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NUCLEAR DECAY SCHEMES OF SOME OF THE ISOTOPES OF TANTALUM

Frank Frederick Felber, Jr.
(Master's Thesis)

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Frank Frederick Felber, Jr.

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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ABSTRACT

A nuclear spectroscopic study of the radiations resulting from the electron capture decay of Ta^{176}, Ta^{177}, and Ta^{178} is reported.

Gamma ray energies were obtained by measurement of internal conversion electron lines with 180-degree - focusing permanent magnet electron spectrographs capable of 0.1% resolution.

The measurements show two rotational transitions in Hf^{176}. Their energies are 88.34 kev and 202.1 kev.

The 112.97-kev transition in Hf^{177} was confirmed as primarily E2 with less than 3% M1 admixture.

Rotational Structure has also been seen in Hf^{178}. Transitions of energies 88.81-, 93.17-, 213.70-, 325.8-, 331.9-, and 427.0-kev have been identified. The 88.81 was found to be a delayed E1 transition of 4.8 sec half-life.

The data are discussed in the light of the Bohr-Mottelson unified model of the nucleus.

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Frank Frederick Felber, Jr.
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I. INTRODUCTION

Since the discovery of the neutron deficient isotopes of tantalum and tungsten by Wilkinson,[1] no accurate spectroscopic measurements have been made on them. These nuclei are in a region between closed shells and a knowledge of the low-lying energy levels might prove interesting for comparison with the Bohr-Mottelson unified model.[2]

Rotational structure arising from the strong coupling of particle and surface motion has already been observed in Hf^{180m}.[3] The first three rotational levels in Hf^{176} which arise from the decay of Lu^{176} have also been identified.[4] Finally, the levels in Hf^{177} populated from Lu^{177} decay have been studied with a crystal spectrometer,[5] thus providing an internal standard for the spectroscopic measurements.

It is of interest to study these levels from the electron capture decay of the tantalum isotopes in order to get a cross check on the assignments. It is of further interest to look for rotational structure arising from the decay of Ta^{178}, in order to make comparisons with a series of even-even isotopes of the same atomic number.

This paper deals with the electron capture decay of W^{178}, its 9.3 min tantalum daughter and the 2.1 hr Ta^{178} isomer. The decay of the other tantalum isotopes of mass 176 and 177 have been studied and will also be discussed.

II. EXPERIMENTAL

A. Chemical

All bombardments were carried out in the 60 inch Crocker Laboratory cyclotron with either alpha particles or deuterons. Standard micro-block target holders were used. The tungsten was produced by bombarding ordinary HfO_2 with 48 Mev alpha particles. The tantalum

activities were made both by 24 Mev deuterons on HfO_2 and by alpha bombardment of Lu_2O_3 at various energies. A spectroscopic analysis performed on the HfO_2 showed only 0.6 percent zirconium impurity, and the Lu_2O_3 used was the spectroscopically pure grade.

The following chemical procedures were used in the research.

Procedure 1. Separation of Carrier-free tungsten from HfO_2

1. Dissolve the target (about 350 mg) in a platinum dish by heating with 6 M HCl - 1 M HF (hereafter called target solvent) for about half an hour.

2. Centrifuge out excess solid material (See note 1).

3. Add just enough $BaCl_2$ to completely precipitate the hafnium as $BaHfF_6$.

4. Centrifuge and set precipitate aside for hafnium recovery.

5. Extract the supernatant liquid with an equal volume of diisopropyl ketone (DIPK). The tungsten stays in the aqueous phase and the tantalum impurity extracts readily into the DIPK.[6]

6. Evaporate the aqueous layer to dryness and take up again with about 5 cc of distilled water.

7. Pour through a column of Dowex-2 anion exchange resin in the chloride form and elute the activity off with 1.3 M HCl (See notes 2 and 3).

8. Evaporate eluate to dryness and take up again in about 3/4 cc of 0.1 M NH_4HSO_4 plating polution adjusted to a pH of 3.6 by addition of some NH_4OH.

9. Plate on 10 mil platinum wire cathode at 100 - 150 milliamperes current for about one hour. The plating procedure has been described previously.[7]

Note 1. Sometimes the target will not dissolve completely. This is probably because high beam currents (10 - 20 μa) cause part of the target to be heated enough to become refractory and therefore very difficult to dissolve.

Note 2. The column contained about 1 cc of wet resin in a cylinder about 5 mm in diameter and about 25 mm long.

Note 3. This step is not specific for tungsten, but since the principal interfering elements are removed before the column step, a reasonably pure tungsten fraction is obtained.

Procedure 2. Separation of Carrier-free tantalum from HfO_2

1. Dissolve the target (~350 mg) in the target solvent in a platinum dish and boil down to about 1/2 to 3/4 cc. Most of the tantalum stays in solution.

2. Centrifuge our precipitate and save for hafnium recovery.

3. Cool the supernatent in ice water and centrifuge again to remove the hafnium which salts out.

4. Extract the clear supernatent solution with an equal volume of DIPK which has previously been equilibrated with the target solvent. (If larger volumes can be tolerated, two extractions may be performed).

5. Wash the organic phase once with an equal volume of the target solvent and discard the washing.

6. Back extract the tantalum into an equal volume of distilled water.

7. Add about 30 - 50 mg of solid $(NH_4)_2 C_2O_4$ to the water and the tantalum is ready for plating.

Note. The plating yield is usually very good except when too much fluoride ion is present in the plating solution.

Procedure 3. Carrier-free separation of tantalum from Lu_2O_3

1. Dissolve target (~100 mg) in 20 - 25 cc of 6 M HCl (See note 1, procedure 1).

2. Dissolve about 5 mg of HfO_2 in 1 ml of conc. HF and add this to the lutetium solution. LuF_3 will precipitate out. The hafnium acts as a holdback carrier.

3. Centrifuge the precipitate and save for lutetium recovery.

4. Evaporate supernatent solution down to about 1/2 to 3/4 cc and carry through steps 3 to 7 of procedure 2.

Procedure 4. Anion exchange separation of hafnium from tantalum[8]

1. Evaporate the tantalum solution obtained from the back extraction step (step 6 procedure 2) down to a few drops.

2. Dilute with about 5 cc of 9.0 M HCl - 0.004 M HF.

3. Pour through column of Dowex-1 anion resin in the chloride form. (See note 2, procedure 1).

4. Elute with 9.0 M HCl - 0.004 M HF. The hafnium comes off in a very sharp band initially and the tantalum stays behind. The separation takes about 1 - 2 seconds.

B. Instrumental

The bulk of the accurate spectroscopic work was carried out using two 180 degree-focusing permanent magnet electron spectrographs with effective fields of 99 and 216 gauss. These have been described previously.[9] The positron spectrum of the 9.3 min Ta^{178} in equilibrium with the tungsten parent was observed on a lens type spectrometer.

Scintillation counting of the gamma rays was accomplished with a 50-channel pulse height analyzer and a Pacific Electro-Nuclear Co. (Penco) 100 channel analyzer, both used with the NaI (thallium activated) crystals as scintillators and Dumont 6292 photomultiplier tubes.

III. RESULTS AND DISCUSSION

A. General

A sample from each bombardment was mounted for traffic counting of the gross activity. The half lives and mass assignments obtained are in good agreement with Wilkinson's work (See ref. 1).

In Table I may be found a summary of the results of the permanent magnet spectrograph runs on the tantalum isotopes. Half life determinations for the stronger lines were made by exposures in the 216 gauss magnet at the intervals indicated for about two days. After the decay of the shorter lived activities, one day exposures were taken every other day for about six days and only the 113- (112.97-) kev transition resulting from the decay of 2.2 day Ta^{177} showed up. No longer-lived tantalum activities were seen on the spectrographic plates.

Electron energy calibration was accomplished in the region around 100-kev by means of the 113-kev transition in Hf^{177}, which has previously been measured very accurately (see ref. 5). The 300-kev region was calibrated by mounting an I^{131} standard and the tantalum source alternately on the same holder so that both sets of lines appeared on the same plate. Great care was taken to insure that both the standard and the source were positioned alike. Any lines which were not recorded on the 99 gauss magnet or which did not have standardizing

lines on the same plate are marked with an ~ sign. These energies are probably good to only 0.3%, while the others are accurate to 0.1%.

Relative intensities are tabulated in the sections where the appropriate isotope is discussed. Electron intensities were determined with a recording densitometer, and they are believed accurate to about 20%. Resolution was not good enough to permit the direct determination of relative photon intensities. However, the decay of the electromagnetic radiation of the sample was followed on the 100 channel analyzer for about nine days and relative photon intensities were obtained by resolution of the decay curves plotted for each individual gamma peak.

The hafnium K X-ray fluorescence yield obtained from Gray's graph[10] is about 95%, so no correction was made for this in determining the intensity of K X-rays.

Table I

Summary of all Conversion Electron Lines Observed

Electron Energy	Intensity* After -- Hrs. 4	8	20**	24	28	36**	44**	$t_{1/2}$ of line	Assignment Subshell	Mass	Comments
16.08	VVW							>8h	K		a,d
22.95	M							>8h	K	176	a,d
23.44	M							<8h	K	178	a,d
27.79	S-M							<8h	K	178	a,d
38.92	VW								none		a,d
39.65	VVW								none		a,d
42.56	W								K L_I L_I***	Hf	a,d
43.09	VVW								K L_I L_{II}	Hf	a,d
44.24	VVW								K L_I L_{III}	Hf	a,d
44.80	W-M								K L_{II} L_{III}	Hf	a,d
46.00	VW								K L_{III} L_{III}	Hf	a,d
47.60	M								K	177	a,d
51.42	VVW								K L_I M_I	Hf	a,d
51.92	VVW								K L_I M_{II}	Hf	a,d
52.50	VVW								K L_{II} M_{III}	Hf	a,d

Table 1 - Continued

Electron Energy	Intensity* After -- Hrs. 4	8	20**	24	28	36**	44**	$t_{1/2}$ of line	Assignment Subshell	Mass	Comments
53.61	VVW								K L_{III} M_{III}	Hf	a,d
53.99	VVW								K L_{III} M_{IV}	Hf	a,d
54.55	VVW								none		a,d
55.21	VVW								K L_{III} N_{I}	Hf	a,d
59.72	VVW								none		a,d
60.62	VVW								none		a,d
70.17	M							>8h	L_{I}		a,d
70.73	VVW							>8h	L_{II}		a,d
71.96	VVW							>8h	L_{III}		a,d
77.05	VW							8h	L_{I}	176	a,d
77.62	VVVS	VVS	VVS	VS	VS	S	S	8h	L_{II}	176	a,b,c
78.78	VVVS	VVS	VVS	VS	VS	S	S	8h	L_{III}	176	a,b,c
79.27	VVW								L_{III}	178	a,d
81.93	VW							2h	L_{I}	178	a,d
82.40	VVS	S	W	VVW				2h	L_{II}	178	a,b,c,f
83.57	VVS	S	W	VVW				2h	L_{III}	178	a,b,c,f
85.94	VS	VS	S	M	M	M	W	8h	M_{II}	176	a,b,c
86.20	VS	VS	S	M	M	M	W	8h	M_{III}	176	a,b,c
87.97	S	S	S	M	M	W	W	8h	N_{III}	176	a,b,c
88.27	VVW								O	176	a,d
90.87	VS	M	VVW					2h	M_{II}	178	a,b,c,f
91.07	VS	M	VVW					2h	M_{III}	178	a,b,c,f
92.05	VVW								none		a,d
92.80	M	VVW						2h	N_{III}	178	a,
93.25	VVVW								O	178	a,
97.15	VVW								none		a,
101.72	W								L_{I}	177	a,d
102.14	S	S	S	S	M	M	M	2d	L_{II}	177	a,b,c
103.36	S	S	S	S	M	M	M	2d	L_{III}	177	a,b,c
110.21	VVW								none		a,
110.66	VVW	VVW	VVW	VVW				2d	$M_{II, III}$	177	a,b,c
112.55	VVW								$N_{II, III}$	177	a,b,c
115.30	VVW								none		a,

Table 1 - Continued

Electron Energy	Intensity* After -- Hrs. 4	8	20**	24	28	36**	44**	$t_{1/2}$ of line	Assignment Subshell	Mass	Comments
116.47	VVW								none		a,
125.18	VVW								none		a,
136.66	M	M	M	W	W	W	VW	8h	K	176	a,b,c
148.32	VS	M	VW	VVW				2h	K	178	a,b,c,f
~151.5	VVW								none		a,e
~164.4	VVW								none		a,e
~166.0	VW								none		a,e
~170.5	VVW								none		a,e
~174.9	VVW								none		a,e
~191.2	VVW								$L_{I, II}$	176	a,e
~192.6	VVW								L_{III}	176	a,e
~197.4	VVW								none		a,e
~201.7	VVW								none		a,e
202.37	VW								L_{I}	178	a,d
203.00	W								L_{II}	178	a,b,c
204.15	VW								L_{III}	178	a,b,c
211.38	VVW								M_{III}	178	a,b
213.3	VVW								none	a,b	
260.42	W	VW	VVW					2h	K	178	a,b,c,f
266.52	W	VW	VVW					2h	K	178	a,b,c,f
~283.3	VVW								none		a,e
314.9	VW								$L_{II, III}$	178	a,b
316.0	VVW								L_{III}	178	a,
320.9	VVW								$L_{I, II}$	178	a,b
361.60	VW	VVW		VVW				2h	K	178	a,b,c,f
415.9	VVW								$L_{I,II}$	178	a,b

Unassigned Transition:- 81.49

Summary of unassigned lines:-

38.92, 39.65, 54.55, 59.72, 60.62, 92.05, 97.15, 110.21, 115.30, 116.47, 125.18, 151.5, 164.4, 166.0, 170.5, 174.9, 197.4, 201.7, 213.3, 283.3

* S = Strong, M = Moderate, W = Weak, V = Very

** Exposed for 8 hrs. All other exposures were 4 hrs.

*** These designate auger electrons; auger energies were calculated by the method of Bergstrom and Hill.[11]

Comments

a. Seen on hafnium + deuteron bombardment

b. Seen on lutetium + 30 Mev alpha bombardment

c. Seen on lutetium + 40 Mev alpha bombardment

d. These lines were resolved only on the 99 gauss magnet. There were no plates exposed in this magnet with samples prepared from the lutetium + alpha bombardments, so direct observation of these lines was not possible.

e. These lines were seen on only one particular hafnium + deuteron experiment because an active source which gave a reading of 15 Roentgens at one inch was prepared. Sources of 1 - 5 R at 1 inch were insufficient to reproduce these lines.

f. The intensity of these lines for a given source strength was less in the 40-Mev than in the 30-Mev alpha bombardment.

B. Tantalum-176

The electron work on this isotope is summarized in Table II. The energy of the 202.1-kev transition is reported accurate to 0.1% because the K-line of this transition was on a plate on which there appeared lines from the I^{131} standard. The L-lines were seen on only one plate and their energies were not quite as good as that of the K-line. The electron binding energies were taken from the compilation of Hill, Church and Mihelich.[12]

Table II

Summary of Electron Data for Ta^{176}

	Electron Energy (kev)	Visual Int.	Numer. Int.	Atomic Subshell	Photon Energy
88.34 ± 0.08-kev transition	22.95	M		K	88.32
	77.05	VW	65	L_I	88.33
	77.62	VVVS		L_{II}	88.37
	78.78	VVVS	49	L_{III}	88.35
	85.94	VS	59	M_{II}	88.32
	86.20	VS		M_{III}	88.32

Table II - Continued

	Electron Energy (kev)	Visual Int.	Numer. Int.	Atomic Subshell	Photon Energy
88.34 ± 0.08-kev transition	87.97	S	$\Sigma N = 16$	N_{III}	88.36
	88.27	VVW		$O_{II, III}$	88.32
202.1 ± 0.2-kev transition	136.66	M		K	202.03*
	191.2	VVW		$L_{I, II}$	202.2
	192.6	VVW		L_{III}	202.2

* This line was given more weight because it was recorded in the 99 gauss magnet and was near standardizing lines.

The $L_I + L_{II} : L_{III}$ ratio in the 88.34-kev transition was found to be 1.3, with most of the $L_I + L_{II}$ conversion taking place in the L_{II}-shell. The theoretical values for E1, E2, E3, M1, M2, and M3 transitions are 3.3, 1.1, 2.2, 113, 4.6, and 0.85, respectively.[13] Therefore, it appears that an assignment of E2 for the multipolarity of this transition is most consistent with the experimental data.

Since the 202-kev transition, when put into the Bohr-Mottelson rotational formula (see Ref. 2)

$$E_{rot} = A [I (I+1)] - B [I (I+1)]^2, \qquad (1)$$

yields values of A and B comparable with those from rotational bands of neighboring even-even nuclei, and since the K : L and L-subshell ratios follow qualitatively those expected from an E2 transition, it is given this assignment. An E2 transition of this energy for an atomic number of 72 should be most highly converted in the L_I - and L_{II} - shells with about half as much in the L_{III}-shell. This was found qualitatively to be the case by visual inspection of the photographic plate. The L_I - and L_{II}-lines were coalesced into one because of insufficient resolution.

Relative photon intensity work is summarized in Table III and the numbers given are believed accurate to about 15%. According to the table, most of the population is to the 2+ level 88 kev above ground,

with the balance populating the 4+ state 202 kev above this. Since the intensity of K X-rays due to K-capture is about the same as that of the 88 kev gamma ray, it appears that there is little or no population of the ground state by electron capture. Positron emission must occur in less than 0.2% of decays, since no definite annihilation peak is observed.

Table III

Photon Data for Ta^{176}

Photon Energy	Integrated Photopeak	Count. Eff.[14]	Escape Peak[15]	Photon Int.	Theor. Conv. Coef.*	Transition Intensity
K X-rays	310	1	16%	360	$\alpha_k(88)=1$ $\alpha_k(202)=0.14$	314**
88	38	0.97	6.6%	42	$\alpha_{tot}=6.6$	328
202	17	0.70	0	24	$\alpha_{tot}=0.2$	29

* K conversion coefficients were obtained from Sliv,[16] while the L-shell conversion coefficients were taken from Rose's tables.[13] The M- and N-conversion coefficients were estimated from the L's wherever necessary.

** This is the intensity of X-rays arising from K-capture only. The X-rays due to K conversion of the 88- and 202-kev transitions have been subtracted from the K X-ray intensity to give this number.

A decay scheme which is consistent with the above data is shown in Fig. 1. The work on the β^- decay of Lu^{176} (see Ref. 4) and the coulomb excitation work[17] have also been included for comparison purposes.

The constants A and B from Eq. 1 have been evaluated. A is 14.54 kev and B is 0.0131 kev.

C. Tantalum-177

Table IV summarizes the electron work done on this isotope. The K- and L-lines of the 112.97 kev transition were used as internal energy standards. The M- and N-lines were too weak to get very accurate readings and therefore were not used for the standardization.

Table IV

Summary of Electron Data for Ta^{177}

	Electron Energy	Visual Intensity	Numerical Intensity	Atomic Subshell	Photon Energy
112.97-kev	47.60	M		K	112.97
transition	101.72	W	22	L_I	113.00
	102.14	S		L_{II}	112.89
	103.36	S	12	L_{III}	112.93
	110.66	VW		$M_{II, III}$	112.91
	112.55	VVW		$N_{II, III}$	112.99

The experimental $L_I + L_{II} : L_{III}$ ratio is 1.8 with much less conversion in the L_I-shell than in either of the other two L-subshells. The theoretical ratios are 3.7, 1.3, 2.4, 117, 6.0 and 1.2 for E1, E2, E3, M1, M2, M3, respectively. The measured and theoretical ratios are consistent with this being an E2 assignment, and M1 admixture must be very low.

It was determined from coulomb excitation[17] that the 113-kev gamma ray is a rotational transition with a g_Ω of 0.2 and a g_R of 0.06. Since the g's are different, a fair amount of M1 admixture might be expected. Using the measured conversion coefficients of Marmier and Boehm (ref. 5), who studied β^- decay of Lu^{177}, one calculates a surprisingly low value of 3% for the maximum amount of M1 radiation. The present work agrees with the Lu^{177} work, and the discrepancy with the coulomb excitation information is unexplained.

The photon data are summarized in Table V. Although it was not seen in the electron spectrum, a 2.2 day component was resolved out of the complex decay of the 200-kev peak of the gamma spectrum. It is about 4% as intense as the 113-kev gamma. Since the counting statistics were considerably poorer for this than the other peaks, limits of error somewhat larger than 15% should be set for the intensity of the 200- (208.34-)* kev gamma ray.

The relative amounts of orbital capture to the different levels are 69% to the ground state, 30% to the 113-kev state and 1% to a state

* Energy determined from Lu^{177} decay

208.3-kev above this. Since no definite annihilation peak was observed, an upper limit of 0.2% can be set for the total amount of positron branching. It was assumed in determining these relative abundances that the decay energy was large enough to use the limiting curve of Brysk and Rose[18] to determine the L_I : K capture ratio. The ratio used was 0.14 and the L_{II} and L_{III} contributions were neglected.

Table V

Photon Data for Ta^{177}

Photon Energy	Integrated Photopeak	Count. Effic.	Escape Peak	Photon Intensity	Theoretical Conversion Coef.	Trans. Int.
K X-rays	68	1	16%	79	$\alpha_k(113)=0.8$ $\alpha_k(208)=0.14$	74
113	5.3	0.94	4%	5.9	$\alpha_{tot}=3.3$	25
208	0.6	0.67	0	0.9	$\alpha_{tot}=0.2$	1.1

Figure II shows the proposed scheme for the electron capture decay of Ta^{177} along with the information on the β^--decay of Lu^{177} and the coulomb excitation work.

D. 2.1 hour Tantalum-178

The summarized electron data appears in Table VI. Six transitions have been assigned to this isotope. Their energies are 88.81-, 93.17-, 213.70-, 325.8-, 331.9-, and 427.0-kev. All the energies are accurate to about 0.1%.

Table VI

Summary of Electron Data for 2.2 hr Ta^{178}

	Electron Energy	Visual Intensity	Numer. Int.	Atomic Subshell	Photon Energy
88.81 ± 0.08 kev transition	23.44	M		K	88.81
	79.27	VVW		L_{III}	88.84
93.17 ± 0.09 kev transition	27.79	S-M		K	93.06
	81.93	VW	42	L_I	93.21
	82.40	VVS		L_{II}	93.15
	83.57	VVS	27	L_{III}	93.14
	90.87	VS		M_{II}	93.25
	91.07	VS		M_{III}	93.19
	92.80	M		N_{III}	93.19
	93.25	VVW		O	93.30
213.70 ± 0.21 kev transition	148.32	VS	3.5	K	213.69
	202.37	VW	0.4	L_I	213.65
	203.00	W	1.0	L_{II}	213.75
	204.15	VW	0.6	L_{III}	213.72
	211.38	VVW		M_{III}	213.40
325.8 ± 0.33 kev transition	260.42	W	1.4	K	325.79
	314.9	VVW		$L_{I, II}$	325.9
	316.0			L_{III}	325.6
331.9 ± 0.33 kev transition	266.52	W	1.0	K	331.89
	320.9	VVW		$L_{I, II}$	331.9
427.0 ± 0.43 kev transition	361.60	VW	0.6	K	426.97
	415.9	VVW		$L_{I, II}$	426.9

The $L_I + L_{II} : L_{III}$ ratio for the 93 kev transition is 1.6, (with very little L_I). Theoretical values for E1, E2, E3, M1, M2, and M3, are 3.3, 1.2, 2.3, 113, 4.9, and 0.9, respectively, making an assignment of E2 most consistent with the data.

The 213-kev transition has an $L_I + L_{II} : L_{III}$ ratio of 2.1, to be compared with 6.5, 2.1, 3.1, 125, 11.6, and 2.8 for E1, E2, E3, M1,

M2, and M3, respectively. Since the background was low in the vicinity of these lines, the L_I- and L_{II}- lines were resolved on the densitometer. The experimental $L_I : L_{II} : L_{III}$ ratio is 0.4 : 1 : 0.6. The theoretical E2 ratio according to Rose[13] is 0.5 : 1 : 0.7. This transition is given an assignment of E2 on the basis of the preceeding information.

No quantitative L-subshell and K : L ratios are available for the 326-, 332-, and 427-kev transitions, so their multipolarities cannot be assigned on this basis. However, it was determined from the gamma spectrum (see following) that the 213-, 326- (or 332-) and 427-kev transitions are all in about the same relative intensity, with the 332- (or 326-) somewhat less intense. Now, in order to get the relative transition intensities determined from the electron spectra to agree with the scintillation work, one must use E2 conversion coefficients throughout. It was also found from the electron work that the 332 is the less intense of the two 300-kev transitions.

The photon data were not obtained the same was as they were for Ta^{176} and Ta^{177}. Instead, a pure sample of Ta^{178} was prepared by bombarding Lu_2O_3 with 18 Mev alpha particles to get only the (α,n) product. The decay of a sample was followed for about ten hours and no other component was found, confirming that the sample was pure. Coincidences were also run on a sample and the results are shown in Table VII. The coincidence apparatus has been described elsewhere.[19]

All the numbers in the table have been corrected for counting efficiency and escape peak. The 213-, 330-, and 427-kev columns have been corrected for E2 conversion, but the 90-kev column has not.

Table VII

Coincidence Intensity Data for 2.1 hr Ta^{187}

	K X-rays	90-kev	213-kev	330-kev	427-kev
Gamma Spectrum (no coinc.)	28	17	21	27	18
Gate: K X-rays	7	7	14	20	11
Gate: 90 kev	7	4	24	20	18
Gate: 213 kev	8	15	--	18	18
Gate: 330 kev	12	13	20	--	15
Gate: 426 kev	7	12	13	12	--

It is evident from the third, fourth, fifth, or sixth row of the table that the 213-, 326-, and 427-kev transitions are in about the same relative intensity. It is also apparent that these transitions as well as the 90-kev radiation are in coincidence. From the evidence that they are in coincidence and the fact that their energies fit the Bohr-Mottelson formula (eq. 1), it can be inferred that the 93.17-, 213.70-, 325.8-, and 427.0-kev transitions form a rotational band up to the 8+ level.

The fact that the intensity of the K X-rays drops when one gates on any member of the rotational bands leads to the conclusion that there is a delay between the electron capture event and the cascade. Furthermore, the intensity of K X-rays one does get is about what is expected from the coincidence between the (E2) conversion of the gammas in the rotational band and the gammas themselves, so it appears that there is no direct electron capture population of any member of the band. Also, the strong 330-kev gamma - K X-ray coincidence coupled with the fact that one of the 300-kev transitions (the 332 ---- see electron data) is less intense than the gammas in the rotational band, suggests that the primary decay is to two levels 332-kev apart. Their relative abundances as determined from the straight gamma spectrum are 40% to the higher energy state and 60% to the lower. The electron data are considerably less reliable, so all relative abundances are based on gamma counting only.

The above information is illustrated graphically in the decay scheme for the 2.1 hr Ta^{178} isomer shown in Fig. III, on the left side. Note that there is an 88.81-kev E1 transition depopulating the delayed state and feeding the cascade. The evidence for this is as follows: The coincidence data show no population of a 90-kev level, so the intensity of the 90-kev radiation, when correction for E2 conversion is made, should be the same as the 213-, 326-, and 427-kev transitions. It is more intense than expected by a factor of about seven. By making the assumption that this transition is as intense as the others, one calculates a total conversion coefficient of about 0.5 for the surplus 90-kev radiation. The theoretical total conversion coefficient for an E1 of this energy is about 0.46. Furthermore, a K-line of an 88.81-kev transition of roughly the same intensity and half live as the K-line of

the 93.17 was observed on a plate exposed in the 99 gauss magnet. The L-lines of the 93.17 were quite prominent, while only a very weak L_{III}-line of the 88.81-kev transition was identified, thus ruling out the possibility of this being an E2. These observations are consistent with the 88.81 being an E1 transition.

A successful attempt was made to determine the half life of the delayed state radiochemically. The hafnium was milked from the tantalum parent according to Procedure number 4. The half life measurement was then made by quickly placing the sample in a single channel gamma analyzer connected to a Speedomax recorder with the chart set to run at a rate of 10 inches per minute. A linear decay curve was obtained in this manner. The half life is 4.8 ± 0.4 sec. According to Moszkowski's single proton formula,[20] (with the statistical factor S = 1) this transition is retarded by a factor of about 1.8×10^{14}.

Two different analyzer settings were used. One to count everything including and above K X-rays, the other to count everything above 100 kev. The same half life was obtained at both settings.

The decay curves are shown in Fig. IV. Fig. V shows the gamma spectrum of the isomer and for comparison purposes, that of the parent Ta^{178}.

Rotational constants were calculated from the first two levels using eq. 1. They are 15.62-kev for A and 0.0136-kev for B.

E. 9.3 min. Tantalum-178

A sample of the 9.3 min Ta^{178} isomer in equilibrium with its tungsten parent was mounted on a wire and placed in a permanent magnet spectrograph of 50 gauss effective field for about 6 days. Only three lines were observed whose energies are 81.70-, 82.34-, and 83.51-kev. These correspond to the L_I-, L_{II}-, and L_{III}-lines, respectively, of a 93.07 ± 0.10-kev transition in Hf^{178}. This is in agreement with the energy of 93.17 ± 0.09-kev found from the decay of the 2.1 hr isomer. The relative visual intensities of the L-lines are what one would expect for an E2 transition.

The sample was then mounted in the 216 gauss magnet and exposed for about 22 days. No lines whose energies lie between 93- and

about 800-kev were seen. Furthermore, no conversion lines resulting from the decay of W^{178} could be identified. Several auger lines were seen, but the energy measurement was not too accurate, since all the lines were bunched in the first fifth of the plate. The energies are probably not much better than 0.5%. The results of this run may be found in Table VIII. Four conversion lines of energies 82.5-, 83.6-, 90.9-, and 92.7-kev corresponding to L_{II}-, L_{III}-, $M_{II, III}$-, and $N_{II, III}$-lines of the 93-kev transition have been identified. These do not appear in the table. A positron spectrum was obtained with a lens type spectrometer and a Fermi-Kurie analysis yielded an end point of 880-kev. Two different spectra were used to determine the relative

Table VIII
Auger Lines from W^{178} - Ta^{178} Mixture

Electron Energy	Process	Auger Energy*	Element
42.4	$K\ L_I\ L_I$	42.60	Hf
42.9	$K\ L_I\ L_{II}$	43.13	Hf
44.6	$K\ L_{II}\ L_{III}$	44.81	Hf
45.9	$K\ L_{III}\ L_{III}$	45.99	Hf
47.1	$K\ L_{III}\ L_{III}$	47.41	Ta
51.7	$K\ L_I\ M_{II}$	51.71	Hf
53.3	$K\ L_{III}\ M_{II}$	53.42	Hf
55.2	$K\ L_{III}\ M_{II}$	55.37	Ta
57.0	--	--	--
60.7	$K\ M_{II}\ M_{II}$	60.62	Hf
62.5	$K\ M_{II}\ N_{II}$	62.56	Hf

*Calculated by method of Bergstrom and Hill[11]

photon intensities which appear in Table IX. First, a sample of pure Ta^{178} was prepared by milking it from the tungsten parent, (this was done by extraction into DIPK) and the ratio of X-rays to 93-kev radiation determined. Then, a ratio of X-rays to 93-kev to 510-kev to 1350-kev radiation was obtained from the mixture. These were normalized at the 93-kev peak, and all radiation above X-rays was assumed to result from tantalum decay.

Table IX
Photon Data for W^{178} - Ta^{178}

Photon Energy	Integrated Photopeak	Count. Eff.	Escape Peak	Photon Int.	Theor. Conv. Coef.	Transition Intensity
X-ray (Mixt.)	298	1	16%	346	--	--
X-ray (Milk'd.)	165	1	16%	190	$\alpha_k 93=1$	180
93	10	.95	6.6%	11	$\alpha_{tot}=6.6$	84
510	1.4	.16	--	8.8	--	4.4*
1350	0.6	.047	--	13	--	13

* This has been corrected for the geometry factor of 2 for annihilation radiation.

The decay scheme which appears on the right side of Fig. III was constructed using the data of Table IX. Neglecting the amount of population of the 93-kev level by positrons and the 1350-kev gamma, one obtains values of 53%, 41%, and 6% for the percentages of total decay to the ground, 93-kev and 1350-kev states, respectively. The total positron branching is about 2%, which disagrees with the theoretical value of 5% obtained from the K : β^+ ratio versus decay energy curve of Freenberg and Trigg.[21]

If the assumption is made that the observed positron energy corresponds to the ground state transition, then the log ft to ground is 4.7 and the log ft to the 2+ level is 4.8. Thus, it appears that the spin of the 9.3 min isomer is 1+. The 6% decay to the 1350 kev level suggests that there is a low spin state at this energy. However, the details of this are not too well understood.

F. General Discussion

According to the Unified model of Bohr and Mottelson, it should be possible to predict the energies of rotational excited states in deformed even-even nuclei by use of the first term of eq. (1) only. This, however, is not strictly the case, although the theory comes rather close in many cases. Use of the second term in eq. (1) has effected a considerable improvement in the ability to predict the energy of the 6+ level. For the two cases in the rare earth region which are known

most accurately, Hf^{180m} and Hf^{178}, the theoretical and experimental 6+ energies agree within experimental error.

Since there aren't too many 6+ levels known, this test cannot be applied too extensively. Another measure of the applicability of the theory is the value of the constant B; small values representing the best agreement. The constants A and B are listed in Table X. Included in the table are the energies of the first and second excited states and the value of $\hbar\omega$, the effective vibrational energy.* There are a few cases for which possible vibrational levels have been reported. These have been included for comparison purposes. The agreement is quite striking. Some of the information in Table X has been summarized in the form of a plot of A, B, and "$\hbar\omega$" versus mass number and appears in Fig. VI.

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*The quantity B is related to A and to an effective vibrational quantum energy $\hbar\omega$, by eqs. VI 3 a and b of ref. 2. This is the $\hbar\omega$ calculated here. Also, the quantities $\hbar\omega_\beta$ and $\hbar\omega_\gamma$ of the equations were assumed equal.

Table X

Rotational Constants for Some Even-Even Nuclei

Nuclide	$E_{2\to o}$	$E_{4\to o}$	A (kev)	B (kev)	Calcd. $E_{6\to o}$	Exptl. $E_{6\to o}$	"$\hbar\omega$"	Possible "Vibrational" Level (β- type or γ- type)	Reference
Sm^{152}	122.0	366.3	21.20	0.144			1020		22
Gd^{154}	122.9	370.9	21.32	0.138			1060	1010 (2+ 2) (γ)	23
Gd^{156}	88.97	288.16	15.01	0.030			1343		24
Dy^{160}	86.3	282.4	14.50	0.019			1600		25
Er^{166}	80.3	264.4	13.45	0.011	544.8	546	1850	1458 (0+ 0) (β)	26
Yb^{170}	84.26	278.0	14.10	0.010			2130		27,28
Hf^{176}	88.34	290.44	14.54	0.013	588	596*	1940		4, This work
Hf^{178}	93.17	306.87	15.62	0.014	632.0	632.7	2130		This work
Hf^{180}	93.3	308.8	15.60	0.008	641.3	641.8	2780		3
W^{182}	100.09	329.36	16.82	0.018			2080	1222 (2+ 2) (γ)	29

*Energy determined from scintillation spectrometer.

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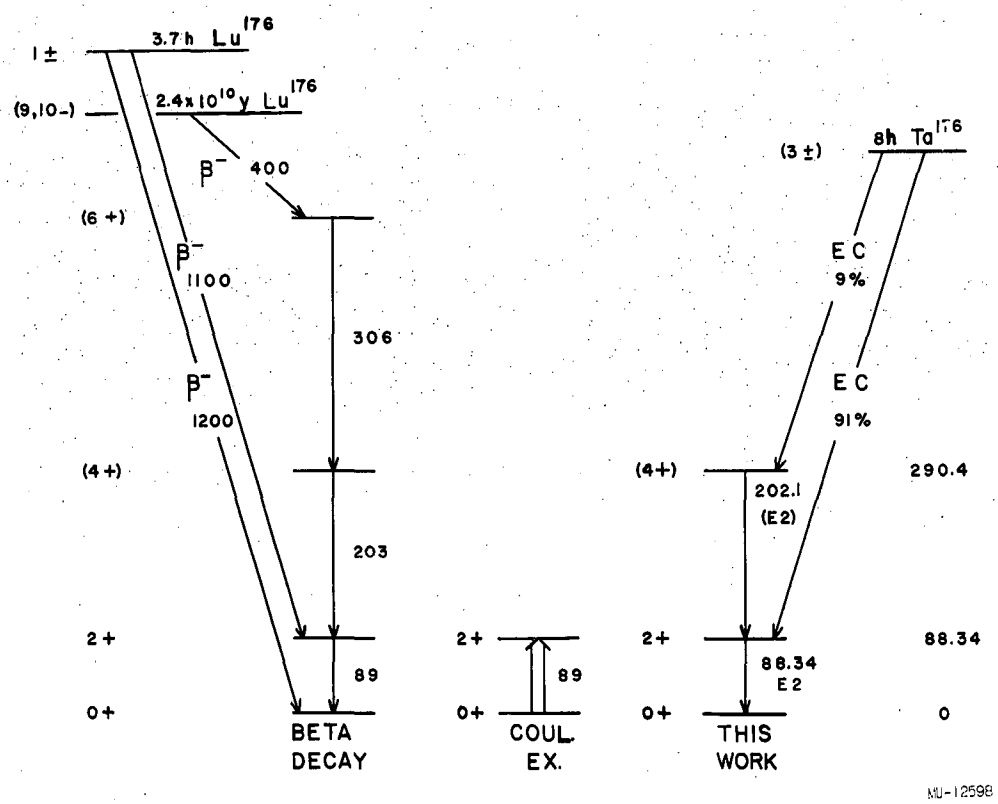

Fig. I Proposed Decay Scheme for Ta^{176}

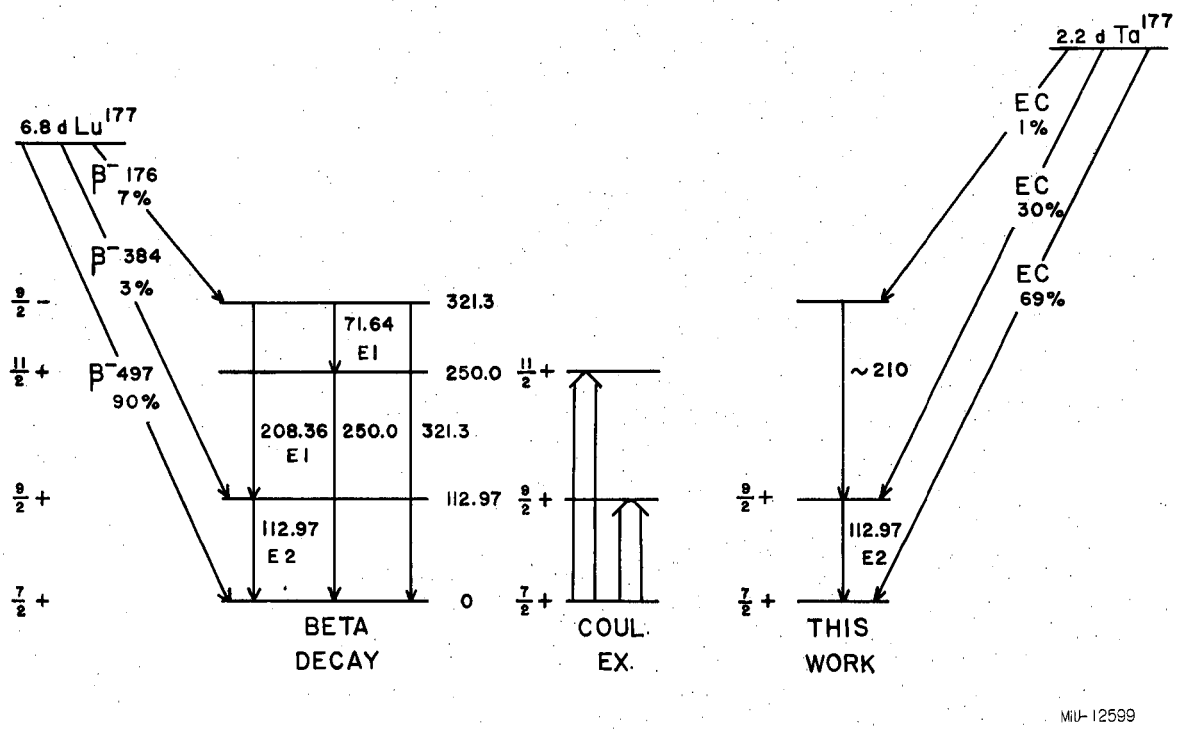

Fig. II Proposed Decay Scheme for Ta^{177}

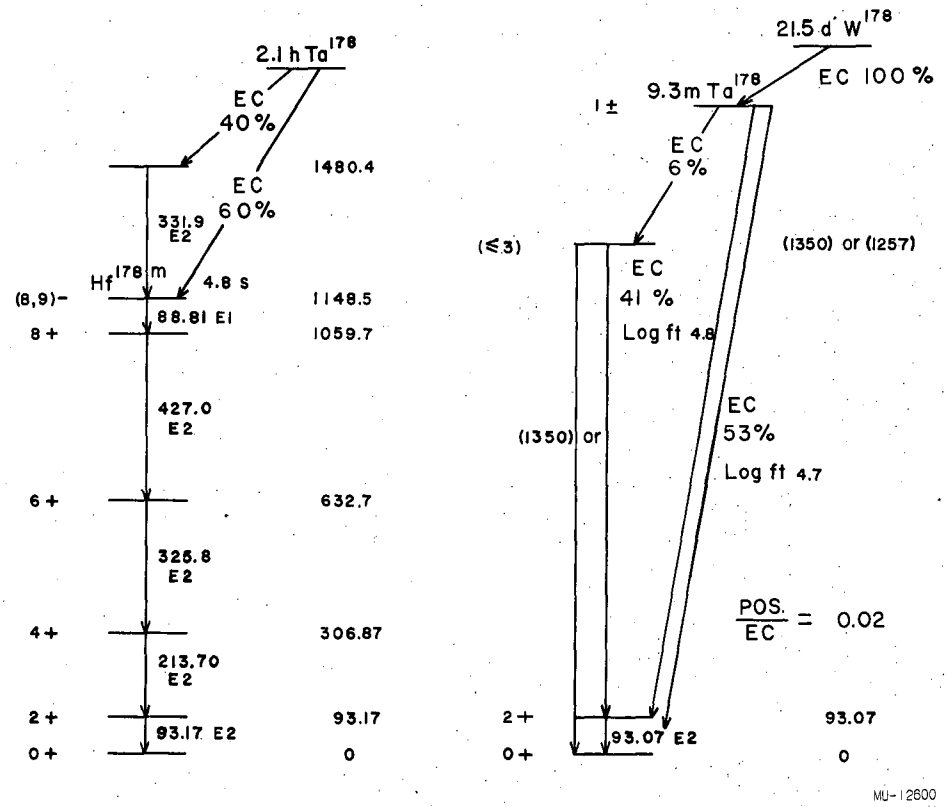

Fig. III Left: Proposed Decay Scheme for 2.1 hr. Ta^{178}. It is interesting to note if one assumes the 331.9-kev transition is of a rotational character and the spin of the 4.8-sec. level in 9, that a value of 16.6-kev obtained for the rotational constant $\hbar^2/2\mathcal{J}$, while that of the ground state rotational band is 15.5-kev. The value 18.4-kev obtains if the spin is assumed to be 8.
Right: Proposed Decay scheme for the 9.3 min. Ta^{178}.

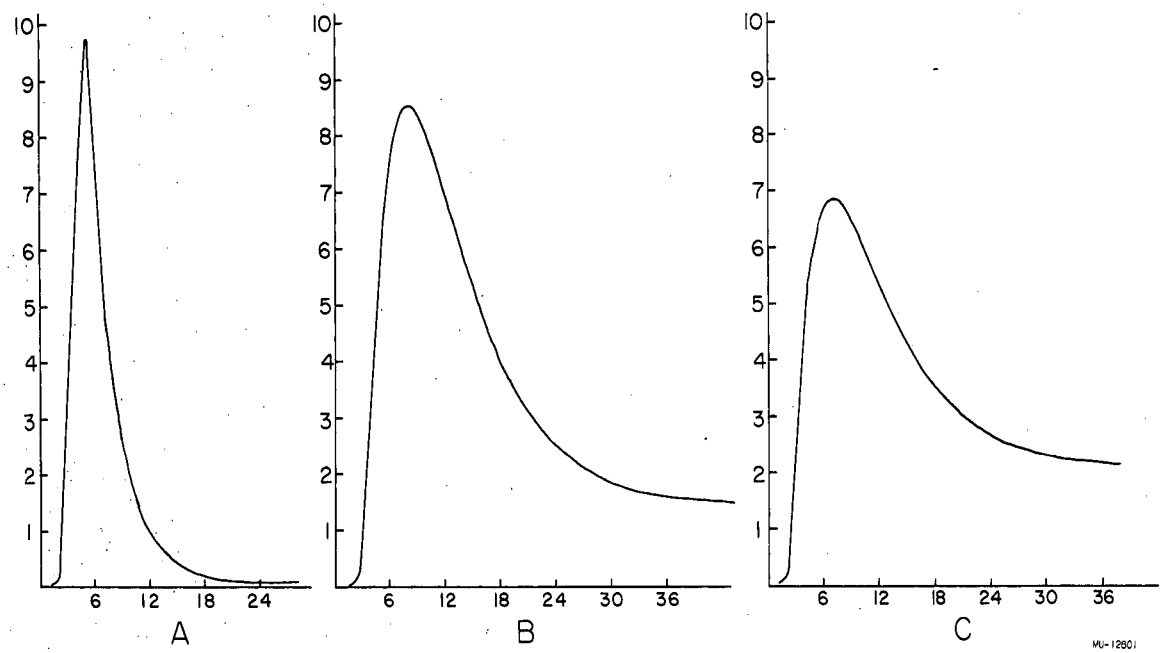

Fig. IV Recorder Chart Trace of the Decay Curves of 4.8 sec Hf^{178m}. The ordinate scales are activity in arbitrary units, while the abscissae are time in seconds.

Curve A was obtained by placing an active source in the detector chamber and then removing it quickly to get the decay curve of the instrument.

Curve B represents the decay curve of the isomer when the single channel analyzer is set to count everything including and above K x-rays.

Curve C is the decay curve of the isomer when the single channel analyzer is set to count everything above 100 kev. The ordinate scale should be half that of A and B.

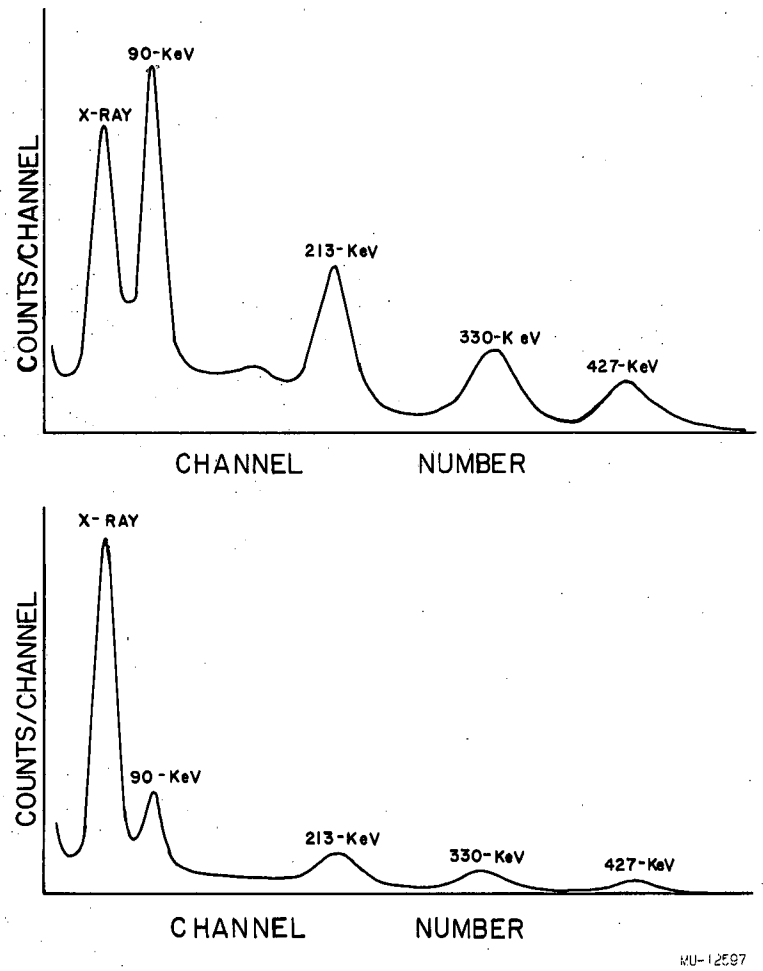

Fig. V Upper Spectrum -- Hf^{178m}
Lower Spectrum -- Ta^{178}

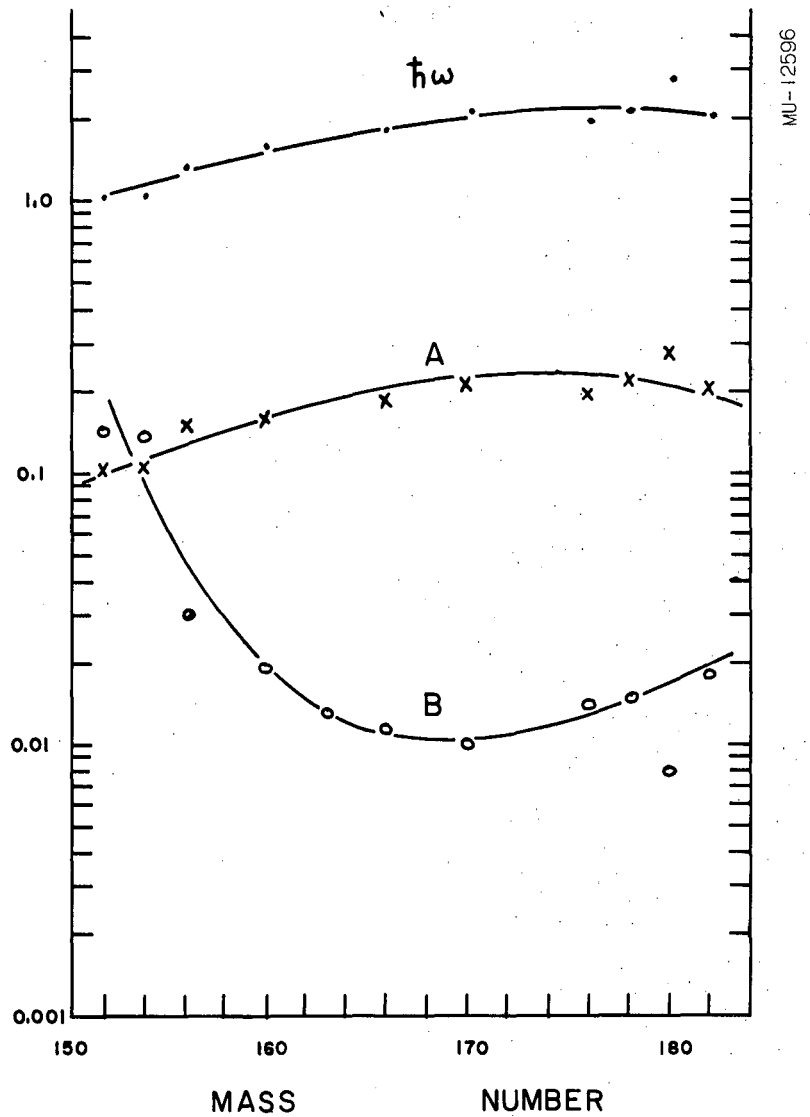

Fig. VI Plot of A and B of eq. (1) and $\hbar\omega = 4\sqrt{A^3/B}$ vs. mass number. Multiply A by 100, B by 1 and $\hbar\omega$ by 1000 to get the proper relation between their ordinates.