Coulomb Excitation of Rare-Earth Nuclei with Alpha Particles*

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Using our doubly charged He beam at 6 Mev we have excited most low-lying (rotational) states in the nuclei between $Z=60$ and $Z=73$. Assignments were made with isotopically enriched targets where available. On the whole the predictions of the "strong coupling" approximation of the unified model are strikingly borne out by our observations. All 2^+ first-excited states of even-even nuclei lying below $\sim$600 kev were observed, some of which were previously unknown. Two excited states were found in most odd-A nuclei as revealed by direct and/or cascade radiation to the ground state as well as coincidence measurements. The positions of the first two rotational levels agree with the simple interval rule in all established cases within the experimental accuracy. The majority of these states are not found in beta-ray spectroscopy. The reduced electric-quadrupole transition probabilities for all these transitions were measured and found to be some 50 to 100 times greater than those expected for an independent particle. Good agreement was found with some values of these quantities available from direct lifetime observations, giving us confidence in the correctness of both the theoretical expression for the total excitation cross section, and the experimental approach for its determination. A systematic decrease in these transition probabilities is evident in even-even nuclei upon approaching the closed-neutron shell at $N=82$; a corresponding increase in the energy of the 2^+ levels reflects the common origin of both phenomena, namely the decrease of the intrinsic deformation of the nuclei. However, the absolute magnitude derived for this deformation depends upon which manifestation is used.

Some evidence for a difference in splitting constants $\hbar^2/2\mathcal{J}$ between even-even, even-odd, and odd-even nuclei is found. Within the rather large uncertainties the transition probabilities do not seem to show a corresponding variation.

I. INTRODUCTION

DURING our initial survey of gamma radiation emitted by nuclei under low-energy alpha-particle bombardment,[1,2] it became apparent that some of the most strongly excited gamma rays occurred in the rare-earth region, mainly between neodymium ($Z=60$) and tantalum ($Z=73$). Some of the energy levels thus excited were known from beta- and gamma-ray spectroscopy; some of them had not been observed previously.

In a few cases the lifetimes of these transitions had been determined by fast coincidence techniques to be about 50 to 100 times faster than predicted on the basis of single-particle estimates. These so-called "fast $E2$" transitions have been known for some time[3]; they are, of course, particularly well suited to study by Coulomb excitation, since the cross section for this $E2$ process is then about 50 to 100 times greater than expected for "normal" $E2$ transitions. In fact, it was pointed out[4] before the discovery of Coulomb excitation that the latter process would especially favor the speeded-up electric quadrupole transitions which play such a prominent role in the unified description of the nucleus.

In one of our previous communications,[5] we have reported preliminary results on the rare-earth region, with particular emphasis on the level spacings observed in nuclei of odd mass number and their bearing on rotational interpretation. In the meantime, we were able to obtain most of the presently available enriched isotopes[6] in this region. This has led to confirmations (and some revisions) of previous level assignments, and has made it possible to obtain absolute cross-section measurements in most cases. From these measurements we can derive reduced transition probabilities, which can be compared with those derived from lifetime measurements.[7,8] Satisfactory agreement in the few cases where overlap of the two methods exists has given us confidence in our general approach. Trends in the sizes of the transition matrix elements bear out the general predictions of the "strong coupling" approximation of the unified model, to be discussed in Sec. IV.

What we observe, in general, can be summarized as follows: only one energy level, the first-excited state with spin 2^+, is excited in even-even nuclei, without exception. In odd-A nuclei, we usually find excitation of two energy levels with presumed spins I_0+1 and I_0+2, respectively, I_0 being the ground-state spin, all parities being the same.[9] Ideally we should then observe three gamma rays, representing transitions from these two states to the ground state, and the (cascade) transition between the two excited states. For technical reasons, however, we see all three gamma rays only in two cases (Eu^{153} and Ta^{181}); most of the time the cascade radiation is hidden by the very much stronger radiation

* A preliminary account of the results in this paper has been presented at the 1955 Annual Meeting of the American Physical Society at New York City [N. P. Heydenburg and G. M. Temmer, Phys. Rev. **98**, 1198(A) (1955)].

[1] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **93**, 351 (1954).

[2] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **93**, 906 (1954).

[3] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[4] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selkab, Mat.-fys. Medd. **27**, No. 16 (1953).

[5] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **94**, 1399 (1954).

[6] Obtained from the Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

[7] F. K. McGowan, Phys. Rev. **80**, 923 (1950); **85**, 151 (1952); **87**, 542 (1952).

[8] A. W. Sunyar, Phys. Rev. **95**, 626 (1954); **98**, 653 (1955).

[9] A. Bohr, dissertation, Copenhagen, 1954 (unpublished); also *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. 17.

of only slightly lower energy from the first excited state. In some cases we only see one gamma ray, which may be associated with one of the three possibilities just mentioned; the other two lines may be hidden by lines from other contaminating isotopes or by the strong characteristic x-radiation excited directly by the incident charged particles, or following the internal conversion of Coulomb-excited gamma radiation. In the detailed discussion which follows in Sec. III we shall present evidence or arguments supporting one or another of these possibilities. In some odd nuclei no lines whatsoever were observed (La^{139}, Pr^{141}, Nd^{143}, Sm^{147}, Sm^{149}).

We have been able to assign all observed lines in $_{60}Nd$, $_{62}Sm$, $_{64}Gd$, and $_{72}Hf$ by means of enriched isotopes; no problem exists in the monoisotopic, odd-A elements $_{65}Tb$, $_{67}Ho$, $_{69}Tm$, and $_{73}Ta$. The case of $_{63}Eu$ is almost certainly settled even though it consists of two about equally abundant isotopes. The element $_{71}Lu$ consists predominantly of isotope 175 (97.4 percent), the balance being occupied by the odd-odd isotope 176. Assignments in this case could be made plausible on the basis of abundance.

This leaves the even-Z elements $_{66}Dy$, $_{68}Er$, and $_{70}Yb$, for which no enriched isotopes are as yet available. We shall nevertheless present our results obtained with natural targets of these elements, since they will become useful as soon as assignments can be made. Because of the systematic behavior of the first-excited states of even-even nuclei[10,11] some fairly consistent identification of gamma rays can be made even for these poly-isotopic elements.

II. EXPERIMENTAL METHOD

A. Alpha-Particle Beam

We have previously described the general features of our experimental setup.[12] Throughout this work we used the doubly charged helium beam produced when singly-charged helium ions lose their second electron upon passing through an oxygen gas stripper[13] mounted some 300 kev below our rf ion source. The nominal beam energy was kept at 6 Mev by running the electrostatic generator at 3.15 Mev. The beam energy is believed to be known to ± 50 kev; the precise energy calibration of the beam must await comparison with some sharp resonances in this energy range determined elsewhere, or the completion of our electrostatic analyzer. The beam energy is known to sufficient accuracy for our present purposes. Most of our previously reported work[1,2,5,14] was done with our He^+ beam which is limited to energies below about 3.6 Mev. Although many of the levels discussed in this paper could be reached with this lower energy beam, the yield of gamma rays in the 200-kev region is increased by a factor of about 40, with correspondingly larger gains in intensity (and hence in accuracy) for higher energy levels, upon shifting to the 6-Mev beam. The beam currents we used ranged from 0.05 to 0.10 microampere, and were integrated in a Faraday cup.

B. Targets

We used thick oxide targets (except for Ta) which were prepared by compressing of the order of 100 mg of the salts into a standard one-half inch wide depression in a small aluminum target cup, thus assuring reproducible thicknesses of material to be traversed by the gamma radiation before it enters the scintillation crystal. An empirically determined self-absorption correction was applied to all observed gamma yields. The geometry was kept rigorously fixed by a tight-fitting Lucite spacer between the target cup and the front face of the aluminum can containing the $1\frac{3}{4}$ in.$\times$2 in. NaI(Tl) crystal. In many cases we inserted from 0.010 in. to 0.025 in. of copper absorber between target and crystal to attenuate strong x-radiation or interfering low-energy gamma radiation from the targets, for which proper correction was, of course, made. For a schematic diagram of our experimental arrangement, we refer to Fig. 1 in reference 12. Since the solid angle subtended by the detector at the target is nearly 2π, no correction was necessary for anisotropy in the gamma emission.

We obtained an empirical conversion factor from the oxides to equivalent pure-element targets by comparing the gamma-ray yields of ordinary HfO_2 and Ta_2O_5 with metallic Hf and Ta targets. The ratio found in both cases was 0.50. We have assumed that the same ratio applies to all rare-earth oxides of the form X_2O_3, where X represents any rare-earth element. Changes in this factor over the interval $Z=60$ to $Z=73$ are expected to be small; consequently, all observed yields were multiplied by 2.0. As noted previously,[14] the only other major effect ascribable to the presence of oxygen in the target is the appearance of a gamma-ray line at 342 kev. We were able to show by means of a Ba_2CO_3 target enriched to 9 percent in O^{18}, that this line results from the $O^{18}(\alpha,n\gamma)Ne^{21}$ reaction, O^{18} having a natural abundance of 0.20 percent. Since the yield of this line was found to be rather independent of element over the rare-earth region, and is of course unaffected by isotopic enrichment, it serves as a valuable intensity marker for the intercomparison of the various targets. In the few instances where a Coulomb-excited line fell close to 340 kev, we were able to perform a satisfactory subtraction of the oxygen contribution. As a check, we also observed these gamma rays from 3-Mev proton excitation.

C. Energy and Intensity Measurements

The gamma-ray energies, determined by calibrating our system with a number of reference sources listed previously,[12] are believed to be reliable to ± 1 percent.

[10] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).
[11] P. Stähelin and P. Preiswerk, Nuovo cimento **10**, 1219 (1953).
[12] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **94**, 1252 (1954).
[13] J. W. Bittner, Rev. Sci. Instr. **25**, 1058 (1954).
[14] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **96**, 426 (1954).

The absolute thick-target yields were obtained as follows: first, a thin source of Au^{198} calibrated in a 4π absolute beta counter at the National Bureau of Standards,[15] was placed at the target position, and the intensity under the photoelectric peak produced by the 411-kev gamma ray was determined. Knowing the absolute disintegration rate of the source we immediately obtain the effective photopeak efficiency of our system for a gamma ray of this energy (8.87 percent). The relative effective photopeak efficiencies for gamma rays of different energies were determined by referring to some semiempirical results obtained by a Swiss group.[16] We can then calculate all the transition probabilities in the manner to be described below. We find good agreement with the known transition probability[17] for the 136-kev transition in tantalum, as well as with those obtained from lifetime measurements where available.

We should like to point out that at no time did we find evidence of continuous gamma radiation (bremsstrahlung)[18] produced by alpha particles in the targets, so that no correction had to be made for such an effect. This is evident from our pulse-height distributions[1,14] and is in keeping with the dependence of the bremsstrahlung cross section on charge and mass of the projectile.[18] Direct background radiation from our generator was also found negligible; the only background for which we had to correct was that produced by Coulomb-excited gamma rays of higher energy than the one of interest.

D. Extraction of Transition Probabilities

The thick-target yields were reduced to thin-target cross sections as follows: We start with the expression for the $E2$ Coulomb excitation cross section as given by Alder and Winther[19]

$$\sigma=(2\pi^2/25)(m^2v_f^2/Z_2^2e^2\hbar^2)g_2(\xi)B(E2), \qquad (1)$$

where $g_2(\xi)$ is a numerically tabulated function of the adiabatic parameter

$$\xi=\frac{Z_1Z_2e^2}{\hbar}\left(\frac{1}{v_f}-\frac{1}{v_i}\right),$$

v_f is the final projectile velocity after collision, v_i is the initial projectile velocity, $B(E2)$ is the sought-after reduced $E2$ (upward) transition probability, ΔE is the energy of the excited state above the ground state, Z_1 and Z_2 are the charges of the projectile and target nucleus, respectively, and m is the projectile mass. The adequacy of expression (1) for the total excitation cross section has been recently demonstrated theoretically[20,21] and supported experimentally.[1,14] For the sake of simplicity we shall use the approximation $\Delta E/E \ll 1$, a condition which is rather well satisfied for our bombarding energy, and level energies below $\sim$300 kev [for $\Delta E>300$ kev, expression (1) was used]; upon inserting numerical constants appropriate for alpha particles, Eq. (1) can be written

$$\sigma(E)\cong(15.1E/Z_2^2)g_2(\xi)B(E2), \qquad (2)$$

where $B(E2)$ is expressed in 10^{-48} cm^4,[22] $\sigma(E)$ in barns, and E in Mev, and ξ is now given by

$$\xi\cong(Z_1Z_2e^2/\hbar v)(\Delta E/2E). \qquad (3)$$

Expression (2) must now be integrated along the track of the slowing-down incident particle. We make the following assumptions[23] concerning the dependence of the stopping power on particle energy and the atomic number Z_2 of the target:

(a) the stopping power, dE/dx, varies as $E^{-\frac{1}{2}}$;

(b) the stopping power *per atom* varies as $Z_2^{\frac{1}{2}}$.

These assumptions are in keeping with experiments on the stopping powers at low energies.[24–26] Since $g_2(\xi)$ is a very steep function of projectile energy, most of the yield is produced at the beginning of the alpha-particle track, and the thick-target yields turn out to be rather insensitive to the details of assumption (a) (see below). Since the range in atomic numbers for the targets studied here is small, not much error can be introduced by any slight departures from assumption (b). If we denote by a subscript zero those quantities referring to the incident particle energy, we obtain the following expression for the thick target yield:

$$Y=N\sigma(E_0)\left[\frac{E_0y(\xi_0)}{(dE/dx)_0}\right], \qquad (4)$$

where $y(\xi_0)$ stands for a numerical integral given by

$$y(\xi_0)=\tfrac{2}{3}\xi_0^{5/3}\frac{1}{g_2(\xi_0)}\int_{\xi_0}^{\infty}\frac{g_2(\xi)}{\xi^{8/3}}d\xi, \qquad (5)$$

and N is the number of target atoms per cc. The factor in brackets occurring in Eq. (4) evidently represents an equivalent target thickness. $\sigma(E)$ is given by Eq. (2) [or Eq. (1) when $\Delta E>300$ kev]. In Table I, we list some representative numerical values of the function

[15] We are indebted to L. Cavallo and H. H. Seliger for this calibration.
[16] Maeder, Müller, and Wintersteiger, Helv. Phys. Acta **27**, 3 (1954).
[17] Alder, Bohr, Huus, Mottelson, Winther, and Zupančič, Revs. Modern Phys. (to be published).
[18] C. Zupančič and T. Huus, Phys. Rev. **94**, 205 (1954).
[19] K. Alder and A. Winther, Phys. Rev. **96**, 237 (1954); see also reference 17.
[20] G. Breit and P. B. Daitch, Phys. Rev. **96**, 1447 (1954).
[21] L. C. Biedenharn and C. M. Class, Phys. Rev. **98**, 691 (1955).
[22] It is understood throughout this paper that the quantity $B(E2)$ has been divided by e^2.
[23] Suggested by T. Huus and B. R. Mottelson (private communication).
[24] C. B. Madsen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 13 (1953).
[25] Chilton, Cooper, and Harris, Phys. Rev. **93**, 413 (1954).
[26] J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 15 (1953).

TABLE I. Numerical values of the thick-target function $y(\xi_0)$ *vs* the adiabatic parameter ξ_0. $y(\xi_0)$ is defined in Eq. (5).

ξ_0	0.0	0.1	0.2	0.3	0.4	0.6	0.8	1.0	1.2	1.4	1.6	1.8	2.0
$y(\xi_0)$	0.400	0.366	0.260	0.213	0.178	0.135	0.108	0.0903	0.0768	0.0674	0.0602	0.0579	0.0540

$y(\xi_0)$ for a series of values of ξ_0.[27] It turns out that if we had assumed a *constant* stopping power instead of assumption (a), the value of $y(0)$, where the maximum discrepancy is to be expected, would have been 0.500 instead of 0.400. The value of $(dE/dx)_0$ we used for tantalum was 197 Mev-cm²/g. Stopping powers for the other targets were computed from this value according to assumption (b) above, and are found in Table II.

Upon inserting (2) into expression (4) we can solve for $B(E2)$; using appropriate numerical constants, we obtain:[22]

$$B(E2)=\frac{AZ_2^2(dE/dx)_0Y}{9.06E_0^2g_2(\xi_0)y(\xi_0)}\times 10^{-48}\ \mathrm{cm}^4, \qquad (6)$$

where A is the atomic weight of the target, $(dE/dx)_0$ the stopping power of the target (for alphas) in Mev-cm²/g, E_0 the incident energy in Mev, and Y is the yield of excited nuclei per incident particle. All other quantities were defined above.

Some remarks are in order concerning the meaning of $B(E2)$. In accordance with a convention adopted in a review article on Coulomb excitation,[17] we shall denote by $\epsilon B(E2)$ for a given gamma ray, the quantity obtained when inserting the experimental thick-target yield of gamma radiation into expression (6). The quantity ϵ, which is of necessity always less than unity, contains the following factors which must still be removed before we arrive at a theoretically meaningful transition probability:

(a) The total internal conversion coefficient α_t; the experimental quantity $\epsilon B(E2)$ must be multiplied by $(1+\alpha_t)$ to obtain the total excitation of a given level. This is an important correction for almost all rare-earth levels we observe. Unfortunately, empirical information in this transition-energy region on K-conversion as well as K/L conversion ratios is scanty,[3,7,8] and theoretical tabulations presently available require rather daring extrapolation.[28,29] A further complication exists in the case of odd-A transitions which involve mixtures of $M1$ and $E2$ radiation; knowledge of this mixture is essential for a determination of internal conversion, and is very scarce in the rare-earth region, especially since many of the transitions are seen only in Coulomb excitation. A number of the transitions have been observed by Huus[30] using conversion-electron detection, so that in principle, at least, total conversion coefficients could and should be determined experimentally.

(b) In the case of odd-A nuclei, where the second rotational state is often excited, a branching correction must also be made. As was mentioned above, the cascade transition from second- to first-excited state is usually not observed, but its strength (corrected for internal conversion) must be added to the direct ground-state transition in order to give a true representation of the total excitation of the second level. In the case of Ta^{181}, for instance, the 167-kev cascade is stronger than the direct 303-kev transition.[17] These branching ratios are of special theoretical interest, since they are connected with the gyromagnetic ratio of collective flow, g_R[4,9] a quantity which is not otherwise obtainable (see Sec. IV-B below). Unfortunately, our results are meager on this score, because of insufficient resolution of scintillation counters. It is hoped that methods capable of higher resolution, such as internal-conversion spectrometry and possibly bent-crystal spectrography will contribute more of these branching ratios. In view of the existing uncertainties discussed above, we only feel justified in reporting values of $\epsilon B(E2)$ *uncorrected* for either (a) or (b) except for the few cases where comparison can be made with values obtained from lifetime measurements.[7,8] Whenever total internal conversion coefficients and branching ratios become available from experiment in the future, the factor ϵ can readily be removed.†

E. Accuracy

A few words should be said concerning the reliability of our quantities $\epsilon B(E2)$. Uncertainties due to counting statistics alone are always negligible. While it is difficult to ascertain the errors inherent in the reduction of our results, we feel that in view of such evidence as the rather good agreement of our results with lifetime measurements, our error in the absolute value of any given transition probability can be conservatively fixed at ±30 percent. The relative error between the isotopes of one element or even neighboring elements is estimated at ±15 percent. In general, lines giving rise to stronger photopeaks are inherently more reliable than weak lines; lines that are more completely resolved from their neighbors also yield better values.

III. RESULTS

In Table II we have collected our experimental results for the rare-earth region. We list, in turn: the element;

[27] We are indebted to T. Huus for furnishing these values.
[28] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951); Rose, Goertzel, and Swift (privately circulated tables).
[29] Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).
[30] T. Huus (private communication).

† *Note added in proof.*—A. W. Sunyar has kindly furnished estimates of the total $E2$ conversion coefficients for our even-even transitions (cf. reference 8). Using these, we have evaluated nuclear deformations for even-even nuclei, which have recently been published in this Journal [Phys. Rev. **99**, 1609 (1955), especially Fig. 1].

TABLE II. Summary of results on Coulomb excitation of rare-earth nuclei by 6-Mev alpha particles. For explanation of symbols used in the column headings, see text. Entries in parentheses are tentative assignments.

Element	A	Abundance (%)	I_0	$E\gamma$ (kev)	ξ_0	$g_2(\xi_0)$	$y(\xi_0)$	$(dE/dx)_0$[a] (Mev-cm²/g)	$\epsilon B(E2)$ (10^{-48} cm⁴)
$_{60}$Nd	145	78.60[b]	7/2	70	0.0925	1.08	0.368	224	~0.03
	146	17.2	0+	455	0.640	0.200	0.126	224	0.84
	148	89.85[b]	0+	300	0.412	0.49	0.171	224	1.50
	150	5.6	0+	128	0.169	0.96	0.283	224	1.24
$_{62}$Sm	148	72.73[b]	0+	562	0.826	0.086	0.105	218	2.06
	150	68.04[b]	0+	337	0.480	0.385	0.154	218	2.32
	152	26.8	0+	122	0.166	0.97	0.288	218	1.36
	154	22.7	0+	82	0.112	1.05	0.350	218	0.48
$_{63}$Eu	(151)	47.8	5/2	310	0.447	0.436	0.162	218	0.64
	153	52.2	5/2	82	0.114	1.05	0.348	218	0.36
				105[c]	0.259	0.80	0.229	218	0.15[c]
				187	0.259	0.80	0.229	218	0.49
$_{64}$Gd	154	2.15	0+	123	0.173	0.96	0.282	213	2.10
	155	72.28[b]	(7/2)	145	0.204	0.90	0.258	213	0.112
	156	20.5	0+	89	0.125	1.02	0.332	213	1.24
	157	69.68[b]	(7/2)	131	0.184	0.93	0.272	213	0.083
	158	24.9	0+	79	0.111	1.06	0.350	213	1.02
	160	21.9	0+	76	0.107	1.07	0.355	213	~1
$_{65}$Tb	159	100	3/2	79[c]	0.194	0.92	0.265	212	0.19[c]
				136	0.194	0.92	0.265	212	0.041
$_{66}$Dy[d]	161 }		(7/2) }						
	162 }		0+ }						
	163 }	97.6	(7/2) }	76	0.110	1.06	0.351	208	0.23[d]
	164 }		0+ }						
	(161) }	43.9	(7/2) }	166	0.240	0.84	0.239	208	0.29[d]
	(163) }		(7/2) }						
$_{67}$Ho	165	100	7/2	94	0.138	1.01	0.318	207	0.54
				206	0.312	0.69	0.206	207	0.036
$_{68}$Er[d]	162 }		0+ }						
	164 }		0+ }						
	166 }	100	0+ }	79	0.118	1.04	0.341	206	0.48[d]
	167 }		7/2 }						
	168 }		0+ }						
	170 }		0+ }						
	(167)	22.9	7/2	172	0.256	0.81	0.230	206	0.081[d]
$_{69}$Tm	169	100	1/2	109[c]	0.182	0.94	0.275	205	1.12[c]
$_{70}$Yb[d]	170 }		0+ }						
	172 }		0+ }						
	173 }	85.7	5/2 }	78	0.120	1.03	0.340	202	0.28[d]
	174 }		0+ }						
	176 }		0+ }						
	(171)	14.3	1/2	110	0.169	0.96	0.286	202	0.20[d]
	(173)	16.1	5/2	180	0.276	0.77	0.221	202	0.086[d]
$_{71}$Lu	175	97.4	7/2	114	0.177	0.96	0.280	201	0.72
				250	0.402	0.51	0.175	201	0.20
	(176)	2.6	>9	180	0.280	0.76	0.220	201	(1.14)
$_{72}$Hf	176	59.45[b]	0+	87	0.137	1.01	0.320	198	0.56
	177	59.08[b]	?	112	0.176	0.96	0.280	198	0.77
				250	0.408	0.50	0.172	198	0.55
	178	31.73[b]	0+	90	0.142	1.00	0.311	198	0.85
	179	53.32[b]	?	119	0.187	0.92	0.272	198	0.67
				260	0.424	0.47	0.169	198	0.056
	180	44.20[b]	0+	93	0.146	0.99	0.309	198	0.78
$_{73}$Ta[e]	181	100	7/2	136	0.218	0.88	0.241	197	0.70
				167[c]	0.504	0.353	0.149	197	~0.13[c]
				303	0.504	0.353	0.149	197	0.15

[a] Stopping power for pure element.
[b] Isotopically enriched target used; all other assignments from natural targets.
[c] Cascade radiation from second rotational state, whose ΔE is used in computation of ξ_0.
[d] Preliminary assignments until enriched isotopes become available; see text.
[e] Metallic target; all others are oxides of the form X_2O_3 (except HfO_2).

mass number of the responsible isotope; relative abundance (either in the natural, or the enriched sample where used); spin of the ground state, I_0; gamma-ray energy in kev (equal to level energy except where otherwise noted); ξ_0; $g_2(\xi_0)$; $y(\xi_0)$; $(dE/dx)_0$ in Mev-cm^2/g; and $\epsilon B(E2)$ in 10^{-48} cm^4.[22] The last five quantities were defined in Sec. II-B. The quantities $\epsilon B(E2)$ listed have been corrected for the presence of oxygen in the targets and brought up to 100 percent, using the appropriate abundances. $(dE/dx)_0$ refers to the stopping power in the pure elements. For targets where no enriched isotopes were available, the abundance correction is made as described below in more detail for the various elements.

A. Lanthanum ($Z=57$)

This element is monoisotopic and consists of the nucleus La^{139}.[31] A careful search failed to reveal the 166-kev transition which is known from the decay of Ce^{139} (or any others). Internal conversion data ascribe an $M1$ character to this radiation.[32] Recent results from low-temperature nuclear alignment of Ce^{139} support this assignment, although a small amount of $E2$ admixture seems indicated.[33] From the negative Coulomb excitation results we can conclude that the $E2$ transition probability (if any) is about two orders of magnitude smaller than in the stronger rare-earth cases, i.e., of the order of the single-particle transition strength. The fact that La^{139} contains a magic number of neutron ($N=82$) is undoubtedly related to this observation.

B. Cerium ($Z=58$)

This element consists entirely of even-even isotopes, the predominant one again having 82 neutrons (Ce^{140}, 88.5 percent) and a known first-excited 2^+ state at 1.61 Mev. We therefore observe no gamma rays from Ce (1.6 Mev is far too high an energy to be excited with 6 Mev alphas) as pointed out previously[2]; the low-energy gamma rays we saw are undoubtedly due to rare-earth impurities.

C. Praseodymium ($Z=59$)

A careful re-examination of monoisotopic Pr^{141} did not yield the 145-kev gamma ray known from the beta decay of Ce^{141}. As in the case of La^{139}, low-temperature nuclear alignment experiments have been carried out on Ce^{141} which establish the transition as $M1$, with an $E2$ admixture of about 4 percent.[34] Pr^{141} is another "magic" nucleus with 82 neutrons. Our negative result again shows the single-particle character of this $7/2^+\rightarrow 5/2^+$ transition, with no effective competition to $M1$ radiation from $E2$ radiation.

[31] Except for a negligible amount of the odd-odd nucleus La^{138} (0.089 percent).
[32] C. R. Pruett and R. G. Wilkinson, Phys. Rev. **96**, 1340 (1954).
[33] Ambler, Hudson, and Temmer (private communication).
[34] Ambler, Hudson, and Temmer, Phys. Rev. **97**, 1212 (1955), and Phys. Rev. (to be published).

D. Neodymium ($Z=60$)

For this element we had enriched targets of both odd-A isotopes Nd^{143} and Nd^{145}, and the even-even isotope Nd^{148}. This permitted us to assign new 2^+ first-excited states to Nd^{148} and Nd^{150} at 300 kev and 128 kev, respectively, as well as to observe the 455-kev state in Nd^{146}. The newly found transitions are shown in Fig. 1. Because of the rapid rate of change of excitation energy with neutron number in this region, we could excite no levels in Nd^{142} and Nd^{144}. A gamma transition of 70 kev, rather weak compared to most rare-earth transitions, was found associated with Nd^{145}, while no gamma rays at all were found to belong to Nd^{143}. The transitions in Nd^{148} and Nd^{150} have since been studed with protons as well.[35]

The trend in the values of $\epsilon B(E2)$ as we go from Nd^{146} to Nd^{150} is worthy of note. The internal conversion corrections are small except for the 128-kev transition whose intensity should be multiplied by a factor of about 2. We see the monotonic increase in the matrix elements as we move away from the closed neutron shell at $N=82$, an effect which can be related to the decrease in excitation energy of the first-excited states (see Sec. IV). A particularly large break in excitation energy occurs between neutron numbers 88 and 90 (300 kev to 128 kev). As we shall see in Sec. III-E this break also exists in samarium.

E. Samarium ($Z=62$)

Figure 2 shows the pulse-height distribution produced by gamma rays of 122 and 82 kev from the first-excited states of the even-even nuclei Sm^{152} and Sm^{154}, respectively, as obtained with a natural target of Sm_2O_3.

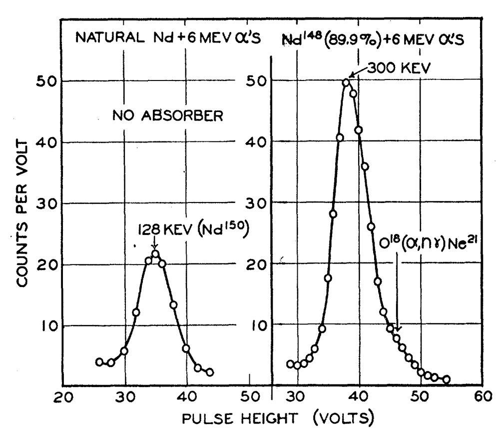

FIG. 1. Coulomb excitation of $_{60}Nd$ with 6-Mev alphas. Pulse-height distributions: ordinary Nd (left) showing 128-kev line from Nd^{150} (5.6 percent); Nd enriched to 89.9 percent Nd^{148} (right) showing 300-kev line in that isotope. Location of 342-kev gamma from $O^{18}(\alpha,n\gamma)Ne^{21}$ is indicated (see Sec. II-B). Both are newly discovered energy levels; 455-kev level in Nd^{146} observed but not shown. No absorber used.

[35] Simmons, Van Patter, Famularo, and Stuart, Phys. Rev. **97**, 89 (1955).

Figure 3 similarly shows the de-excitation gamma radiation of the first-excited 2^+ states at 562 kev and 337 kev in enriched targets of Sm^{148} and Sm^{150}, respectively. Although the 337-kev line is superimposed upon the 342-kev "oxygen" line (see Sec. II–B), its intensity can easily be found by subtracting the oxygen contribution of a neighboring rare-earth oxide. The dotted curve illustrates the magnitude of this contribution. Our observation of these four lines again confirms the spin and parity of the first-excited states in these nuclei as 2^+ and illustrates the systematic trend of their excitation energies. A particularly large jump in excitation energy is again found between neutron number 88 and 90 (337 kev to 122 kev), just as in neodymium (Sec. III-D). Additional evidence has been pointed out for a major rearrangement at neutron number 88, such as an abrupt change in the ratio of second to first-excited state energies in even-even nuclei from a value near 2 to a value near 3.33,[36] the latter being the ratio predicted by the "strong coupling" approximation of the unified model (see Sec. IV).

Although we had available enriched targets of both odd-A isotopes of Sm ($A=147$ and $A=149$), we observe no gamma rays associated with these nuclei. We can set an upper limit on the intensity of any levels in the odd nuclei of Sm of about 2 percent of that of the levels in the even-even nuclei.

It is again interesting to note the trend in the quantity $\epsilon B(E2)$ for the four even nuclei. We must remember the internal conversion correction (Sec. II-E), which raises $\epsilon B(E2)$ for the 122-kev transition by a factor of about 2, and that for the 82-kev transition

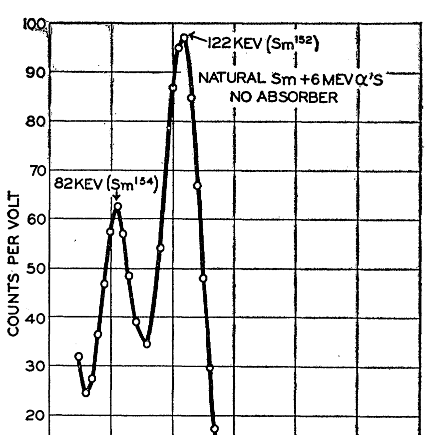

FIG. 2. Coulomb excitation of $_{62}Sm$ with 6-Mev alphas. Pulse-height distribution for ordinary Sm showing first-excited state gamma transitions at 82 kev in Sm^{154} (22.7 percent) and at 122 kev in Sm^{152} (26.8 percent). No absorber used.

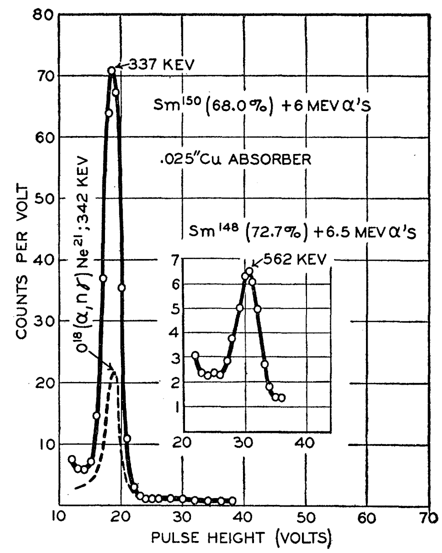

FIG. 3. Coulomb excitation of $_{62}Sm$ with alphas. Pulse-height distributions: enriched Sm^{150} (68.0 percent)+6-Mev alphas showing 337-kev transition (left); enriched Sm^{148} (72.7 percent) +6.5-Mev alphas showing 562-kev transition (right). Dashed curve indicates amount of 342-kev radiation contributed by the $O^{18}(\alpha,n\gamma)Ne^{21}$ reaction (see Sec. II-B). 0.025 in. Cu absorber used.

by a factor of about 8. The transition probability is then seen to vary inversely with the excitation energy.

The case of Sm^{152} is of special importance, because the lifetime of the 122-kev transition has actually been measured by fast coincidence techniques.[8] From the value of the half-life ($=1.4\times10^{-9}$ second), corrected for internal conversion (Sunyar estimates $\alpha_t=1.27$[8]), and Weisskopf's formula for the probability for electric quadrupole radiation,[37] we obtain a *downward* $B(E2)$ of 0.66.[38] This corresponds to an *upward* $B(E2)$ of 3.3 the statistical factor $(2I_f+1)/(2I_i+1)$ being 5 in this case. Using our value for $\epsilon B(E2)$ of 1.36, we obtain $B(E2)=3.1$ after multiplication by $(1+\alpha_t)$; this is in very gratifying agreement with the value obtained from the lifetime measurement.

F. Europium ($Z=63$)

Europium consists of two odd-A isotopes, Eu^{151} (47.8 percent) and Eu^{153} (52.2 percent). Our early investigation of Eu targets[2] revealed three gamma rays at 82 kev, 105 kev, and 187 kev. Figure 4 shows the gamma-ray spectra observed through varying thicknesses of absorber. By using two NaI crystals and two single-channel pulse-height selectors in coincidence, one set on the photopeak of the 82-kev line and the other at varying pulse-height settings, we obtained a coincidence maximum at about 105 kev, showing that some of the 82-kev radiation was indeed in cascade with the 105-kev gamma ray, and establishing the decay scheme shown in the insert of Fig. 4. This represents a well-

[36] G. Scharff-Goldhaber and J. Weneser, Phys. Rev. 98, 212 (1955).

[37] V. F. Weisskopf, Phys. Rev. 83, 1073 (1951).

[38] $E2$ transition probabilities always expressed in units of 10^{-48} cm^4.

developed rotational spectrum, as we shall discuss below. Although we have shown that all three gamma rays belong to the same nucleus we could not identify the responsible isotope, since enriched Eu^{153} is as yet unavailable. However, in the meantime the 82-kev transition has been looked for and found to be very weakly excited in the conversion-electron spectrum following the beta decay of Sm^{153},[39,40] so that the assignment of the system of levels to Eu^{153} is almost certain. Note that the quantity $\epsilon B(E2)$ for the 105-kev cascade radiation is computed using a value of ξ_0 appropriate for an excitation energy of 187 kev.

Recently we discovered an additional gamma ray of 310 kev, which we have temporarily assigned to the isotope Eu^{151}, mainly because some slight evidence for a 0.265-kev gamma ray (determined by absorption) following the decay of Gd^{151} is found in the literature.[41] Note that the two Eu isotopes are also straddling the gap between $N=88$ and $N=90$, a fact which might be related to the existence of a strong rotational spectrum in one but not the other nucleus.

G. Gadolinium ($Z=64$)

Figure 5 shows the gamma-ray spectra from ordinary Gd and the odd-A isotopes Gd^{155} and Gd^{157}. The 123-kev line belongs to Gd^{154},[42] whose natural abundance is only 2.15 percent. The large composite peak at around 80 kev is due to the even-even isotopes Gd^{156}, Gd^{158}, and Gd^{160}, whose first-excited state energies are known to be 89 kev, 79 kev,[42] and 76 kev,[30] respectively. The shifts in the peak position, and changes in peak width as well as height in the three targets of Fig. 5 are satisfactorily accounted for by their isotopic compositions; for instance, the target enriched in Gd^{155} contains 17.7 percent Gd^{156}, 2.9 percent Gd^{158}, and 0.81 percent Gd^{160}. This explains the narrower half-width of the photopeak from this target, as well as its peak value of 87 kev. We were able to obtain transition probabilities for all isotopes but Gd^{152} (0.20 percent) and Gd^{160}; the latter has a value of about the same order as Gd^{158}, but could not be accurately determined with the enriched targets at our disposal.

The two new lines in the odd-A isotopes at 131 kev and 145 kev represent an unexpected departure from the so-called "strong coupling" situation in rotational spectra; unexpected because we found well-developed rotational levels in odd-A nuclei on either side of $_{64}Gd$, namely in $_{63}Eu^{153}$ and $_{65}Tb^{159}$. The transition probabilities in Gd^{155} and Gd^{157} are about twenty times weaker than in their even-even neighbors, and the companion lines expected in odd nuclei are missing. Using a ground-state spin $I_0=7/2$,[43] the predicted ratio of second- to first-rotational state energy is 2.22;‡ if the discovered lines represent the first rotational states, the companion states should lie at 291 kev and

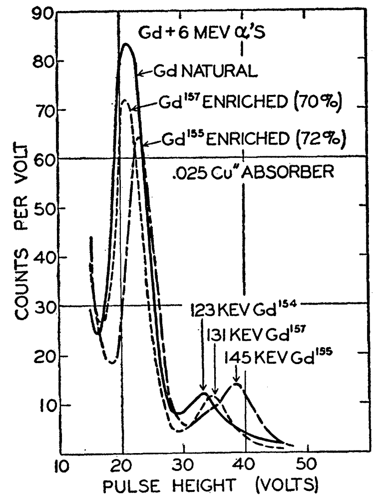

FIG. 5. Coulomb excitation of $_{64}Gd$ with 6-Mev alphas. Pulse-height distributions: ordinary Gd showing 123-kev state in Gd^{154} (2.15 percent); enriched Gd^{155} (72.3 percent) showing state at 145 kev; enriched Gd^{157} (69.7 percent) showing state at 131 kev. Large composite peak around 80 kev is due to varying amounts of Gd^{156} (89 kev), Gd^{158} (79 kev), and Gd^{160} (76 kev) in the three targets. Note low intensity of 131-kev and 145-kev lines (see Sec. III-G). 0.025 in. Cu absorber used.

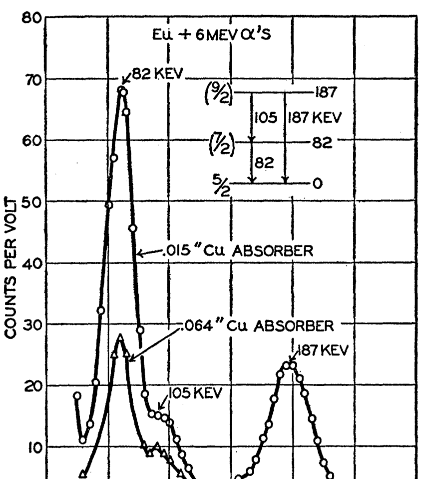

FIG. 4. Coulomb excitation of $_{63}Eu$ with 6-Mev alphas. Pulse-height distribution for ordinary Eu shows lines at 82 kev, 105 kev, and 187 kev. 0.015 in. Cu absorber used; low-energy portion of spectrum also shown as seen through 0.064 in. Cu absorber. Coincidence experiment establishes decay scheme shown as insert. Excited isotope is probably Eu^{153} (see Sec. III-F), 310-kev line also observed (not shown).

[39] N. Marty, Compt. rend. **238**, 2516 (1954).
[40] E. L. Church (private communication).
[41] R. E. Hein and A. F. Voigt, Phys. Rev. **79**, 783 (1950).
[42] E. L. Church and M. Goldhaber, Phys. Rev. **95**, 626 (1954).

[43] $I_0 \geqslant 5/2$ according to K. Murakawa, Phys. Rev. **96**, 1543 (1954).

‡ *Note added in proof.*—Recent hyperfine structure evidence shows the spin of the odd Gd nuclei to be 3/2 [F. A. Jenkins and D. R. Speck, Bull. Am. Phys. Soc. **30**, No. 5, 27 (1955)]; the appropriate ratio is then 2.40, and the situation is similar to Tb^{159} (Sec. III-H).

322 kev, respectively. Our search of this energy region revealed none, although it should be kept in mind that because of the low intensity of the first rotational states, these second states would probably be undetectable. A more attractive way out of the dilemma is to assume that the observed lines really represent *second* rotational states. This would immediately remove the difficulty of low transition probability. A glance at Table II shows their intensities to be of the order of the intensities found for second rotational states. However, we must still account for the first rotational states, which would be expected at 59 kev, and 65 kev, respectively.§ We found no direct evidence for such radiation. However, these low-energy gamma rays would be very highly converted; furthermore, the nearness of the very strong Gd K x-ray at about 44 kev complicates the detection of such gamma rays. We have carefully examined the profile of the K x-ray in the target enriched in Gd^{155} where a 65-kev line might be expected, and conclude that such a line might indeed be hidden in the tail of the x-ray peak. A strong piece of evidence in this direction is the fact that the K x-ray line from the odd-A targets was considerably ($\sim$50 percent) stronger than from the ordinary Gd target. Since the direct excitation of target x-rays is of course independent of isotopic composition, and since approximately the same visible amount of gamma radiation producing K x-rays following internal conversion exists in all three Gd targets, the excess K x-radiation from the odd-A targets is most easily explained by the presence of a strongly converted gamma transition which is itself hidden in the wing of the x-ray peak. For a similar situation where the facts are actually established we refer to the next element, Tb^{159} (Sec. III-H). The most direct confirmation of the correctness of our interpretation of the odd Gd spectra will, as in the case of Tb, have to come from an observation of internal conversion electrons.

The lifetime of the 123-kev transition in Gd^{154} is known directly.[8] Again using Sunyar's estimate for the total internal conversion coefficient ($\alpha_t=1.42$),[8] we calculate an upward transition probability $B(E2)=3.4$. From our excitation data we obtain a value of 5.1. This represents a larger discrepancy than found in the other cases where comparison is possible. The most likely reason for this is to be found in the fact that our result here was obtained from natural Gd, of which Gd^{154} constitutes only 2 percent.

H. Terbium ($Z=65$)

The gamma-ray spectrum we observe for monoisotopic Tb^{159} is shown in Fig. 6. The two transitions at 79 kev and 136 kev do not fit a rotational scheme; however, the energy difference of 57 kev together with the 136-kev gamma ray constitute a perfect rotational spectrum, with an energy ratio of 2.39. The theoretically expected ratio for the case of ground-state spin $I_0=\frac{3}{2}$ is 2.40. It turns out that Huus[30] finds a very strong conversion-electron line at 57 kev in Tb. We have checked the ratio of intensities of the 79-kev and 136-kev gamma ray and found it to remain constant between 3-Mev and 6-Mev bombarding energy. Had the 79-kev gamma ray originated at a 79-kev level, this intensity ratio would have changed by a factor of about three in favor of the 136-kev radiation. This proves that the 79-kev line originates at the 136-kev level and is the cascade to the 57-kev level. Hence the level scheme shown in the insert of Fig. 6 is established. It represents a perfect example of a rotational spectrum. In computing $\epsilon B(E2)$ for the 79-kev cascade radiation, we used the value of ξ_0 appropriate for an excitation of 136 kev.

The reason for our inability to detect the 57-kev gamma radiation is its high internal conversion and its nearness to the 46-kev x-radiation, which is itself enhanced by the internal conversion process. In spite of very careful examination of the 57-kev region with critical absorbers we were not able to detect the radiation from the first rotational state. The cascade radiation is so clearly resolved in this case for two reasons: (a) the second rotational state has a comparatively low energy so that it is strongly Coulomb-excited; (b) the ground-state spin of $\frac{3}{2}$ is the lowest one giving rise to an ordinary rotational spectrum,[9] with the maximum second-to-first state energy ratio of 2.40. This means that we have a difference between cascade and first-rotational state energy of 40 percent. Note that we were also able to see the cascade in the next most favorable cause of Eu^{153} ($I_0=5/2$), where the latter difference is 29 percent (Sec. III-F).

I. Dysprosium ($Z=66$)

Our results for this element were obtained from a natural target pending the availability of enriched isotopes. Contributions from Dy^{156} and Dy^{158} can be

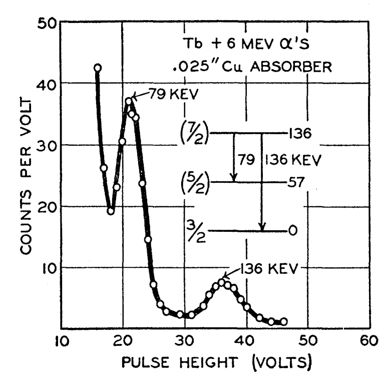

FIG. 6. Coulomb excitation of $_{65}Tb^{159}$ with 6-Mev alphas. Pulse-height distribution shows gamma rays at 79 kev and 136 kev, whose relative intensity is *independent* of bombarding energy, establishing decay scheme shown as insert (see Sec. III-H). 0.025 in. Cu absorber used.

§ *Note added in proof.*—With $I_0=3/2$, these values are 55 and 60 kev, respectively.

neglected because of their low abundances (0.05 and 0.09 percent, respectively). This leaves a total of about 53 percent for the even-even isotopes 162 and 164, and about 44 percent for the odd-A isotopes 161 and 163. The only excited state known definitely in Dy is the 86-kev level in Dy^{160} whose lifetime is known,[7] but which we did not observe in natural Dy (only 2.3 percent Dy^{160}). We find a strong, undoubtedly composite peak at around 76 kev, which we shall temporarily assume to belong to both even-even isotopes (Dy^{162} and Dy^{164}) and both odd-A isotopes for the purpose of calculating $\epsilon B(E2)$ It is reasonable to expect little change in the position of the first-excited states of the even-even nuclei in view of such behavior of these states in elements on both sides of Dy (see Gd and Hf, Secs. III-G and III-O). Hence there is little doubt that the line at 166 kev must belong to one or both odd-A isotopes, and represents their second rotational state. We temporarily make our abundance correction as if both Dy^{161} and Dy^{163} share the 166-kev level. We assume a tentative value of $I_0=7/2$ for these nuclei.[44]

J. Holmium ($Z=67$)

Figure 7 shows the gamma spectrum observed in monoisotopic Ho^{165}, and its level scheme. This again represents a well-developed rotational spectrum for a nucleus of spin 7/2. The 94-kev first-excited state is probably the one previously found in the decay of Dy^{165}, although its spin assignment is more likely to be 9/2 than 5/2.[45]

K. Erbium ($Z=68$)

Our results for this element are again based on bombardment of a natural target because of unavailability of enriched isotopes. The element mainly consists of

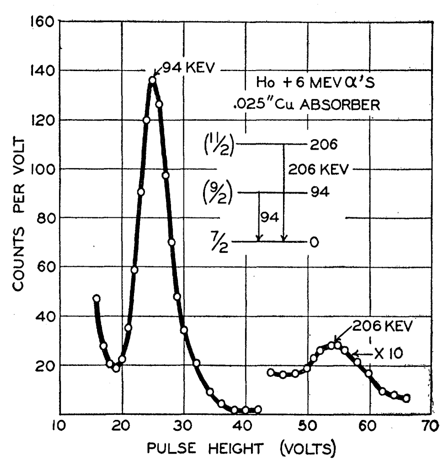

FIG. 7. Coulomb excitation of $_{67}Ho^{165}$ with 6-Mev alphas. Pulse-height distribution shows transitions at 94 kev and 206 kev. Decay scheme shown as insert. 0.025 in. Cu absorber used.

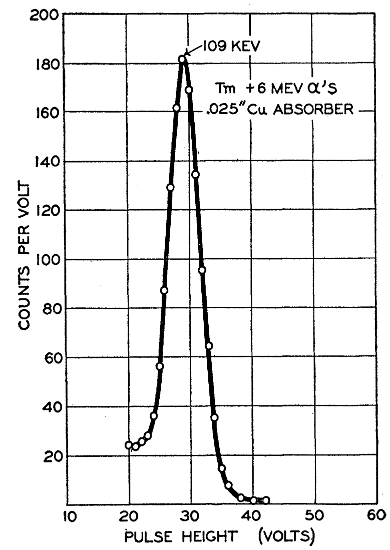

FIG. 8. Coulomb excitation of $_{69}Tm^{169}$ with 6-Mev alphas. Pulse-height distribution shows a single peak at 109 kev, probably representing a cascade transition from a 120-kev level (see Sec. III-L). This nucleus has $I_0=\frac{1}{2}$. 0.025 in. Cu absorber used.

the three even-even isotopes Er^{166}, Er^{168}, and Er^{170}, and the single odd isotope Er^{167}. As in the case of dysprosium discussed above, one expects the first-excited states of the even-even nuclei to lie close together around 80 kev. We again associate the level at 172 kev with the second rotational level of the single odd-A nucleus Er^{167}, having spin $I_0=7/2$. Using the appropriate ratio of 2.22, the first rotational level should occur at 78 kev. Hence, we shall ascribe the large photopeak at 79 kev to *all* isotopes of Er until further clarification. The only known level in any abundant even-even isotope of Er is the 81-kev state of Er^{166}, whose lifetime is also known. We calculate the *upward* transition probability from McGowan's lifetime (1.7×10^{-9} sec)[7] to be 5.3, whereas we get from our excitation data a value of $B(E2)=4.4$, using Sunyar's estimate for the total conversion coefficient ($\alpha_t=8.1$). This near agreement shows that the photopeak at 79 kev must be due to most isotopes of Er in approximate proportion to their abundances. If, for example, the 79-kev peak belonged entirely to Er^{166} (33.4 percent natural abundance), we would calculate $B(E2)=13.2$. In fact, lifetime measurement of a 91-kev state in Er^{164} [8] yields a transition probability of the same order [$B(E2)up=5.05$], supporting this conclusion.

L. Thulium ($Z=69$)

The pulse-height spectrum for the monoisotopic element Tm^{169} is shown in Fig. 8. A single peak at 109 kev is observed. Huus[30] observes two conversion-electron peaks corresponding to transitions at 111 kev and 119 kev. He believes from a study of the shape of the excitation of the 111-kev radiation, that the latter originates from a level at about 120 kev, indi-

[44] K. Murakawa and T. Kamei, Phys. Rev. **92**, 325 (1953).
[45] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

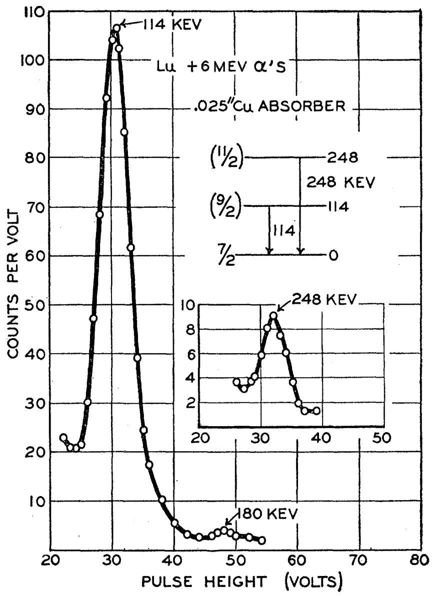

FIG. 9. Coulomb excitation of $_{71}Lu^{175}$ with 6-Mev alphas. Pulse-height distribution shows transitions at 114 kev and 248 kev (at lower gain), leading to decay scheme shown in the insert (see also reference 50). Small peak at 180 kev is arbitrarily assigned to Lu^{176} (2.6 percent). 0.025 in. Cu absorber used.

cating a first-excited state at about 9 kev. This scheme receives some support from existing data on the decay of Yb^{169}.[46] Our scintillation detector could not resolve two lines at 111 kev and 119 kev. However, from the position of the peak we seem to excite predominantly the (cascade?) radiation of 109 kev. Now the ground-state spin of Tm^{169} is $\frac{1}{2}$, and hence we do not expect a conventional rotational spectrum.[9] We have previously studied three of these spin $\frac{1}{2}$ nuclei[47]; some very precise data have recently appeared concerning W^{183}, having spin $\frac{1}{2}$.[48] All these cases illustrate the special features of the rotational system when decoupling of the odd-particle spin from the intrinsic nuclear deformation axis takes place. The nearness of the first-excited state to the ground state fixes the value of the decoupling parameter a[9] at about -0.75 and yields a "splitting constant" $\hbar^2/2\mathfrak{I}$ in keeping with that of even-even neighbors. Our transition probability for the 109-kev transition was computed using a value of ξ_0 appropriate for an excitation energy of 120 kev.

M. Ytterbium ($Z=70$)

The case of Yb is similar to those of Dy and Er. Again only natural targets were available. Only one excited state is known, the 84.1-kev level of Yb^{170}, whose lifetime has been measured.[49] The four even-even isotopes of mass numbers 170 through 176 are all fairly abundant and are again expected to have first-excited states around 80 kev. The fact that the composite peak lies at 78 kev is evidence for the existence of states at somewhat lower energies in Yb^{172}, Yb^{174}, and Yb^{176}. Pending further work with separated isotopes, we shall once again divide the photopeak at 78 kev among these four even nuclei and the odd-A isotope Yb^{173}, whose spin is 5/2. We ascribe the gamma ray at 180 kev to the second rotational state of Yb^{173}, so that the first rotational state should lie at about 79 kev. The other odd-A isotope Yb^{171} has spin $I_0=\frac{1}{2}$, so that we anticipate unconventional behavior as in Tm^{169} (see Sec. III-L). We have arbitrarily assigned the line at 110 kev to Yb^{171} for the present time. Using the total internal conversion estimate from Sunyar's tabulation[8] ($\alpha_t=7.4$) and the lifetime of the 84-kev transition in Yb^{170},[49] we calculate an upward $B(E2)$ of 5.07, whereas we get a value of 2.3 from our data. This discrepancy seems to indicate that the line at 78 kev predominantly belongs to only some of the five isotopes which we have assumed to share its intensity equally; in fact only about 40 percent of the isotopes (by abundance) would account for the observed strength.

N. Lutetium ($Z=71$)

The pulse-height distribution for this element is shown in Fig. 9. The abundance of the odd-A nucleus Lu^{175} accounts for 97.4 percent of the element, the remaining 2.6 percent belonging to the odd-odd nucleus Lu^{176}. The photopeaks at 114 kev and 250 kev constitute another example of a well-developed rotational spectrum for a nucleus of spin $I_0=7/2$. Gamma rays of nearly the same energy have recently been found in the decay of Yb^{175},[50] the only case where a second rotational state of an odd-A element was seen in beta decay.

We arbitrarily and tentatively assign the 180-kev line to odd-odd Lu^{176}, since both energy and intensity of this line are not unreasonable for this nucleus. If this identification is borne out by future measurements, it would constitute the only example of a rotational transition in an odd-odd nucleus in the "strong coupling" region.

O. Hafnium ($Z=72$)

For the element hafnium we had available enriched isotopes of the two odd-A nuclei Hf^{177} and Hf^{179}, as well as of the even-even nucleus Hf^{176}. First-excited 2^+ states were known in Hf^{176} and Hf^{180}; their lifetimes are also known.[7,8] From our three enriched targets, coupled with an ordinary Hf target, we were able to determine the quantities $\epsilon B(E2)$ for Hf^{176}, Hf^{178}, and Hf^{180}. From the lifetimes and estimates of total internal conversion coefficients,[8] we calculate the following *upward* transi-

[46] Martin, Jensen, Hughes, and Nichols, Phys. Rev. **82**, 579 (1951).
[47] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **95**, 861 (1954).
[48] Murray, Boehm, Marmier, and DuMond, Phys. Rev. **97**, 1007 (1955).
[49] Graham, Wolfson, and Bell, Can. J. Phys. **30**, 459 (1952).
[50] J. P. Mize (private communication); also Mize, Bunker, and Starner, Bull. Am. Phys. Soc. **30**, No. 3, 71 (1955).

tion probabilities:

$$Hf^{176}: B(E2)=4.9 \; ; \; Hf^{180}: B(E2)=4.4.$$

Using our Coulomb-excitation results, we obtain

$$Hf^{176}: B(E2)=4.3; \quad Hf^{180}: B(E2)=5.1,$$

in rather satisfactory agreement with the former values.

The gamma-ray spectra obtained from the odd-A isotopes of Hf are shown in Figs. 10 and 11, together with the inferred level schemes. The photopeaks in the 100-kev region show the contributions of the residual amounts of the even-even isotopes, as well as the first rotational states of the odd isotopes. Note the shift of the lower-energy peak from 90 kev in the Hf^{177} sample, whose even-even constituent is predominantly Hf^{178} (31.7 percent), to 93 kev in the Hf^{179} sample, whose even-even component is almost pure Hf^{180} (44.2 percent). The peaks shown in the 300-kev region in Figs. 10 and 11 represent, in addition to the ever-present 342-kev line from O^{18} (see Sec. II-B), the second rotational states of Hf^{177} and Hf^{179} at 250 kev and 260 kev, respectively.[51] Their intensities are seen to differ considerably (about a factor of 10) simply by comparing them with their neighboring "oxygen lines," which are of identical height. Unfortunately, the ground state spins of the odd Hf isotopes are not known,[52] but if they are the same for both nuclei, the difference in the strengths of the second rotational states is rather unexpected. At the same time, the first-excited state transitions in the two odd nuclei are of quite comparable intensities. Recent precision spectroscopy

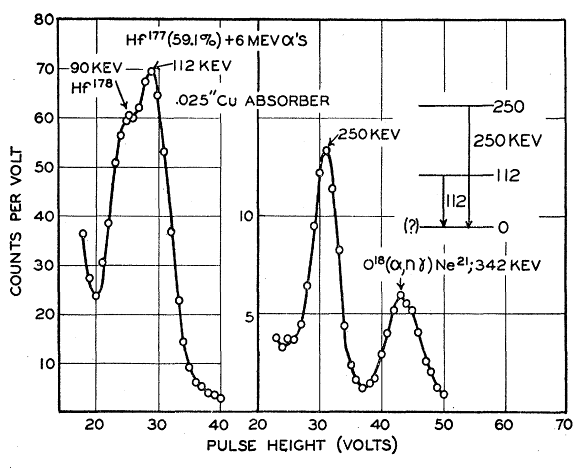

FIG. 10. Coulomb excitation of $_{72}$Hf with 6-Mev alphas. Pulse-height distribution for enriched Hf^{177} (59.1 percent) shows lines at 112 kev and (at lower gain) 250 kev belonging to Hf^{177}. One also sees 90-kev transition in Hf^{178} (31.7 percent) and 342-kev line due to $O^{18}(\alpha,n\gamma)Ne^{21}$ reaction (see Sec. II-B). Decay scheme for Hf^{177} shown as insert (see also reference 53). Note relative heights of 250-kev and "oxygen" peaks (compared to Fig. 11). 0.025 in. Cu absorber used.

[51] Lines at somewhat lower energies have been reported by McClelland, Mark, and Goodman, Phys. Rev. **97**, 1191 (1955).

[52] E. Rasmussen, Naturwiss. **23**, 69 (1935).

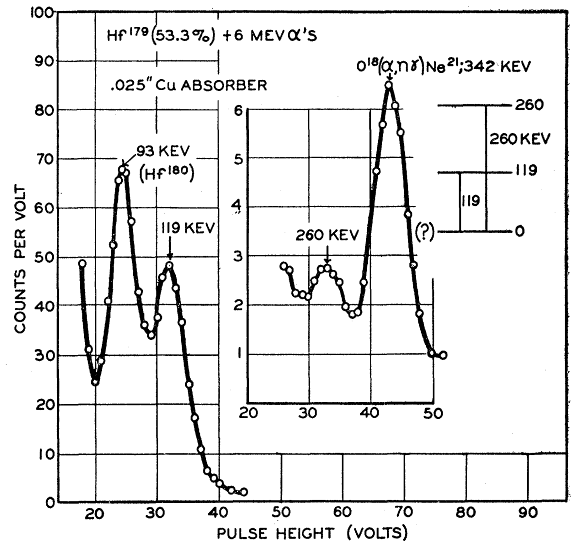

FIG. 11. Coulomb excitation of $_{72}$Hf with 6-Mev alphas. Pulse-height distribution for enriched Hf^{179} (53.3 percent) shows lines at 119 kev and (at lower gain) 260 kev belonging to Hf^{179}. One sees 93-kev transition in Hf^{180} (44.2 percent) and 342-kev line due to $O^{18}(\alpha,n\gamma)Ne^{21}$ reaction (see Sec. II-B). Decay scheme for Hf^{179} shown as insert. Note relative heights of 260-kev and "oxygen" peaks (compare to Fig. 10). 0.025 in. Cu absorber used.

of Lu^{177} by Marmier and Boehm[53] has confirmed the two levels in Hf^{177}, although some discrepancies exist concerning multipolarities of gamma-ray transitions. Using their very accurate energy values of 112.97 kev and 249.69 kev for the first and second rotational states of Hf^{177}, we can obtain a value for the ground-state spin I_0 as well as for the splitting constant $\hbar^2/2\mathcal{J}$ from the expression for rotational states (see Sec. IV). The spin value is 3.76, which is close to 7/2 and deviates from that half-integer in the direction to be expected from small vibration-rotation interaction effects,[4,9] which tend to depress higher rotational states slightly; in fact, an upward shift of 1.5 kev of the second rotational states level would yield a spin of exactly 7/2. The splitting constant in this interpretation is 11.87 kev, which is less than the 15 kev obtained from the neighboring 2^+ states at 90 kev. This again represents a trend in odd-even alternation which is observed throughout the rare-earth region, as will be seen in Fig. 13. On the other hand, Marmier and Boehm[53] favor a $p_{\frac{3}{2}}$ configuration for the ground state in keeping with shell-model predictions. If we use their spin assignments of 5/2 and 3/2 for first and second excited states, respectively, we can insert their energy values into the expression for an anomalous rotation spectrum[9] and extract values for the decoupling parameter a and splitting constant $\hbar^2/2\mathcal{J}$. They are:

$$a=1.98; \quad \hbar^2/2\mathcal{J}=27.95 \text{ kev}.$$

The latter value is definitely too large to accord with neighboring even-even nuclei; in fact the 2^+ states

[53] P. Marmier and F. Boehm, Phys. Rev. **97**, 103 (1955).

should then be found at around 180 kev, instead of the observed 90 kev. Sunyar quotes an *upper* limit for the lifetime of the 112-kev transition in Hf^{177} of 5×10^{-10} second.[8] When combined with his estimate for the total internal conversion coefficient of 2.7, this leads to *lower* limits for the upward $B(E2)$, depending upon the assumed spins, as follows:

(a) $B(E2)>2.2$ for $7/2\rightarrow9/2$;

(b) $B(E2)>5.2$ for $1/2\rightarrow5/2$.

Our excitation data yield $B(E2)=2.8$, and seem to favor choice (a).

While these arguments certainly do not constitute proof of a ground-state assignment different from $p_{\frac{1}{2}}$, we feel that they strongly support a conventional rotation spectrum starting with ground-state spin 7/2 (or possibly 5/2). A direct measurement of the spins of Hf^{177} and Hf^{179} is sorely needed.

P. Tantalum ($Z=73$)

This element is well-known to possess a simple rotational spectrum.[54] We have measured the intensities of the 136-kev and 303-kev transitions mainly as an internal check and normalization point for comparing intensities and transition probabilities. The 167-kev cascade was seen only feebly, but we were able to obtain a pronounced coincidence peak at 167 kev when examining the coincidence spectrum between 136-kev radiation and the rest of the gamma spectrum, as described above under europium (Sec. III-F). It is interesting to note that the K x-radiation emitted from the tantalum target at about 61 kev, as shown in Fig. 1 of a previous communication,[1] is almost entirely accounted for by the internal K-conversion of the 136-kev gamma ray, so that no more than a few percent of the x-ray peak can be ascribed to direct target excitation. This explains the differences in behavior of the excitation curves given in Fig. 2 of the previous reference[1] for proton and alpha-particle bombardment: the x-radiation maintains a constant ratio to the gamma radiation for alpha particles (even up to 6 Mev), whereas it increases with energy relative to the gamma ray in the case of proton bombardment.

IV. DISCUSSION

A. Review of the Unified Model

The sixteen stable elements from praseodymium through tantalum which we have studied in this paper constitute a continuous series of nuclei more or less removed from the closed shells for both protons at $Z=50$ and $Z=82$, and for neutrons at $N=82$ and $N=126$. It is just in this region that nuclei are expected to depart farthest from the spherical shape owing to the presence of a large number of nucleons outside of closed shells. The existence of large static quadrupole moments in this region of mass numbers has long pointed in this direction; the occurrence of electric-quadrupole transitions with probabilities up to two orders of magnitude greater than encompassed by the transition of a single particle, as revealed by Coulomb excitation or lifetime measurements, points in the same direction. As Bohr and Mottelson point out,[4] the existence of rotational energy levels with positions given by the simple quasi-molecular expression

$$E=(\hbar^2/2\mathcal{J})[I(I+1)-I_0(I_0+1)], \qquad (7)$$

connected by fast $E2$ transitions, is a direct consequence of the large deformations known to exist in these nuclei, "large" being quantitatively interpreted as large compared to the zero-point oscillations of the nuclear surface shape, inherent in this quantum phenomenon. If this condition is fulfilled, a preferred axis of deformation (z-axis) exists in the nucleus, and the peculiar quantized flow of nuclear matter leading to a rather simple rotational spectrum is an inevitable consequence.

Since we have a preferred axis, we can define an intrinsic measure of the departure of the nuclear surface from the spherical shape with respect to that axis, the so-called intrinsic quadrupole moment Q_0. This quantity can be defined for all nuclei, even if their spin is 0 or $\frac{1}{2}$. The quantity $\mathcal{J}$ occurring in Eq. (7) above is proportional to Q_0^2, as is also the quantity $B(E2)$ measured in our experiments. Thus large deformations imply low-lying excited states and large transition probabilities. It turns out both from our work and Sunyar's that the values of Q_0 one obtains from (7) are always larger, by a factor of about two, than those obtained from $B(E2)$, pointing to the need for refinement in the simple model. A similar discrepancy can also be discerned in the region of $A\sim100$.[55,56]

B. Coulomb Excitation and the Unified Model

The rather straightforward way in which transition probabilities are obtained from Coulomb-excitation cross sections, and the complete uniformity with which all stable nuclei are treated, make this method probably the most desirable one available today for the study of the properties of the first few excited states of nuclei. In addition to its special suitability for the measurement of fast $E2$ transitions, as pointed out in the introduction, it is a favorite for the investigation of experimental features having a bearing on the unified model in one further respect: it is in the very nature of the unified model to predict trends and properties transcending any specific nuclear species. Hence, the possibility of studying all nuclei under precisely the same conditions eliminates many sources of error inherent in the conventional methods of nuclear spectroscopy where

[54] T. Huus and C. Zupančič, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 28, No. 1 (1953).

[55] K. W. Ford, Phys. Rev. 95, 1250 (1954).

[56] G. M. Temmer and N. P. Heydenburg, Phys. Rev. 98, 1308 (1955) and to be published.

moving from one nucleus to the next may entail an entirely different decay scheme, caused by vast differences in beta-decay energy, degree of forbiddenness of the beta transitions, spins and parities of parent nuclei, not to mention the complete lack of these pieces of information in a large number of cases.

The absence of positive excitation results for the three nuclei having 82 neutrons (La^{139}, Ce^{140}, Pr^{141}) illustrates the lack of enhanced electric-quadrupole transitions for nearly spherical nuclei.

Recently some low-lying gamma-ray transitions in Eu^{153} and Gd^{155} were found to have electric-dipole character.[40,42] A few words are in order concerning the complete absence of these transitions in our work. The intrinsic Coulomb-excitation probability of $E1$ transitions[19,57] is about three hundred times *greater* than the quadrupole excitation for transitions of $\sim$100 kev and $Z=64$; however, the reduced $E1$ transition probability $B(E1)$ is expected to be very much *smaller* than that of a single proton. Remembering that the $E2$ transitions we observe are about one-hundred times stronger than those of a single particle, we would not expect to observe $E1$ transitions in spite of their being *a priori* favored by the Coulomb-excitation process. A number of $E1$ transitions have recently been discovered in the strong-coupling region beyond Pb^{206} with transition probabilities about three orders of magnitude below the single-particle strength.[58] Our negative result can be used to set a limit of about this order of magnitude for any $E1$ transitions leading to the ground state.

1. *Even-Even Nuclei*

Starting with Nd^{146}, we were able to excite all first-excited 2^+ states lying below about 600 kev, the latter limit being dictated merely by the intrinsic Coulomb-excitation mechanism as embodied by the function $g_2(\xi)$ defined in Sec. II-D. We have added a number of new cases to the growing systematics of the *positions* of first-excited states of even-even nuclei. A plot summarizing the results in this paper on even-even nuclei excited with 6-Mev alpha particles is shown in Fig. 12. The excitation energy of the first-excited state is plotted as a function of the neutron number N. The sharp rise toward the left where we approach the closed shell at $N=82$ is very apparent. A number of the levels were previously unknown (open circles). In the cases of Dy, Er, and Yb no detailed results are as yet available, as discussed in Sec. III, and the points are shown at positions representing an average over the even-even isotopes. A plot of the reciprocal of the transition probabilities would have a very similar appearance to the curve in Fig. 12. Because of the existing uncertainties in total internal conversion coefficients we refrain from presenting such a plot; we have already pointed out the monotonic variations with neutron number which exist

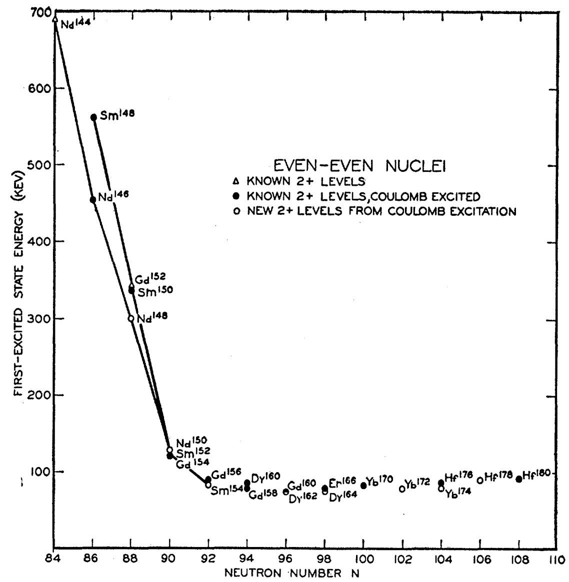

FIG. 12. Summary of results on even-even rare-earth nuclei. Energies of first-excited (2^+) states *vs* neutron number. (△)—known states, not Coulomb-excited; (●)—known states, Coulomb-excited; (○)—new states discovered by Coulomb excitation. Levels for elements where no enriched isotopes are available (Dy, Er, Yb) shown at an average position. See also references 10 and 11.

in the quantity $B(E2)$ in the region of rapid change, i.e., neutron number $N=86$ to $N=92$, as represented by elements Nd and Sm. Here the internal conversion correction is not too important. The same trend can still be discerned in the three even-even Gd nuclei, again after proper allowance is made for internal conversion.

On the whole, the transition probabilities observed here are from 50 to 100 times larger than one calculates from the single-particle formula of Weisskopf.[37]

2. *Odd-A Nuclei*

In the nuclei of odd mass number that we have examined we were usually able to observe or infer the existence of two excited states, as is expected from the $E2$ character of the excitation process and the spins and parities associated with rotational levels. No rotational energy levels in the odd nuclei (except in Ta) were known when we began this investigation. There is a difference in the reliability of our results between odd-Z (odd-even) and odd-N (even-odd) nuclei, because of the fact that the former all occur as monoisotopic elements (with the exception of Eu) and permit unique assignments to be made. In Table III, we have collected the information on level spacings for all odd nuclei, listing the first-excited state energy E_{I_0+1}, the second-excited state energy E_{I_0+2}, their observed ratio ρ_{exp} and predicted ratio ρ_{theor} for the given (or assumed) ground-state spin I_0. It must be pointed out that much greater precision in energy determination than is available with scintillation counters is required in order to have

[57] C. J. Mullin and E. Guth, Phys. Rev. 82, 141 (1951).
[58] Stephens, Asaro, and Perlman, Phys. Rev. 96, 1568 (1954).

TABLE III. Summary of results on rotational level positions in odd-A nuclei of the rare-earth region. Only nuclei having "conventional" spectra are listed ($I_0>\frac{1}{2}$). ρ_{exp} stands for experimental ratio of second-to-first excited state energies; ρ_{theor} is obtained from Eq. (7). Values of ground state spin I_0 in parentheses are assumed. This table supersedes Table I of reference 5.

Nucleus	I_0	E_{I_0+1} (kev)	E_{I_0+2} (kev)	ρ_{exp}	ρ_{theor}
$_{63}Eu^{153}$	5/2	82	187	2.28±0.04	2.29
$_{65}Tb^{159}$	3/2	57[a]	136	2.39±0.05	2.40
$_{66}Dy^{161,163}$ [b]	(7/2)	76	166[b]	2.18±0.04	2.22
$_{67}Ho^{165}$	7/2	94	206	2.19±0.04	2.22
$_{68}Er^{167}$ [b]	7/2	79	172[b]	2.18±0.04	2.22
$_{70}Yb^{173}$ [b]	5/2	78	180[b]	2.31±0.04	2.29
$_{71}Lu^{175}$	7/2	114	250	2.19±0.04	2.22
$_{72}Hf^{177}$	(7/2)	112	250	2.23±0.04	2.22
$_{72}Hf^{179}$	(7/2)	119	260	2.18±0.04	2.22
$_{73}Ta^{181}$	7/2	136	303	2.23±0.04	2.22

[a] Inferred from 79-kev cascade radiation (see Sec. III-H); conversion electrons directly observed by T. Huus (private communication).
[b] Conclusions, based on systematics, are tentative (see appropriate sections above for details).

a sensitive test of the rotational structure, especially if such small departures from the simple formula (7) as are expected from vibration-rotation interaction effects (about 1–2 kev in this region) are to be detected. The case of Hf^{177}, discussed in Sec. III-O above, is a case in point.[53] Very precise conversion-line data are also available in some odd transuranic elements (see, for instance, the case of Np^{237} as known from the alpha decay of Am^{241}).[59,60] However, our results on level positions certainly bear out the predictions of the unified model in zeroth approximation very well. No more than two levels were observed in any odd-A nucleus.

There is considerable variation in the values of $\epsilon B(E2)$ for second rotational states, as well as in the ratio of second to first-excited state transition probabilities. As we have pointed out above, the possibility (and even certainty) of $M1$ cascade competition in returning to the ground state will profoundly affect the value of $\epsilon B(E2)$ for the second rotational states. In fact, in the famous case of Ta^{181}, where the composition of the 136-kev transition has been experimentally determined by angular correlation measurements,[61] the $M1$ component outweighs the (enhanced) $E2$ component by about 6 to 1; the cascade between second and first-rotational state probably represents a similar situation. Now if we calculate the branching ratio for the de-excitation of a second rotational state, via cascade or direct crossover, to the ground state, from the theoretical expressions furnished by the unified model for $E2$ transitions only, we find that the cascade path is weaker than the crossover, the exact extent depending on spins, transition energies, and conversion coefficients. Any indication that the observed cascade transition is stronger than the crossover is evidence of additional transition probability provided by $M1$ radiation. The two cases where we are able to observe the cascade radiation directly illustrate this point. The cascade at 105 kev in Eu^{153} is rather weak compared to the 187-kev crossover transition, as can be seen from Table II. [Fig. 4 gives a distorted impression of this ratio, because of the effect of the Coulomb-excitation correction contained in $g_2(\xi)$.] The situation is reversed in the case of Tb^{159}, where the 79-kev cascade is about five times stronger than the 136-kev crossover. Internal conversion correction increases this ratio even further. Note that the actual value of $\epsilon B(E2)$ for the 136-kev transition in Tb^{159} is only about one tenth that for the 187-kev transition in Eu^{153}. We see that the true transition probabilities to these second rotational states, as represented by the sum of the crossover and cascade radiations, are more nearly equal. This is to be expected, since this probability is supposedly a function of Q_0^2 only, and the deformations in the two nuclei in question are certainly comparable. In the remaining odd-A nuclei, where we do not observe the cascade directly, we can infer its importance from the size of $\epsilon B(E2)$ for the direct $E2$ cross-over transition. When the latter is strong, the cascade transition is weak which means, as we pointed out above, that the contribution of $M1$ radiation to the cascade is reduced. From the variation in $\epsilon B(E2)$ for the second rotational states (see Table II) we see that this contribution can vary considerably from nucleus to nucleus. According to the unified model[4] the $M1$ transition probability is governed by the quantity $(g_\Omega-g_R)^2$ where g_Ω and g_R stand for the gyromagnetic ratios of the intrinsic particle configuration and the collective rotational flow, respectively; they also determine the static magnetic moment in the ground state. Variations in $(g_\Omega-g_R)$ are presumably to be ascribed to fluctuations in g_Ω, since g_R is not expected to change radically from one nucleus to the next.

Strictly speaking we should diminish $\epsilon B(E2)$ of the first-rotational state by the amount contributed by cascade from the second-rotational state, which leads to the same gamma ray. In practice, however, this gives rise to an unimportant correction because of the relatively weak excitation of the second state when using alpha particles, whose $g_2(\xi)$ falls off rapidly with excitation energy ΔE (see relative photopeak heights in Figs. 4, 6, 7, 9, and 10).

This discussion of odd-A nuclei should not be terminated without recalling the few instances where the possibility of discrepancy with the general behavior expected from the unified model exists at present.

(a) As mentioned in Sec. III-D, the case of Nd^{145} is exceptional in that we find a weak, but low-energy transition at 70 kev with no apparent companion lines. The complete absence of any observable transitions, such as in Nd^{143}, Sm^{147}, and Sm^{149} is more in keeping with the approach to a closed-shell structure in this region; the low energy line in Nd^{145}, while not necessarily

[59] Milsted, Rosenblum, and Valadares, Compt. rend. **239**, 259 (1954).
[60] P. P. Day, Phys. Rev. **97**, 689 (1955).
[61] F. K. McGowan, Phys. Rev. **93**, 471 (1954).

representing a transition to the ground state, must nevertheless be considered an exception.

(b) Several interpretations of the situation in the odd isotopes of Gd have been presented in Sec. III-G. However, until further direct evidence becomes available concerning possible first excited states of Gd^{155} and Gd^{157}, we must class these nuclei as exceptions, especially since they are bracketed both as to proton number and neutron number by nuclei showing standard rotational bands (Eu^{153} and Tb^{159}).

(c) In Sec. III O we have pointed to the fact that the second rotational states in Hf^{177} and Hf^{179} at 250 kev and 260 kev, respectively, differ in transition probability by about an order of magnitude. This factor is subject to very little error. At the same time the first rotational transitions (reflecting the nuclear deformation) are quite comparable. Unfortunately, the spins of the ground states of these nuclei are not known with certainty.[52] If they are the same, as might be indicated by the similarity of the level positions, the difference observed for the second states would, in the framework of the unified model, imply very different $M1$ transition probabilities for the cascade transitions in the two nuclei (see beginning of this section). However, the spins and coupling schemes might well be different in the two isotopes (as in Yb^{171} and Yb^{173}).

In addition, we should keep in mind the definite discrepancy which exists between the values of Q_0 obtained from the position of rotational states, and the values derived from the size of the reduced transition probabilities: nuclei seem more deformed when judging from the location of their excited states, i.e., the latter lie at an energy which is lower by a factor of about four than would be expected from the observed transition strengths.

3. *Comparison of Even and Odd Nuclei*

An important question in connection with collective effects concerns possible even-A—odd-A differences in deformation. Spectroscopic quadrupole moments obviously cannot throw any light on that subject. As we pointed out earlier, one approach uses level positions as an index of deformation. In Fig. 13, we have plotted the "splitting constants" $\hbar^2/2\mathcal{J}$ for all rare-earth nuclei of interest, distinguishing even-even, even-odd, and odd-even nuclei, by appropriate symbols. The spins of the odd nuclei we used are either known, or assumed in accordance with the entries given in Table III. For the elements Dy, Er, and Yb we have obtained the splitting constants from what we expect to be the second rotational states, in the odd nuclei, as discussed in Sec. III. Tentative values are represented by open symbols. There seems to be a grouping into the three above-mentioned classes: even-odd nuclei have splitting constants falling consistently lowest; odd-even nuclei are next with somewhat higher $\hbar^2/2\mathcal{J}$; finally, even-even nuclei are lying highest. All three groups show a tendency to coalesce toward the higher neutron numbers. Incidentally, the fact that this plot has the neutron number N as abcissa (and no proton parameter) does not affect these conclusions, since most of the low odd-A points are bracketed in Z on both sides; for instance, low $_{65}Tb^{159}$ occurs at the *same* neutron number ($N=94$) as $_{64}Gd^{158}$ and $_{66}Dy^{160}$ which lie higher, etc.

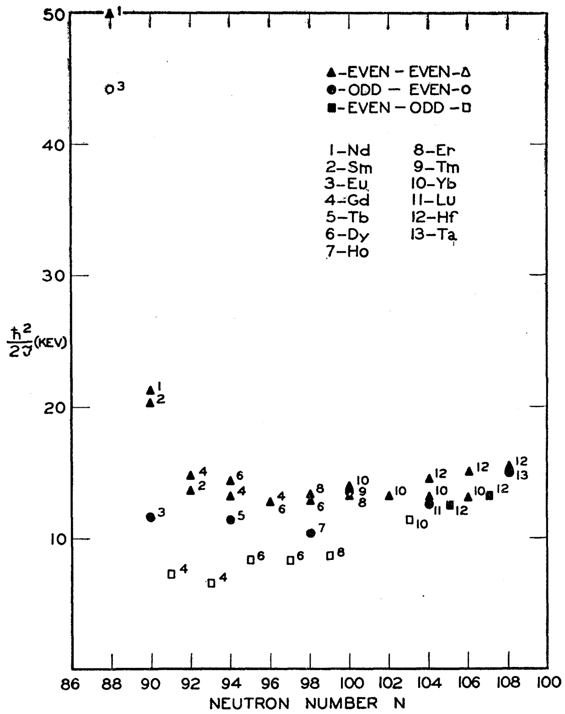

FIG. 13. Summary of results on rotational-level parameters in rare-earth nuclei. "Splitting constants" $\hbar^2/2\mathcal{J}$ *vs* neutron number, as obtained from Eq. (7). Solid symbols represent definitive results, open symbols represent tentative assignments. (△)—even-even nuclei; (⊡)—even-odd nuclei; (⊙)—odd-even nuclei. Nuclei are identified by numbers in the figure.

Let us now examine the other index of deformation, the reduced transition probabilities. As we have seen above, the quantity $B(E2)$ to the first-rotational states in odd-A nuclei is least influenced by cascade or branching effects, and hence reflects most accurately the quadrupole-transition probability, and intrinsic quadrupole moment Q_0. The expression for the reduced *upward* transition probability to the first rotational state of an odd-A nucleus of spin I_0 is given by

$$B(E2)=(15/16\pi)e^2Q_0^2[I_0/(I_0+1)(I_0+2)]; \quad (8)$$

for the values of I_0 which occur in the rare earth nuclei (3/2, 5/2, 7/2) this quantity is about 0.05 $e^2Q_0^2$. For the $0^+\rightarrow2^+$ transition in even-even nuclei, we get a value 0.10 $e^2Q_0^2$ (from a different expression). Hence, according to the unified model, we expect a variation of about a factor of two between even and odd nuclei in $B(E2)$ having identical intrinsic quadrupole moments. An examination of the experimental values $\epsilon B(E2)$ in Table II, reveals the following trends after we make (admittedly crude) estimates of internal conversion effects (which differ in odd and even nuclei because of the presence of an additional $M1$ component in the former): within the rather wide limits imposed by the conversion uncertainties, the data indicate no systematic variations between odd and even Q_0 values, although the intrinsic factor of two in $B(E2)$ mentioned above seems to be present. The case of Hf, which is particularly reliable, illustrates this point. Furthermore, no difference between odd-proton and odd-neutron nuclei is found. Although the uncertainties involved in these considerations are considerable, we believe that nuclear deformations inferred from transition probabilities do not exhibit a behavior of the type illustrated in Fig. 13 for the deformations indicated by level positions.

V. CONCLUSIONS AND SUMMARY

We have studied many nuclei in the so-called "strong coupling" region, where large nuclear deformations should most nearly justify the assumptions made in deriving from the unified model the relations having to do with rotational states and transitions in nuclei. A thorough understanding of the situation in this limited domain is expected to be helpful in disentangling nuclear properties in those regions where one or more of these assumptions are no longer valid, namely where "intermediate coupling" or "weak coupling" regimes prevail. It is therefore gratifying to find that, on the whole, the unified model is successful in accounting for most of the main features which are observed when strongly deformed nuclei are excited by the electric field of relatively slow charged particles. By way of recapitulation, we here collect together those features:

(1) Over-all uniformity in behavior is found as we move from one nucleus to the next, without much regard to specific constitution such as evenness or oddness of proton of neutron number, spin, etc.

(2) We find systematic occurrence of rotational states obeying a simple quasi-molecular interval rule. In keeping with the $E2$ character of Coulomb excitation we observe one excited state only, having spin 2^+ in even-even nuclei, and generally two excited states in odd-A nuclei. While no individual spin assignments were made to the two states in the various cases, we know that their parities must equal the ground state parity, and that the values of the spins must be limited to fall within two units of the ground-state spin I_0. The particular sequence I_0, I_0+1, I_0+2, demanded by the unified model, could be definitely established only by angular distribution measurements. However, the intensities of cascade and crossover radiations in most cases seem to point to different spins for the two excited states. The fact that none of the second-excited states are found to be directly excited in beta decay points toward some degree of forbiddenness for such a transition, and a spin difference of at least two units between these states and the ground state. All this still allows a sequence which is the inverse of the one given above, but we shall favor the normal sequence on the basis of agreement with the theory.

(3) We find systematic occurrence of electric-quadrupole transitions having probabilities of the order of one hundred times those which would be accounted for by the $E2$ transition of a single proton. Significant variations of this quantity are found where variations in the excited-state positions occur as well, in such a direction as to support the view that both reflect a decrease in nuclear deformation upon approaching a closed-shell configuration.

However, a few examples of possibly exceptional behavior in odd-A nuclei have been found and were summarized at the end of Sec. V-B(2).

The main contribution of Coulomb excitation to the understanding of the rotational level structure probably lies in the odd-A nuclei; essentially no lifetime information is available for them, and their level structure is generally inaccessible by other means. Accurate measurements of level energies, branching ratios and internal conversion coefficients (to determine $M1$–$E2$ mixture ratios) can yield a wealth of information concerning details of nuclear structure not otherwise obtainable. It is evident from our results that much remains to be done along these lines.

We have found to our surprise that some of the simple properties encountered in the strong-coupling region can still be found for nuclei of much lower mass number, especially in the even-even nuclei. Some results on Rh, Pd, Ag and Cd have already been published[47,56] while additional information on even lighter but still deformed nuclei will be the subject of a subsequent publication.

VI. ACKNOWLEDGMENTS

We are greatly indebted to the following persons for their cooperation in supplying us with the indispensable materials of this research: to C. P. Keim (Oak Ridge National Laboratory) for all enriched isotopes used; and to F. H. Spedding (Ames Laboratory), and E. L. Weise (National Bureau of Standards) for a number of high-purity rare-earth oxides. A number of people have kindly informed us of results prior to publication concerning decay schemes of various nuclei covered in this research. Finally, it is a pleasure to recall the many fruitful discussions with A. Bohr, B. R. Mottelson, and T. Huus during a most pleasant stay at the Institute for Theoretical Physics in Copenhagen.