# Coulomb Excitation of Medium-Weight Nuclei

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We have studied gamma-ray transitions in nuclei from titanium to cesium which are Coulomb-excited by alpha particles of energy up to 7 Mev. We obtained energies and reduced transition probabilities for most  $2^+$  first excited states of the even-even nuclei, using isotopically enriched targets. A number of these states had not been observed previously. Agreement is obtained with the lifetime values from resonance fluorescence in the two cases where such a comparison is possible at present ( $Ge^{72}$  and  $Ge^{74}$ ). All even-even transitions are found to be systematically faster than the single-proton estimate by a factor ranging from 10 to 50. being always slowest near closed shells. Their interpretation as vibrational excitations of nuclei with spherical equilibrium shapes leads to values for the inertial parameters  $B_2$  and surface tensions  $C_2$  (in the vibrational Hamiltonian) showing strong shell structure effects. A number of thick-target excitation functions between 3 and 7 Mev are in excellent accord with semiclassical E2 theory. No obvious systematic behavior is encountered in the odd-A nuclei, although most transitions contain considerably enhanced E2 components. Coincidence and angular distribution measurements were performed for Se<sup>77</sup> and Ru<sup>101</sup> to establish decay schemes and spins of excited states. The 17.5-second E3 isomer in Se<sup>77</sup> was excited indirectly via a higher state. We encountered no rotational spectra with the possible exception of Se<sup>77</sup>.

## I. INTRODUCTION

HE well-known systematic trend in the positions of first excited states of even-even nuclei<sup>1,2</sup> confined our early Coulomb excitation efforts to those regions of the nuclear chart where these energy levels could be effectively excited with our alpha-particle (He<sup>+</sup>) beam of up to about 3.5 Mev.<sup>3-5</sup> Because of the almost exponential decrease of the Coulomb excitation cross section with an increase of the adiabatic parameter

$$\xi \cong \frac{Z_1 Z_2 e^2}{\hbar v} \left(\frac{\Delta E}{2E}\right),\tag{1}$$

where  $Z_1$  and  $Z_2$  are charges of projectile and target nuclei, respectively,  $\Delta E$  is the level energy, and E and v are the energy and velocity of the incident particle, respectively, we were then limited to states lying below about 250 kev for  $Z_2 > 60$ , and to states below 500 kev for lighter nuclei. Since the lowest known state of an even-even nucleus with  $Z_2 < 60$  lies at 362 kev (44Ru<sup>104</sup>), most even-even nuclei in that region were then inaccessible to us. A number of the lighter odd-mass nuclei, however, having excited states below about 400 kev, were the subject of an earlier study.<sup>5</sup> Our success in obtaining substantial currents of doubly-charged helium ions (at twice the energy) from our electrostatic generator effectively doubled the region of excitation energies  $\Delta E$  which could now be reached [see Eq. (1)]. Some results on Rh and Ag,<sup>6</sup> and Pd and Cd<sup>7</sup> using this beam

were the subject of earlier publications. For intensity reasons, this beam was also used in our work on the rare-earth region, where a brief description of our gas stripper for the production of He<sup>++</sup> ions may be found.<sup>8</sup>

In this article we shall give a complete account of a more or less systematic investigation of both even and odd-mass nuclei extending from titanium (Z=22) to Cs (Z=55), using a beam of He<sup>++</sup> ions with energy between 6 and 7 Mev, and targets containing enriched isotopes<sup>9</sup> whenever possible. We have had the use of 52 of these isotopes, without which most of this work would not have been possible. Our measurements for most of the even-even nuclei have been presented in the form of a plot<sup>10</sup> (see Sec. IV-A and Fig. 9). These measurements consist mainly in the determination of gamma-ray energies, and the evaluation of absolute gamma-ray yields from thick targets. We have made use of some angular distributions to restrict the spin assignments for excited states. In two cases of odd-A nuclei (Se77, Ru<sup>101</sup>) we made coincidence measurements to establish decay schemes.

Because of the negligible contribution of the internal conversion process to the de-excitation of most of the states observed, as well as of the above-mentioned excellent agreement with the semiclassical Coulombexcitation formalism,<sup>11</sup> we can obtain rather reliable values for the reduced electric-quadrupole (E2) transition probabilities B(E2). Measurements of these transition probabilities are equivalent to lifetime deter-

<sup>&</sup>lt;sup>1</sup>G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).

<sup>&</sup>lt;sup>2</sup> P. Stähelin and P. Preiswerk, Nuovo cimento 10, 1219 (1953). <sup>3</sup>G. M. Temmer and N. P. Heydenburg, Phys. Rev. 93, 351 (1954)

<sup>&</sup>lt;sup>4</sup> N. P. Heydenburg and G. M. Temmer, Phys. Rev. 93, 906

<sup>&</sup>lt;sup>4</sup> N. F. Heydenburg, 2014
<sup>6</sup> G. M. Temmer and N. P. Heydenburg, Phys. Rev. 96, 426 (1954). Values of transition probabilities B(E2) in Table II of this reference have been revised (see Table II of this article).
<sup>6</sup> N. P. Heydenburg and G. M. Temmer, Phys. Rev. 95, 861 (1954).

<sup>7</sup> G. M. Temmer and N. P. Heydenburg, Phys. Rev. 98, 1308 (1955).

<sup>&</sup>lt;sup>8</sup> N. P. Heydenburg and G. M. Temmer, Phys. Rev. 100, 150 (1955). Table I of this reference contains a numerical error; the value of  $y(\xi_0)$  for  $\xi_0=0.1$  should read 0.332. <sup>9</sup> Obtained from the Stable Isotopes Division, Oak Ridge

National Laboratory. <sup>10</sup> G. M. Temmer and N. P. Heydenburg, Phys. Rev. 99, 1609

<sup>(1955).</sup> Revised versions of Figs. 1 and 2 of this reference are given in N. P. Heydenburg and G. M. Temmer, Annual Review of Nuclear Science [Annual Reviews, Inc., Stanford (to be published)], Vol. 6.

<sup>&</sup>lt;sup>11</sup> K. Alder and A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 29, Nos. 18 and 19 (1955).

minations lying in the region of 10<sup>-11</sup> second or less, where few (if any) reliable electronic measurements have been carried out to date.12 Information on the transition probabilities in this mass-number region must therefore come either from Coulomb excitation, or from resonance fluorescence of gamma rays, a method whose scope has also been greatly expanded recently,<sup>13,14</sup> yet is inherently limited by source strength requirements. In fact, there are only two cases so far in which the two methods are susceptible of comparison (Ge<sup>72</sup> and Ge<sup>74</sup>) and the results are in good agreement. This satisfactory check on the absolute magnitude of the Coulomb excitation cross section, coupled with our extensive excitation curves for thin targets in lighter elements up to 3.8 Mev,<sup>5</sup> as well as some thick target curves up to 7-Mev alpha-particle energy (see Figs. 7 and 9) lead us to trust the essential reliability of the Coulomb excitation process for the determination of electromagnetic properties of low-lying nuclear states. An even more stringent test of the simple theory is provided by the excellent agreement of theory and experiment on the angular distribution of gamma radiation following Coulomb excitation.<sup>15</sup>

In some of the lightest nuclei, the possibility of course always exists that some of the gamma radiation originated in a nuclear inelastic scattering encounter, i.e., in compound-nucleus formation. Evidence of this type of contribution was found earlier<sup>5,16</sup> and is quite in evidence when using protons as projectiles.<sup>17</sup> From the shape of the excitation curve it is usually possible to infer the presence of gamma-ray yield over and above that demanded by Coulomb excitation proper, although confusion might conceivably arise on occasion. In the very light elements, the lifetimes deduced from Coulomb excitation could then represent lower limits rather than actual values.

## **II. EXPERIMENTAL METHOD**

We have previously described most of the experimental features of this work.<sup>16</sup> Our doubly-charged He beam is used exclusively, as is a 2 in. $\times 1\frac{3}{4}$  in. NaI(Tl) crystal mounted on a DuMont 6292 photomultiplier tube, located so as to intercept a solid angle of almost  $2\pi$  behind the target. As was already stated, a number of isotopically enriched targets served to assign energy levels and to obtain more reliable yield data. We were able to operate with as little as 8 milligrams of total target substance. The absolute efficiency of our system is accurately known from the calibration with 411-kev gamma radiation from Au<sup>198,18</sup> Appropriate corrections for photopeak efficiencies relative to 411

kev are made in accordance with semiempirical procedures<sup>19</sup> as well as some computed efficiencies.<sup>20</sup> We believe our gamma-ray energies to be generally accurate to  $\pm 1\%$ . The transition probabilities for the stronger transitions are estimated to be known to  $\pm 15\%$ ; the relative probabilities of sequences of isotopes of the same element are undoubtedly known to greater accuracy, whereas weak transitions (usually of highlying states) are not known quite as well.

No correction was necessary for bremsstrahlung from the targets, whose contribution is negligible compared to that found under proton bombardment.<sup>21</sup> Background conditions are particularly favorable with our generator, which has been used exclusively with an alpha-particle beam for about five years, and no subtraction was necessary other than of the contributions from the  $O^{18}(\alpha, n\gamma) Ne^{21}$  reaction<sup>5,10</sup> in the few cases where oxide targets had to be used, and of the Compton distributions due to any higher energy gamma rays from the target. For elements with atomic number above 40, we inserted a 10-mil copper absorber which almost eliminated the K x-rays resulting from K-shell ionization while it has a negligible effect on most gamma rays observed.

For a detailed discussion of the process by which we obtain the transition probabilities from thicktarget yields, we refer to our article on rare-earth nuclei.8

For our coincidence studies we used an additional and essentially identical single-channel analyzing system with a 1 in  $\times$  1 in. crystal, whose output was put in coincidence with the output from our regular channel in a circuit whose resolving time was  $\sim 0.5$ microsecond. The two crystals were located about one inch from the target and were shielded from each other by a one-eighth inch lead plate to eliminate Comptonrecoil coincidences. The counting rate was kept low enough to limit the accidental coincidence rate to less than 20% of the total counting rate. Since no quantitative information was sought in these coincidence experiments, the absolute efficiencies of the counters in this geometry were of no interest.

Our angular distribution measurements consisted of readings at 0° and 90° to the beam axis; any spurious anisotropies were eliminated by placing a gamma-ray source at the target position.

## III. RESULTS

### A. Even-Even Nuclei

Because of their more systematic behavior as to position, spin and parity (2+), and transition probability to their first excited states, we shall discuss the even-even nuclei separately. Only one gamma ray was observed from each nucleus, without exception. In Table I we have collected our experimental results for

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<sup>&</sup>lt;sup>12</sup> See, e.g., C. F. Coleman, Phil. Mag. 46, 1135 (1955)

 <sup>&</sup>lt;sup>13</sup> See, e.g., F. R. Metzger, Phys. Rev. 101, 286 (1956).
 <sup>14</sup> F. R. Metzger, J. Franklin Inst. 261, 219 (1956).

F. K. McGowan and P. H. Stelson, Phys. Rev. 99, 127 (1955).
 N. P. Heydenburg and G. M. Temmer, Phys. Rev. 94, 1254 (1954)

 <sup>&</sup>lt;sup>17</sup> Mark, McClelland, and Goodman, Phys. Rev. 98, 1245 (1955).
 <sup>18</sup> Calibrated sources through the courtesy of L. Cavallo and H. H. Seliger of the National Bureau of Standards.

<sup>&</sup>lt;sup>19</sup> Maeder, Müller, and Wintersteiger, Helv. Phys. Acta 27, 3 (1954).
 <sup>20</sup> M. J. Berger and J. Doggett, Rev. Sci. Instr. 27, 269 (1956).
 <sup>21</sup> P. H. Stelson and F. K. McGowan, Phys. Rev. 99, 112 (1955).

where

even-even nuclei from titanium through tellurium. Results for palladium and cadmium were previously reported,<sup>7</sup> but have been revised, as have most values of the transition probabilities used in Fig. 2 of reference 10. A more complete and revised plot is given in Fig. 9. We list, in turn: the element; mass number A; gammaray energy in kev (always equal to the excitation energy  $\Delta E$  in even-even nuclei); the reduced (upward) transition probability (divided by  $e^2$ ) B(E2) in  $10^{-48}$ cm<sup>4</sup>; the equivalent mean radiative lifetime for the downward quadrupole transition,  $\tau_{\gamma}(E2)$ , in seconds; the latter was obtained from the expression for E2

TABLE I. Summary of Coulomb excitation results for even-even nuclei of intermediate mass. We list: element, mass number A, transition energy  $E_2^*$  in kev, the reduced E2 transition probability transition energy  $D_2^{-1}$  meet, the reduced  $D_2$  transition probability B(E2) in barns<sup>2</sup>, the mean lifetime for decay  $\tau_{\gamma}(E2)$  in  $10^{-12}$  sec, the favored factor F, and the vibrational parameters  $B_2/B_2^{**}$  (dimensionless) and  $C_2$ (Mev). For the definition of the last three quantities, see Eqs. (10), (11), and (12), and text. Enriched isotopes used in all cases except Zn<sup>66</sup>, Ru<sup>100</sup>, Ru<sup>102</sup>.

Element	A	<i>E</i> 2+ (kev)	B(E2) (10 <sup>-48</sup> cm <sup>4</sup> )	$ au_{\gamma}(E2)$ (10 <sup>-12</sup> sec)	F	$B_2/B_2^*$	C2 (Mev)
<sub>22</sub> Ti	46 48	890 990	0.056	13 14	11	19 32	37 77
<sub>26</sub> Fe	56	854	0.10	8.7	15	15	36
₃₀Zn	64	1000	0.11	3.7	14	15	61
	66	1040	0.087	4.0	11	18	86
32Ge	70	1020	0.098	3.9	10	18	94
	72	830ª	0.16	6.2	18	13	46
	74	593	0.25	23	27	12	22
	76	566	0.23	29	24	13	24
34Se	74	635	0.21	19	23	15	32
	76	567	0.43	17	44	8.1	14
	78	615	0.36	13	36	8.7	20
	80	654	0.23	15	22	13	34
	82	880	0.056	13	5.2	39	190
42Mo	94	871	0.29	2.8	23	11	67
	96	778	0.31	4.5	23	11	57
	98	786	0.27	5.0	20	12	70
	100	528	0.66	15	47	8.0	21
₄₄Ru	100	540	0.30	30	22	18	48
	102	473	0.63	26	44	10	20
	104	362	1.04	69	71	8.7	9.8
$_{46}Pd^b$	104	550	0.46	17	31	13	36
	106	510	0.59	20	39	11	27
	108	424	0.78	40	51	9.6	17
	110	370	1.04	57	66	8.3	11
₄8Cd <sup>b</sup>	110	654	0.41	8.3	26	13	58
	112	620	0.46	9.7	28	12	49
	114	550	0.55	15	33	11	37
	116	508	0.62	20	36	11	32
52Te	120	560	0.55	14	31	13	48
	122	570	0.47	14	26	14	58
	124	608	0.39	13	21	16	77
	126	662	0.32	10	17	18	100
	128	750	0.28	6.2	15	18	140
	130	850	0.26	3.5	13	18	170

• This represents the second excited state; first excited state  $0^+$ . b Values for B(E2) for this nucleus have been revised and supersede those in reference 7.

transitions<sup>22</sup> (neglecting internal conversion):

$$\frac{1}{\tau_{\gamma}(E2)} = \frac{4\pi}{75} \left(\frac{\Delta E}{\hbar c}\right)^5 \frac{1}{\hbar} B(E2) \downarrow, \qquad (2)$$

$$B(E2)\downarrow = \frac{1}{5}B(E2)\uparrow \tag{3}$$

for the case of the  $0^+ - 2^+$  transitions encountered in all even-even nuclei observed. In the next column we list the so-called favored factor F, which provides a convenient measure of the departure from single-particle strength of these transitions, and is simply the ratio of observed B(E2) to the single-particle estimate of that quantity given by23,24

$$B(E2)_{\rm s.p.\uparrow} = \frac{45}{100\pi} R_0^4 S \cong 3 \times 10^{-5} A^{4/3} S \text{ barn}^2, \quad (4)$$

where the value of  $R_0$  used is given by

$$R_0 = 1.20A^{\frac{1}{3}} \times 10^{-13} \,\mathrm{cm},$$
 (5)

and S is a statistical factor depending on the initial and final spins<sup>24</sup>; if we assume two particles to be responsible for the transition in this estimate, S differs from unity by less than 20% regardless of particle configuration, and we take S to be equal to unity. Finally, the last two columns show two parameters,  $B_2$  and  $C_2$ , derived from the experimental results using the unified model as applied to vibrational quadrupole excitations.<sup>22,25</sup> B<sub>2</sub> is an inertial parameter measuring the mass transport in the quadrupole vibration, and is given in units of the parameter which would obtain for irrotational motion,  $B_2^*$ ; it is therefore a dimensionless quantity.  $C_2$  is a measure of the nuclear surface tension, expressed in Mev, and represents the effective restoring force in the vibrational motion. We postpone until later a discussion of the significance of the quantities,  $B_2$  and  $C_2$ , and merely list below convenient numerical relations connecting theory and experiment:

$$\frac{B_2}{B_0^*} = \frac{7.14 \times 10^{-3} Z^2}{E_0 + B(E_2) A^{1/3}} = \frac{242 C_2}{E_0^2 + A^{5/3}},\tag{6}$$

$$C_2 = 2.95 \times 10^{-5} Z^2 A^{4/3} \frac{E_{2+}}{B(E2)}.$$
 (7)

 $E_{2+}$  is the energy of the first-excited state, in Mev, and B(E2) is expressed in units of  $10^{-48}$  cm<sup>4</sup> (barn<sup>2</sup>);  $C_2$  is then given in Mev, while  $B_2/B_2^*$  is dimensionless.

### B. Odd-A Nuclei

Some results on odd-A nuclei were published previously<sup>5-7</sup>; we have obtained some detailed information

<sup>22</sup> A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 27, No. 16 (1953).
<sup>23</sup> V. F. Weisskopf, Phys. Rev. 83, 1073 (1951).
<sup>24</sup> S. A. Moszkowski, Phys. Rev. 89, 474 (1953); Sharp, Kennedy, Sears, and Hoyle, Chalk River Report CRT-556, 1954 (unpublished). (unpublished).

Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. (to be published).

on a number of additional nuclei, establishing decay schemes and determining the partial E2 lifetimes of their excited states. The decay of excited states differing in spin by one unit from the ground state spin will generally take place by a mixture of magnetic dipole (M1) and E2 radiation, of which only the E2 part is excited. We detected coincidences in some cases where two or more excited states were reached. Because of the more complicated nature of the ground-state configurations in odd-A nuclei, as well as of the lack of information concerning the spins of many excited states, they are not susceptible of systematic discussion such as is possible for even-even nuclei. Each nucleus is essentially a separate problem, and even though our method of examining these nuclei is extremely uniform and views all nuclei in exactly the same manner, nuclei differing from each other by no more than by the addition of two neutrons are sometimes found to exhibit completely different spectra. The transition probabilities seem to vary considerably, usually being several times larger than for single-particle transitions, although some are found to be comparable to this natural unit. No correlation of these properties with the odd or even character of the proton number is apparent. We summarize in Table II, in a manner analogous to Table I for the even-even nuclei, our experimental results for odd-A nuclei. No values for  $B_2$  or  $C_2$  are presented, since no clearly vibrational states have been identified. A column has been inserted giving the ground-state spins and parities (the latter as assigned from the shell model), as well as the spins and parities of the excited states if known; probable but unconfirmed values are given in parentheses. The values for the partial E2 lifetimes for decay are dependent upon the values of the spins in the initial and final states, because Eq. (3) for odd nuclei becomes

$$B(E2)\downarrow = \frac{2I_0 + 1}{2I^* + 1}B(E2)\uparrow,\tag{8}$$

where  $I_0$  is the ground state spin, and  $I^*$  is the spin of the excited state in question. The range of values for  $\tau_{\gamma}(E2)$  indicated in some cases then correspond to the range of spins  $I^*$  a priori allowed by E2 selection rules. As indicated in Sec. II, a few measurements of gamma-ray anisotropies served to restrict the excitedstate spins, using the general arguments based on the extensive systematic investigations of angular distribution by Stelson and McGowan.<sup>15</sup>

Where more than one state is excited, the values of B(E2) shown have been corrected for cascade decay in the lower states (reduction), and for branching decay in the upper state (increase). Cascade radiation is not listed; for complete branching information in these cases, we refer to the detailed discussion of individual elements below. In no case did we correct for internal conversion; in a few of the very low-lying states this correction could be appreciable. Knowing the total internal conversion coefficient  $\alpha_t$ , our values of B(E2) should be multiplied by  $(1+\alpha_t)$ , while our values of  $\tau_{\gamma}(E2)$  should be divided by the same factor.

The "favored factor" F was computed using the single-particle estimate in Eq. (4). We used the appropriate statistical factors S for the spins listed in Table II, assuming one-particle configurations. S may differ appreciably from unity in some cases.

## C. Odd-Odd Nucleus

We have excited a state in the only available, stable odd-odd nucleus V50.9 The results are given at the end of Table II.

We shall now discuss individual nuclei in some detail.

TABLE II. Summary of Coulomb excitation results for odd-A (and odd-odd) nuclei or intermediate mass. We list: element, mass number A, transition energy  $\Delta E$  in kev, ground state spin and parity  $I_0$ , excited state spin and parity  $I^*$  (tentative values in parentheses), the reduced E2 transition probability  $\epsilon B(E2)$  in barns<sup>2</sup> (uncorrected for internal conversion), the *partial* mean lifetime for decay  $\tau_{\gamma}(E2)$  in sec (power of ten multipliers in parentheses), and the favored factor F. The latter is computed using the appropriate single-particle statistical factors S(e.g., Na<sup>23</sup>). Enriched isotopes used in all cases except Mo<sup>95</sup>.

	Ele- ment	A	$\Delta E$ (kev)	I <sub>0</sub>	<i>I</i> *	$\epsilon B(E2)$ (10 <sup>-48</sup> cm <sup>4</sup>	) $\tau_{\gamma}(E2)$ (sec)	F
Ì					A. Odd-	A		
	11Na 22Ti 23V 25Mn 26Fe	23ª 47ª 51ª 55ª 57ª	446 160 320 128 137	3/2+ 5/2- 7/2- 5/2- 1/2-	5/2+ (7/2)- (5/2)- 7/2- 5/2-	0.016 0.040 0.0055 0.075 0.057	4.3 (-10) 2.6 (-8) 3.3 (-9) 4.2 (-8) 8.9 (-8)	45 210 34 320 15
	29Cu	63 65	690 990 815 1150	3/2- 3/2- 3/2- 3/2- 3/2-	?- ?- (5/2)-	0.010 0.029 0.0087 0.027	$\begin{array}{c} 0.26 - 1.0 ( -10) \\ 1.5 \ -4.5 ( -12) \\ 1.3 \ -5.2 ( -11) \\ 2.3 ( -12) \end{array}$	6.5 46 5.5 40
	30Zn	67ª	182	5/2-	5/2-	0.032	1.3(-8)	18
	32Ge	73	68 815	9/2+ 9/2+	(11/2)+ ?+	0.084 0.062	6.6(-7)	450 <sup>d</sup>
	33As	75	200 283 574 814	3/2- 3/2- 3/2- 3/2- 3/2-	5- 5- 5-	0.025 0.071 0.072 0.066	•••• ••• •••	31 90 90 80
	84Se	77	244 457	1/2- 1/2-	(3/2)- (5/2)-	0.14 <sup>b</sup> 0.25°	1.4(-9) 4.9(-11)	36 43
	$_{42}Mo$	95	204	5/2+	(3/2)+	0.070	2.2(-9)	95
	₄₄Ru	99 101	89.5 127 307 522	5/2+ 5/2+ 5/2+ 5/2+ 5/2+	1/2+, 3/2+ (9/2)+ (7/2)+ ?+	0.054 0.061 <sup>b</sup> 0.036° 0.041	0.9-1.8(-7) 6.8(-8) 1.1(-9) 2-9(-11)	20, 70 9.0 70 75
	45Rh	103ª	295 357	1/2- 1/2-	$\frac{3/2^{-}}{5/2^{-}}$	0.25 0.37	2.9(-10) 1.1(-10)	44 43
	₄₅Pd	105ª	266 433	5/2+ 5/2+	?+ ?+	0.040 0.17	•••	70 25
	47Ag	107ª 109ª	318 413 305 400	1/2- 1/2- 1/2- 1/2-	3/2- 5/2- 3/2- 5/2-	0.15 0.21 0.17 0.24	$\begin{array}{c} 3.3 (-10) \\ 9.7 (-11) \\ 3.6 (-10) \\ 1.0 (-10) \end{array}$	25 23 28 26
	₄₃Cd	111ª 113ª	340 290	1/2+ 1/2+	$\frac{3/2^+}{3/2^+}$	0.19 0.46	1.9(-10) 7.0(-10)	30 70
					B. Odd-O	dd		
	23V	50	225	6+	(7)+	0.011	1.3(-8)	27•

Values for B(E2) for this nucleus have been revised and supersede those Values for B(D2) for this increasing average for revised and superseder to the in references 6 and 7.
 Value corrected (decreased) for cascade decay from upper state.
 Value corrected (increased) for cascade decay through lower state.
 Value very sensitive to I\* (statistical factor S).
 Statistical factor S in two-particle estimate equals 100/273.

(1) Titanium (Z=22).—We have previously reported results on the excitation of the odd-A nucleus Ti<sup>47</sup> (160-kev level) with 3-Mev alpha particles.<sup>5</sup> We examined enriched metallic targets of all Ti isotopes. Ti<sup>46</sup> shows the known first excited state at 890 kev<sup>26</sup>; Ti<sup>48</sup> reveals two gamma-ray transitions, one of which (990 kev) is associated with the known first excited state at 990 kev.<sup>26</sup> The line at 750 kev shows an excitation function which is steeper than that of the 990-kev transition, indicating that it must arise from a nuclear reaction. We believe that it originates in the reaction  $Ti^{48}(\alpha, n\gamma)Cr^{51}$ ; this belief is supported by the fact that we observed a gamma-ray transition of precisely the same energy when bombarding a target of V<sup>51</sup> with 3-Mev protons. The neutron groups from the reaction  $V^{51}(p,n)Cr^{51}$  have been observed,<sup>27</sup> and we undoubtedly see the 750-kev transition from the  $V^{51}(p,n\gamma)Cr^{51}$ reaction.

After bombarding the Ti<sup>46</sup> target for a few hours with 6-Mev alpha particles, we found a positron activity with a half-life of 42 minutes, which was evidently  $Cr^{49}$ resulting from the Ti<sup>46</sup>( $\alpha, n$ )Cr<sup>49</sup> reaction.

An enriched target of Ti<sup>47</sup> showed gamma transitions at 730 kev and 1.02 Mev; there was considerable neutron background present, and it was not possible to establish Coulomb excitation behavior in these lines. It is *a priori* possible that they arise in one or more of the following processes: Ti<sup>47</sup>( $\alpha,n\gamma$ )Cr<sup>50</sup>; Ti<sup>47</sup>( $\alpha,p\gamma$ )V<sup>50</sup>; Ti<sup>47</sup>( $\alpha,\alpha'\gamma$ )Ti<sup>47</sup>. All of these product nuclei are stable species, and cannot be identified by radioactivity. The first excited state of even-even Cr<sup>50</sup> is not known, but is expected to lie higher than 730 kev from the systematics; the 1.02-Mev gamma ray may come from that state. No excited states are known in V<sup>50</sup> other than the 225kev state reported below, and none are known in Ti<sup>47</sup> between 160 kev and 1.40 Mev.<sup>26</sup>

We observed a transition at 1.44 Mev in an enriched  $Ti^{49}$  target, which probably corresponds to the first excited state of this nucleus.<sup>26</sup>

No excited state was found in enriched Ti $^{50}$ , which has no known state below 1.5 Mev. $^{26}$ 

(2) Vanadium (Z=23).—We have previously studied the odd-A nucleus V<sup>51</sup> (320 kev). Figure 1 shows the gamma spectrum observed from the odd-odd nucleus V<sup>50</sup> under 3-Mev alpha bombardment, along with the spectrum for ordinary vanadium.<sup>28</sup> The enrichment of the target in V<sup>50</sup> was 22%<sup>9</sup> (natural abundance 0.24%). The 320-kev transition from V<sup>51</sup> is of course also in evidence, as is the 342-kev line from O<sup>18</sup>( $\alpha$ , $n\gamma$ )Ne<sup>21</sup> which is always found in oxide targets.<sup>5</sup> (The target material was V<sub>2</sub>O<sub>5</sub>.) A thick-target excitation function



FIG. 1. Pulse-height spectra obtained for targets of  $V_2O_5$  with 3-Mev alpha particles. Open circles are for ordinary vanadium showing the 320-kev state of  $V^{51}$ ; full circles are for  $V^{50}$  enriched to 22% showing additional line at 225 kev. The intensity of the 342-kev line associated with  $O^{18}$  is indicated.

showed that the 225-kev radiation was emitted from a level whose excitation energy  $\Delta E$  was considerably less than 320 kev, so that it is safe to conclude that there is an excited state of V<sup>50</sup> at 225 kev.

The high ground-state spin of  $V^{50}$   $(I_0=6^+)$  allows spin values for the excited state at 225 kev between 4<sup>+</sup> and 8<sup>+</sup>. It is not unlikely that the spin of the state be 7<sup>+</sup> in view of the fact that the configuration of  ${}_{23}V_{27}{}^{50}$  might be  $(f_{7/2})^3(f_{7/2})^{-1}$ , according to the shell model. The transition strength seems to be several times that expected for a two-particle transition; the statistical factor S was computed for a  $[(f_{7/2})^3]_{7/2}$  $\rightarrow [(f_{7/2})^3]_{7/2} E2$  transition, the other particle remaining in the same state. Since this nucleus has stable isobars (Ti<sup>50</sup>, Cr<sup>50</sup>) to either side, only Coulomb excitation or scattering experiments can give information on its excited states.

Upon bombarding V<sup>51</sup> with 6-Mev alpha particles, a strong 154-kev transition appeared, whose excitation function was considerably steeper than that of the Coulomb-excited 320-kev transition. We ascribe this line to the V<sup>51</sup>( $\alpha,n\gamma$ )Mn<sup>54</sup> reaction, the 154-kev transition coming from the first-excited state of Mn<sup>54</sup> (oddodd). The only other possible reaction, V<sup>51</sup>( $\alpha,p\gamma$ )Cr<sup>54</sup> leads to an even-even nucleus, whose first-excited state is known to lie at 0.84 Mev.<sup>26</sup>

(3) Chromium (Z=24).—Six-Mev alpha-particle bombardment of ordinary metallic chromium produced gamma rays at 150, 511, 420, 940, and 1360 kev, of which only the last three remained (slightly enhanced) in an enriched target of  $Cr^{52}$ . We believe that the latter

<sup>&</sup>lt;sup>26</sup> Way, King, McGinnis, and van Lieshout, *Nuclear Level Schemes* (A=40-A=92) (U. S. Atomic Energy Commission, Washington, D. C., 1955). We should like to record our appreciation to the Nuclear Data group for producing this invaluable compilation.

<sup>&</sup>lt;sup>27</sup> Stelson, Preston, and Goodman, Phys. Rev. 80, 287 (1950). <sup>28</sup> Similar results were reported simultaneously by Fagg, Geer, and Wolicki, reference 36.



FIG. 2. Pulse-height spectrum for Fe<sup>56</sup>. 854-kev line represents the gamma ray from the well-known first-excited state of this nucleus. Obtained with 6-Mev alpha particles. Steep rise at low energy is due to Fe<sup>57</sup>(123 kev).

originate in the  $Cr^{52}(\alpha,n\gamma)Fe^{55}$  reaction, where they correspond to the first three known states in Fe<sup>55</sup>. The latter nucleus has a *K*-capture decay of 2.9-year halflife, and was therefore not observed. We do not think it likely that the two gamma rays of highest energy come from the  $Cr^{52}(\alpha,p\gamma)Mn^{55}$  reaction, where levels of similar energy exist, because (a)  $(\alpha,n)$  reactions seem to be greatly favored over  $(\alpha,p)$  reactions at our energies in this region of *Z* (see also Ti, V, Mn, and Co); (b) we should certainly have observed the first excited state of  $Mn^{55}$  at 128 kev in preference to the higher states. The first excited state of  $Cr^{50}$  is not known, but was probably not observed because of the low abundance of this nucleus (4.4%) in ordinary Cr.

The 511-kev annihilation line belonged to Fe<sup>53</sup>, a 9minute positron emitter produced by the  $Cr^{50}(\alpha,n\gamma)Fe^{53}$ reaction. This leaves the 150-kev transition, which may only come from an excited state of Fe<sup>53</sup> (none are known), if we omit  $(\alpha,p)$  reactions from our consideration.

(4) Manganese (Z=25).—We have previously reported extensive studies on the 128-kev level in Mn<sup>55,5</sup>. We have found no definite evidence for Coulomb excitation of higher states, mainly because of the considerable amount of compound-nucleus formation at the higher bombarding energies necessary to reach higher states. We have observed a prominent 150-kev gamma ray with steep excitation which we ascribe to an excited state of Co<sup>58</sup>, formed by the Mn<sup>55</sup>( $\alpha, n\gamma$ )Co<sup>58</sup> reaction; the only other possible reaction, Mn<sup>55</sup>( $\alpha, p\gamma$ )Fe<sup>58</sup> leads to an even-even nucleus with a first excited state at 805 kev.<sup>26</sup> (5) Iron (Z=26).—We have previously studied the

radiations from Fe<sup>57</sup> in an enriched target.<sup>5</sup> Figure 2 shows the well-known first excited state of Fe<sup>56</sup> at 850 kev, observed from a natural Fe target (abundance of Fe<sup>56</sup>: 91.7%). Our value for the mean lifetime of the transition ( $8.7 \times 10^{-12}$  sec) is in agreement with the lower limit obtained by Ilakovac from the resonance fluorescence of this transition<sup>29</sup> ( $\tau_{\gamma} > 6 \times 10^{-12}$  sec). We obtained a thick-target excitation curve between 4.0 and 6.5 Mev in excellent agreement with *E2* theory. Radiation from the 2<sup>+</sup> states in Fe<sup>54</sup> and Fe<sup>58</sup> was not observed, but could undoubtedly be excited with enriched targets.

(6) Cobalt (Z=27).—Upon bombarding monoisotopic Co<sup>59</sup> with 6.5-Mev alpha particles, we observed gamma rays at 350, 511, and 1150 kev; the 350-kev line may either be due to a slight amount of oxygen in the metallic target, or may represent a transition in Cu<sup>62</sup>, excited by the  $\operatorname{Co}^{59}(\alpha,n\gamma)\operatorname{Cu}^{62}$  reaction; the latter nucleus was detected by its known positron decay with 9.7-minute half-life<sup>26</sup> (accounting for the 511-kev line). The 1150-key transition could represent either the (Coulomb) excitation of the known first level of Co<sup>59 26</sup> or the de-excitation of the first excited state (?) of Ni<sup>62</sup>, which is produced in the  $Co^{59}(\alpha, p\gamma)Ni^{62}$  reaction. In view of the prevalence of the  $(\alpha, n)$  reaction over the  $(\alpha, p)$  reaction in this region of the periodic table at our bombarding energies, we believe the former possibility to be the more likely one. A continuous background,



FIG. 3. Composite pulse-height distributions for the even-even isotopes of germanium. Enrichment of individual targets and bombarding energies used are given in the insert.

<sup>29</sup> K. Ilakovac, Proc. Phys. Soc. (London) A67, 607 (1954).

presumably due to the neutron reaction, prevented us from determining a reliable cross-section value.

(7) Nickel (Z=28).—A weak indication of 1.4-Mev radiation was detected from an enriched target of metallic Ni<sup>58</sup>, which probably comes from the first-excited state.<sup>26</sup> We examined no other isotopes of this element.

(8) Copper (Z=29).—Enriched targets of Cu<sup>63</sup> and Cu<sup>65</sup> revealed quite similar spectra consisting of two transitions each: 690 kev and 990 kev in Cu<sup>63</sup>, and 815 kev and 1.15 Mev in Cu<sup>65</sup>. Inelastic proton groups leading to these states have also been found at the Rice Institute.<sup>30</sup> Only the higher energy transitions were known from previous beta decay studies. Re-examination of the decay of Zn<sup>63</sup> revealed the line at 690 kev,<sup>31</sup> and further search for the lower-energy transition in Cu<sup>65</sup> is in progress.<sup>32</sup> Our partial decay lifetime  $\tau_{\gamma}(E2)$ for the 990-kev state in Cu<sup>63</sup> lies between 1.5 and  $5.9 \times 10^{-12}$  sec (for allowed spins 1/2 to 7/2); resonance fluorescence yields  $\sim 4 \times 10^{-13}$  sec.<sup>29</sup> This shows that the decay must take place predominantly by M1 radiation, the ratio of E2 to M1 radiation lying between  $\sim 0.10$ and 0.36. The mixture eliminates the possibility of spin 7/2 for this state, and reduces the upper limit of  $\tau_{\gamma}(E2)$  to  $4.5 \times 10^{-12}$  sec. After bombardment of Cu<sup>65</sup> with 6.5-Mev alpha particles we detected the 68-minute positron activity of Ga<sup>68</sup>, resulting from the Cu( $\alpha$ ,n)Ga<sup>68</sup> reaction.

(9) Zinc (Z=30).—We have already reported our findings in the odd-A isotope  $Zn^{67.5}$  We have observed transitions in the even-even nuclei  $Zn^{64}$  (1.00 Mev, enriched target) and  $Zn^{66}$  (1.04 Mev).

(10) Gallium (Z=31).—No further study.<sup>3</sup>

(11) Germanium (Z=32).—We have previously reported work on the surprising low-energy transition at 68 kev in Ge<sup>73</sup>.<sup>5</sup> We have been able to observe the 0<sup>+</sup>-2<sup>+</sup> transitions in the even-even nuclei Ge<sup>70</sup> (1020 kev), Ge<sup>72</sup> (830 kev), Ge<sup>74</sup> (593 kev), and Ge<sup>76</sup> (566 kev) using enriched metallic targets. The first and last of these represent previously unknown first excited states. The pulse-height distributions are shown in Fig. 3. It should be recalled that the first excited state of Ge<sup>72</sup> has character 0<sup>+ 26</sup> and was therefore not observed in Coulomb excitation. In Table III we compare our results for the mean lives of the transitions in Ge<sup>72</sup> and Ge<sup>74</sup> with those obtained by Metzger from resonance fluorescence

TABLE III. Comparison of lifetimes obtained by Coulomb excitation and resonance fluorescence for Ge<sup>72</sup> and Ge<sup>74</sup>. Metzger's fluorescence data from reference 13 and private communication.  $\tau_{\gamma}(E2)$  is the mean life.

		$\tau_{\gamma}(E2)$ (1	0 <sup>-12</sup> sec)
Nucleus	$E_{2}^{+}$ (kev)	Coulomb exc.	Res. fluor.
<sup>32</sup> Ge <sup>72</sup> 32Ge <sup>74</sup>	830 593	$6.2 \pm 0.5$ $2.3 \pm 0.2$	$4.6 \pm 1.2$ $1.9 \pm 0.3$

<sup>30</sup> Reference 26 and J. P. Schiffer (private communication).

<sup>31</sup> R. van Lieshout (private communication).



FIG. 4. Comparison of level schemes of  $As^{75}$  as obtained from radioactivity measurements and Coulomb excitation by 6-Mev alpha particles. To the left is shown the scheme proposed by Schardt and Welker (reference 33); to the right is our level scheme. Resolution does not permit identification with given level or levels in the triplet at about 280 kev.

experiments. The agreement between these two entirely different approaches is satisfactory. A thick-target excitation curve was obtained for  $Ge^{76}$ , between 4.0 and 6.5 Mev, in good agreement with E2 theory.

(12) Arsenic (Z=33).—Arsenic consists of only one isotope, As<sup>75</sup>. We observed four gamma-ray transitions, which from their excitation functions seem to correspond to states of the corresponding energies. Figure 4 shows a comparison of the level scheme observed by us with that found by Schardt and Welker<sup>33</sup> from decay of Ge<sup>75</sup> and Se<sup>75</sup>. Because of the limited resolution available with NaI crystals, we are not able to decide the exact correspondence of levels at around 280 kev.

(13) Selenium (Z=34).—We were able to study all stable isotopes of Se with enriched targets. The eveneven nuclei each yielded one state, several of which had not been observed previously. The trend in both their excitation energies and transition probabilities is very pronounced and can be seen in Figs. 11, 12, and 13. An excitation curve for Se<sup>78</sup> between 4.5 Mev and 6.8 Mev showed good agreement with E2 theory.

The odd-A nucleus  $Se^{77}$  turned out to be one of the most interesting cases we studied. Figure 5 shows the pulse-height distribution observed, as well as the coincidence peak we obtained when one channel was kept fixed on the line at 244 kev. This established the cascade nature of the 211-kev and 244-kev transitions. From the identical shapes of the excitation curves for the 457-kev and 211-kev transitions we can conclude that both originate from a state at 457 kev.

In addition to these major features, we found that we excited the known isomeric 160-kev transition. With the beam turned off, we were able to obtain the decay curve shown in Fig. 6. The half-life of 17.5 seconds agrees well with previous determinations. Since this transition is known to be of the E3 type, it was of interest to ascertain whether it was excited directly from the ground state, or indirectly via some higher excited state. The latter alternative was confirmed by determining the

<sup>33</sup> A. W. Schardt and J. P. Welker, Phys. Rev. 99, 810 (1955).

<sup>&</sup>lt;sup>32</sup> R. H. Nussbaum (private communication, May, 1956).



FIG. 5. Pulse-height spectrum for  $Se^{77}$  excited by 6.5-Mev alpha particles. Circles indicate the singles spectrum; squares represent the coincidence spectrum obtained when one channel was kept fixed on the 244-kev peak. Peak at 34 kev was shown to be an impurity x-ray. 86-kev peak is doubtful and not likely to be the cascade feeding the isomeric transition (see Fig. 6).

excitation curve of the delayed activity, and finding it to have a shape similar to the excitation curve for the 244-kev transition, and not at all the shape of E3 Coulomb excitation. Both the 244-kev and 457-kev transitions yielded excitation curves in very good agreement with E2 theory between 4.5 and 6.5 Mev. The small peak at 86 kev shown in Fig. 5 may be the cascade transition feeding the isomeric state. Only a little over 1% of the de-excitation of the 244-kev state<sup>34</sup> (or 2% when including internal conversion) takes



FIG. 6. Decay scheme of Se<sup>77</sup> states excited in Coulomb excitation with 6-Mev alpha particles. The decay curve for the 17.5-sec isomer indirectly excited is shown at the bottom. Spin assignments in parentheses are based on angular distribution measurements and are tentative. The isomer is probably fed from the 457kev state by a slow *E*1 transition (not shown) rather than by the 86-kev transition from the 244-kev state (see discussion of Se<sup>77</sup> in text). No evidence for direct *E*3 excitation was found.

<sup>34</sup> There is some question as to whether the isomer is fed from the 244-kev or the 457-kev state; see the following.

place via the isomeric state. From the known lifetime of the 160-kev transition we can compute the reduced E3 transition probability to be

$$B(E3) \uparrow = 1.03 \times 10^{-4} \text{ barn}^{3}$$

Using the semiclassical expression for E3 Coulomb excitation given by Alder and Winther<sup>11</sup> we find a cross section of 4.7 microbarns; our observed cross sections for E2 excitation of the 244-kev and the 457-kev states are 9 and 11 millibarn, respectively; hence we see that even the 1% cascade feeding of the isomeric state still exceeds any possible direct E3 Coulomb excitation by a factor of 20, so that there is no evidence for the direct process in Se<sup>77</sup>.

Since the ground-state spin of Se<sup>77</sup> is  $1/2^-$ , only states of  $3/2^-$  and  $5/2^-$  can be excited by E2 transition. We have investigated the angular distribution of the 244kev and 457-kev gamma rays in the hope of selecting between these two possibilities. We measured the relative intensities at 0° and 90° to the beam, and found the anisotropies given in Table IV. We also show the anisotropy for the pure E2 transition from the 2<sup>+</sup> state of Se<sup>78</sup>. These measurements support the tentative spin

TABLE IV. Angular distribution of gamma radiation from Se<sup>77</sup> and Se<sup>78</sup>. A is the anisotropy defined by  $A = [W(0)/W(\pi/2)] - 1$ . Bombarding energy  $E_0$  in Mev.

Nucleus	$E_0$ (Mev)	$\Delta E$ (kev)	A	Transition
<sup>34</sup> Se <sup>78</sup> <sup>34</sup> Se <sup>77</sup>	6.00 6.00 5.00	615 457 244	$+0.50\pm0.03$ $+0.50\pm0.03$ $-0.03\pm0.03$	$E2$ $E2^{a}$ $M1 - E2^{a}$

\* Tentative assignment.

assignments shown in the insert of Fig. 6. They are similar to the spin sequences in  $\mathrm{Rh}^{103}$ ,  $\mathrm{Ag}^{107}$ , and  $\mathrm{Ag}^{109}$  as determined by McGowan and Stelson.<sup>15</sup> Since the isomeric state has character  $7/2^+$ , it is hardly conceivable that an M2 transition from the  $3/2^{-}$  state at 244 kev can compete with the M1+E2 decay to the ground state, even to the extent of 1%. However, the very small anisotropy of the 244-kev radiation was measured at 5 Mev, where (1) the adiabatic parameter  $\xi$  is larger and the *a priori* angular effects are enhanced, and (2) the cascade effects from the 457-kev state are reduced. This low anisotropy favors mixed radiation and hence a spin of  $3/2^{-}$ . If the spin were  $5/2^{-}$ , the cascade to the isomeric state would be electric dipole as is found in the case of silver<sup>35</sup> for the second Coulombexcited state. The branching ratio (cross-over/cascade) is found to be 0.92, which also speaks in favor of the (1/2, 3/2, 5/2) sequence of spins, since we would expect an excess of crossover radiation if M1-E2 mixtures were allowed in both decay paths. We are thus forced to the conclusion that our excitation function for the isomeric activity is probably in error, and that the isomer must be fed by an E1 transition from the state

<sup>35</sup> T. Huus and A. Lundén, Phil. Mag. 45, 966 (1954).

at 457 kev. Assuming that this occurs via a 300-kev cascade (not detected because of its very low intensity), we can estimate the reduced electric dipole matrix element B(E1) from the branching ratio. We find that  $B(E1)=2.3\times10^{-6}$  barn, which is about  $2\times10^{-4}$  times the single-particle estimate. This in keeping with the observations of most low-energy electric dipole transitions.<sup>36</sup>

The  $\epsilon B(E2)$  values given in Table II have been corrected for the *decrease* of the first-excited-state excitation due to cascade feeding, as well as for the *increase* in second state excitation due to the same effect. The factor  $\epsilon$  merely reflects the fact that the small internal conversion corrections were not made. The peak seen at about 30 kev in Fig. 5 was found to be an impurity x-ray, probably of barium.

(14) Bromine (Z=35).—No further study; see reference 26.

(15) Krypton (Z=36).—By using a gas cell separated from the main vacuum by a thin nickel foil ( $\sim$ 300 kev thick) we were able to examine the gamma spectra of krypton and xenon. In Table V we list the gamma-ray transitions observed, as well as the probable isotopic

TABLE V. Coulomb excitation of natural krypton. Isotopic assignments are based on systematics and relative abundances, and are tentative.  $I_0$  is ground state spin;  $I^*$  is excited state spin.

A	Relative abundance (percent)	Eγ (kev)	I <sub>0</sub>	<i>I</i> *
80	2.3	610	0+	2+
82	11.5	775	0+	2+
83	11.5	457	9/2+	?+
84	57.0	880	0+	2+

assignments based on abundance considerations, as well as existing decay schemes. One point of interest was the observation of a 770-kev transition, probably associated with  $Kr^{82}$ . From the decay studies of  $Rb^{82}$ and  $Br^{82\ 37}$  no conclusion could be drawn concerning certain level orders near the ground state, and we were able to rule out the possibility of a first excited state at 550 kev in  $Kr^{82}$ . We hope to be able to carry on further measurements on this noble gas with enriched target material.

(16) Rubidium (Z=37).—We re-examined ordinary rubidium in the form of RbCl with 3-Mev alpha particles and confirmed our previously reported gamma ray at 155 kev<sup>3</sup>; this gamma ray has recently been assigned to the isotope Rb<sup>85,38</sup> and is associated with a known level in that nucleus.<sup>26</sup> A transition at 163 kev was found in Cs (see the following), and we took care to



FIG. 7. Pulse-height spectrum for ordinary ruthenium bombarded with 6.0-Mev alpha particles. Transitions to the firstexcited  $2^+$  states in the more abundant even-even isotopes can be seen, as well as some low-energy lines associated with the odd-*A* isotopes Ru<sup>99</sup> and Ru<sup>101</sup>, as assigned with isotopically enriched targets. Peak *BS* represents backscattering. Insert shows level scheme for Ru<sup>99</sup>, spin assignments being based on *E2* excitation, and long life and no gamma radiation in the decay of Tc<sup>99</sup>.

eliminate the possibility of cesium contamination in rubidium and vice versa.

No further results are reported for the elements strontium (Z=38) through niobium (Z=41).

(17) Molybdenum (Z=42).—We studied separated isotopes of Mo<sup>94</sup>, Mo<sup>96</sup>, Mo<sup>98</sup>, and Mo<sup>100</sup>, yielding firstexcited 2<sup>+</sup> states at 871, 778, 786, and 528 kev, respectively. Anomalous behavior in the position of these states can be seen between Mo<sup>96</sup> and Mo<sup>98</sup>, where the energy increases although we are going in the direction of larger neutron numbers. The transition probabilities seem to show the same "kink" (see Fig. 11). We measured the anisotropy of the 528-kev gamma radiation from Mo<sup>100</sup> with 6-Mev alphas and found it to be  $0.72\pm0.05$ , which agrees with the thick-target systematics<sup>15</sup> for pure E2 transitions. The excitation curve for this transition showed the theoretical shape from 3 to 7 Mev. The 205-kev transition associated with Mo<sup>95</sup> was previously reported.<sup>3</sup> Several of the even-even transitions had not been observed previously.

(18) Ruthenium (Z=44).—Figure 7 shows the spectrum obtained from a target of natural ruthenium. We examined separated targets of Ru<sup>104</sup>, Ru<sup>99</sup>, Ru<sup>101</sup>. Figure 8 shows an excitation curve for the 362-kev transition in Ru<sup>104</sup>, the lowest 2<sup>+</sup> state of any mediumweight nucleus. Because of the great strength of this transition, we were able to follow the yield over a factor of over 20 000. The agreement with E2 theory is seen to be excellent. The assignments of transitions to the other even isotopes Ru<sup>100</sup> and Ru<sup>102</sup> were made on the basis of abundance, and systematics.<sup>26</sup> The odd nuclei

<sup>&</sup>lt;sup>36</sup> See Fig. 3 of M. Goldhaber and J. Weneser, *Annual Review* of *Nuclear Science* (Annual Reviews, Inc., Stanford, 1955), Vol. 5, p. 1.

<sup>5.</sup> p. 1. <sup>37</sup> N. Benczer and C. S. Wu, Bull. Am. Phys. Soc. Ser. II, 1, 41 (1956).

<sup>&</sup>lt;sup>38</sup> Fagg, Geer, and Wolicki, Bull. Am. Phys. Soc. Ser. II, 1, 165 (1956).



FIG. 8. Thick-target excitation curve for  $Ru^{104}$  with alpha particles. The curve is the theoretical, semiclassical (except for WKB correction) E2 curve modified for thick target by the procedure outlined in reference 8. Normalized at 5.5 Mev. Extreme yields differ by a factor of about 20 000. Statistical errors are represented by the size of the circles.

of Ru showed low-lying levels, only one of which is associated with Ru<sup>99</sup> (89.5 kev). (See insert of Fig. 7.) It is clear that since the well-known, long-lived betaemitter Tc<sup>99</sup> (2×10<sup>5</sup> years) shows no gamma rays,<sup>39.</sup> the spin of the excited state of Ru<sup>99</sup> is limited to the values 1/2 and 3/2.

The spectrum obtained from  $\operatorname{Ru}^{101}$  is shown in Fig. 9, with the decay scheme given in the insert. The figure also shows the coincidence spectrum obtained when one channel was fixed on the 180-kev peak. The high state at 522 kev was also found to belong to  $\operatorname{Ru}^{101}$ . The *B* values given in Table II have been corrected for cascade decay as described for Se<sup>77</sup> above. The branching ratio (crossover/cascade) was found to be 1.43. The tentative spin assignments given in the figure are based on measurements of the anisotropies for the 127-kev and 303-kev transition listed in Table VI.<sup>40</sup> No anisot-

TABLE VI. Angular distribution of gamma radiation from Ru<sup>99</sup>, and Ru<sup>101</sup>. A is the anisotropy defined by  $A = [W(0)/W(\pi/2)] - 1$ . Bombarding energy  $E_0$  in Mev.

Nucleus	<i>E</i> <sup>0</sup> (Mev)	$\Delta E$ (kev)	A	Transition
44Ru <sup>99</sup> 44Ru <sup>101</sup> 44Ru <sup>101</sup>	2.50 2.50 6.50	89.5 127 307	$^{+0.03}_{-0.02} \pm 0.03$ $^{+0.22}_{-0.04_{5}} \pm 0.03$	$M1 - E2^{a} \\ E2^{a} \\ M1 - E2^{a}$

<sup>a</sup> Tentative assignment.

ropy was observed for the 89.5-kev radiation in Ru<sup>99</sup>, which still allows the two possible spin values mentioned above. The large branching ratio, favoring crossover radiation, seems to speak in favor of mixed M1-E2 radiation in both decay paths from the 307-kev state, and supports the spin assignments.<sup>40</sup>

Recently the 89.5-, 127-, and 180-kev transitions may have been observed in the decay of some long-lived radioactivities in rhodium.<sup>41</sup>

(19) Rhodium, Palladium, Silver, Cadmium (Z=45, 46, 47, 48).—We have previously published results for these elements based on enriched isotopes.<sup>6,7</sup> The values given in Tables I and II for the transition probabilities are revised and supersede our previous values. Further work on these isotopes has also been reported.<sup>15,21</sup> In



FIG. 9. Pulse-height spectra for Ru<sup>101</sup> bombarded with 6-Mev alpha particles. Open circles represent singles spectrum; triangles show coincidence spectrum obtained when one channel was kept fixed on the 180-kev peak. Line at 522 kev also belongs to this isotope. Level scheme is shown in the insert. There is evidence that the level at 307 kev is a doublet, one member of which gives rise to the cascade, the other to crossover radiation only (see reference 40). Branching information holds if only one level exists at 307 kev. Spin assignments are based on angular distribution measurements and are tentative.

them only by coincidence detection. They also have evidence that the state at 307 kev is really a doublet, one member of which decays by direct ground state transition only, the other only by cascade via the 127-kev state. This would of course vitiate our results for branching ratio and transition probability for the higher state. These authors also find additional transitions in  $Pd^{105}$ . We are indebted to Dr. McGowan for informing us of these results prior to publication.

to publication. <sup>41</sup> Hisatake, Jones, and Kurbatov, Bull. Am. Phys. Soc. Ser. II, 1, 271 (1956).

<sup>&</sup>lt;sup>39</sup> Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

<sup>&</sup>lt;sup>40</sup> F. K. McGowan and P. H. Stelson (private communication) have observed a number of additional transitions in Ru<sup>101</sup>, most of

Fig. 10 we show one more excitation curve, for the 370-kev transition in Pd<sup>110</sup>, showing excellent agreement with E2 theory over a large energy interval.

(20) Indium, Tin (Z=49, 50).—No lines observed.

(21) Antimony (Z=51).—Upon bombarding natural antimony metal with 6.5-Mev alpha particles, which consists of two odd isotopes, Sb<sup>121</sup> and Sb<sup>123</sup>, we detected a line at 154 kev, and a broad unresolved peak at 564 kev. The former corresponds to a known level in Sb<sup>123 39</sup>; the broad peak showing about twice the width of the 511-kev annihilation line from Na<sup>22</sup>, can be accounted for by two gamma rays at 506 kev and 575 kev, coming from known levels at these energies in Sb<sup>121,39</sup>

(22) Tellurium (Z=52).—We had available all six even-even isotopes of this element in enriched metallic form, and observed all first-excited states as listed in Table I. The odd isotopes Te<sup>123</sup> and Te<sup>125</sup> were studied elsewhere.42

(23) Iodine (Z=53).—This monoisotopic element,  $I^{127}$ , was briefly re-examined<sup>4</sup> in the form of CaI<sub>2</sub>, PbI<sub>2</sub>, and KI, with both 3-Mev and 6-Mev alpha particles and transitions at 58, 200, 365, and 480 kev were observed. They are not listed in Table I. Other work on iodine<sup>43</sup> revealed many more lines under proton bombardment, of which only the ones at 58 and 200 key seem to correspond with our results. Mathur's decay scheme of  $Xe^{127}$  <sup>44</sup> shows the first three levels of  $I^{127}$  at



FIG. 10. Thick-target excitation curve for Pd<sup>110</sup> with alpha particles. The curve is the theoretical, semiclassical (except for WKB correction) E2 curve modified for a thick target by the procedure outlined in reference 8, normalized at 5.0 Mev. Extreme yields differ by a factor of about 5000. Statistical errors are represented by the size of the circles.





FIG. 11. Systematics of level energies and transition probabilities for even-even nuclei between Ti and Te. This is a revised and more complete version of Fig. 2 of reference 10. For definition of the deformation parameters  $\beta_E$  (open circles) and  $\beta_B$  (full circles) see Eqs. (9). Numbers in the figure are atomic numbers Z for the various nuclei.

56, 200, and 368 kev, in good agreement with our findings.

(24) Xenon (Z=54).—Using the gas-cell technique described for krypton above, we obtained the Coulomb excitation spectrum from natural xenon. Table VII shows the transitions along with tentative isotopic assignments based on systematics, abundance, and known levels.39

(25) Cesium (Z=55).—We re-examined this monoisotopic element, Cs<sup>133</sup>, in the form of CsCl with 3-Mev alpha particles, and found two lines at 81 kev<sup>3</sup> and 163 kev, the first of which corresponds to a level known from the beta decay of Xe<sup>133</sup> and Ba<sup>133</sup>.<sup>39</sup>

(26) Barium (Z=56).—We did not re-examine this element.4

Elements beyond barium have been previously discussed.8

## IV. DISCUSSION

#### A. Even-Even Nuclei

In Fig. 11, which is a revised and expanded version of Fig. 2 in reference 10, we have plotted the quantities  $\beta_{E^2}$  and  $\beta_{B^2}$ , defined as follows (for even-even nuclei)<sup>22</sup>:

$$\beta_{B}^{2} = \frac{2.43 \times 10^{5}}{A^{5/3} \Delta E}; \quad \beta_{B}^{2} = \frac{8.48 \times 10^{4} B(E2)}{Z^{2} A^{4/3}}, \qquad (9)$$

where  $\Delta E$  is given in kev, and B(E2) in  $10^{-48}$  cm<sup>4</sup>. Irrespective of the detailed interpretation of these socalled deformation parameters, they constitute a useful means of presenting the information on both level positions  $\Delta E$  and reduced transition probabilities B(E2), removing rather general effects of nuclear size,

TABLE VII. Coulomb excitation of natural xenon. Isotopic assignments are based on systematics and relative abundances, and are tentative.  $I_0$  is ground state spin;  $I^*$  is excited state spin.

A	Re	lative abundand (percent)	$e E_{\gamma}$ (kev)	Ιο	<i>I</i> *
13	0	4.1	530	$0^+$	2+ 2+
129,	2	26.9	670	$0^+$	2+
13	4	10.4	870	0+	2+

<sup>&</sup>lt;sup>44</sup> H. B. Mathur, Phys. Rev. 97, 707 (1955).

mass, and charge. We used the electromagnetic nuclear radius given by Eq. (5) above. A striking degree of correlation is seen to exist between the two quantities  $\beta$ , extending throughout the region of medium-weight even-even nuclei.

The relations (9) above hold rigorously only for strongly deformed nuclei according to the unified model,<sup>22</sup> i.e., for nuclei having axially symmetric equilibrium deformations leading to rotational states. This so-called strong-coupling approximation has been found to be remarkably successful in certain rather sharply-defined regions of the nuclear chart, mainly for neutron numbers between 90 and 112, and again greater than 136. In those regions a plot of the type given in Fig. 11 above is directly meaningful in terms of the unified model, the quantities  $\beta$  representing nuclear deformation parameters interpretable in terms of the nuclear ellipsoidal shape.<sup>22,45</sup>

For even-even nuclei of the medium-heavy elements, convincing evidence has been presented<sup>46</sup> for the existence of a spherical equilibrium shape for these nuclei, and the consequent applicability of the formalism of the unified model bearing on vibrational excitations. The main empirical points here are the spins of second excited states  $(0^+, 2^+, 4^+)$ , their location at roughly 2.2 times the energy of the first excited states, the absence or weakness of M1 radiation in  $2^+-2^+$  transitions between second and first excited states, the absence or retardation of crossover radiation from the second states to the ground states, and the enhanced nature of the E2transitions to the first-excited states.45 A glance at Table I and the quantity F, the favored factor, reveals that the reduced transition probabilities are from 10 to 40 times larger than for a single-proton transition, attesting to a considerable amount of cooperative motion of the quadrupole type in these nuclei. All these facts point to the occurrence of E2 surface excitations (phonons) having angular momentum 2, and obeying the usual selection rules for a harmonic oscillator. Attempts have been made to introduce higher-order terms in  $\beta^2$  in the collective Hamiltonian to account for the departure from the expected 2.0 energy ratio of second- to first-excited state, without sacrificing the benefit of the above-mentioned selection rules.<sup>46,47</sup>

The parameters which are appropriate to a description of vibrational states occur in the collective Hamiltonian<sup>22</sup>:

$$H_{\text{coll}} = \frac{1}{2} \sum_{\lambda} (B_{\lambda} |\dot{\alpha}_{\lambda\mu}|^2 + C_{\lambda} |\alpha_{\lambda\mu}|^2),$$

where the summation is extended over all orders of excitation, the  $\alpha_{\lambda\mu}$  and  $\dot{\alpha}_{\lambda\mu}$  are the amplitudes of deformation and their time derivatives, respectively, and the quantities  $B_{\lambda}$  and  $C_{\lambda}$  are mass transport and

surface tension parameters, respectively, associated with the vibration of order  $\lambda$ . We limit ourselves to quadrupole excitations ( $\lambda = 2$ ). For the excitation probability of the first vibrational 2<sup>+</sup> state (one-phonon excitation), Bohr and Mottelson<sup>22</sup> have given the expression

$$B(E2)(0^+ \rightarrow 2^+) = 5 \left(\frac{3}{4\pi} ZeR_0^2\right)^2 \left(\frac{\hbar}{2(B_2C_2)^{\frac{1}{2}}}\right). \quad (10)$$

Rather than obtain the surface tension parameter  $C_2$ from the surface energy term appearing in the semiempirical mass formula,<sup>22</sup> it is preferable to obtain it empirically along with  $B_2$  from relation (10) and the one for the frequency of the quadrupole vibration<sup>22</sup>:

$$\omega_2 = (C_2/B_2)^{\frac{1}{2}}.$$
 (11)

The first-excited state energy  $E_{2^+}$  is simply associated with  $\hbar\omega_2$ .<sup>25</sup> Note that no assumption as to the detailed collective mass flow has been made. A useful limiting case is represented by the irrotational flow of an incompressible liquid drop, for which the parameter  $B_2$ is given by<sup>22</sup>

$$B_2^* = (3/8\pi)AMR_0^2, \tag{12}$$

where M is the nucleon mass, and  $R_0$  is given by Eq. (5). The empirical values  $B_2$  are conveniently expressed as the ratios  $B_2/B_2^*$ . The solutions of Eqs. (10) and (11) for  $C_2$  and  $B_2/B_2^*$  in the form of simple numerical relationships have already been given in Sec. III-A, Eqs. (6) and (7); our empirical values for these quantities are listed in the last two columns of Table I. It is noteworthy that the effective surface tension  $C_2$  is a strong function of neutron number, and decreases monotonically as we move away from closed shells, reaching values much lower than estimated from the liquid-drop model. Values in excess of this estimate can be found near closed shells. Figure 12 shows a plot on



FIG. 12. Summary plot of surface tension parameters  $C_2$  for even-even nuclei between Ti and Te. The logarithm of  $C_2$  is plotted against neutron number N. Also shown is the estimate for  $C_2$ derived from the liquid drop model and the semiempirical mass formula (see text). Numbers in the figure are atomic numbers Z for the various nuclei. Note strong shell structure effects. For definition of  $C_2$ , see Eqs. (7), (10), (11), and (13).

 <sup>&</sup>lt;sup>45</sup> A. Bohr and B. Mottelson, Kgl. Danske Videnskab Selskab,
 Mat.-fys. Medd. 30, No. 1 (1955).
 <sup>46</sup> G. Scharff-Goldhaber and J. Weneser, Phys. Rev. 98, 212

<sup>(1955).</sup> <sup>47</sup> L. Wilets and M. Jean, Phys. Rev. 102, 788 (1956).

our empirical values of  $C_2$  vs neutron number, along with the estimate furnished for  $C_2$  by the liquid drop model<sup>22</sup>:

$$C_2 = 4.90A^{\frac{3}{2}} - 0.114 \frac{Z^2}{A^{\frac{3}{2}}}$$
 Mev, (13)

where use has been made of the empirical surface energy term  $4\pi R_0^2 S = 15.4A^{\frac{3}{2}}$  Mev derived from nuclear binding energies by Rosenfeld.<sup>48</sup> The inertial parameters  $B_2$ everywhere exceed the values corresponding to irrotational flow, the factor being smallest far from closed shells. Figure 13 shows a plot of  $B_2/B_2^*$ .

The occurrence of the sharply defined transitions between "strong coupling" and "weak coupling" regions points to the existence of a critical variable; Bohr and Mottelson<sup>45</sup> give an estimate in terms of the critical moment of inertia  $\mathcal{G}_{\text{crit}}$ , defined empirically by the familiar relation for the first rotational state of an even-even nucleus

$$E_2^{**} = 3\hbar^2 / \mathcal{G}_{\text{erit.}}$$
(14)

Their latest estimate for  $\mathcal{G}_{\text{crit}}$  is  $\mathcal{G}_{\text{crit}} = 0.23 \mathcal{G}_{\text{rig}}$ , where

$$\mathcal{G}_{\rm rig} = \frac{2}{5} M A R_0^2 (1 + 0.31 \beta^2 + \cdots)$$
(15)

is the moment of inertia of a rigid ellipsoid of constant density. Only for values of  $E_{2^+}$  appreciably less than  $E_{2^*}$  should we expect rotational spectra. A numerical estimate of this critical first-excited-state energy is

$$E_{2^{+}} \cong (922/A^{5/3})$$
 Mev. (16)

It is interesting to examine this limit in various regions of the nuclear chart; a number of the observed 2<sup>+</sup> states in the medium-heavy nuclei fall somewhat below this limit. However, in all the cases where the information is available, the typical symptoms of vibrational rather than rotational excitation are found (see above). (Mg<sup>24</sup> represents an exception, with an energy ratio  $E_4+/E_2+$ = 3.0, but there  $E_{2^+}$  is less than one third the critical value  $E_{2^+}^*$ .)

We shall briefly mention below the possibility of rotational behavior in some odd mass nuclei of this region.

Finally, the excitation of higher excited states of known spin 2<sup>+</sup> in even-even nuclei is of great interest in connection with the various theoretical attempts<sup>46,47</sup> at an understanding of the low-lying level system. This is rendered difficult experimentally because the ratio of second- to first-excited state energies in these nuclei is found to be very nearly 2.2<sup>46</sup>; it is further known that the cascade between second and first excited states is usually much more intense than the direct crossover to the ground state, so that the problem is one of detecting a very weak gamma-ray (cascade) in the presence of a very strong one (first-excited state transition) from which it differs in energy by only about 20% at best.

<sup>48</sup> L. Rosenfeld, *Nuclear Forces* (North-Holland Publishing Company, Amsterdam, 1948), p. 24.



FIG. 13. Summary plot of inertial parameters  $B_2$  for even-even nuclei between Ti and Te.  $B_2/B_2^*$  is plotted against neutron number N.  $B_2^*$  is the inertial parameter for irrotational flow [defined in Eq. (12)]. Note considerable deviations from unity, as well as shell structure effects. Numbers in the figure are atomic numbers Z for the various nuclei. For definition of  $B_2$ , see Eqs. (6), (10), and (11).

Disturbing first-excited-state radiation from neighboring even-even nuclei (even in highly enriched targets) will usually contribute to obscuring that energy region. The predicted first-order transition probability to such a second state of course vanishes, since according to the vibrational interpretation a two-phonon absorption is involved. It might, however, be possible to detect the excitation by coincidence measurements provided that rather large bombarding currents at fairly high energy are available.

### B. Odd-A Nuclei

There is very little we can say in a general way concerning the nuclei of odd mass in this region. Because of the great variation of ground-state configurations, it is not surprising that each odd-A nucleus seems to require separate description. Many of the E2 transitions observed are much stronger than the single-particle strength. This usually manifests itself in terms of an enhanced admixture of E2 radiation to the usually predominant M1 radiation occurring in many of these transitions between states of equal parity, differing in spin by less than two units. The vibrational interpretation of these enhanced transitions is rather complicated, since one must now take into account the coupling of the phonons to the ground state configuration<sup>25</sup>; this is possible for the limiting cases of weak and strong coupling, but rather poorly understood for the case of intermediate coupling, which unfortunately seems to be the rule rather than the exception. An empirical way to decide what régime of coupling prevails for any particular case is to evaluate a dimensionless coupling parameter q in terms of the quantities  $\hbar\omega_2$  [see Eq. (11)] and  $C_2$  for a neighboring even-even nucleus:

$$q = \left(\frac{5}{16\pi}\right)^{\frac{1}{2}} \left(\frac{K}{(\hbar\omega_2 C_2)^{\frac{1}{2}}}\right),$$
 (17)

where K is  $\sim 40$  MeV, and to compare its value to 1.

We have already discussed the existence of an effective critical first excited state energy in even-even nuclei; although we saw that no rotational spectra are known to exist even in the nuclei where the criterion (16) is just satisfied, it is possible that the addition of an odd nucleon may provide enough additional polarizing force to distort the nuclear core into a nonspherical equilibrium shape. Between mass numbers 100 and 110 definite strong-coupling symptoms have been reported for three nuclei of character 1/2-, Rh<sup>103</sup>, Ag<sup>107</sup>, and  $Ag^{109.6}$  The case of spin 1/2 is slightly anomalous,<sup>22</sup> and in the fitting of the rotational spectrum there occurs an additional constant, the decoupling parameter a. Since only two transitions are excited in these nuclei, the test of their rotational nature (in the sense of strongly deformed nuclei) is not too stringent, as far as level positions are concerned. Nevertheless, both the absolute magnitudes and ratios of transition probabilities point toward possible rotational behavior.

In our present work, we have encountered another odd nucleus with spin  $1/2^-$  having properties similar to the three nuclei mentioned above, including an E3 isomeric transition and two prominently excited states. Since three of the even-even nuclei of selenium satisfy criterion (15), and the coupling parameter  $q \gg 1$  [see Eq. (17)], it may not be unreasonable to expect some rotational behavior in Se<sup>77</sup>. If we accept the spin sequence (1/2, 3/2, 5/2) as supported by our angular distribution measurements, we can solve for the quantities a and  $\hbar^2/2g$  from the level energies at 244 and 457 kev:

$$a=0.31; \hbar^2/2g=61.9 \text{ kev}.$$

The latter splitting constant corresponds to a firstexcited 2<sup>+</sup> state in a neighboring even-even nucleus of 370 kev. The lowest observed 2<sup>+</sup> state is in Se<sup>74</sup> at 567 kev; this seems to indicate an increased effective moment of inertia due to the odd neutron contribution. The ratio of excitation probabilities is 1.8 which is not too far from the expected ratio of 1.50. The very small E1 matrix element connecting the 5/2<sup>-</sup> and 7/2<sup>+</sup> states could be ascribed to the operation of the selection rule applying to the quantum number K,<sup>22</sup> there being a change of 3 units between the 5/2<sup>-</sup> member of the ground state rotational band (K=1/2) and the 7/2<sup>+</sup> state (K=7/2).

If we carry this analysis a step further, assuming that the mixed cascade radiation from the 5/2 to the 3/2state at 211 kev contains the theoretically predicted amount of E2 component, we can compute the mixture ratio  $\delta^2 = \text{Int}(E2)/\text{Int}(M1)$  as well as the reduced magnetic-dipole transition probability B(M1), expressed in units of the square of the nuclear magneton:

$$\delta^2 = 0.055; \quad B(M1) / \left(\frac{e\hbar}{2Mc}\right)^2 = 0.013$$

The latter quantity can be compared to the singleparticle M1 estimate, ascribing the transition entirely to the odd neutron, and we obtain

$$B(M1)/B(M1)_{\rm s.p.} = 9.6 \times 10^{-3}.$$

We have found no case of ground state spin  $I_0 > 1/2$ where any indication of a simple rotational spectrum exists, for A < 153. In fact, we have found a nonrotational spectrum in Eu<sup>151</sup> (N=88) and a perfectly developed rotational band for Eu<sup>153</sup> (N=90),<sup>49</sup> although both have  $I_0=5/2$ .

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<sup>&</sup>lt;sup>49</sup> N. P. Heydenburg and G. M. Temmer, Phys. Rev. 104, 981 (1956), following paper.