(\vec{t}, α) reaction on actinide nuclei and the observation of ²⁴³Np

E. R. Flynn, D. L. Hanson, and R. A. Hardekopf

University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87545

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The (t,α) reaction has been carried out on the actinide targets ²³⁴U and ²⁴⁴Pu using a 17 MeV beam of 80% polarized tritons. The residual nucleus ²³³Pa has been previously studied and serves to calibrate the trends of the analyzing power for known spin states. Twenty-two energy levels were observed in ²⁴³Np, a nucleus previously unreported, and a number of spin assignments are suggested. The observed analyzing powers are very similar to those previously measured in the lead nuclei, permitting the use of empirical spin determinations over a wide range of nuclei.

NUCLEAR REACTIONS ²³⁴U, ²⁴⁴Pu (\bar{t}, α), $E_t = 17$ MeV; measured $\sigma(E_{\alpha}, \theta)$, $A_y(E_{\alpha}, \theta)$; enriched targets DWBA analysis.

I. INTRODUCTION

The actinide nuclei play a fundamental role in the study of nuclear spectroscopy. They assume an important role in the theories of deformed nuclei because of their large quadrupole deformations. They are also of practical interest because of their fission properties. More recently the interest in super heavy nuclei has made the studies of actinide nuclei even more appropriate because of the large overlap of the theories which predict super heavy nuclei and the properties of actinide nuclei. Techniques which lead to increased knowledge of this nuclear region are thus very desirable. There is also a need to extend the range of study to new actinides further from the known line of stability, as these provide a further test of the theories which attempt to explain the known region and extrapolate to unknown regions.

Experimental limitations have previously led to a rather sharp cutoff in knowledge of nuclei on the neutron-rich side of the actinides. In particular, there has been little experimental effort in studying the proton hole states in the actinide nuclei. The high level density requires good energy resolution while the high nuclear charge requires reasonable beam energies. The $(d, {}^{3}\text{He})$ reaction has not been utilized effectively in this region because of the general lack of high resolution deuteron beams with energies above 30 MeV. The (t, α) reaction offers the advantage of a high Q value (approximately 12-15 MeV) over the $(d, {}^{3}\text{He})$ (approximately -2 MeV) and thus becomes practical on Van de Graaff accelerators. This reaction, however, suffers from the lack of distinctive angular distributions for the assignment of angular momentum transfers. Complete reliance must then be placed on the fingerprint patterns¹ for the determination of spin values.

The recent introduction of a polarized triton source² permits a further degree of investigation in the (t, α) reaction: the use of analyzing powers for spin determination. This technique has now been successfully applied in the lead isotopes³ and the rare earths.⁴ Large asymmetries are seen throughout the nuclei examined with particularly good differentiation of spin-orbit partners. It is the purpose of the present paper to investigate the usefulness of the (\bar{t}, α) reaction in the heaviest nuclei, the actinide region. The $^{234}U(t,\alpha)^{233}$ Pa reaction was examined because of the previous knowledge of ²³³Pa from (t, α) studies, ¹ from (³He, d) reactions⁵ and from decay experiments.⁶ This then serves to calibrate the usefulness of the analyzing powers. The ²⁴⁴Pu(\bar{t}, α)²³⁴Np reaction leads to a completely new nucleus ²⁴³Np and thus serves as an example of the application of the (\overline{t}, α) reaction. In neither of these cases is any attempt made to provide a detailed theoretical understanding of the level schemes (Ref. 1 already provides this in some detail for ²³³Pa) but only to illustrate the value of the (t, α) reaction in this nuclear region.

II. EXPERIMENTAL TECHNIQUES

The experiment was carried out using a beam of 17-MeV polarized tritons² from the Los Alamos Scientific Laboratory's FN Van de Graaff facility. The average beam polarization was 0.8 with a typical beam current of 50 nA. Measurements were made from 15° to 60° in 10° steps, with spin up and spin down runs taken at each angle. A monitor detector was used to measure the elastic triton scattering, measuring uniformity between individual runs and relative cross sections be-tween various targets. The targets consisted of

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FIG. 1. Spectrum at 50° of the ${}^{234}U(\bar{t}, \alpha)$ 233 Pa reaction. Both spin up and spin down are shown.

uranium and plutonium oxides highly enriched isotopically and of 188 and 102 $\mu g/cm^2$ respective thicknesses.

The reaction α particles were momentum analyzed in a quadrupole-dipole-dipole-dipole (Q3D) spectrometer⁷ and detected in a 1 m helical focalplane detector.⁸ Particle separation of α particles from other groups was excellent and no leakthrough of other particles was seen. Typical energy resolutions were 15 keV for the plutonium target and 18-20 keV for the uranium, where these are target thickness dominated.

Energy calibration was provided by the ²⁰⁶Pb(t, α) reaction to known levels in ²⁰⁵Tl.³ Exposures were made at identical fields for both the ²⁰⁶Pb and ²⁴⁴Pu targets in order to carefully establish the mass of the lowest observable state of ²⁴³Np.



FIG. 2. As in Fig. 1 except for ²⁴⁴Pu (\bar{t}, α) ²⁴³Np.

This nucleus has never been observed before and the (t, α) Q value could only be estimated. The magnetic field was chosen such that a 500 keV error in this estimate would have still placed the ground state region on the detector and, as the results below show, this proved to be more than adequate.

III. RESULTS

Spectra taken at 50° for both spin up and spin down for the 234 U target are shown in Fig. 1. Figure 2 contains similar spectra for the 244 Pu target. The results are tabulated in Tables I and II where excitation energies, cross sections at 50°, and possible spin assignments are given. In the case of 233 Pa, a comparison to previous results is also presented. Cross-section angular distributions for a number of selected states of 233 Pa and 243 Np are shown in Figs. 3 and 4, respectively. The analyzing powers calculated according to the formula

$$A_{y} = \frac{N_{\bullet} - N_{\bullet}}{P_{\bullet}N_{\bullet} + P_{\bullet}N_{\bullet}}$$

are given in Figs. 5 and 6. Here N_{\star} and N_{\star} are the peak areas for spin up and spin down, respec-

		rom Ref. b	$K^{n}[Nn_{2}\Lambda]$	<mark>+</mark> [530]	 _[530]	$\frac{3}{2}^{+}[651]$	$\frac{3}{2}$ [651] + $\frac{1}{2}$ [400]	$\frac{1}{2}$ + [400]	$\frac{1}{2}^{+}[400]^{f}$		$\frac{3}{2}^{+}[402]$	$\frac{3}{2}^{+}[402]$	× .					<mark>3</mark> [514])	$\frac{1}{2}$ [660] ¹	<u>+</u> [[541])											
		Γ	٦щ		1 - ~	+640	$\frac{13}{2}$ + $\frac{1}{2}$ +	+			+ 2 4-	+						- - 		(} -											
បរ	f. b	5 MeV)	Theo.	63	107	28	77 + 247	39	2 ^f		163	10						51	44^{1}												
do/	Rei	(60°, 15	Exp.	79	80	58	321	54	00	19	143	27	20	36	6	21	9	74												·	
		E_{x}	Ref. b	0	57	104	v	с С	290	347	444	474	543	569	690	736	863	980		1065											
		Known	Ref. a	2 + 0	57	104,109	163, 169	202	300	(366)	448,454		554	585				(385)													
		S	(for N = 23)	0.043	0.061	0.023 ^e	0.48	0.077 ^e	0.008,0.01	0.38	0.26	(0.02)	0.19	0.20	0.006	0.004	0.006	0.033, (0.074)		0.035	0.057	0.031,0.012	0.019	0.040	0.032	0.024	0.032, 0.052				
	Α.	suggested	م ∎	। ।		4 + 1	- +	4-1	(} *_, \ *_)	'~ * ~	+	קי	+ 1 *	+ + +	(<mark>-}</mark> -)	(<u>-</u>	(- <u></u>]-)	<u>₹</u> +,(<u></u> , 1)		2 <mark>+</mark> +	++	-+ + 2 2 2 2 2 2 2 2 2 2 2	+	+ -f~	+ +	+ ••• * ••	- - - - - - - - - - - - - - - - - - -	1			
		$d\sigma/d\Omega$	[μb/sr (50°)]	69	74	54	296	(45)	11	22	154	(27)	26	41	10	л С	10	85		41	35	19	12	25	42	32	43	28	32	18	
		E,	(keV)	0	55	107	171	205 ^d	296	355	454	488	555	586	703	742	872	966		1073	1176	1233	1267	1308	1386	1417	1486	1557	1625	1680	e 6.
		Level	No.	0	1	7	3,4	2	9	7	6	10	12	13	14	15	18	20		22	23	24	25	26	27	28	29	30	31	32	Reference

TABLE I. $^{24}U(t, \alpha)^{23}Pa$.

^bReference 1. ^c These authors used energies from Ref. 6 for these levels. ^dPoorly resolved. ^eUsing J^{π} from Ref. 1. ^f These assumptions are based on the current work.

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Level No.	E_x (keV)	dσ/dΩ [μb/sr (50°)	A_y suggested J^{π}	<i>S</i> (for <i>N</i> = 23)
0	0	8	$l - \frac{1}{2}$	
1	76	175	$\frac{1}{2}^+, \frac{3}{2}^-$	0.34,0.10
2	105	113	$\frac{3}{2} - \frac{1}{2}$	0.07,0.22
3.	175	82	<u>-</u> , <u></u> +	0.04,0.04
4	251	95	5+	0.08
5	295	194	<u>\$</u> , +	0.11, 0.38
6	330	20	<u>3</u> +	0.03
7	• 380	5	2	•
8	400	16	(7)	0.007
9	422	7	(<u>`</u> \$_)	0.004
10	532	47	(1 ⁺)	0.08
11	580	19	() () () ()	0.009
12	675	51	$(\frac{1}{2}, \frac{1}{2})$	0.08
13	710	12	$l^{2} + \frac{1}{2}$	
14	772	9	$l = \frac{1}{2}$	
15	808	10	4	
16	853	11	$l=\frac{1}{2}$	
17	1044	69	$l + \frac{4}{3}$	
18	1128	14	4	
19	1173			
20	1268	36		
21	1391			
22	1430			

TABLE II. 244 Pu $(\vec{t}, \alpha)^{243}$ Np.

tively, and P_{\star} and P_{\bullet} are the corresponding beam polarizations. The latter were measured before and after each run and the two readings averaged.

Since it cannot be declared with absolute certainty that the lowest excitation energy state seen in ²⁴³Np is the ground state, all energies presented in Table II are relative to the highest energy alpha group seen. An extrapolation of ground state masses of the lighter neptuniums would suggest a Q value of 12.5 ±0.1. The observed Q value of the highest energy alpha group was 12.405 ±0.010 MeV, which corresponds to a ²⁴³Np mass of 243.064 330 u for this state. There is thus no significant deviation in the observed Q value for this level and the expected ground state value. This, of course, still is not sufficient evidence for having observed the ground state.

IV. DISTORTED WAVE ANALYSIS AND SPIN DETERMINATIONS

Distorted wave (DW) analysis of the differential cross sections and analyzing powers are required to extract spectroscopic strengths and to assist in spin identification. The code DWUCK⁹ was used for this purpose with triton¹⁰ and α -particle¹¹ potentials from the literature, except for a triton spin-orbit term of 6-MeV depth, and these are summarized in Table III. The fits to the differential cross sections are shown in Figs. 3 and 4 for a number of representative cases. As can be seen from these figures, it is difficult to assign ltransfers from such shapes although some differentiation between low and high l transfers based on their slopes is possible. The spectroscopic values obtained from these fits and the relation

$d\sigma/d\Omega = NS \sigma_{DWUCK}$

are given in Tables I and II. The value N is quite dependent upon the optical potentials chosen and varies between a value of 23 and 54 (Ref. 3) in the lead region depending upon the choice of potentials. Here we choose N=23 for the values shown in Tables I and II.

The calculation of polarization by the code DWUCK follows the method described by Bassel *et al.*¹² The polarization is defined as the average projection of the transferred angular momentum I along $\vec{k}_a \times \vec{k}_b$ where \vec{k}_a and \vec{k}_b are the relative momentum of the incoming and outgoing particles, respectively. In terms of the matrix elements for the transfer of a definite angular momentum

$$\beta_{sj}^{im} = \frac{i^{-i}k_{b}^{2}}{(2l+1)^{1/2}\sqrt{4\pi}} \frac{m_{a}}{m_{b}}$$

$$\times \int d\mathbf{\bar{r}}_{bB} \int d\mathbf{\bar{r}}_{aA} \phi_{bB}^{(-)*}(\mathbf{\bar{k}}_{b}, \mathbf{\bar{r}}_{bB})$$

$$\times f_{Isj, m}(\mathbf{\bar{r}}_{bB}, \mathbf{\bar{r}}_{aA})$$

$$\times \phi_{aA}^{*}(\mathbf{\bar{k}}_{a}, \mathbf{\bar{r}}_{aA}),$$



FIG. 3. Angular distributions for the 234 U (t, α) 233 Pa reaction for a selected number of states. The lines are DWBA predictions.

the polarization is given by

$$\pi_{isj} = \frac{2}{l} \frac{\sum_{m} [(l+m+1)(l-m)]^{1/2} \mathrm{Im}(\beta_{si}^{im}\beta_{sj}^{im+1*})}{\sum_{m} |\beta_{sj}^{im}|^2}.$$

If the above, $f_{1sj,m}$ is the form factor for the reaction which for pickup reactions would be computed using a Woods-Saxon well. The ϕ 's are the distorted waves also calculated in a Woods-Saxon well but for the incoming and outgoing particles.



FIG. 4. As in Fig. 3 except for the 244 Pu (t, α) 243 Np reaction.

The calculation of analyzing powers in the code DWUCK is performed in the same manner as calculating the differential cross section with optical model parameters supplied for t, α and the captured proton including spin-orbit terms where necessary. Spin factors must also be supplied to the code. The resulting analyzing power is then calculated in the time reversed channel which is a standard output of DWUCK. It is necessary to use this channel since the polarization of the inci-





FIG. 5. Analyzing power results for the $^{234}U(\vec{t}, \alpha)$ 233 Pa reaction. Also shown are DWBA calculations (solid lines) and empirical shapes (dashed lines) for a number of cases. The empirical cases are from the 208 Pb (\vec{t}, α) 207 Tl reaction or from known spin cases in the present reaction.

dent beam cannot be input. In codes where the time-reversed channel is not calculated, the calculation is performed assuming a time-reversed reaction. Either procedure gives the correct analyzing power, or polarization. The Madison convention is assumed. The bound state wave functions are taken as the proton single particle states appropriate to this region of the actinides (see Ref. 13, p. 839). The quadrupole deformation parameter ν_2 is about 0.25 for the U-Pu nuclei which suggests a single particle energy of about -6 to -7 MeV for 93 pro-



FIG. 5. (Continued)

tons. In this region there is little ambiguity in the choice of form factors as the Nilsson model calculations indicate in Ref. 13. The significant orbitals which are used in the DW calculations are the $1g_{7/2}$, $2d_{5/2}$, $2d_{3/2}$, $3s_{1/2}$, $1h_{3/2}$, $1i_{13/2}$, $2f_{7/2}$, $2f_{5/2}$, $3p_{3/2}$, $3p_{1/2}$, and $2g_{3/2}$. The only choice in form factors for a given j^{τ} state would occur if principal quantum numbers would mix and the Nilsson calculations do not indicate this to be likely in this region.

All calculations were done with a spherical form factor which is all that is allowed in the normal DWUCK code. This introduces some errors for these deformed nuclei although it is the stan-



FIG. 6. As in Fig. 5 except for the ²⁴⁴Pu (\bar{t}, α) ²⁴³Np reaction and using empirical results from the uranium and lead results.

dard procedure and fits data well in the rareearth region (see Ref. 4). Some of the differences between data and DW fit may be due to this approximation.

Comparisons to analyzing power measurements are given in Figs. 5 and 6 for a number of cases. In general, the fits give a qualitative representation of the data where the spins are known. The quality of the fits is comparable to the lead and rare earth region for the larger cross-section states so there would appear to be no new strong coupled channel effects occurring in this region. Both the distorted wave Born approximation (DWBA) results and the data suggest no strong

Т

-1/2* EMP

---- 3/2" EMP



FIG. 6. (Continued)

variation in the magnitude of the analyzing power between lead and the actinide regions. The figures thus contain both a comparison to DWBA and the empirical shapes of lead.³ In addition, the empirical shapes of known spin states in ²³³Pa are used to suggest spin assignments in ²⁴³Np where the spin values were not observed in the lead case.

V. DISCUSSION

The analysis of Ref. 1 may be used in comparing the present data to theoretical expectations. No attempts have been made in the present analysis to perform rotational model calculations but only to explore the possible value of analyzing power measurements. Table I contains the expected theoretical energies and cross sections given by Thompson *et al.*¹ The experimental cross sections quoted by these authors and the ones given in Table I are very close, indicating, as suggested by the DWBA, that $\sigma(15 \text{ MeV}, 60^\circ) \approx \sigma(17 \text{ MeV}, 50^\circ)$ for the (t, α) reactions. Thus for the purposes of a quantitative discussion, these theoretical values seem appropriate.

A. 233pa

There appears to be good agreement between the spin assignments of Thompson *et al.*¹ and the analyzing power measurements for the lowest ten states. For the levels at 107 and 205 keV only l $+\frac{1}{2}$ and $l-\frac{1}{2}$, respectively, could be assigned because of poor DWBA fit or inadequate resolution. The 171-keV doublet is dominated by the $\frac{1}{2}$ * spin as indicated by the analyzing power value. We are able to make suggestive spin assignments for a number of levels above this, but it is difficult to attach these to definite Nilsson orbits. This is especially true in the region of 1 MeV where a considerable number of particle-phonon coupled states are expected.

The 998-keV level was suggested by Ref. 1 as possibly the $11/2^{\circ}9/2^{\circ}[514]$ state although the analyzing power measurements indicate it more likely is a $9/2^*$ as its A_v is similar to that of the 107-keV state. In this region one does expect a $9/2^{*}$ state from the $9/2^{*}1/2^{+}[660]$ configuration of about the strength observed. This strength is estimated from the occupation probabilities V^2 given by Chasman *et al.*¹³ and von Egidy *et al.*¹⁴ and C_{J}^{2} coefficients by Erskine et al.¹⁵ Here we use S $=2V_{J}^{2}C_{J}^{2}$ and assume no Coriolis coupling so the result is only qualitative. This result predicts a cross section of 44 μ b/sr or about one half of that observed but comparable to the alternative prediction based on a $11/2^{-}9/2^{-}[514]$ configuration. Thus we strongly favor the $9/2^+$ spin assignment.

We also disagree with the tentative spin assignment of the 1073-keV level from Thompson *et al.*¹ The A, values suggest a j = l + 1/2, not j = l - 1/2, and most likely $5/2^*$. It is unclear which band this state would belong to. Finally, the level at 296 keV also appears as a $5/2^*$ and is at the proper energy and has approximately the correct intensity to be the $5/2^*1/2^*[400]$ orbital.

TABLE III. Optical model parameters for DWBA calculations.

	V (MeV)	γ _r (fm)	<i>a</i> , (fm)	W (MeV)	<i>r</i> _i (fm)	<i>a</i> _i (fm)
triton	150.3	1.24	0.707	10.6	1.42	0.816
α	225.8	1.304	0,515	24.3	1.304	0.515
Þ	a	1.25	0.65			A

^aAdjusted to fit binding energy with $\lambda_{so} = 32$.

B. ²⁴³Np

The most likely spin assignments for ²⁴³Np are shown in Table II. ²³⁹Np is the heaviest neptunium isotope for which reaction data is available,¹⁴ little information is known on ²⁴¹Np and nothing on ²⁴³Np. In ²³⁹Np the lowest band is the $5/2^{+}[642]$ and examination of the C_J^2 coefficients indicates that only the $9/2^+$ and $13/2^+$ members would be excited with sufficient strength in the (t, α) reaction to be observed here. Neither the weak state labeled as 0 in Table II nor the strong state 76 keV above this appear to be described by these spins. The $A_{\rm v}$ curve of Fig. 6 is best described by the empirical $3/2^*$ fit but this does not rule out other j=l-1/2 possibilities because of the poor statistics associated with the weak nature of this state. Thompson $et al.^1$ note that for increasing neutron number in the U isotopes, the deformation increases and the ground state Nilsson configuration changes. A similar phenomena may also be occurring here, and examination of the proton single particle levels as a function of deformation (see, e.g., Ref. 13) indicates that the $5/2^{+}[642]$ orbital drops rapidly with increasing deformation. The $5/2^{-}[523]$ band changes little with deformation and may indeed become the ground state band here. Possible candidates for the state 0 here are the $5/2^{-1}$ and $9/2^{-1}$ members of this band with a detailed Coriolis coupled calculation required to indicate if the observed strength would agree with this suggestion.

The $1/2 \cdot [400]$ orbital rises rapidly with increasing deformation and examination of the C_{J}^{2} coeffi-

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cients for this orbital suggests strongly that it may belong to level 1 as seen here. In fact there are two possibilities for this $1/2^{+}$ state, the 76and 295-keV levels noted in Table II. The alternative spin assignment for these levels as $3/2^{-}$ is expected from the $1/2^{-}[530]$ configuration which lies in the vicinity of the $1/2^{+}[400]$ in ²³⁹Np, although somewhat higher. The 105-keV level also fits well with a $3/2^{-}$ assignment. At 175 keV the A_{y} values suggest a possible spin of $7/2^{-}$ which would agree with the expected presence of a $7/2^{-}$ $1/2^{-}[530]$ state in this vicinity. The $5/2^{-}1/2^{-}[530]$ member is expected to be quite weak.

These qualitative discussions indicate that the expected strong configurations from the Nilsson model conform with the experimental observations. They also suggest that the ground state band of the light neptunium isotopes is no longer the ground state in ²⁴³Np, an effect similar to that observed for the uranium isotopes. It is clear that a complete Coriolis coupled calculation will be necessary to confirm these interpretations, but the present data, with the spin restrictions demanded by the A_y measurements, should serve as excellent input to such a calculation.

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