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Nuclear Spectroscopy of Neutron-Deficient Rare Earths (Tb through Hf)†

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A survey has been made of electron-capturing nuclides produced by irradiating rare earths (Gd through Hf) of various isotopic enrichments with 22-Mev protons. Ion-exchange chemistry was performed, conversion-electron spectra were analyzed, and the various activities were cataloged. At least ten new activities, Tb^{153}, Tb^{155}, Tb^{156m}, Dy^{155}, Ho^{156}, Ho^{160m}, Ho^{162}, Er^{168m}, Yb^{171m}, and Lu^{169} were found and studied. Eighteen previously known activities also were examined. Multipolarities consistent with the observed conversion electron intensity ratios are proposed. The positions of levels of even-even nuclei as a function of both proton and neutron number are discussed.

INTRODUCTION

THE rare-earth radionuclides produced by neutron capture have been studied extensively in the past several years, but little has been done with the neutron-deficient rare earths. It is of value to study in detail all the isotopes of a given atomic number, and thus be able to correlate data on energy levels with one fixed parameter (i.e., atomic number) to demonstrate the effects of varying neutron number. It has been shown in the study of single-particle levels[1] that such data are indeed interesting. In addition, a great deal of data has accumulated from the Coulomb excitation experiments[2] in the rare-earth region. These data suffer from one handicap: lack of precision, since for technical reasons an inherently low-resolution scintillation counter is usually employed. Experiments with internal-conversion electrons[2] from transitions produced by Coulomb excitation have been performed; these too suffer from the fact that the desired "carrier-free" source is precluded for intensity reasons. The Coulomb excitation experiments usually cannot allow a mass assignment for polyisotopic elements. If one were able to do good spectroscopy of these nuclides, including mass assignments, the value of these Coulomb experiments would be enhanced, since these two techniques are corollary. In addition, it is possible to observe rotational levels in radioactive nuclei which are not available for Coulomb excitation. See for example, the section on levels of Tb^{157}.

Other level data are becoming available: for example, the radioactive decay chains beginning with the spallation products of high-energy interactions, in particular, the investigations of Nervik and Seaborg,[3] who irradiated Ta with 340-Mev protons. In some cases, these decay chains have been given unique mass assignments by direct mass measurements.[4] Photoexcitation[5,6] is another promising attack, one which should give rise to levels not obtainable in ordinary beta decay processes, in particular levels depopulated by $M2$ transitions. Other promising methods are excitation with electrons[7] or fast neutrons.[8]

What is needed is a survey of the radioactive decay chains in this region in order to show the way for more detailed and definitive experiments on known activities. It is the purpose of this paper to describe such a survey. We have been successful in producing high specific-activity sources for use in permanent-magnet spectrographs. We have attempted to obtain the genetic relationships of all the gamma-ray transitions present. We have analyzed several hundred spectrograms, each containing a large number of conversion lines. In spectra of such complexity, many overlaps of conversion lines are observed. These overlaps are usually resolvable by use of isotopically enriched targets, variable energy of irradiation, and half-life data. Figure 1 shows some typical spectra of various ages.

Rare earths (20 to 30 mg of the oxide) were irradiated in the ORNL 86-inch cyclotron[9] at proton-beam energies ranging from 12 Mev, which should produce the (p,n) reaction only, up to the maximum energy of 22 Mev, which in most cases should produce the $(p,3n)$ reaction. Irradiations of one to eight hours were made at a beam current of 70 microamperes. Ion-exchange columns[10] were used to separate the activity desired from the target material. The desired fractions were put into the chloride form, dissolved in pyridine, and electroplated onto 10-mil Pt wires. This method of preparing rare-earth sources has proved to be reliable and effective.[11] We were able to obtain spectrograms of

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‡ Operated for the U. S. Atomic Energy Commission by Union Carbide Nuclear Company.
[1] M. Goldhaber and D. H. Hill, Revs. Modern Phys. **24**, 179 (1952); Gillon, Gopalakrishnan, de-Shalit, and Mihelich, Phys. Rev. **93**, 124 (1954).
[2] See the comprehensive review by Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. **28**, 432 (1956).
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[4] M. C. Michel and D. H. Templeton, Phys. Rev. **93**, 1422 (1954).
[5] C. L. Hammer and M. G. Stewart, Phys. Rev. **106**, 1001 (1957).
[6] S. H. Vegors, Jr., and P. Axel, Phys. Rev. **101**, 1067 (1956).
[7] B. Waldman and W. C. Miller, Phys. Rev. **82**, 305 (1951).
[8] A. A. Ebel and C. Goodman, Phys. Rev. **93**, 197 (1954); Martin, Diven, and Taschek, Phys. Rev. **93**, 199 (1954).
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[10] T. H. Handley and E. L. Olson, Phys. Rev. **94**, 968 (1954).
[11] Mihelich, Ward, and Jacob, Phys. Rev. **103**, 1285 (1956).

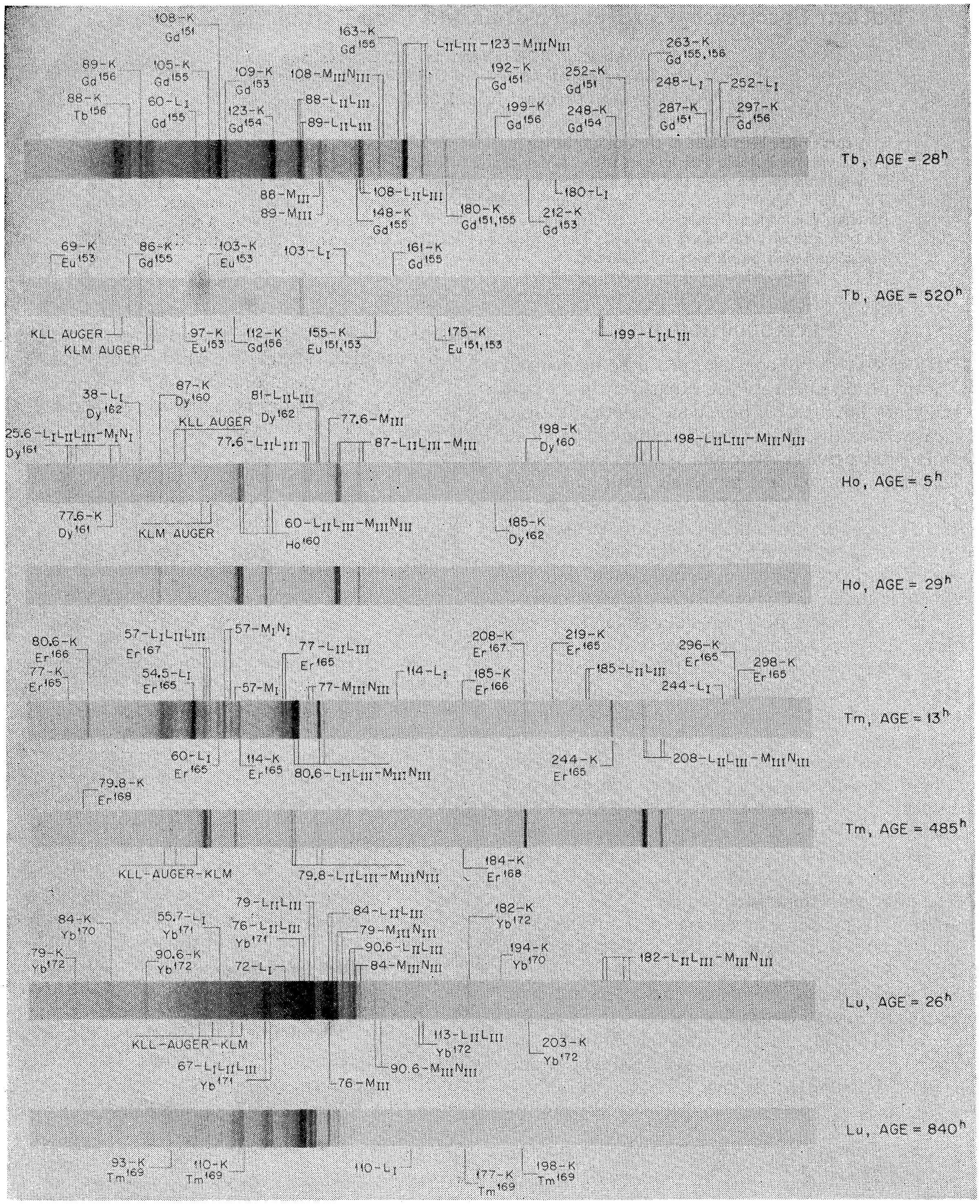

FIG. 1. Conversion-electron spectra of rare earths of various ages produced by 22-Mev protons (127-gauss magnet).

excellent quality with these sources.§ The activation rate and time for chemical processing permitted the observation of activities with half-lives ranging from $\frac{1}{2}$ hour to many years. In all cases we have examined electron-capturing nuclei.

The spectrograph employed was a standard con-

§ *Note added in proof.*—More reliable electrodeposition has been achieved using the method of R. Ko, Nucleonics **15**, 72 (1957).

version-electron spectrograph with a magnetic field of 127 gauss; electrons of energy between 10 and 380 kev were recorded. For a given source, successive exposures were made and the relative decay rates of the various lines determined photometrically. Hence, if the absolute decay rate of any line was known, then the absolute decay rates of the others can be determined. Figure 2 shows some typical half-life curves of terbium activities.

The photographic detectors were either "*AA*" or *No*-screen x-ray film. The "*AA*" emulsion, although less sensitive by a factor of two, has improved contrast and granularity properties.

Scintillation-counter spectra were obtained for three fractions from the ion-exchange column (target element Z, $Z+1$, and $Z-1$); a single-channel analyzer was used in obtaining these spectra. Some hitherto unreported activities were obtained in this fashion. For the reactions leading to carrier-free nuclides, the most prolific is $(p,2n)$, followed by (p,n) and $(p,3n)$.

All the rare earths from Gd through Hf were irradiated, and in each case the chemically separated products were studied in the spectrograph. We shall discuss each successful target in turn, and shall present the data which are now available. Data now being obtained will be published at a later date.

All assigned multipolarities are consistent with the measured K/L and L ratios.[12]

Finally, we shall discuss some of the regularities in the level positions in these nuclei, in particular the variation of first excited level position as a function of neutron number for a given Z, and the behavior of the ratio of possible second to first excited state energies as a function of neutron number for a given Z.

EXPERIMENTAL RESULTS

Irradiation of Gadolinium to Produce Terbium Isotopes

A full-energy irradiation of natural Gd produces a conversion electron spectrum extremely rich in lines, since there are seven stable isotopes of Gd. Several identical properties of the various radioactivities of $Z=65$ made the analysis of the data difficult. Only by repeated irradiations of Gd targets of varying isotopic abundances[13] were we able to catalog the radiations correctly. The half-life standard used was that obtained from a Geiger counter decay curve for pure Tb^{155}, which was found to have a half-life of 5.6±0.1 days. In all cases, the half-lives in parentheses in the section headings are our measured values, unless otherwise noted. Some of the facts adding to the complexity of the analysis were as follows:

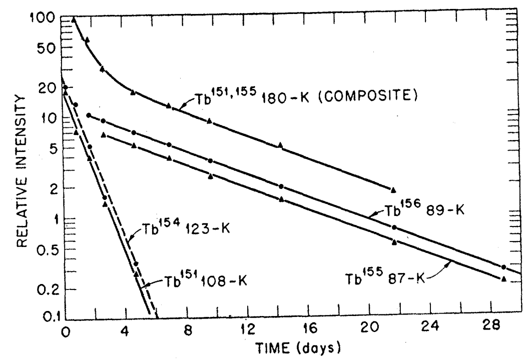

FIG. 2. Decay curves of conversion-electron lines of terbium activities produced by proton irradiation of gadolinium.

1. Tb^{155} and Tb^{156} have similar half-lives (5.6 days).
2. Tb^{151} and Tb^{154} have almost identical half-lives (21±2 hr).
3. There are gamma-ray transitions of identical energy in the decay of Tb^{155} and Tb^{156} (263±0.2 kev).
4. There are gamma-ray transitions of identical energy in the decay of Tb^{151} and Tb^{155} (180±0.2 kev).
5. There is an exact overlap of the K and L_{III} conversion lines of an 88-kev isomeric transition in Tb^{156} and the K conversion line of an 87-kev transition (Gd^{155}) and L_{II} conversion line of an 89-kev transition (Gd^{156}), respectively.

Since various levels in some of the stable Gd isotopes have been well established, these data could be used to assign masses to the activities we produced. For example, levels in Gd^{154} and Gd^{155} are fed by β decay of Eu^{154} [14] and Eu^{155}.[15] In addition, accurate determinations of the energies of the capture gamma rays in Gd^{156} and Gd^{158} have been made by Church and Goldhaber.[16] Hence, when we observe transitions identical to any of these, we have ascertained the mass assignment for the decay chain in question.

Tb^{154}(8 hr+22 hr)→Gd^{154}.—Handley and Lyon[17] have assigned half-lives of 7.5 and 17.5 hours to this isotope. We observe internally converted transitions of 123 and 248 kev apparently identical to those observed in the decay of Eu^{154}. Handley and Lyon report that the ratio of the "7.5"-hr activity to that of "17.5" hr is only 1/500. We may observe some of the shorter lived activity in our studies.

Tb^{155}(5.6 days)→Gd^{155}.—Handley and Lyon[17] had set limits of <10 minutes or >5 years for the half-life of Tb^{155} in their experiments on enriched Gd targets. However, we obtained conversion lines apparently

[12] M. E. Rose, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North Holland Publishing Company, Amsterdam, 1955).

[13] Obtained from Division of Stable Isotopes of the Oak Ridge National Laboratory.

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[15] E. L. Church and M. Goldhaber, Phys. Rev. **95**, 626(A) (1954); Dubey, Mandeville, and Rothman, Phys. Rev. **103**, 1430 (1956); F. Boehm and E. N. Hatch, Bull. Am. Phys. Soc. Ser. II, **2**, 231 (1957); B. Harmatz and J. W. Mihelich (unpublished).

[16] E. L. Church and M. Goldhaber, Phys. Rev. **95**, 626(A) (1954).

[17] T. H. Handley and W. Lyon, Phys. Rev. **99**, 1415 (1955).

TABLE I. Conversion electron data for radioactivities of terbium produced by proton bombardment of gadolinium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Tb^{151}(20 hr)→Gd^{151}							
108.3	1000[b]	~145[c]	~145[c]	145	100	w	$M1+E2$
180.3	155	~25			w		
192.2	50	~10					
252.1	155	~20					
287.6	110	~15					
Tb^{153}(62 hr)→Gd^{153}							
41.5		~415	~360	~400			Superimposed on Auger lines
51.7		~75	~10	~10	~25		
68.1	45	w[d]			w		
87.5	180						
102.1	145	25					
109.8	1000	210			55	w	
174.4	55	~10					
195.2	45	<10					
212.2	195	30					
249.8	60						
Tb^{154}(8 hr and 22 hr)→Gd^{154}							
123.2	1000[b]	...	310	310	155	45	$E2(2+\rightarrow0+)$
248.1	75	...	15[c]	~5	~5	w	$E2(4+\rightarrow2+)$[e]
347.0	6		w				$E2(6+\rightarrow4+)$[f]
Tb^{155}(5.6 day)→Gd^{155}							
18.8			>75	>85	115	45	
21.0			>20	>25	25	~10	
31.3			25	25	~15		
45.3		40	20	30	25	w	
60.1		180	55	55	60	25	$M1+E2$
86.7	1000[b]	230	55	80	80	25	$E1$
101.2	55	~10			w		
105.4	815	145[c]		?[d]	35	10	$E1$
149.0	315	45			~15	w	$M1$
160.8	65	w					$M1$
161.5	255	35					$M1$
163.5	420	60			~15		$M1$
180.4	515	75			20	w	
182.0	~15	w					
221.0	~30	?[d]			w		
239.7	~15	w					
262.9	125	20			w		
341.3	~15						
368.3	w						
Tb^{156m}(5.5 hr) isomeric transition in Tb							
88.40	~130	...	1000	940	470	130	$E3$
Tb^{156}(5.6 day)→Gd^{156}							
89.10	1000[b]	...	870	910	430	120	$E2(2+\rightarrow0+)$
111.9	100	~20			w		
155.2	45	~8					
199.4	360		85[c]	50	40	~10	$E2(4+\rightarrow2+)$[e]
262.7	~25	w					
296.7	~13						$(6+\rightarrow4+)$[f]
356.6	w						
422.2	w						

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "w" indicates a weak line.
[c] L_{I} and L_{II} lines not completely resolved.
[d] Conversion line is a composite of two different lines.
[e] Probable.
[f] Possible.

identical to those of transitions in Gd^{155} arising from the decay of Eu^{155}. These transition energies are 19, 45, 60, 86, and 105 kev. By superimposing spectrograms, one is able to make a very sensitive test as to the "identicality" of sets of conversion lines. Hence, it was evident that we were indeed producing Tb^{155}. To confirm our results, we irradiated two targets with 12-Mev protons to produce the (p,n) reaction alone; the targets were enriched in masses 155 and 156, respectively. Although neither target was enriched to a high degree, the relative intensity of the Tb^{155} and Tb^{156} transitions were consistent with the isotopic enrichment factors. Tb^{155} has a half-life the same as that of Tb^{156} and somewhat similar decay energies; hence it is not surprising that it was overlooked in the previous experiments.

Subsequently, a source of Tb^{155}, free of Tb^{156}, was made in the following fashion. A target of Dy (enriched to 16.8% in mass 156 from the normal isotopic abundance of 0.05%) was irradiated with a full-energy proton beam. The Tb^{155} was produced as the daughter

TABLE II. Conversion electron data for radioactivities of dysprosium produced by proton bombardment of terbium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Dy^{157}(8.2 hr)→Tb^{157}							
60.76	···	180	~20	~20	65	20	$M1+E2(I_0+1\rightarrow I_0)$[d]
82.98	≥300	150	w	w	40	10	$M1+E2(I_0+2\rightarrow I_0+1)$[d]
143.9	20	···	~5	~5			$E2(I_0+2\rightarrow I_0)$[d]
182.5	75	15					
265.5	~10						
326.6	1000[b]	145	···	···	35	10	
Dy^{159}(134 day)→Tb^{159}							
57.98	···	1000	~140[c]	125	305	80	$M1+E2$

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "w" indicates a weak line.
[c] L_{I} and L_{II} lines not completely resolved.
[d] Possible.

of the short-lived Ho^{155} and Dy^{155}, and was chemically extracted from the target. Table I lists the conversion-electron data, and it may be remarked that the results obtained by analyzing the spectra of the enriched Gd targets and the Dy^{156} target are in complete agreement.

The level scheme of Gd^{155} is complex. The electron capture of Tb^{155} leads to levels of several hundred kev excitation, whereas the β^- decay of Eu^{155} apparently does not proceed to any level above that of 105.5 kev. Scintillation-counter studies are now being made.

Tb^{156}(5.5 hr and 5.6 days)→Gd^{156}.—Handley and Lyon[17] have assigned these two half-lives to mass 156. Heydenburg and Temmer[18] have obtained a level of 89 kev in Gd^{156} by Coulomb excitation. In addition, Church and Goldhaber[16] have observed capture gamma rays of 88.8 and 198.7 kev. Handley and Lyon reported that the 5.5-hr activity decayed with a weak "beta" activity. It develops that this radiation is an isomeric transition of 88.4 kev in Tb^{156}, of $E3$ character.[19] The analysis of many spectrograms obtained in quick sequence make our assignment unambiguous. A growth and decay of the ground-state activity of this nucleus was observed. A search was made for the transition de-exciting the first excited level of Dy^{156} which might be fed by beta decay. This transition would be of particular interest since in this case the neutron number is 90; the position of this level should be appreciably higher for this lower neutron number. However, it is possible that the conjectured β decay is of low energy and hence goes to the ground state of Dy^{156}. There is some evidence for a level of 138 kev in Dy^{156}, as observed in the electron-capture decay of Ho. Table I displays our electron line and transition data.

Our scintillation-counter data are in good agreement with those of Handley and Lyon, except for the fact that not all of the transitions they observed occur in isotopes of mass 156. As regards mass 156, it is probable that the levels 2^+ and 4^+ and possibly 6^+ are being populated; in addition, a number of high-energy transitions (greater than 1 Mev), which have small energy differences among themselves indicate that possibly a set of closely spaced levels separated from the ground state by an excitation energy of roughly 1 Mev are being populated. There may be some similarity here to the level scheme of W^{182} studied with the beta emitter Ta^{182}.[20] Recent work indicates the existence of 25 γ-ray transitions in Gd^{156}, as observed from a study[21] of the beta decay of Eu^{156}.

Tb^{151}(20 hr)→Gd^{151}.—A copious yield of Tb^{151} was obtained. The mass assignment has been made on the basis of yields from different targets of varying mass enrichments. Rasmussen *et al.*[22] have assigned a 19-hour alpha activity to Tb^{151}. The electron data are given in Table I.

Tb^{153}(62 hr)→Gd^{153}.—The presence of this activity is established by the observation of the daughter activity, Gd^{153}, which decays to levels in Eu^{153}. This conclusion is consistent with the yields from various mass-enriched targets. Subsequent to this work, Schultz[23] has reported on the activities of Gd^{151} and Gd^{153} produced by alpha bombardment of Sm. Our results are in complete accord with hers.

We should remark here on our negative results. We did not observe a transition of 145 kev in Gd^{155} corresponding to the one observed by Heydenburg and Temmer[18] in Coulomb excitation. There was no indication of any activity due to Tb^{157} or Tb^{158}. An intense source of Dy^{157} showed no daughter (Tb^{157}) activity. Gd targets enriched in masses 158 and 160 were irradiated with protons but no activities of mass greater than 156 (except Tb^{160}) were observed.

Certain remarks regarding the accuracy of the data in Tables I through VI should be made at this time.

[18] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **100**, 150 (1955).
[19] J. W. Mihelich and B. Harmatz, Phys. Rev. **106**, 1232 (1957).
[20] J. W. Mihelich, Phys. Rev. **95**, 626(A) (1954); Murray, Boehm, Marmier, and DuMond, Phys. Rev. **97**, 1007 (1955).
[21] F. Boehm and E. N. Hatch, Bull. Am. Phys. Soc. Ser. II, **1**, 390 (1956).
[22] Rasmussen, Thompson, and Ghiorso, Phys. Rev. **89**, 33 (1953).
[23] V. A. Schultz, University of California Radiation Laboratory Report UCRL-3594, 1957 (unpublished).

TABLE III. Conversion electron data for radioactivities of holmium produced by proton irradiation of dysprosium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Ho^{156}($\sim$1 hr)$\rightarrow Dy^{156}$							
138.1	[c]		85[d]	83	w		$E2(2+\rightarrow 0+)$
Ho^{160}(5.0 hr) Isomeric Transition in Ho							
60.09	...	...	920	1000	510	160	$E3$
Ho^{160}(22 min ?)$\rightarrow Dy^{160}$							
87.00	470	...	770	790	390	100	$E2(2+\rightarrow 0+)$
197.5	125	...	32[d]	17	13		$E2(4+\rightarrow 2+)$[e]
297.6	3.5		w				$E2(6+\rightarrow 4+)$[f]
Ho^{161}(2.5 hr)$\rightarrow Dy^{161}$							
25.65		$\sim$800	$\sim$700[d]	1000	[c]	240	
77.52	[c]	$\sim$350	585[d]	585	350	85	$M1+E2$
103.2	225	$\sim$50					
175.4	$\sim$50						
Ho^{162}(67 min)$\rightarrow Dy^{162}$							
38.2		790	$\sim$40[d]	40	[d]		$M1+E2$
57.7		275			75	25	
80.80	320	...	990	1000	550	150	$E2(2+\rightarrow 0+)$
184.8	380	...	120[d]	70	60	w	$E2(4+\rightarrow 2+)$[e]
282.8	55						$E2(6+\rightarrow 4+)$[f]
Ho^{164}(37 min)$\rightarrow Dy^{164}$							
73.0	$\sim$340	...	975	1000			$E2(2+\rightarrow 0+)$
Ho^{164}(37 min)$\rightarrow Er^{164}$							
91.3	$\sim$250	...	500	420			$E2(2+\rightarrow 0+)$

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "w" indicates a weak line.
[c] Conversion line is a composite of two different lines.
[d] L_{I} and L_{II} lines not completely resolved.
[e] Probable.
[f] Possible.

The true transition energies are within $\pm 0.15\%$ of our values. The estimate of the uncertainties in the intensity data is more difficult to make. In general, the most prominent conversion line is assigned an intensity value of 1000 arbitrary units. A series of spectrograms of various exposures allows us to obtain intensity measurements on lines of greatly different intensities; a single spectrogram does not allow comparing of lines with an

TABLE IV. Conversion-electron data for radioactivities of thulium produced by proton irradiation of erbium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Tm^{165}(29 hr)$\rightarrow Er^{165}$							
47.2	...	135[c]	<115[d]	105	...	...	
54.5		<1275[d]			390	105	
60.5		180			40	w	
77.3	50		210	220	110	30	$E2$
113.7	300	55			15		
219.3	130	30			w		
243.3	1000	155			$\sim$40	w	
296.5	70	w					
297.8	110	25			w		
347.3	30	w					
356.9	20	w					
Tm^{166}(7.7 hr)$\rightarrow Er^{166}$							
80.7	230		1000	980	495	145	$E2(2+\rightarrow 0+)$
154.6	10						
184.7	165		50[c]	33	20	w	$E2(4+\rightarrow 2+)$[e]
194.8	18						
215.4	13						
Tm^{167}(9.6 days)$\rightarrow Er^{167}$							
57.10	...	325	250[c]	270	180	60	$M1+E2$
208.3	1000		800	545	385	110	$E3$
Tm^{168}(87 days)$\rightarrow Er^{168}$							
79.86	250	...	1000	970	465	140	$E2(2+\rightarrow 0+)$
184.6	180		w	w			$E2(4+\rightarrow 2+)$[e]
198.7	$\sim$170						
448.0	$\sim$25						

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "w" indicates a weak line.
[c] L_{I} and L_{II} lines not completely resolved.
[d] Conversion line is a composite of two different lines.
[e] Probable.

TABLE V. Conversion-electron data for radioactivities of lutetium produced by proton irradiation of ytterbium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Lu^{170}(1.9 day)→Yb^{170}							
84.2	235		940	1000	440	125	$E2(2+\to 0+)$
193.5	35		~10[c]	~5	*w*		$E2(4+\to 2+)$
Lu^{171}(8.1 days)→Yb^{171}							
55.6		140			[d]		
66.7		140	250[a]	230			
75.8	~90		910	1000	490	140	$E3$
Lu^{172}(6.7 days)→Yb^{172}							
78.7	>190		930	1000	500	150	$E2(2+\to 0+)$
90.6	~105		210	190	100	~30	$E2$
112.8	~35		27[c]	20	~12	*w*	$E2+(M1)$?
181.5	210		65[c]	45	~30	~8	$E2(4+\to 2+)$[e]
203.8	40		14[c]	8	*w*		$E2$
270.5	19	*w*					
324.6	11	*w*					
373.1	9	*w*					
Lu^{173}(1.4 year)→Yb^{173}							
78.8	>570	1000	~160[c]	160	340	90	
100.9	620	230	~30[c]	30	60	20	
171.5	~25						
272.7	80						
Lu^{174}(165 days)→Yb^{174}							
76.6	>210		1000	~1000	450		$E2(2+\to 0+)$

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "*w*" indicates a weak line.
[c] L_{I} and L_{II} lines not completely resolved.
[d] Conversion line is a composite of two different lines.
[e] Probable.

intensity ratio much greater than 10. In comparing the intensities of various lines with the one of 1000 arbitrary units, the following estimates of uncertainty may be applied: for conversion lines of less than 40-kev kinetic energy, intensities given are lower limits; conversion lines of 75 or more units have an uncertainty of 20%; conversion lines of less than 75 intensity units have an uncertainty as great as 40%. Intensity ratios of well-resolved lines of similar energy have an uncertainty of considerably less than 20%; K/L ratios, for moderately intense lines of greater than 40-kev energy, somewhat less than 20% as is the case for L ratios where the separation is not complete.

Terbium Target to Produce Dysprosium Isotopes

Terbium is a monoisotopic element of mass 159. Dy^{157} decays with a half-life[24] of 8.2 hours, and photons were observed with a scintillation counter.[24]

Our spectrograms indicated electron lines corresponding to several transitions, as tabulated in Table II.

One may postulate an interesting level scheme, assuming a ground state spin (I_0) of $\frac{3}{2}$. There is the possibility of two rotational levels[25] at 60.76 kev (I_0+1) and 144 kev (I_0+2). The 144-kev level is depopulated by a 83.0-kev $M1$ (very small amount of $E2$), and a relatively weak crossover of 144 kev which may be of $E2$ character. It is of interest to note that in the Coulomb excitation experiments[18] on Tb^{159}, the ratio of the intensity of the cascade and crossover transitions is large (>5). In addition, there may be a level at 326.6 kev from which transitions proceed to all three levels in the lower band.

We have observed a transition of 58.0 kev following the electron capture of Dy^{159}. This transition, too, is of $M1+E2$ multipole order with the $M1$ fraction considerably greater than the $E2$ fraction. This level has been obtained by Coulomb excitation, and has also been observed in the β decay[26] of Gd^{159}.

Dysprosium Target to Produce Holmium Isotopes

A 30-mg oxide target was irradiated for two hours at maximum beam energy (22 Mev). Within three hours after the completion of the cyclotron run, the chemical

TABLE VI. Conversion-electron data for radioactivities of hafnium produced by proton irradiation of lutetium.

Transition (kev)	K	L_{I}	L_{II}	L_{III}	M	N	Remarks[a,b]
Hf^{173}(24 hr)→Lu^{173}							
123.9	800	95	~20[c]	25	*w*		$E1$
135.3	280	~40	~40[c]	40	35	~10	$M1+E2$
140.0	1000	145	~20[c]	20	45	~12	$E1$ or $M1+E2$
162.3	35	~5					
297.8	45	~6	~1	~1			
307.3	8	<2					
312.1	13	~2					
357.9	~3	*w*					

[a] Multipole assignments are made on the basis of K/L and L ratios.
[b] Intensity data are arbitrarily normalized to 1000 units for the most prominent line in each activity. Comparison may not be made between data for different nuclides. "*w*" indicates a weak line.
[c] L_{I} and L_{II} lines are not completely resolved.

[24] T. H. Handley and E. L. Olson, Phys. Rev. **90**, 500 (1953).
[25] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1957).
[26] Jordan, Cork, and Burson, Phys. Rev. **92**, 315 (1953); R. Ballini and R. Barloutaud, J. phys. et radium **17**, 534 (1956); N. Marty, Compt. rend. **241**, 385 (1955).

separation was performed, the electrolysis completed, and the source placed in the spectrograph. We were able to take many spectra of short duration in succession. Our conversion-electron data are presented in Table III.

Ho^{164} is known to decay by electron capture, as well as beta decay, with a reported half-life of between 37 and 42 minutes.[26] We were able to detect the conversion lines of the previously reported activity and confirm some of the energy measurements of Brown and Becker[27] (73.0 and 91.3 kev).

Ho^{156}(~1 hr)→Dy^{156}.—As mentioned before, there is evidence for a Ho^{156} activity of 1-hr half-life which decays by electron capture to a 138-kev level in Dy^{156}. The mass assignment is made from the observation that this transition is observed only in targets enriched in Dy^{156} but not in targets enriched in Dy^{158}. There seem to be no other transitions of appreciable intensity associated with this activity. It must be admitted, however, that this assignment is not completely firm, but is probable.

Ho^{160m}(5.0 hr) and Ho^{160}(22 min ?)→Dy^{160}.—The decay of Ho^{160} by electron capture populates levels in Dy^{160}, some of which are the same as those fed by the beta-decay[28] of 73-day Tb^{160}. Gamma-ray transitions which are apparently common to both beta decay and electron catpure are those of 87, 197, 297, 391, 890, and 970 kev. Nervik and Seaborg,[3] and Handley,[29] observed additional gamma rays of 650 and 730 kev in the decay of Ho^{160}. It is worth noting that the mass assignment is certain since Nervik and Seaborg have made a mass measurement of the parent of Ho^{160} (Er^{160}). In addition, we have rechecked this point by irradiating an enriched sample of Dy in which the normal 2.6% Dy^{160} was enriched to 64%.

The transitions of 87 and 197 kev in Dy^{160} are apparently $E2$, and probably depopulate levels of 2^+ and 4^+ character. The transition of 297.6 kev is possibly $E2$, proceeding from the 6^+ level.

A transition of 60.1 kev with a half-life of 5.0 hr, converts in Ho^{160}. Hence, we are dealing here with an isomeric transition in Ho^{160}, most probably of $E3$ multipole order. Obviously, this transition, or an as yet unseen low energy (less than 15 kev) transition, is responsible for the half-life. It appears that $E3$ transitions[5,19] are not uncommon in the rare earth region.

Our spectra show no evidence of any growth of the intensities of the transitions converting in Dy; the decay rate of these lines appears to be identical (within 5%) to that for the lines of the isomeric transition. Hence, one is left with two alternatives: either (1) there is no electron capture from the isomeric state and the ground state of Ho^{160} has a short half-life, very likely that of 22 min. as reported by Handley[29]; or (2) the isomeric state does undergo electron capture to levels in Dy^{160}, in which case the half-life of the ground state of Ho is either short or very long compared to 5 hr, or is 5 hr $+5\%$.

Ho^{161}(2.5 hr)→Dy^{161}.—Electron lines corresponding to transitions of 25.6 and 77.5 kev are observed to decay with a half-life of 2.5 hr and are accordingly assigned to mass 161, since Handley and Olson[30] have observed an activity of such a half-life presumably from mass 161. Cork *et al.*[31] and Smith *et al.*[32] have reported the following transitions: 25.6, 74.9, and 78 kev, which are internally converted in Dy^{161}, following the beta decay of Tb^{161}. It seems reasonable that the 25.6-kev transition, at least, is common to both decay processes. We have confirmed this point by producing Tb^{161} as the daughter of the Gd^{161} β^- decay and chemically separating the Tb activity and examining the internal conversion electron spectrum. Only the 25.6-kev transition is common. Therefore, one can conclude that it probably proceeds from the first excited state of Dy^{161}. Heydenburg and Temmer[18] have observed, by Coulomb excitation, a transition of 76 kev in Dy, presumably in an odd-mass isotope.‖

Ho^{162}(67 min)→Dy^{162}.—We have listed the transitions assigned to mass 162 in Table III. The mass assignment is made on the basis of yields from targets enriched in various masses. Mass 163 is possible, but the work of Handley and Olson[33] indicates that Ho^{163} has a very short or a very long half-life.

The conversion ratios of the transitions of 80.8 and 184.8 kev indicate that they are most likely of $E2$ multipole order.

We have assigned a gamma-ray transition of 38.2 kev to Ho^{162}, despite the fact that Brown and Becker[27] assigned a transition of the same energy to Ho^{164}, produced by (γ(22 Mev), n) on Ho^{165}. The decay rate of this transition is definitely consistent with a half-life of 67 min., and isotopically enriched targets have proven that this transition is not due to an activity of mass 164, but is in an activity associated with mass 162. One might postulate that Brown and Becker produced Ho^{162} by a (γ,$3n$) reaction, but then one has to explain the absence in their data of some of the more intense transitions from Ho^{162} (e.g., 80.8 kev).

[27] H. Brown and R. L. Becker, Phys. Rev. **96**, 1372 (1954).
[28] L. Shartovalov, Izvest. Akad. Nauk. Ser. Fiz. U.S.S.R. **17**, 503 (1953); Burson, Jordan, and Leblanc, Phys. Rev. **94**, 103 (1954); H. Jaffe, University of California Radiation Laboratory Report UCRL-2537 (unpublished); Keshishian, Kruse, Klotz, and Fowler, Phys. Rev. **96**, 1050 (1954); M. A. Clark and J. W. Knowles, Bull. Am. Phys. Soc. Ser. II, **2**, 231 (1957).
[29] T. H. Handley, Phys. Rev. **94**, 945 (1954).
[30] T. H. Handley and E. L. Olson, Phys. Rev. **93**, 524 (1954).
[31] Cork, Brice, Schmid, and Helmer, Phys. Rev. **104**, 481 (1956).
[32] Smith, Hamilton, Robinson, and Langer, Phys. Rev. **104**, 1020 (1956).
‖ *Note added in proof.*—N. P. Heydenburg and G. F. Pieper, Phys. Rev. **107**, 1297 (1957) report levels at 46 and 103 kev in Dy^{161} as observed by coulomb excitation. We observed a transition of 103.2 kev but saw no indication of transitions of 57 or 46 kev. The situation is not yet entirely clear.
[33] T. H. Handley and E. L. Olson, Phys. Rev. **92**, 1260 (1953).

Dysprosium Target to Produce Dy^{155}

In the Dy fraction of this target, an activity was observed which is attributed to Dy^{155}, produced by a (p,pn) reaction on the 0.052% abundant Dy^{156}. We observe, in addition to the 8.2-hr Dy^{157},[34] an activity of about 20-hr half-life which consists of x-rays and photons of 230 kev. Photon peaks of the same energy as those observed in the decay of Tb^{155} grow in and then decay in a manner consistent with the conclusion that they are fed by an activity of about 20 hr and then decay with a daughter half-life of several days. Since the source, of low intensity, could not be produced carrier-free, no spectrographic data were obtained.

Holmium Target to Produce Erbium

An irradiation of the maximum available proton energy (22 Mev) was made on a holmium target in an attempt to produce the 75-min activity[33] by the $(p,3n)$ reaction, as well as to check for any nuclear gamma rays from Er^{165}. Intense Auger lines in Er but no conversion lines of less than 380 kev were observed.

Erbium Target to Produce Thulium Isotopes

Stable Er exists in six masses, ranging from Er^{162} to Er^{170}. The activities[35] due to masses 165, 166, and 167 have been determined by Michel and Templeton,[4] who performed mass separations of the active isotopes. The half-lives stated here for these isotopes are those reported by them. No evidence for the decay of the Tm isotopes of mass 161, 162, 163, or 164 was observed. In addition, the well-known[36] Tm^{170} (120-day) activity was identified. Our conversion-electron data are presented in Table IV.

Tm^{165}(29 hr)[4]$\rightarrow$$Er^{165}$.—This level scheme is indeed a formidable one. Handley and Olson[33] reported gamma-ray transitions of 0.0205, 0.808, 1.16, and 1.38 Mev. We observe a large number of internally converted transitions as noted in Table IV.

Tm^{166}(7.7 hr)[4]$\rightarrow$$Er^{166}$.—The electron capture of Tm^{166} leads to levels in stable Er^{166}, some of which also are reached by the beta decay of the two isomers[37] of Ho^{166}. In each case high-lying levels are populated. Some of the transitions observed in the decay of Tm^{166} are apparently not present in the decay of the Ho^{166} activities.

Tm^{167}(9.6 day)[4]$\rightarrow$$Er^{167}$.—Nervik and Seaborg[3] observed gamma rays of 49, 115, 202, 515, and 720 kev. In addition, Heydenburg and Temmer[18] observed a level at 172 kev by Coulomb excitation in a presumably odd-mass Er isotope (possibly mass 167) which was postulated to be the second excited level. Hence, it follows that the first excited level might be at 78 kev for this nucleus of spin $\frac{7}{2}$.[38] The only low-energy transitions we observe are those of 57.1 and 208.3 kev. It is of interest to note that a 210-kev level in Er with a half-life of 2.5 seconds has been excited with high-energy photon irradiation[5] and $Er(n,\gamma)$ activation.[39] Our conversion data indicates that the transition of 208 kev is of $E3$ multipole order,[19] and is apparently from the same level as that produced by the (γ,n) experiments.[5]

We have strengthened the mass assignment by producing this activity by irradiating Yb with protons and chemically extracting Tm. In this case the ratio of the yield of Tm^{167} compared to Tm^{168} is increased relative to that for the $Er+p$ irradiation.

Tm^{168}(87 day)$\rightarrow$$Er^{168}$.—We observed conversion electrons decaying at a rate consistent with the reported half-life of 87 days[34] for Tm^{168}.

More recent experiments have shown the existence of several more internally converted gamma-ray transitions and higher energy gamma rays of 745 and 820 kev. There is in addition a metastable level with a half-life of 0.1 μsec, which is about 1 Mev above the ground state. This level scheme is now under investigation with delayed coincidence techniques.

Recent studies of the capture gamma rays following resonant neutron capture[40] have shown a spectrum of photons similar to that of Tm^{168}. Our mass assignments confirm the fact that the capture is due to Er^{167}.

Ytterbium Target to Produce Lutetium Isotopes

One should expect a complex spectrum of activities arising from a full energy proton irradiation of natural Yb, which consists of seven stable isotopes. Such was the case. Table V presents our conversion electron data.

The Lu^{170} activity was identified by the decay to a level in Yb^{170}, this same level being reached by the well-known[36] β^- decay of Tm^{170}. The half-life of Lu^{172} has been reported[41] as 6.7 days and we have used the lines of the 182-kev transition in this activity as one of our half-life standards. Nethaway *et al.*, investigated the β^- decay of Tm^{172}, an alternate path to Yb^{172}.[42] We have produced Lu^{169}($T_{\frac{1}{2}}$=1.5 days) which decays to levels in Yb^{169} for we then observe the well-known ground-state activity of Yb^{169}. The conversion lines of the Yb^{169}(30.6 day)[43] were used as a half-life standard for the longer lived lines. The measured decay rates of Lu^{172}($T_{\frac{1}{2}}$=6.7

[34] T. H. Handley and E. L. Olson, Phys. Rev. **90**, 500 (1953).
[35] G. Wilkinson and H. G. Hicks, Phys. Rev. **75**, 1370 (1949).
[36] Graham, Wolfson, and Bell, Can. J. Phys. **30**, 459 (1952); Pohm, Lewis, and Jensen, Phys. Rev. **95**, 1523 (1954).
[37] Graham, Wolfson, and Clark, Phys. Rev. **98**, 1173(A) (1955); Milton, Fraser, and Milton, Phys. Rev. **98**, 1173(A) (1955).
[38] B. Bleaney and H. E. D. Scovil, Proc. Phys. Soc. (London) **A64**, 204 (1951).
[39] E. der Mateosian and M. Goldhaber, Phys. Rev. **76**, 187 (1949); M. Goodrich, Oak Ridge National Laboratory Report ORNL-940, 1951 (unpublished); Campbell, Kahn, and Goodrich, Oak Ridge National Laboratory Report ORNL-1164, 1951 (unpublished).
[40] Fenstermacher, Hickoff, and Schultz, Bull. Am. Phys. Soc. Ser. II, **2**, 41 (1957).
[41] G. Wilkinson and H. G. Hicks, Phys. Rev. **81**, 540 (1951).
[42] Nethaway, Michel, and Nervik, Phys. Rev. **103**, 147 (1956).
[43] Cork, Brice, Martin, Schmid, and Helmer, Phys. Rev. **101**, 1042 (1956).

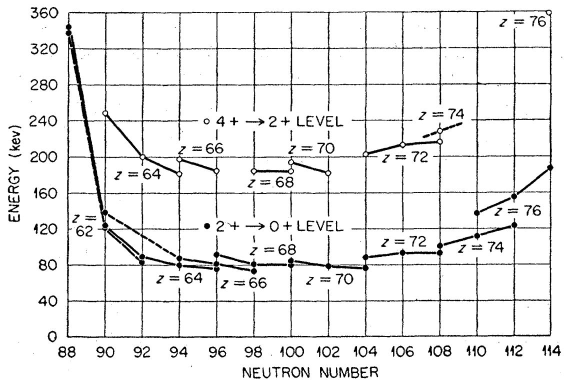

FIG. 3. Energies of first and postulated second excited states in even-even nuclei, for $88<N<114$ with Z as a parameter.

days) and Yb^{169}($T_{\frac{1}{2}}=30.6$ days) are consistent with each other.

In general, the lines may be attributed to activities of the following half-lives: 1.5, 1.9, 6.7 and 8 days; and in addition, half-lives on the order of 150 and 450 days. The latter two values have been confirmed by Geiger-tube counting.

We may remark at this time that we did not find any evidence for the 3.7-hr Lu^{176m} activity[44] which one might expect to be produced by $Yb^{176}(p,n)Lu^{176}$.

Heydenburg and Temmer[18] observed levels of 78 and 110 kev by Coulomb excitation. They assigned the 78-kev composite peak to the first excited states of the four even-A Yb isotopes and Yb^{173}. The 110-kev peak was assigned to Yb^{171}, which has a ground state spin of $\frac{1}{2}$ and thus would be expected to have an anomalous level pattern.

Our data suggest excited levels of 84.2 and 277.8 kev in Yb^{170}, levels of 78.65 and 260.2 kev in Yb^{172}, and a level of 76.6 kev in Yb^{174}, and possibly 78.8 kev in Yb^{173}.

The transitions assigned to mass 171 are those with decay rates corresponding to the half-life of 8 days quoted by Wilkinson and Hicks.[35] In this activity, there is a transition of 66.65 kev ($M1+E2$) which may proceed between rotational levels. A transition of 66.6 kev following the β^- decay of Tm^{171} has been observed by Smith *et al.*[45] Therefore, the mass seems to be well established.

We confirm the half-life[35] of Lu^{173}($\sim$1.4 yr) which decays by electron capture to Yb^{173}. On the basis of preliminary¶ scintillation counter data, it appears that the transitions assigned this activity in Table V are the same as those observed in a well-aged Hf^{173} source produced by $Lu^{175}(p,3n)$, which decays to Lu^{173}.

As regards the Hf^{173}(24-hr)[41] activity, preliminary experiments indicate the existence of a number of gamma-ray transitions. There appears to be at least one well developed rotational sequence.

SYSTEMATICS OF THE LEVELS OF EVEN-EVEN NUCLEI

In Fig. 3 we have plotted the energies of the first excited states as a function of neutron number with the atomic number as parameter. It is clear that in the region of neutron number 94 to 108, the value of $\mathcal{I}$, the moment of inertia (where $\mathcal{I}=I(I+1)\hbar^2/2E$, E being the energy of the level of spin I),[25] has a dependence on the proton number for a given neutron number. This is particularly apparent at a neutron number of 96, where the atomic numbers are 64, 66, and 68. The lowest energy observed was that of 73.0 kev for ${}_{66}Dy_{98}{}^{164}$. Curves of a similar nature have been presented by Scharff-Goldhaber[46] and Hollander.[47]

TABLE VII. Tabulation of transitions and ratios of level energies for postulated rotational levels in even-even nuclei as observed in this investigation.

Nuclide	Energy (kev)	(Second excited level / First excited level)[a]	(Third excited level / First excited level)[b]
Gd^{154}	123.2 248.1 (347.0)[c]	3.01	5.83
Gd^{156}	89.1 199.4 (296.7)	3.25	6.57
Dy^{156}	(138.1)		
Dy^{160}	87.0 197.5 (297.6)	3.27	6.69
Dy^{162}	80.8 184.8 (282.8)	3.28	6.79
Dy^{164}	73.0		
Er^{164}	91.3		
Er^{166}	80.7 184.7	3.29	
Er^{168}	79.9 184.6	3.31	
Yb^{170}	84.2 193.5	3.30	
Yb^{172}	78.7 181.5	3.31	
Yb^{174}	76.6		
Hf^{176}	88.4 202.0	3.29	
Hf^{178}	93.3 213.5	3.29	

[a] Level designations are probable.
[b] Level designations are possible.
[c] There are no multipole data for the transitions in parentheses.

[44] J. W. Mihelich and E. L. Church, Phys. Rev. **85**, 690 (1952).
[45] Smith, Robinson, Hamilton, and Langer, Phys. Rev. **107**, 1314 (1957).
¶ *Note added in proof.*—Recent data make these conclusions definite.
[46] G. Scharff-Goldhaber, Phys. Rev. **103**, 837 (1956).
[47] J. M. Hollander, Phys. Rev. **103**, 1590 (1956).

We have plotted in Fig. 3 the positions of *postulated* second excited states of even-even nuclei. These assignments are not "certain" but are "reasonable" in view of the $E2$ multipole character of the transitions, the transition intensities, and the absence of any transitions corresponding to the sum or difference of the two transitions in question. We present those data, not as proof for any given nuclear model, but as an indication of what the level structure may be.

The data for higher lying levels is more uncertain. The transitions we designate as *possible* $6+ \rightarrow 4+$ transitions are real transitions; the placing of the conversion lines in a given activity is done only after careful consideration of activation yields, half-lives, etc. The true level designations await further (and difficult) experiments. It is not out of place to point out that, however, the rotational levels which may be indicated in these nuclei do obey rather well the $I(I+1)$ energy relation.

SUMMARY

We have presented data which should be of use in level studies of neutron-deficient rare-earth nuclei. In particular, we have established the following new activities: Tb^{153}, Tb^{155}, Tb^{156m}, Dy^{155}, Ho^{156}, Ho^{160m}, Ho^{162}, Er^{168m}, Yb^{171m}, and Lu^{169}. We have confirmed and improved the data on Tb^{151}, Tb^{154}, Tb^{156}, Dy^{157}, Dy^{159}, Ho^{160}, Ho^{161}, Ho^{164}, Tm^{165}, Tm^{166}, Tm^{167}, Tm^{168}, Lu^{170}, Lu^{171}, Lu^{172}, Lu^{173}, Lu^{174}, and Hf^{173}. Tables VII and VIII lists those transitions proceeding between rotational levels observed in this survey. The occurrences of several new isomers are discussed. We have plotted the positions of excited levels of even-even nuclei and are planning to extend our data toward isotopes of lower neutron numbers, i.e., nuclei which have less deformation, as well as to nuclei of higher atomic number. Scintillation-counter studies are now being made on a number of the longer lived activities, and will be reported at a later time. We are continuing this survey using recently available enriched isotopes, and are obtaining precise energy measures of transitions up to 1600 kev.

TABLE VIII. Transitions proceeding between possible rotational levels for odd-A nuclei studied in this investigation.

Nuclide	Energy (kev)	Ground state spin	Remarks
Gd^{151}	108.3		
Gd^{155}	60.1	$\frac{3}{2}$[a]	
Tb^{157}	60.76 83.0 143.9	$\frac{3}{2}$[b]	$E_2/E_1 = 2.37$
Tb^{159}	58.0	$\frac{3}{2}$[c]	
Dy^{161}	77.5	$\frac{3}{2}$,[d] $\frac{5}{2}$[e]	See section on Ho^{161}
Er^{165}	(22.9) 54.5 77.3	$\frac{1}{2}$[b]	{23.0-kev transition very weak. Anomalous level spacing.
Yb^{171}	66.7	$\frac{1}{2}$[e]	
Yb^{173}	78.8	$\frac{5}{2}$[e, f]	
Lu^{173}	135.3 162.3 297.8	$\frac{7}{2}$[b]	$E_2/E_1 = 2.20$

[a] D. R. Speck, Phys. Rev. **101**, 1725 (1956); W. Low, Phys. Rev. **103**, 1309 (1956).
[b] Deduced from energy ratios.
[c] J. E. Mack, Revs. Modern Phys. **22**, 64 (1950).
[d] M. Murakawa, J. Phys. Soc. Japan **11**, 804 (1956).
[e] A. H. Cooke and J. G. Park, Proc. Phys. Soc. (London) **A69**, 282 (1956).
[f] K. Krebs and H. Nelkowski, Z. Physik **141**, 254 (1955) and Ann. Physik **15**, 124 (1954).

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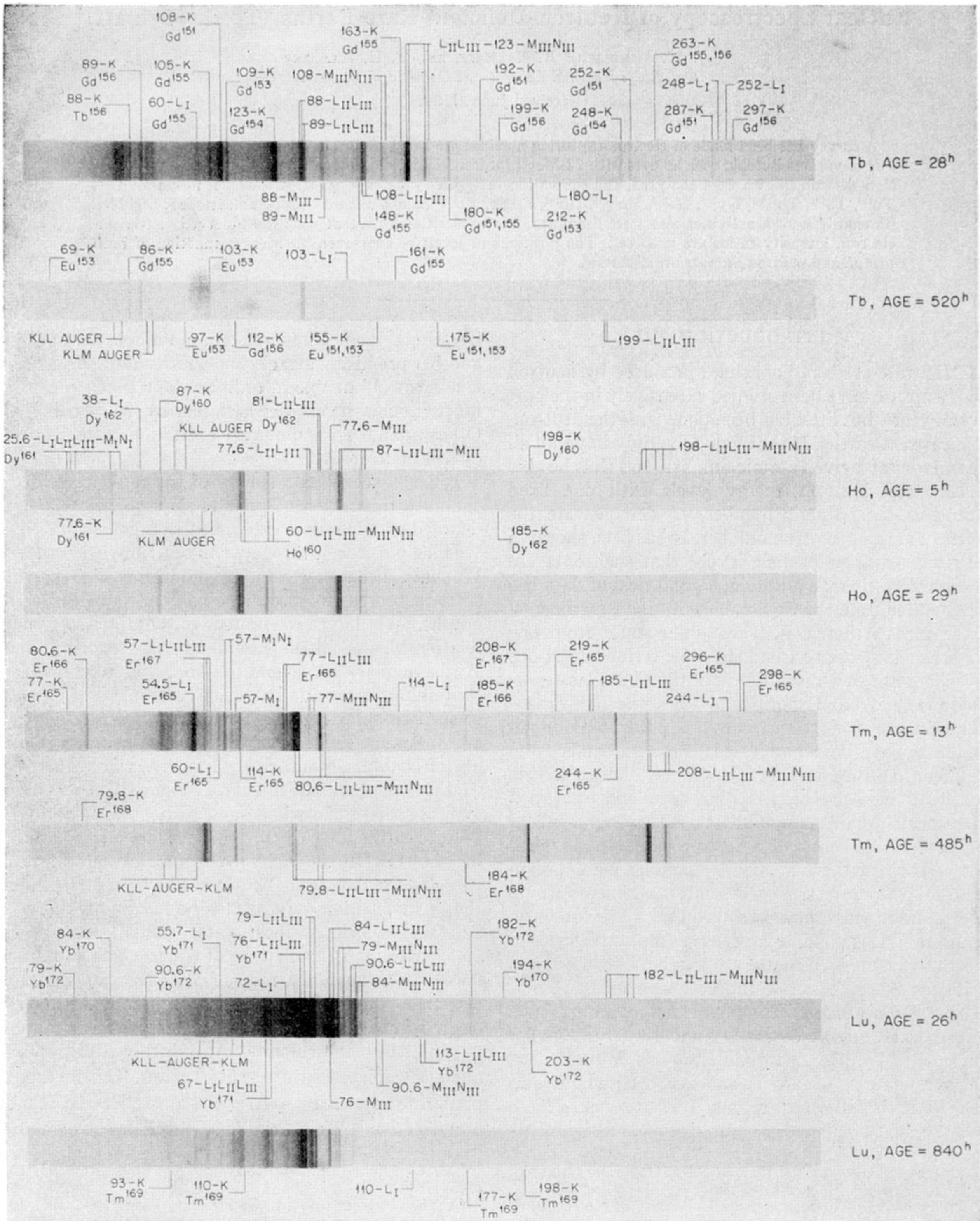

FIG. 1. Conversion-electron spectra of rare earths of various ages produced by 22-Mev protons (127-gauss magnet).