large-area, low-background β-ray counting system was developed to obtain sufficient sensitivity. Information on the radiative widths of the known resonances below 120 keV was derived. The cross-section average found for the Maxwellian temperature distribution assumed for a stellar interior (kT= 30 keV) is $\frac{1}{3}$ mb. This value is quite compatible with expectations based on current nucleosynthesis theory.

INTRODUCTION

THE ^{208}Pb(n, γ) cross section at stellar interior temperatures is particularly important in determining the past history of the heavy elements found in the solar system.[1] It is the most abundant of the nuclei at the end of the s-process[2] neutron-capture chain. These recycle through α decay as indicated below:

$$^{206}\mathrm{Pb}(n,\gamma)^{207}\mathrm{Pb}(n,\gamma)^{208}\mathrm{Pb}(n,\gamma)^{209}\mathrm{Pb}(\beta)$$
$$\rightarrow{}^{209}\mathrm{Bi}(n,\gamma)^{210}\mathrm{Bi}(\beta+\alpha)^{206}\mathrm{Pb}.$$

The r process[2] also leads directly to these nuclei. It also produces a substantial number of trans-bismuth elements. Of these latter in the solar system only ^{232}Th, ^{238}U, and a little ^{235}U have survived the past 4.5×10^9 years in appreciable quantities. The rest have decayed by well known chains of successive α and β radioactivity to additional lead and bismuth. Thus the lead and heavier elements we find on earth and in meteorites should preserve through their isotopic abundances, evidence of the extent and duration of the processes leading to the formation of our heavy elements.

The high abundance of ^{208}Pb produced in the s process of neutron capture is directly related to its extraordinarily low cross section. This in turn is related to closed major nuclear shells of 82 protons and 126 neutrons. Experimentally this has made even the detection of the ^{208}Pb(n, γ) reaction very difficult for neutrons in the relevant energy range.[3] It is imperative to determine the ^{208}Pb(n, γ) cross section in order to aid in the unscrambling of the contributors to the solar system ^{208}Pb abundance.

METHOD AND APPARATUS

To improve the sensitivity and discrimination against spurious events we adapted the classic induced-radioactivity method to measuring the ^{208}Pb(n, γ)^{209}Pb yield. The ^{209}Pb product nuclei decay by weak β-ray emission ($E_{\max}$=635 keV, mean life 285 min). The absence of a γ ray in the decay required pressing and rolling the activated samples into thin foils and the development of a high-efficiency, large-area, low-background β-ray counter. As such thin foils were far from uniform, the β-ray self-shielding of each foil was determined by activation in a known thermal neutron flux. To obtain quantitative β-counting results, the thermal ^{208}Pb(n, γ)^{209}Pb cross section was also redetermined.[4]

The β-ray counter we developed used thin discs of plastic scintillator[5] (0.125-cm$\times$11-cm diam) as the detecting material. Such a disc was mounted on a 1-cm-thick quartz plate atop a 5-in. photomultiplier and the outer face covered with a thin (0.001 cm) light-

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

[1] D. D. Clayton, in Proceedings of the Symposium on the Origin and Distribution of the Elements, Paris, 1967 (unpublished).

[2] E. M. Burbidge, G. R. Burbidge, W. A. Fowler, and F. Hoyle, Rev. Mod. Phys. **29**, 547 (1957).

[3] R. L. Macklin and J. H. Gibbons, Phys. Rev. **159**, 1007 (1967); see, also, Astrophys. J. **150**, 721 (1967).

[4] This was done by J. Emery of Oak Ridge National Laboratory, who is publishing his results separately.

[5] NE-102 from Nuclear Enterprises, Inc.