Further studies of pairing excitations in actinide nuclei: ^{233}U, ^{237}Pu, ^{235}Np, ^{241}Am, ^{224}Ra, and ^{238}Pu†

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(Received 30 October 1973)

The behavior of a pairing excitation, previously reported in even actinide nuclei, has been explored with two odd-neutron (^{235}U and ^{239}Pu) and two odd-proton (^{237}Np and ^{243}Am) targets as well as the even-even targets ^{226}Ra and ^{240}Pu. It is found that this behavior is similar to that previously found in even-even actinide nuclei.

[NUCLEAR REACTIONS ^{226}Ra, ^{235}U, ^{237}Np, ^{239}Pu, ^{243}Am, ^{240}Pu(p, t) measured $\sigma(\theta)$, deduced excitations and cross sections of pairing excited states.]

I. INTRODUCTION

Some three years ago the systematic observation of an excited 0^+ state was reported from a survey of (p, t) reactions on even actinide targets.[1] Such a 0^+ state was observed for all targets at ~15% of the ground-state cross section. It has been suggested[2] that this could be the results of a quadrupole splitting of the pairing interaction where the removal of a prolate pair would lead to the normal ground state of the final nucleus while the removal of an oblate pair leaves the excited 0^+ state. This qualitative picture seemed consistent with the experimental observation that the excited 0^+ state was not seen in the pair-adding (t, p) reaction,[3] since theavailable low-lying oblate orbitals are generally fully occupied.

In the present experiments we undertook to study the coupling of this pairing excitation to an odd neutron and an odd proton. The experimental conditions were identical to the ones described in Ref. 1 and the measurements were extended to targets of ^{235}U, ^{239}Pu, ^{237}Np, and ^{241}Am as well as the additional even targets of ^{226}Ra and ^{240}Pu.

II. EXPERIMENTAL METHODS

Targets of the various elements were prepared by use of the Argonne isotope separator. A layer of 50–150 μg/cm^2 of the target isotope was deposited on laminated 30–40 μg/cm^2 carbon films by use of the retarded beam of the separator. In general the deposit was focused to a 0.5-mm ×5-mm line to minimize the amount of radioactivity handled. The only exception to this procedure was the preparation of the ^{226}Ra targets. These were prepared by vacuum sublimation of $RaCl_2$ or RaF_2 to a thickness of about 25 μg/cm^2 since it appeared that use of these compounds minimized the emanation of ^{222}Rn from the targets. All data were taken at a proton energy of 16.5 MeV using the Argonne FN tandem and Enge split-pole magnetic spectrograph. The target thicknesses were determined by measuring forward-angle proton elastic scattering yields. In general the experimental procedures were similar to those given in Ref. 1.

III. EXPERIMENTAL RESULTS

Table I lists the excitation energies, cross sections at 15°, and assignments for the states observed in the various reactions. Figure 1 illustrates typical triton spectra for the ^{235}U(p, t)-^{233}U reaction, and Fig. 2 illustrates the angular distribution of the cross sections populating the various members of the ground-state rotational band in the ^{237}Np$(p, t)^{235}$Np reaction. Figures 3 and 4 illustrate the angular distributions for the $\Delta L = 0$ transitions observed for the various targets.

As can be seen by inspection of the figures, the unique character of the $\Delta L = 0$ angular distribution makes assignment of these transitions unambiguous; however, this is not true for the $\Delta L = 2$ and $\Delta L = 4$ cases.

IV. DISCUSSION OF RESULTS

For ^{239}Pu, with spin $\frac{1}{2}^+$ and having the $\frac{1}{2}^+[631]$ orbital at ground, the (p, t) reaction was found to populate the 145-keV $J^\pi = \frac{1}{2}^+$ level of the $\frac{1}{2}^+[631]$ orbital in ^{237}Pu. Only one other $\Delta L = 0$ transition

TABLE I. Levels observed in (p,t) reactions.

Residual nucleus	E_x (keV)	J^π	ΔL	$\left.\frac{d\sigma}{d\Omega}\right\|_{15°}$ (μb/sr)
^{224}Ra	0	0^+	0	198 ± 26
	84 ± 2	2^+ [a]	2	31 ± 4
	251 ± 3	(4^+) [b]	4	19 ± 3
	477 ± 2			6 ± 1
	918 ± 3	0^+	0	16 ± 3
	1223 ± 4	0^+	0	12
	1627 ± 3			13 ± 3
	1761 ± 4			8 ± 1
	1949 ± 4			2 ± 1
^{233}U	318 ± 1.5	$\frac{7}{2}^-$	0	49 ± 6
	352 ± 2		2	7 ± 2
	(396)			5 ± 4
	502 ± 1.5	$\frac{7}{2}^-$	0	250 ± 27
	567 ± 2		2	27 ± 3
	646 ± 3		4	13 ± 2.5
	819 ± 2	$\frac{7}{2}^-$	0	32 ± 4
	865 ± 3			3 ± 0.5
	923 ± 2		(2)	11 ± 2
	982 ± 2		(2)	15 ± 2.5
	1824 ± 3	$\frac{7}{2}^-$	0	18 ± 2
	2021 ± 4			12 ± 2
	2070 ± 3	$\frac{7}{2}^-$	0	11 ± 2
^{235}Np	0	$\frac{5}{2}^+$	0	240 ± 21
	32 ± 1	$(\frac{7}{2}^+)$ [b]	2	25 ± 3
	77 ± 2	$(\frac{9}{2}^+)$ [b]	(2,4)	21 ± 3
	133 ± 2	$(\frac{11}{2}^+)$ [b]	4	11 ± 1
	201 ± 1.5	$(\frac{13}{2}^+)$ [b]	4	7 ± 3
	834 ± 3.5	$\frac{5}{2}^+$	0	17 ± 2
	962 ± 2			9 ± 2
	998 ± 1		2	4 ± 1.5
	1024 ± 1.5		2	4 ± 1.5
	1260 ± 1.5	$\frac{5}{2}^+$	0	13 ± 2
^{235}Np	1293 ± 6		2	3 ± 1
	1818 ± 2	$\frac{5}{2}^+$	0	7 ± 2
^{237}Pu	145 ± 1.5	$\frac{1}{2}^+$	0	182 ± 16
	198 ± 5	$\frac{5}{2}^+$ [a]	2	31 ± 5
	222 ± 5	$(\frac{7}{2}^+)$ [b]	(2,4)	10 ± 2
	304 ± 4	$(\frac{9}{2}^+)$ [b]	(4,6)	10 ± 2
	407 ± 1			4 ± 1
	800 ± 2	$\frac{1}{2}^+$	0	37 ± 2
	851 ± 4.5		2	8 ± 1.5
	998 ± 5			11 ± 2
	1025 ± 3		(2,4)	7 ± 1.5
^{238}Pu	0	0^+	0	150 ± 15
	45 ± 1	2^+ [a]	2	41 ± 3
	146 ± 2	4^+ [a]	4	24 ± 1
	306 ± 2	6^+ [a]	6	4 ± 0.5
	945 ± 3	0^+	0	16 ± 1
	985 ± 2		2	8 ± 1
	1032 ± 2		2	9 ± 1
	1134 ± 4	(0^+)	(0)	2 ± 0.5
	1252 ± 2			2 ± 0.5
^{241}Am	0	$\frac{5}{2}^-$	0	245 ± 20
	42 ± 2	$\frac{7}{2}^-$ [a]	2	46 ± 5
	94 ± 2	$\frac{9}{2}^-$ [a]	(2,4)	28 ± 4.5
	158 ± 1.5	$\frac{11}{2}^-$ [a]	4	13 ± 3
	234 ± 1.3	$(\frac{13}{2}^-)$ [b]	4	4 ± 1
	952 ± 1	$\frac{5}{2}^-$	0	17 ± 2.5
	982 ± 2			4 ± 1
	1136 ± 3			2 ± 1
	1550 ± 4	$(\frac{5}{2}^-)$	(0)	8 ± 1

[a] Spin and parity assignment made by comparison with known level energies given in *Table of Isotopes*, edited by C. M. Lederer, J. M. Hollander, and I. Perlman (Wiley, New York, 1967), 6th ed.

[b] Spin and parity assignment made on the basis of fit to the rotational energy sequence.

was seen, and this to a state at 801 keV with 19% of the intensity of the transition to the 145-keV state.

For ^{235}U$(p,t)^{233}$U the situation is more complex. The target spin is $\frac{7}{2}^-$($\frac{7}{2}^-$[743] orbital) and the transition proceeds strongly to a state, presumably the $\frac{7}{2}^-$[743] in ^{233}U, at 502 keV excitation. Also seen is a state at lower excitation, 318 keV, which is probably the $\frac{7}{2}$ member of the $K=\frac{5}{2}$ band, ($\frac{5}{2}^-$[743]), Coriolis mixed with the $\frac{7}{2}^-$[743] state. A calculation of the Coriolis mixing indicates that these states should be mixed to ~25%. Another $\Delta L=0$ transition is seen to a state at 820 keV with 11% of the summed strength of the first two, and two weaker states at 1824 and 2070 keV with ~5% strength each.

For the two odd-proton targets the principal $\Delta L=0$ transition was observed to proceed to the ground state of the final nucleus with the excited $\Delta L=0$ strength split between 835-keV (7%) and 1261-keV (5%) states for ^{235}Np and a 953-keV (7%) state, as well as a possible state at 1550 keV (3%), for ^{241}Am.

For the odd-proton nuclei the excited $\Delta L=0$ transition comes about as high above the ground state as in the corresponding even-even nuclei.

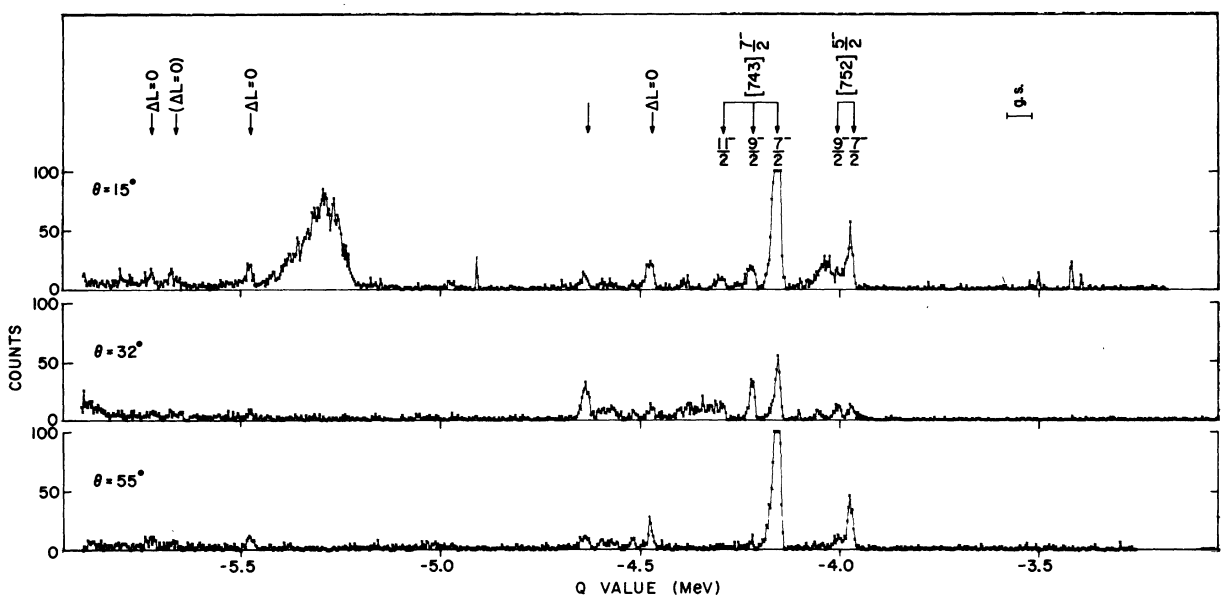

FIG. 1. Triton spectra observed in the $^{235}U(p,t)$ reaction at 16.5 MeV.

This is demonstrated in Fig. 5 where the energy of these states is shown together with all other known even-neutron actinide nuclei.

For odd-neutron nuclei the excited $\Delta L = 0$ fragment appears to be much closer to the lowest $\Delta L = 0$ transition. The systematic trend in Q values is plotted in Fig. 6 for the uranium and plutonium isotopes.

It is clear from this figure that it is the Q values for the lowest $\Delta L = 0$ transition that show an anomaly for odd neutron number; the excited-state Q values do not show such an anomaly. While the major part of the displacement of the Q values of the lowest $\Delta L = 0$ transition id sue to blocking and the nonuniform spacing of the single-particle orbitals, it is not readily apparent why these same effects should cancel exactly in the transitions to the excited state.

We have included on Fig.6 the Q values calculated from mass tables[4] for the $^{237}U(p, t)^{235}U$ reaction for the lowest $\Delta L = 0$ transition between the $\frac{1}{2}^+[631]$ levels which are at essentially ground-state energies in both nuclei. We have also included the calculated Q value to the state at 771 keV in ^{235}U which was assigned as having $J^\pi = \frac{1}{2}^+$ by Bjornholm *et al.*[5] Braid *et al.*[6] in the $^{234}U(d, p)^{235}U$ and $^{236}U(d, t)^{235}U$ populated a state at this energy (768 ± 3 keV), and tentatively assigned this level to the β vibrational band built on the $\frac{1}{2}^+[631]$, noting that the cross section for populating this level was ~50% of that of the $\frac{1}{2}^+[631]$.

However, this is what one might expect from an explanation in which the paring excited state involves removal of quanta far away from the Fermi surface. The blocking on one of the valence orbits by an odd neutron causes a serious perturbation in the paring energy as one may see in the anomalous behavior of the Q value for the lowest $\Delta L = 0$

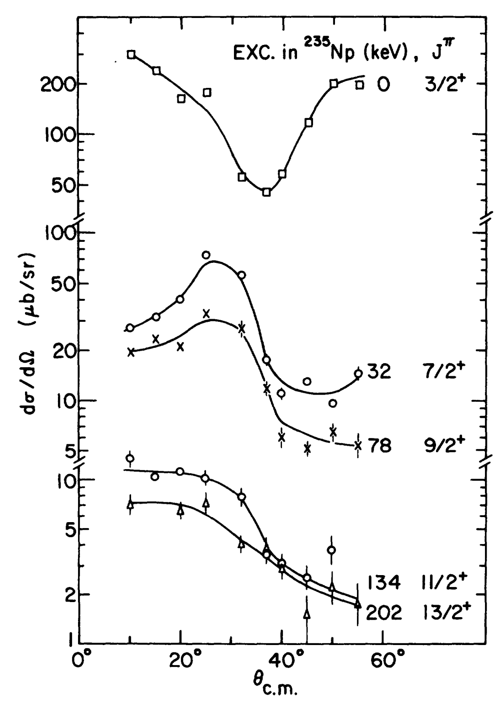

FIG. 2. Angular distributions observed for populating the members of the ground-state rotational band in ^{235}Np.

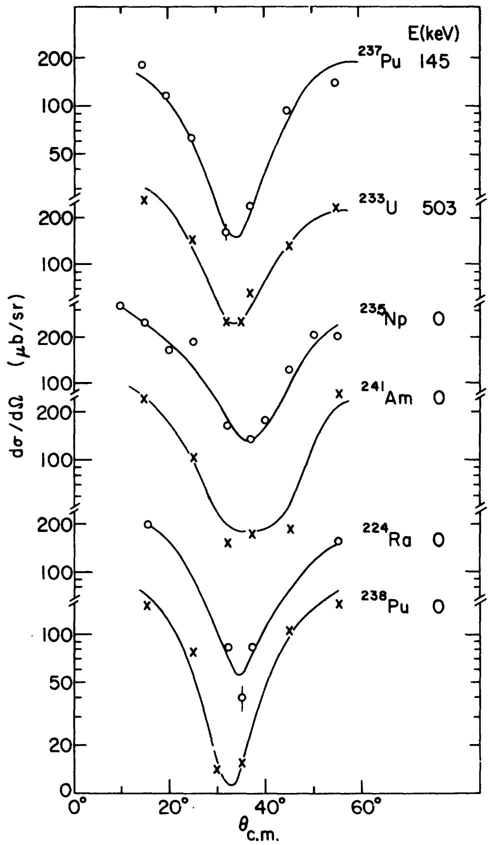

FIG. 3. Angular distributions observed for the major $\Delta L = 0$ transitions.

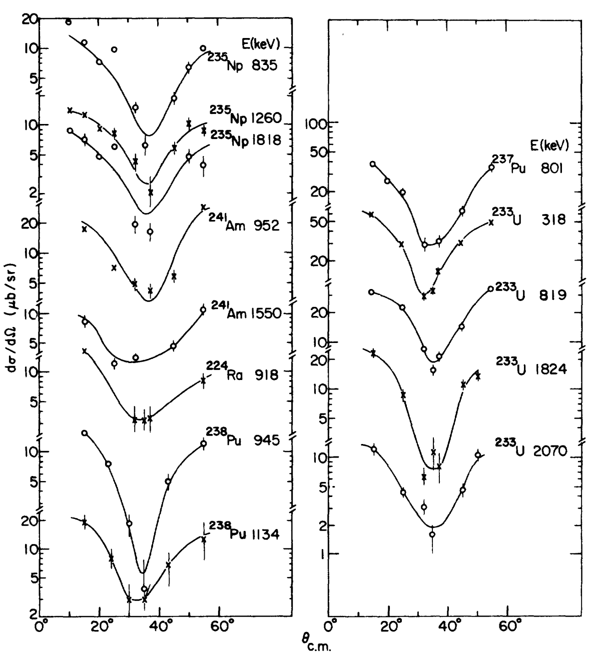

FIG. 4. Angular distributions observed for the excited $\Delta L = 0$ transitions.

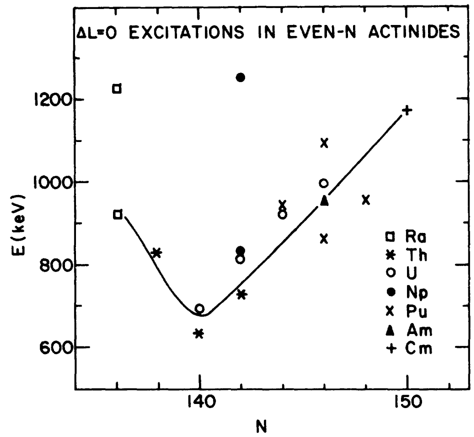

FIG. 5. Excitation energies of the excited $\Delta L = 0$ transitions in even-N actinide nuclei.

transition. No trace of this anomaly is found in the states of higher excitation, either in ^{233}U, ^{235}U, or ^{237}Pu, suggesting that the odd-neutron orbit plays no role in this excitation. Clearly this result is consistent with the qualitative picture of nonuniform pairing,[2,3] but as yet there is no quantitative fit with theory. One's confidence in the detailed model would be greater if it could also describe the systematic trend in the energy of the pairing excited states, i.e., those states populated by the higher excitation $\Delta L = 0$ transitions.

The fact that no such anomaly is seen with an odd proton is consistent with the assumption that the excited $\Delta L = 0$ state seen in a two-neutron transfer reaction does not involve the proton valence orbits differently for the ground state and pairing excited state.

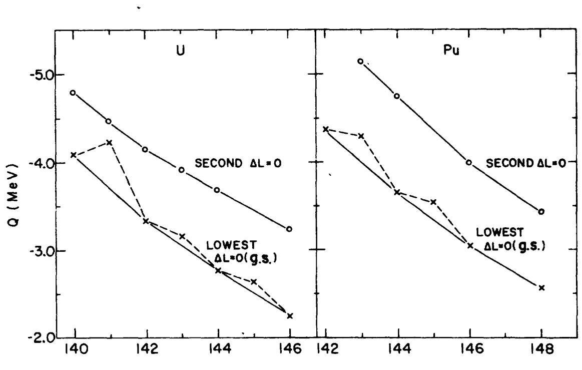

FIG. 6. Q values for the first two $\Delta L = 0$ transitions of the (p,t) reactions in actinide nuclei. It should be noted that the value for the $^{237}\text{U}(p,t)^{235}\text{U}$ reaction is calculated from the data for the $^{235}\text{U}(t,p)^{237}\text{U}$ reaction (Ref. 7) and the excitation energies of the $\frac{1}{2}^+[631]$ state in ^{235}U (Ref. 5).

V. INTERPRETATION OF CROSS SECTIONS

A distorted-wave Born-approximation (DWBA) calculation with the code TWOPAR[7] indicates that the magnitude of the cross sections for the $\Delta L = 0$ transition depends on the form factors and therefore the exact description of the states involved. However, our calculations indicate that the dependence of the $\Delta L = 0$ cross sections on Q value is similar for states with different form factors, and is affected strongly by the Coulomb barrier of the outgoing triton. In order to correct for the change in cross sections due to differences in Q value we have used the code TWOPAR to calculate the DWBA cross sections for each of the $\Delta L = 0$ transitions observed in these experiments and those given in Ref. 1.

The distorted-wave calculations were done using as typical form factors those derived from wave functions of a pair of nucleons in either the $4s_{1/2}$ or $3f_{7/2}$ orbitals coupled to $\Delta L = 0$. The orbital binding energies were adjusted to give the correct ground-state Q value. It was found that over the excitation energy range of our experiments the Q dependence of these calculated cross sections was the same using either form factor.

TABLE II. $\Delta L = 0$ "reduced cross sections" calculated by TWOPAR.

Residual nucleus	Excitation E (keV)	Expt $\frac{d\sigma}{d\Omega}\Big\|_{60°}$ (μb/sr)	Beam E (MeV)	S_{4s}	R
^{224}Ra	0	155 ± 25	16.5	0.85	
	918	8 ± 2		0.07	0.08 ± 0.03
	1223	13 ± 2		0.14	0.18 ± 0.03
^{228}Th	0	210 ± 40	17.0	1.23	
	830	42 ± 9		0.36	0.29 ± 0.06
^{230}Th	0	304 ± 40	17.0	2.08	
	636	73 ± 9		0.75	0.36 ± 0.05
	1590	14 ± 4		0.23	0.12 ± 0.02
^{232}U	0	140 ± 35	17.0	1.39	
	692	21 ± 6		0.28	0.21 ± 0.08
^{233}U	318	49 ± 6	16.5	0.55	0.20 ± 0.03
	502	220 ± 25		2.65	
	819	22 ± 3		0.31	0.11 ± 0.02
	1824	13.5 ± 2		0.40	0.15 ± 0.02
	2070	10 ± 2		0.42	0.15 ± 0.02
^{234}U	0	260 ± 30	17.0	1.86	
	812	27 ± 6		0.28	0.15 ± 0.04
^{236}U	0	260 ± 30	17.0	1.61	
	920	37 ± 8		0.35	0.22 ± 0.05
^{235}Np	0	200 ± 20	16.5	2.22	
	834	9.5 ± 2		0.15	0.07 ± 0.01
	1260	11.5 ± 2		0.27	0.12 ± 0.015
	1818	4 ± 1		0.20	0.09 ± 0.01
^{237}Pu	145	137 ± 20	16.5	2.36	
	800	34 ± 5		0.80	0.34 ± 0.06
^{238}Pu	0	160 ± 20	16.5	1.84	
	945	12 ± 2		0.25	0.13 ± 0.02
	1134	2 ± 0.5		0.05	0.03 ± 0.01
^{240}Pu	0	190 ± 30	17.0	1.41	
	862	52 ± 10		0.58	0.40 ± 0.10
	1091	20 ± 5		0.27	0.18 ± 0.04
^{241}Am	0	275 ± 40	16.5	3.05	
	952	32 ± 9		0.55	0.18 ± 0.03
	1550	11 ± 3		0.35	0.11 ± 0.02
^{246}Cm	0	170 ± 30	17.0	1.37	
	1176	25 ± 6		0.36	0.26 ± 0.06

The measured cross sections (at 60° shown in column 3) divided by the calculated one, using the $4s_{1/2}$ form factor, are shown in Table II in the column labeled S_{4s}. The calculations using the $3f_{7/2}$ form factor yielded uniformly lower values of this "reduced cross section," $S \equiv \sigma_{exp}/\sigma_{DWBA}$, by $20 \pm 5\%$ than those with the $4s_{1/2}$ form factor. Only the latter are shown. The column labeled R is the average value of these "reduced cross sections" for the various $\Delta L = 0$ transitions in each nucleus divided by the "reduced cross section" for the major $\Delta L = 0$ transition in that nucleus. Also tabulated is the beam energy used in each measurement.

It should be realized that the previous calculation is only a correction for the binding energies and kinematics of the reaction, and will only serve to approximate the form factors, which of course arise from a coherent sum of many states. Also, while the ratios of the $\Delta L = 0$ cross sections in a given nucleus are quite accurate, the absolute values of the ground-state cross sections have a much greater error.

The optical potentials used in the above calculations are:

	V_0(MeV)	r_0(fm)	a(fm)	W(MeV)	r_0'(fm)	a_0'(fm)	V_{so}(MeV)	r_{so}(fm)	a_{so}(fm)
p	57.0	1.26	0.6	10.0	1.26	0.6	0	0	0
t	155.0	1.29	0.7	0	0	0	40.0	1.29	0.7

In the case of a target such as ^{237}Np which has $j \neq 0$, the transfer reactions can proceed by admixtures of L waves. This may perhaps be observed in the transition to the ground state of ^{235}Np in Fig. 2. The $\Delta L = 0$ transition is somewhat more asymmetric than is usual and appears to have a high cross section at about 25° where the $\Delta L = 2$ transitions are at a maximum, indicating a possible $L = 2$ admixture.

Also, in a simple model with no L mixing the ratios of the cross sections for states reached by each ΔL value are determined by Clebsch-Gordan coefficients only, and

$$\left(\frac{\sigma_{7/2}}{\sigma_{9/2}}\right) = \frac{(\langle \frac{5}{2}, \frac{5}{2}, 2, 0 | \frac{7}{2}, \frac{5}{2}\rangle)^2}{(\langle \frac{5}{2}, \frac{5}{2}, 2, 0 | \frac{9}{2}, \frac{5}{2}\rangle)^2} = 2.84,$$

$$\left(\frac{\sigma_{11/2}}{\sigma_{13/2}}\right) = \frac{(\langle \frac{5}{2}, \frac{5}{2}, 4, 0 | \frac{11}{2}, \frac{5}{2}\rangle)^2}{(\langle \frac{5}{2}, \frac{5}{2}, 4, 0 | \frac{13}{2}, \frac{5}{2}\rangle)^2} = 3.33.$$

As can be seen in Fig. 2 the ratio of $(\sigma_{7/2}/\sigma_{9/2})$ is not constant and averages 1.8 over the entire angular range. In fact it only approaches the theoretical limit at 25°. Similarly, the ratio of $(\sigma_{11/2}/\sigma_{13/2}) \approx 2$ rather than 3.33, indicating that the simple model of pute L-wave transitions is not correct. This reaction may be an interesting testing ground for two-step transitions.

TABLE III. Q values for major $\Delta L = 0$ transitions.

Target	Q (MeV)	Target	Q (MeV)
^{226}Ra	-2.816 ± 0.015	^{237}Np	-3.816 ± 0.015
^{230}Th	-3.55 ± 0.015	^{239}Pu	-4.163 ± 0.015
^{232}Th	-3.07 ± 0.015	^{240}Pu	-3.692 ± 0.015
^{234}U	-4.099 ± 0.015	^{242}Pu	-3.045 ± 0.015
^{235}U	-4.145 ± 0.015	^{244}Pu	-2.560 ± 0.015
^{236}U	-3.330 ± 0.015	^{243}Am	-3.407 ± 0.015
^{238}U	-2.765 ± 0.015	^{248}Cm	-2.894 ± 0.015

VI. REACTION Q VALUES

We have measured the Q values for each of our targets and in Table III we have tabulated the Q values observed for the major $\Delta L = 0$ transition in each nucleus; we have included for completeness those given in Ref. 1. These are ground-state transitions for all nuclei except ^{235}U and ^{239}Pu.

VII. SUMMARY

It is obvious from the data presented in Table II that the transitions to the pairing excited states have between 15 and 50% of the strengths of the major $\Delta L = 0$ transitions in all actinide nuclei observed including even-even, even-odd, and odd-even targets when corrected for the reaction Q dependence. This is especially noteworthy since the range of nuclides studied has been extended from $N = 136$ (^{224}Ra) to $N = 150$ (^{246}Cm).

A second major observation is that these excited $\Delta L = 0$ transitions have a smooth Q dependence and appear far less affected by blocking of valence nucleons than do the major transitions.

We wish to express our appreciation to J. Lerner for preparing the isotopically purified targets.

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

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[7]We wish to thank Dr. B. Bayman for providing this computer program.