of the sum of the 1.199-Mev beta end point and the 1.290-Mev gamma energies is 2.489±0.010 Mev or 2.673±0.011 mMU.

ACKNOWLEDGMENTS

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Coulomb Excitation of Elements of Medium and Heavy Mass*

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Yields of gamma rays from thick targets of In, I^{127}, Ta^{181}, $Re^{185,187}$, $Ir^{191,193}$, $Hg^{198,199,200,202}$, Th, and U were investigated under proton or alpha-particle bombardment in the energy range 2 to 4 Mev. By using the theory for electric excitation, reduced matrix elements for all electrically excited radiations were computed and compared with single-particle estimates. The results are interpreted in terms of the strong-coupling collective model of Bohr and Mottelson and compared with the systematic trends in the region of the closed shell at 126 neutrons.

A 512-kev gamma ray excited in In by proton bombardment must result from a nuclear reaction. From excitation curves, gamma rays in I^{127} at 60, 208, 392, 438, 631, 751, and 941 kev appear to arise from Coulomb excitation. No cascade radiations are observed. The radiations from Ta, Re, Ir, and Hg show the effects of a transition from the strong collective rotational motion to a collective vibrational nuclear motion or single-particle excitation as one approaches the closed neutron shell. Only the first excited states of Th^{232} and U^{238} were excited with alpha particles. No evidence for other levels was observed with proton excitation. The results agree within experimental error with other excitation measurements and with radiative lifetime measurements.

INTRODUCTION

COULOMB excitation has recently been exploited to produce a large volume of experimental data on the properties of low-lying excited states of nuclei throughout the mass table.[1] Because the nature of the electromagnetic interaction is well understood, one can make quantitative deductions of the magnitudes of radiative matrix elements from the experimental data. These results may then be compared with the same quantities deduced from direct radiative lifetime measurements. The theory of Coulomb excitation has been treated semiclassically[2] as well as rigorously.[3] The semiclassical approximation developed by Alder and Winther[2] is sufficiently accurate for the work reported here. Coulomb excitation studies have provided a striking confirmation of the predictions of the unified nuclear model of Bohr and Mottelson.[4] That model is most successful in the regions between closed shells where the collective effects in the nucleus produce large distortion, enhanced quadrupole moments, and enhanced radiation matrix elements.

Some of the experiments reported here were undertaken to supplement investigations of level properties by inelastic neutron scattering.[5] At the present time, inelastic neutron scattering cannot be used to determine spins and parities, although it can excite a much wider variety of states than can electric excitation. I^{127} was selected because the results from (n,n') scattering were quite ambiguous and it was hoped that Coulomb excitation would clarify the problem.

Verifications of the theory for excitation had been made[6] for some medium-mass elements and it was generally assumed that the direct nuclear interaction would not play any appreciable role in nuclei as heavy as In when bombarded with protons in the energy range 2–3 Mev. After startling deviations from theory were observed in indium, excitation studies were made

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[1] A review paper by Alder, Bohr, Huus, Winther, and Zupancic Revs. Modern Phys. (to be published) summarizes the experimental results. See also summary papers by G. Temmer and N. Heydenburg, Phys. Rev. **100**, 150 (1955) and Huus, Bjerregaard, and Elbek, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **30**, No. 17 (1956). P. H. Stelson and F. K. McGowan, Phys. Rev. **99**, 112 (1955) and McClelland, Mark, and Goodman, Phys. Rev. **97**, 1191 (1955).

[2] K. A. Ter-Martirosyan, J. Exptl. Theoret. Phys. (U.S.S.R.) **22**, 284 (1952). K. Alder and A. Winther, Phys. Rev. **91**, 1578 (1953); **96**, 237 (1954).

[3] G. Breit and P. B. Daitch, Phys. Rev. **96**, 1447 (1954); Biedenharn, McHale, and Thaler, Phys. Rev. **100**, 376 (1955).

[4] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953); A. Bohr, dissertation, Copenhagen 1954 (unpublished); also A. Bohr and B. Mottelson in *Beta- and Gamma Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 17.

[5] J. J. van Loef and D. A. Lind, Phys. Rev. **101**, 103 (1956).

[6] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **96**, 426 (1954).

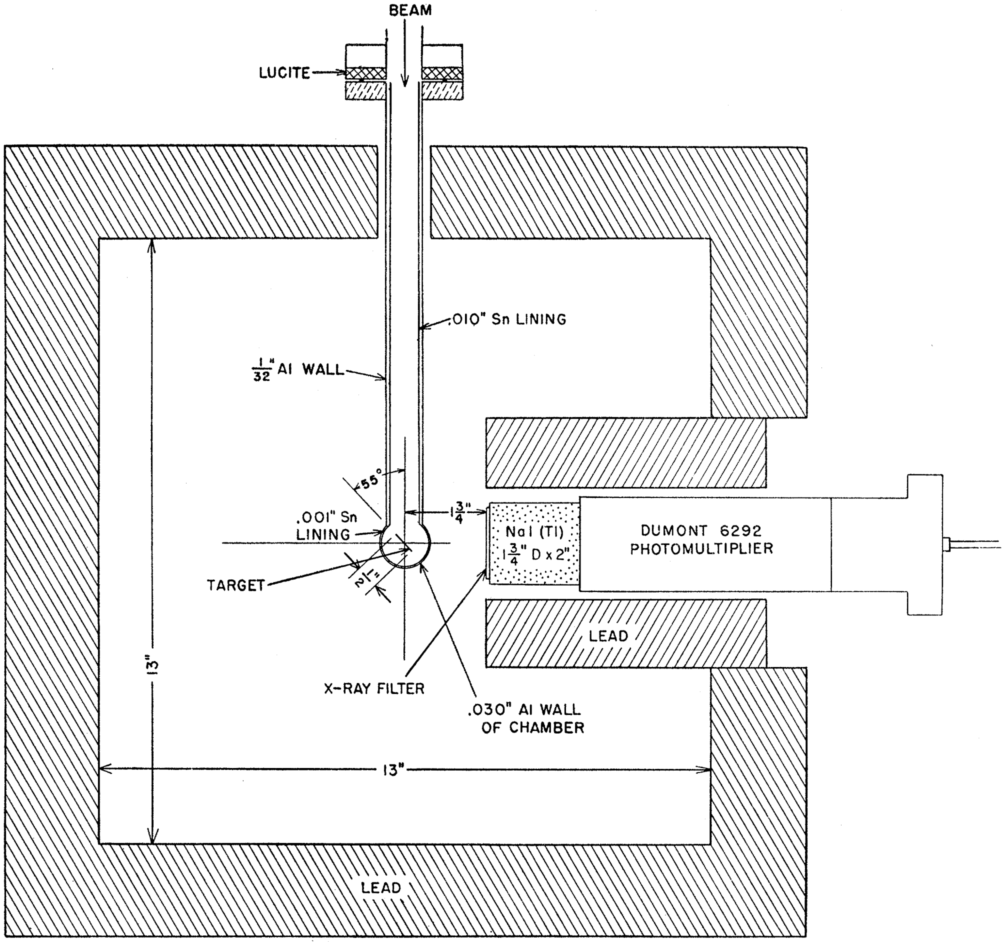

FIG. 1. Top view of target chamber and detector assembly. The extensive shielding was necessary to cut out the room background. Electron suppressor and beam-defining apertures were located off the top of the figure.

on I^{127} as well as the heavier elements to verify that excitations were actually by the Coulomb field. The elements Ta, Re, Ir, and Hg were selected because they span a range of neutron number approaching a closed shell. Data were not available on several of these elements. Ta provided a check of our techniques because the results for it are well known. These elements cover the transition from the well-developed rotational states[4] to the vibrational collective modes expected to exist near the closed shells.[7] Systematics[8] of the level schemes and transition probabilities made in the rare-earth region could be extended to the closed shell at 126 neutron number. Th and U lie in the next strong-coupling region and were expected to show the well-developed rotational spectrum. In addition, a vibrational mode with spin 2+ should lie at an energy sufficiently low to be excited by protons of the energy available. There is also evidence from alpha-decay studies[9] for a low-lying 1− state in these nuclei. An unsuccessful attempt was made to observe either of these states in Th and U. Our results on the first 2+ state are reported, however.

EXPERIMENTAL PROCEDURE

The experiment consisted simply of bombarding a target with charged particles and measuring the intensity of the de-excitation gamma-ray spectrum with a scintillation spectrometer. The charged-particle beam accelerated by the Wisconsin electrostatic generator was analyzed with 0.1% resolution by an electrostatic analyzer.[10] The arrangement of the target area is shown in Fig. 1. The flat targets were positioned at 55° to the beam so that the gamma rays could be observed at 90°. This position seemed most suitable from the standpoint of keeping to a minimum the absorbing material between target and detector. It would have been preferable to observe at 55° or 145°, where the Legendre polynomial $P_2(\theta)$ becomes zero and the intensity is very nearly the average value over the sphere. Corrections were made for anisotropy, using data obtained here for angular distributions or

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[7] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 14 (1952); K. W. Ford and C. Levinson, Phys. Rev. **100**, 1 (1955).

[8] A. W. Sunyar, Phys. Rev. **98**, 653 (1955); G. M. Temmer and N. P. Heydenburg, Phys. Rev. **99**, 1609 (1955).

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published elsewhere.[11] The target crystal distance was set at 4.4 cm in order to make variations in the source-detector distance have negligible effect on the absolute observed yield. Excitation curves were taken at 1.8 cm because only relative yields were needed. For angular distributions, the crystal was suspended from above with its axis at 10 cm from the target so it could be rotated through ±135° inside the lead shield house.

Thick targets were used throughout the experiment, except in the case of In where yields from thick and thin targets were compared. These consisted of metallic foils when possible or of powders contained in shallow 0.010-inch tin cups 0.5 inch in diameter. The powders were melted or pressed to produce a compact target which would not disintegrate under bombardment. Target materials of the highest available purity were used but some difficulty was always experienced because of the presence of foreign material in or on the target. A diffusion pump and liquid air trap were connected between the last beam-defining aperture and the target to minimize organic deposits at the spot where the beam strikes the target. Graded filters were placed between the target and detector to reduce the bremsstrahlung and x-ray background.

The gamma spectrometer was a conventional NaI(Tl) crystal 4.4 cm in diameter by 5 cm long mounted on a Dumont 6292 photomultiplier. The spectrometer was calibrated for gamma detection efficiency with the following gamma sources: Ce^{144}, Hg^{203}, Na^{22}, and Cs^{137}. The absolute strength of the sources was determined by measuring the number of pulses under the complete pulse-height spectrum when a narrow collimated beam of radiation was passed through the NaI detector. The detector was sufficiently long so that 80% or more of the beam was absorbed; thus the errors in the absorption coefficients[12] introduced little error into the final result. A Co^{60} standard prepared in this way was compared with a Co^{60} standard source obtained from the National Bureau of Standards; the agreement in strength was within 5%. The Na^{22} sources were checked independently by measuring gamma-gamma coincidence rates. All the intensity measurements were internally consistent within 5% but a larger uncertainty of 12% has been assigned because a check of source strengths made at the Oak Ridge National Laboratory[13] failed to agree within the expected 5%.

Data were taken with alpha particles or protons, depending on the level being excited, and in some cases the same levels were observed with both particles. Excitation curves were taken for almost every line investigated. When one is certain that the radiations observed are Coulomb-excited, this is not necessary, but the data presented here bear out the fact that this precaution should always be observed.

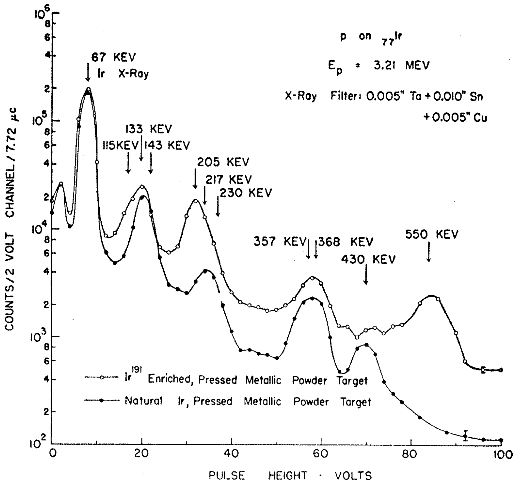

FIG. 2. Spectra of natural iridium metal and an enriched target under proton bombardment. Contamination is responsible for the lines at 205, 430, and 550 kev as well as the general increase in background for the enriched target. The machine background was from 5 to 10% of the spectrum shown.

ANALYSIS OF DATA

Gamma-ray intensities were determined by comparing the full-energy peak area with the photopeak efficiency curves obtained experimentally with the standard sources. The machine and bremsstrahlung backgrounds were obtained by making runs with tin or lead targets, both of which have a low yield of Coulomb-excited line radiation. The entire spectral intensity, after subtraction of background and proton bremsstrahlung, could be accounted for by the photopeaks and associated Compton spectra of the various radiations excited in the target. The photopeak widths were dictated by the energy *versus* resolution curves. Even when lines were not resolved, it was frequently possible to carry out a unique separation.

Figure 2 shows the spectra obtained from the Ir targets. The similar level schemes of Ir^{191} and Ir^{193} cause unresolved groups of photopeaks at about 140, 220, and 360 kev. These peaks are too broad to be accounted for by single gamma rays and are resolved as shown in Fig. 3, where only the parabolas representing photopeaks are drawn. The line at 115 kev follows a Coulomb excitation function and agrees with the isotopic enrichments, but is probably spurious; the 205-kev line is an impurity line and is not Coulomb-excited. The results from the enriched target bear out this analysis and aid in the identification of the isotope responsible for each line. It should be noted that in some cases the isotopic enrichment factors were used to determine relative magnitudes of the composite photopeaks. This was done only after it had been verified for

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[11] F. K. McGowan and P. H. Stelson, Phys. Rev. **99**, 127 (1955).

[12] G. R. White, National Bureau of Standards Report 1003, 1952 (unpublished).

[13] We are indebted to Dr. P. H. Stelson of the Oak Ridge National Laboratory for providing us with an independent calibration of our sources. The method used there was to measure the intensity in a known geometry with NaI detectors for which the efficiency had been computed.

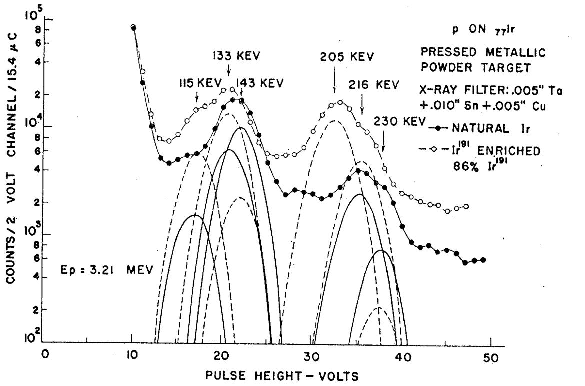

FIG. 3. Resolution of composite photopeaks in the Ir spectrum into constituent photopeaks. The positions and magnitudes of the photopeak parabolas were adjusted to give the best fit. In some cases it was necessary to use the isotopic abundances also, but in most nuclei they merely provided a check on the quantitative decomposition. The excitation curve of yield *versus* proton energy permits one to eliminate the spurious line at 205 kev.

lines excited in the different isotopes that the intensities followed accurately the reported enrichment factors. The energies of the lines making up a composite peak compared well with the values available from other experiments on the level structure of the element.

The actual thick-target gamma-ray yields per microcoulomb of bombarding ions are given in Table I. The accuracy of the results ranges from 20 to 35%. The error in the efficiency is 15%; the error in the separation of the photopeak yield may be 5–10%. An isotropic distribution was assumed unless data were available to correct for the anisotropy. In no case was the correction more than 12%.

Calculation of Transition Probabilities

The reduction of experimental yields to transition probabilities was carried out by using the semiclassical expression of Alder and Winther[2] for σ_{ex}. If the cross section is expressed in terms of the incident projectile energy, E, in the laboratory system one has

$$\sigma_{\text{ex}}(E)=\left(\frac{4\pi^2}{25}\right)\frac{M_1k(kE-\Delta E)}{Z_2^2e^2\hbar^2}B_{\text{ex}}(E2)g_{E2}(\xi),$$

$$\xi=\frac{Z_1Z_2e^2}{\hbar^2}\left(\frac{m}{2}\right)^{\frac{1}{2}}\left[\frac{1}{(kE-\Delta E)^{\frac{1}{2}}}-\frac{1}{(kE)^{\frac{1}{2}}}\right], \tag{1}$$

where $B(E2)$ is the reduced matrix element for an $E2$ transition and $g_{E2}(\xi)$ a tabulated function; $k=M_2/(M_1+M_2)$, and M_1 and M_2 are the masses of the projectile and target, respectively; ΔE is the excitation energy. The observed thick target yield is given by

$$Y(E_i)=KN_p\int_0^{E_i}\frac{\sigma_{\text{ex}}(E)}{\sigma_S(E)}dE, \tag{2}$$

where N_p is the number of particles incident on the target; K is a factor which gives the fraction of the excitations of a specified state which lead to the gamma rays under observation, and $\sigma_S(E)$ is the stopping cross section for the target material. Values of $\sigma_S(E)$ were obtained from the compilation of Fuchs and Whaling.[14] Interpolations were made by assuming that $\sigma_S(E)$ varies as $Z^{1/3}$, and the energy dependence was obtained from Rosenblum's formula,[15] but the values are known to no better than 10%. The theoretical thick-target yield curves were computed from $\sigma_S(E)$ and Eqs. (1) and (2). With the exception of a few cases noted in the discussion, all excitation curves agreed quite well with the theoretical values. In odd nuclei in which two levels are excited, the contribution of cascade transitions from higher states to the observed yield must be accounted for. Usually this contribution was sufficiently small to be undetectable in the excitation curves. Excitation curves provide a check on the accuracy of the spectral decomposition techniques since the relative intensities of different lines vary widely with bombarding energy.

The calculations of the value of $B(E2)$ can be made from the slope of the thick-target yield curve and the value of $\sigma_S(E)$ once the value of the factor K in Eq. (3) is known. Total internal conversion coefficients needed to compute K were taken from tables prepared by Rose and collaborators.[16] However, these have been corrected, as suggested by both theoretical and experimental work,[17] for the finite nuclear size. Data on conversion coefficients from radioactivity studies and from measurements of conversion electrons in Coulomb excitation[18] have been used, together with the theoretical values corrected for finite nuclear size and shielding, to arrive at values which are most consistent with all the data available. These are listed in Table I. In some cases the estimates of Sunyar[19] have been adopted to facilitate comparison with measured lifetimes. It may also be noted that much better agreement is obtained for $E2$ to $M1$ mixing ratios from conversion coefficient data and branching ratios or angular distributions when the nuclear size corrections are adopted.

RESULTS

The results are compiled in Table I. The gamma-ray energies and the bombarding particle are indicated; the

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[14] R. Fuchs and W. Whaling (unpublished compilation, 1954).

[15] S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 779 (1953); S. Rosenblum, Ann. Phys. **10**, 408 (1928).

[16] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951). See M. E. Rose in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers Inc., New York, 1955), Chap. 14; also circulated tables, "L Shell and Low-Energy K Shell Conversion Coefficients," by Rose, Goertzel, and Swift, 1955 (unpublished).

[17] A. H. Wapstra and G. J. Nijgh (private communication); L. A. Sliv, Z. Eksptl. Theoret. Fiz. **21**, 770 (1951); L. A. Sliv and M. A. Listengarten, Z. Eksptl. Theoret. Fiz. **22**, 29 (1952).

[18] Huus, Bjerregaard, and Elbek, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **30**, No. 17 (1956).

[19] A. W. Sunyar, Phys. Rev. **98**, 653 (1955).

TABLE I. Summary of results. Values for the yield are given for several bombarding energies. See text for discussion of the excitation functions. α is the total internal conversion coefficient. Column 9 gives the ratio of the observed decay transition matrix element to the single-particle value $B(E2)_{sp}$. $\tau_\gamma(E2)$ is the mean radiative lifetime for the $E2$ contribution to the transition computed from $B(E2)_{ex}$. Q_0 is the intrinsic quadrupole moment of the ground state computed by assuming the Bohr-Mottelson collective model with strong coupling.

Nucleus	Gamma-ray energy kev	Bombarding particle	Incident energy E_p or E_α Mev	Yield of gamma rays per microcoulomb[a] $\times 10^{-4}$	α_T	σ_{exc} cm² $\times 10^{+27}$	$B(E2)_{ex}$ / e^2 cm⁴ $\times 10^{+50}$	$B(E2)_d$ / $B(E2)_{sp}$	$\tau_\gamma(E2)$ sec $\times 10^{+10}$	Q_0 cm² $\times 10^{+24}$
In^{115}	512±12	p	3.50	10.5	0	0.545	18.2	55		
			2.50	0.090		0.0075	0.99	3.0		
I^{127}	60±2	α								
	201±4	α								
	208±7	p	3.20	8.00	0.14	0.180	4.4	12		
			2.50	2.85		0.117	4.5	12		
	392±8	p	3.20	1.55	0	0.058	2.32	6.1		
			2.50	0.24		0.016	1.49	3.9		
	438±10	p	3.20	0.45	0	0.0133	0.61	1.6		
			2.50	0.094		0.0059	0.58	1.5		
	631±10	p	3.20	2.45	0	0.112	11.6	31		
			2.50	1.85		0.020	10.6	28		
	751±25	p	3.20	0.76	0	0.041	7.4	20		
			2.80	0.17[b]		0.017	8.9	24		
	941±50	p	3.20	0.090	0	0.037	23	62		
			2.80	0.024[b]		0.021	54	140		
Ta^{181}	137±3	p	2.90	56.2[c]	2.09	3.56	183	239	2.18	6.59
			2.00	11.9[c]		1.72	186	244	2.14	6.64
	165±4	p	2.90	4.76[c]	1.08					
	302±4	p	2.90	3.14[c]	0.079	0.480	50	54	0.227	6.74
			2.00	0.21[c]		0.080	47	51	0.241	6.54
Re^{185}	125±4	p	3.20	28.2	2.85	1.74	83	98	42.4	4.2
	158±5	p	3.20	7.35	1.41					
	280±10	p	3.20	0.90	0.09	0.53	41	39	2.0	5.0
Re^{187}	135±4	p	3.20	32.0	2.18	2.54	123	146	19.8	5.1
	163±5	p	3.20	5.9	1.28					
	300±10	p	3.20	1.11	0.07	0.60	50	48	0.66	5.5
Ir^{191}	133±4	p	3.20	18.4	2.47	1.02	51	52	57	3.1
	216±6	p	3.20	7.4	0.66					
	356±8	p	3.20	6.8	0.05	0.67	78	60	0.36	5.3
Ir^{193}	143±4	p	3.20	12.7	2.02	1.05	53	53	40	3.2
	230±6	p	3.20	1.07	0.56					
	368±8	p	3.20	3.38	0.04	0.25	32	25	0.75	3.4
Hg^{198d}	411	p	3.20	1.02[c,b]	0	0.24	44	13	0.76	2.2
Hg^{199}	159±4	p	3.20	6.79	1.03[e]	0.27	17	8	150	1.6
	209±5	p	3.20	2.48	0.7[e]	0.094	6.4	5	64.0	1.3
Hg^{200}	375±7	p	3.20	3.46[c]	0.05	0.34	55	15	1.1	2.4
Hg^{202d}	439	p	3.20	2.36[c,b]	0	0.20	43	12	0.57	2.2
Th^{232}	50±5	α	3.33	0.39[b]	340	17.0	335	80		5.7
			2.46	0.072[b]		8.0	317	75		5.6
U^{238}	45±3	α	3.33	0.21[b]	700	24.7	490	120		6.9

[a] Gamma-ray yields were computed from intensities measured at 90° to the beam by assuming isotropic angular distribution.

[b] Error ±50%.

[c] Yields corrected for angular distribution with data given by F. K. McGowan and P. H. Stelson, reference 11.

[d] Computed from the 425±7 kev gamma ray observed in natural Hg. The yield was 3.02×10^4 gamma rays/ uc for proton energy of 3.20 Mev (see $Hg^{198,\,202}$ discussion).

[e] Internal conversion coefficient adopted from Sunyar, reference 19.

errors in the gamma-ray energies arise in part from instrumental effects and in part from the decomposition of complex spectra. The observed yields at specified bombardment energies are given. The excitation cross sections and the reduced matrix elements, calculated under the assumption that electromagnetic excitation is responsible, are also shown. In indium and perhaps in I^{127} the assumption of pure electric excitation may be invalid. Excitation curves were taken for most of the radiations investigated, and the agreement with the theoretical electric $E2$ curves is indicated by the fact that $B(E2)_{ex}$ is independent of bombarding energy. $\tau(E2)$ is the mean radiative lifetime for the $E2$ contribution to the transition. It is the reciprocal of $T(E2)$, the radiative transition probability, which is given by

$$T(E2)=\left(\frac{4\pi}{75}\right)\frac{1}{\hbar}\left(\frac{\omega}{c}\right)^5\frac{2I_i+1}{2I_f+1}B(E2)_{ex}, \tag{3}$$

where ω is the angular frequency of the radiation, and I_i and I_f are the spins of ground state and excited state, respectively. The ratio of the reduced matrix element for decay to the single-particle estimate given by Blatt and Weisskopf[20] is presented. No statistical factors have been incorporated into the single-particle estimate, but

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[20] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

TABLE II. Results on mixing ratios computed from crossover to cascade intensities. The experimental intensity ratio is given as R. The values of Q_0 are taken from Table I. μ_n is the magnetic moment in nuclear magnetons. g_K is the intrinsic particle gyromagnetic ratio, and g_R the ratio for the rotational motion.

	I_0	R Cascade to crossover intensity ratio	$\delta_{12}{}^2$ [a]	$Q_0\times10^{+24}$ (cm²)	μ_n [b]	$\lvert g_K-g_R\rvert$	g_K	g_K (single particle)	g_R [c]
Ta^{181}	7/2	1.52	0.19	6.62	2.1	0.45	0.70	0.49 $(g_{7/2})$	0.25
Re^{185}	5/2	8.17	0.008	4.6	3.17	1.08	1.53	1.92 $(d_{5/2})$	0.53
Re^{187}	5/2	5.36	0.02	5.3	3.20	1.15	1.63	1.92	0.52
Ir^{191}	3/2	1.09	0.03	3.1	0.17	0.47	0.32 −0.10	0.08 $(d_{3/2})$	−0.17 0.42
Ir^{193}	3/2	0.316	0.30	3.2	0.17	0.02	0.12 0.11	0.08	0.10 0.13

[a] $\delta_{12}{}^2$ is the $E2/M1$ intensity for cascade transitions.
[b] Values of magnetic moments are taken from H. Kopfermann, *Kernmomente* (Akademische Verlags, Frankfurt am Main, 1956).
[c] We have taken sign of $|g_K-g_R|$ positive ($Q_0/\delta>0$) to give a smaller value of g_R except for Ir isotopes where both values are shown.

the ratio $B(E2)_d/B(E2)_{sp}$ provides a measure of the collective nature of the nuclear oscillation. The conversion coefficients used are also listed. The values of the static quadrupole moment, Q_0, are computed from the reduced matrix elements using the results of the collective model of Bohr and Mottelson.[4]

The intensity ratio for crossover to cascade radiation from the second excited state of odd-A nuclei provides a means of computing the $E2$–$M1$ mixing ratio of the cascade radiation. One must assume that the collective model gives the ratio of the crossover to cascade probabilities for $E2$ radiation correctly. The agreement between values of Q_0 derived from excitation of first and second states substantiates that assumption. In Table II are listed these data and values of $\delta_{12}{}^2$, the ratio of intensities of $E2$ to $M1$ radiation. The values of the gyromagnetic ratios for the collective motion, g_R, and the intrinsic motion, g_K, are calculated as well. Because one can determine only $|g_K-g_R|$ in this manner, the positive value was assumed because it gave the most consistent value of g_R except in the cases of $Ir^{191,193}$. For those isotopes both signs were used. Equation (4) gives the relation used.

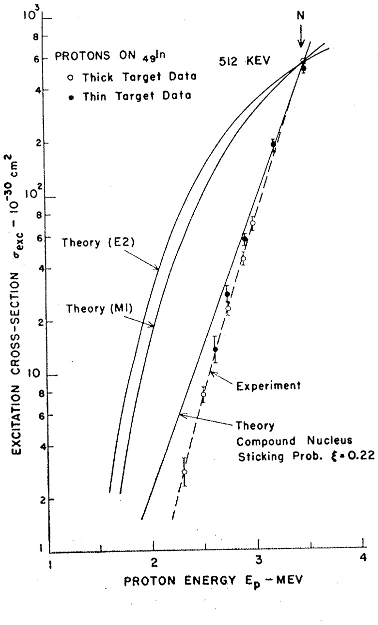

FIG. 4. Cross section for excitation of the 512-kev gamma ray from In^{115} under proton bombardment. Note that the cross sections calculated from thick and thin targets agree. The curves labeled Theory ($E2$) and Theory ($M1$) were calculated from the Coulomb excitation functions of Alder and Winther. Curves for $E1$, $E2$, and $E3$ do not differ significantly when normalized at N. The full curve based on compound nucleus formation using a sticking probability, ξ, of 0.22 and a nuclear radius, $R=1.5A^{1/3}\times10^{-13}$ cm is shown. The value of ξ was picked to normalize the curves at N.

$$\delta^2=\frac{3}{20}\frac{1}{I(I+2)}\left[\frac{Q_0\Delta E\,M_0}{\hbar^2(g_K-g_R)}\right]^2 \tag{4}$$

for $I+1\rightarrow I$, $I_0\neq1/2$. The values of g_K for a single particle are incorrect since they are calculated for a spherical well. The spheroidal well wave functions appropriate for these nuclei should be used to calculate these values.

Tantalum

Tantalum was studied to check the experimental technique. The three well-known gamma rays at 137±3 kev, 165±4 kev, and 302±4 kev were observed. The thick-target yields were compared with those obtained by Stelson and McGowan.[1] The absolute error in our yield measurements is 15–20% while they assign an error of 10–20%. The observed yields are consistently 15 to 30% lower than their values. It has been established that this discrepancy arises entirely from the method of absolute calibration of the gamma detector. The ratio of cascade to direct transitions from the 303-kev state was found to be 2.9 as compared to the value of 3.37 which they found. However the value of the $E2$:$M1$ mixing ratio $\delta^2=0.19$ is in excellent agreement with the result from angular distribution measurements.

Indium

Thick and thin targets of natural indium (isotopic abundances: In^{115}−95.8%, I^{113}−4.2%) were bom-

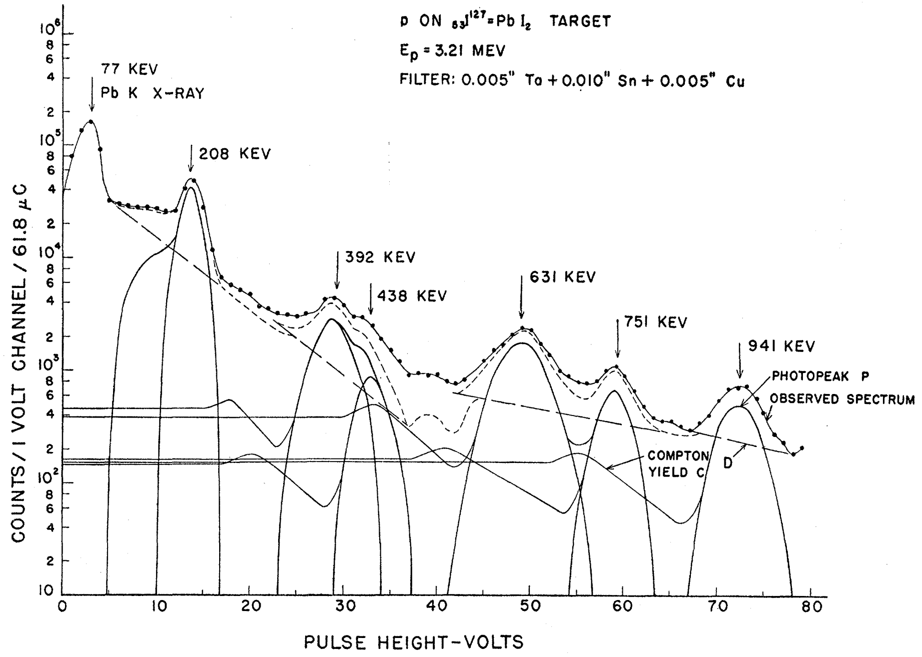

FIG. 5. Pulse-height spectrum from a thick PbI$_2$ target bombarded by protons. The region on the low-energy side of the 208-kev line suggests a possible cascade from the 392-kev level to the 208-kev level. The small peak at A is probably an impurity line. The decomposition of the spectrum into the parabolic photopeaks and the associated Compton spectra is shown. The 631-kev peak is too broad; possibly it is a composite line.

barded with protons and the single gamma ray at 512±12 kev, was observed. Temmer and Heydenburg[21] and Mark, McClelland, and Goodman[22] have reported a gamma ray at 500 kev in the proton bombardment of In which they assign to In^{115}. Varma and Mandeville[23] have established a level scheme from the decay of Cd^{115}. They find a level at 595 kev but none in the region of 500 kev. The yield and angular distribution of the 512-kev radiation were measured and an excitation curve was obtained from which the excitation cross sections shown in Fig. 4 were computed. Cross sections obtained from thick and thin targets agree. The experimental and theoretical electric excitation curves normalized at point N show a startling discrepancy. No other electromagnetic interaction would give a better fit, so the cross section for a (p,n) reaction was calculated assuming the statistical model for nuclear reactions.[20] The cross section is given by the relation

$$\sigma(p,n)=\sigma_c\xi, \tag{5}$$

where

$$\sigma_c=\pi\lambda\!\!\!^{-2}\sum(2l+1)T_l, \tag{6}$$

and ξ is a sticking probability; σ_c is the cross section for compound nucleus formation and T_l is the transmission factor for the lth partial waves. σ_c was computed for protons assuming a nuclear radius $R=1.5A^{1/3}\times10^{-13}$ cm. The sticking probability $\xi=0.22$ was chosen to make the cross section agree with experimental value at the normalizing point. The experimental excitation curve indicates that the reaction proceeds via the compound nucleus. If this is true, the only likely reactions would be (p,n) or (p,γ) reactions in In^{115}. Sn^{115} and Sn^{116} are stable so no activities would be produced. At the present time no assignment can be made for this line.

Iodine

Six gamma rays of energies 208±7, 392±8, 438±10, 631±10, 751±25, and 941±50 kev were observed from I^{127} under proton bombardment of PbI$_2$ targets. See Fig. 5. Gamma rays at 60±2 kev and 201±4 kev were clearly observed when iodine was bombarded with alpha particles. Excitation curves for all the gamma rays of iodine have been obtained and are shown in Fig. 6. With the exception of the lines at 392 and 941 kev, the agreement with theory is sufficiently good to indicate that electric excitation is probably responsible. The energy level scheme for I^{127} is not well understood, so a clear interpretation of the spectra is impossible. However, inelastic neutron scattering data[5] indicates that levels at energies equal to the observed gamma-ray energies are present in I^{127}. No appreciable intensity of cascade radiation has been observed. Bergström[24] and Mathur,[25] studying the beta decay of X^{127}, have observed levels at 56, 202, and 365 kev with cascade radiation between the three excited states. Possibly the anomaly on the low side of the 208-kev line is cascade radiation from the 392- to 208-kev levels. The intensity of cascade transitions may be low because they are highly converted. A search was made for neutrons from

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

[22] Mark, McClelland, and Goodman, Phys. Rev. **98**, 1245 (1955).

[23] J. Varma and C. E. Mandeville, Phys. Rev. **97**, 977 (1955).

[24] I. Bergström, Arkiv. Fysik. **5**, 191 (1952).

[25] H. B. Mathur, Phys. Rev. **97**, 707 (1955).

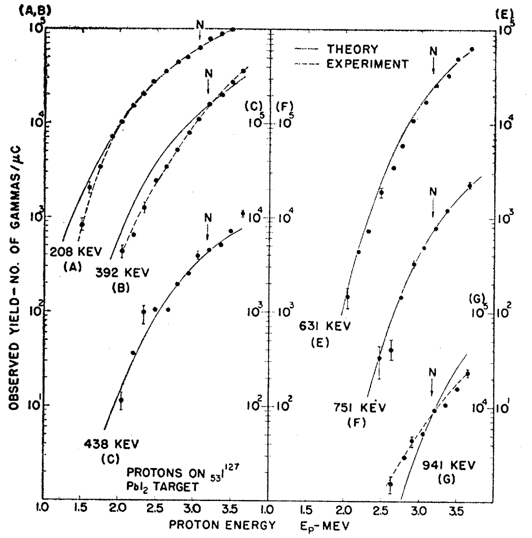

FIG. 6. Observed yields of gamma rays from I under proton bombardment compared with theoretical thick target yield curves for $E2$ excitations. The normalization is at the point N. Except for the 392- and 941-kev lines, the agreement is quite good and suggests at least that Coulomb interaction and not direct nuclear interaction is responsible for the excitation.

a possible (p,n) reaction but none were found. Furthermore, the radiations obtained by neutron and proton bombardment are the same; this indicates that they must arise from I^{127}. Excitation cross sections and reduced radiative transition matrix elements were calculated for all the transitions observed. The ratio of the reduced matrix element to the single-particle estimate range from the order of 1 to 30 and more. It is suggested that perhaps different levels are excited in the beta decay and in these observations. The discrepancy between 365 kev observed by Bergström and the value of 392 kev is too great to assign to instrumental error. Furthermore, levels at 370, 418, and 655 kev are reported[26] in the beta decay of Te^{127}. It is quite possible that the level scheme is much more complicated and that the resolution is not sufficient to see any but the most intense lines. It is certain, however, that the lines observed are not the result of cascade transitions from higher levels.

Re^{185}

Energy level diagrams for the Re, Ir, and Hg isotopes which were studied are shown in Fig. 7. The spin assignments in brackets are made on the basis of the strong-coupling rotational model. Only the low-lying states and transitions observed in electric excitation experiments are shown. We observed levels at 125±4 kev and 280±10 kev. These levels have been electrically excited previously by McClelland *et al.*[1] and Huus *et al.*[1] The ratio of the excitation energies is within a few percent of the ratio expected for rotational levels in the strong coupling approximation. Although the values of Q_0 determined from the two excited states, 4.2 and 5.0×10^{-24} cm^2, respectively, are consistent within the estimated experimental error, they are somewhat below the spectroscopic value of 7.9×10^{-24} cm^2 given by Mack.[27] From excitation of the first state, McClelland *et al.*[1] give a Q_0 value of 4.3×10^{-24} cm^2 while Huus *et al.*[1] set limits of 4.4 and 8.1×10^{-24} cm^2, depending upon the $M1$ and $E2$ mixing ratio. The ratio of the number of cascade to crossover gamma rays is 8.2 ± 2.0 from which a value of $\delta_{12}{}^2=0.008$ was obtained. The results obtained here are in reasonable agreement with recent results of Bernstein and Lewis[28] and Fagg.[29]

Re^{187}

Studies[30] of the radioactive decay of W^{187} indicate a level at about 134 kev; both conversion electrons and radiations from this level have been studied.[1,28,29] Another level at 320 kev has also been observed.[31] In the present work, electrically excited levels were identified at 135±4 and 300±10 kev. The ratio of the level energies is that expected for rotational levels. The values of Q_0, 5.1 and 5.5×10^{-24} cm^2, calculated from the two levels are to be compared with the spectroscopic value of 7.3×10^{-24} cm^2. McClelland *et al.*[1] quote a value at 4.1×10^{-24} cm^2 for Q_0, determined from the first excited state, and Huus *et al.*[1] give a value of 5.2×10^{-24} cm^2. The ratio of crossover to cascade gamma intensities, 5.36 ± 1.5, is in good agreement with the results of other workers.[29,31]

Ir^{191}

Composite gamma rays with energies of 133, 219, and 360 kev have been observed by Temmer and Heydenburg[32] in their electric excitation study of natural Ir. Huus *et al.*[1] give a measured value of 129 kev for the first excited state and assign a value of 3.3×10^{-24} cm^2 to Q_0. In the present experiments, levels were excited at 133±4 and 356±8 kev which agree with earlier electric excitation experiments as well as with radioactive decay studies.[30] The ratio of the excitation energies differs by about 10% from the ratio predicted for rotational levels. Sunyar's[19] value of 4.4 for the internal conversion coefficient is considerably larger than the value adopted here. The K/L ratio of 3.9 for the 133-kev radiation obtained from observations of conversion electrons[28] leads to the value we have adopted. This result is consistent with a mixing ratio

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[26] J. W. Starner, Los Alamos Scientific Laboratory (private communication). *Note added in proof.*—See Knight, Mizo, Starner, and Barnes, Phys. Rev. **102**, 1592 (1956).

[27] J. Mack, Revs. Modern Phys. **22**, 64 (1950).

[28] E. M. Bernstein and H. W. Lewis, Bull. Am. Phys. Soc. Ser. II, **1**, 41 (1956) and private communication.

[29] L. W. Fagg (private communication) and Wolicki, Fagg, and Geer, Phys. Rev. **100**, 1265(A) (1955).

[30] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1955).

[31] H. Mark and G. Paulissen, Phys. Rev. **99**, 1654 (1955).

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

$\delta^2=0$ and is in agreement with the similar result from the cascade to crossover intensity ratio. The value of Q_0 computed from the lower state was 3.1×10^{-24} cm^2 while that from the upper state was 5.3×10^{-24} cm^2. The difference is somewhat greater than the experimental error. The quadrupole mean radiative lifetime for the 133-kev state, $\tau(E2)=5.7\times10^{-9}$ sec, is consistent with the upper limit of 2.3×10^{-8} sec given by Sunyar.[19] The intensity ratio of cascade to crossover radiation from the second state is 1.09±0.25.

Ir^{193}

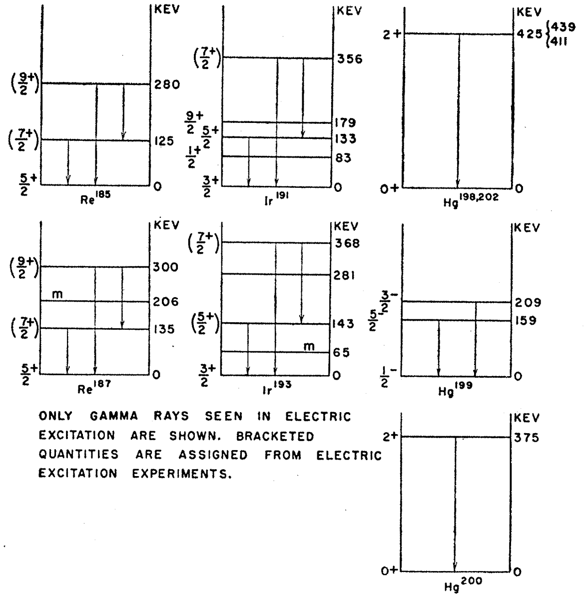
Gamma rays were observed at 143±4, 230±6, and 368±8 kev, and their excitation curves were in good agreement with theoretical electric excitation curves for an $E2$ transition. The energies are consistent with those reported by Temmer and Heydenburg.[32] Cork *et al.*[33] report a 139-kev level from Os^{193} decay studies. For the first excited level, Huus *et al.*[1] determined a Q_0 of 2.7×10^{-24} cm^2 which is somewhat below the values of 3.2 and 3.4×10^{-24} cm^2 found in these experiments. The cascade to crossover observed intensity ratio is 0.32±0.08. This value is not consistent with that found for the other odd-A nuclei in this region.

$Hg^{198,202}$

Excitation of the isotopes of mercury was studied using targets of HgO, Hg amalgamated on the surface of spectroscopically pure Pb, and HgO enriched to 65% in Hg^{199}. The known first excited states of the even isotopes of Hg are listed in Table III. The excitation of the first two levels of Hg^{199} was observed in the enriched target. A line at 368 kev and a composite line at 425 kev were observed. The 368-kev line was assigned to Hg^{200} and the 425-kev line was assumed to be a composite line of 411 kev from Hg^{198} and 439 kev of Hg^{202}. Since this work was completed, Barloutand, Grjebine, and Riou at Saclay have reported an investigation of Hg using separated isotopes.[34] Our matrix elements $B(E2)/e^2$ are approximately one-half the values they obtain. The small abundance of Hg^{196} and the absence of evidence for excitation of Hg^{204} [34] permit the assignment of the observed lines to Hg^{198}, Hg^{199}, Hg^{200}, and Hg^{202}. The composite yield at 425 kev was separated by placing known photopeak profiles at 411 and 439 kev and adjusting the heights to fit the composite peak. Known abundances were also used to calculate the cross sections. The error in the cross section may be 50% or more. Metzger and Todd[35] give a value of $\tau(E2)$ equal to 3.2×10^{-11} sec for the 411-kev level in Hg^{198}, and the value determined in the present experiments is 8.7×10^{-11} sec. For the 439-kev level in Hg^{202}, Metzger[36] gives $\tau(E2)=3.4\times10^{-11}$ sec compared to 6.5×10^{-11} sec determined here.

TABLE III. Known first excited states in even Hg isotopes.

Isotope	Level kev	Natural abundance percent
Hg^{196}	426	0.146
Hg^{198}	411	10.02
Hg^{200}	368	23.13
Hg^{202}	439	29.8
Hg^{204}		6.85



ONLY GAMMA RAYS SEEN IN ELECTRIC EXCITATION ARE SHOWN. BRACKETED QUANTITIES ARE ASSIGNED FROM ELECTRIC EXCITATION EXPERIMENTS.

FIG. 7. Level schemes for isotopes of Re, Ir, and Hg. Transition arrows indicate gamma rays observed under proton bombardment. Other known levels in the same energy region are included. The 425-kev gamma ray from natural Hg is assumed to be a composite of 411- and 439-kev gamma rays from Hg^{198} and Hg^{202}, respectively. The bracketed spin values are assigned from the systematics of rotational states.

Hg^{199}

Gamma rays were observed with energies of 159±4 and 209±5 kev. The good agreement between the observed and theoretical excitation curves identifies these gamma rays as decay radiations from electrically excited states. Heydenburg and Temmer [32] have previously excited a 163-kev gamma ray. Levels at these energies have been found in radioactive decay studies by Bergström *et al.*[37] From lifetime measurements, Sunyar[19] gives $\tau(E2)=0.7\times10^{-8}$ sec for the 159-kev state which compares reasonably well with the value 1.5×10^{-8} found here. However, Sunyar's value of $\tau(E2)=6.2\times10^{-10}$ sec for the 209-kev state is a factor of ten smaller than the value of 6.4×10^{-9} sec determined in the present

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[33] Cork, LeBlanc, Nester, Martin, and Brice, Phys. Rev. **90**, 444 (1953).

[34] Barloutand, Grjebine, and Riou, Compt. rend. **242**, 1284 (1956).

[35] F. R. Metzger and N. B. Todd, Phys. Rev. **95**, 853 (1954).

[36] F. R. Metzger, Phys. Rev. **98**, 200 (1955).

[37] Bergström, Hill, and DePasquali, Phys. Rev. **92**, 918 (1953).

experiments. The internal conversion coefficients used by Sunyar were adopted. Because of the intense x-ray spectrum, the yield of any 50-kev cascade gamma ray could not be measured, but the relative magnitude of the 159- and 209-kev yields and the agreement of the experimental and theoretical excitation curves imply a small cascade to crossover ratio. De-Shalit *et al.*[38] give a ratio of 0.25 to 0.30. Correcting for the 50-kev yield would increase the cross section for the 209-kev level by about 25%, but would not significantly reduce the discrepancy in $\tau(E2)$.

Hg^{200}

From Tl^{200} decay studies, Bergström *et al.*[37] assigned an energy of 368 kev to the first excited state of Hg^{200} with $I=2+$. A gamma ray which exhibited an electric excitation function was observed at 375±7 kev. The observed lifetime was 1.2×10^{-10} sec; no independent measurements of this value are available.

Thorium

An investigation of thorium and uranium was undertaken to see if the higher 2+ level predicted by the collective model[4] or the 1− level could be observed. While this work was in progress, Stelson and McGowan[1] reported a very weakly excited gamma ray at 760 kev under 5-Mev proton bombardment. Even at our peak proton energy of 3.6 Mev, we could not observe the 760-kev radiation. A number of different samples of Th metal and ThO_2 gave quite different spectra; the spurious lines were attributed to trace amounts of impurity elements. With alpha particles it was possible to observe the 50-kev first excited state and to obtain a cross section from which the reduced matrix element was calculated. The conversion coefficient, α_T, was estimated from tables of L shell conversion coefficients; the total M-shell conversion coefficient was assumed to be approximately 1/3 of the total L-shell conversion coefficient.[19] The value of 340 for α_T is subject to considerable error; nevertheless the reduced matrix element obtained is 80 times the single-particle value. Our value for Q_0 differs from that of Temmer and Heydenburg[39] but the difference lies only in the choice of α_T. They have taken a value $\alpha_T=1000$.

Uranium

Natural uranium as metallic foil and U_3O_8 were used for targets under the alpha-particle bombardment. The radiation at 45±3 kev corresponds to transitions from the first rotational 2+ state to the ground state in U^{238}. The error in the yield may be 50%. If one uses a value of 700 for α_T, the matrix element becomes 120 times the single-particle value. Again the discrepancy with Temmer and Heydenburg[39] is entirely accounted for by the value taken for α_T.

DISCUSSION

The results presented for the heavy nuclei (Ta and above) have been interpreted by the collective model using the strong-coupling approximation even though the sequence from Ta through Hg represents a transition from the region of the well-developed rotational states to the region near a closed shell where the deformation is expected to be small and hence the coupling weak. Th and U fall in the strong-coupling region again. The results may be compared with the systematic trends observed by Temmer and Heydenburg.[39] Even though the use of the strong-coupling formulas is not justified throughout the range from Ta to Hg, the values so obtained show a trend to smaller Q_0 as one approaches the closed shell at 126 neutrons. The values of the nuclear deformation fall along the curve published by Temmer and Heydenburg.[39] The even Hg isotopes have first excited states above the limit set by Bohr and Mottelson[40] for the existence of a rotational spectrum. This limit is given as $E_1<9.22\times10^5/A^{5/3}$ kev; for Hg it is about 300 kev. Osmium lies at about the limit, and our results indicate that iridium already shows considerable deviation from the strong-coupling model. Furthermore the value of $B(E2)/B(E2)_{sp}$ as seen in Table I shows a sharp decrease between Ir^{193} and Hg.

The measurements on indium indicate that some care needs to be taken in interpreting Coulomb excitation results for protons at intermediate mass unless excitation curves are also available. Excitation ratios by two different charged particles should be used to confirm that the interaction is via the Coulomb field. The excitation of iodine is difficult to understand because seven levels are excited and because the results from neutron excitation and Coulomb excitation are in essential agreement as to the levels but are in serious disagreement with the results from radioactive decay. Until additional experimental information is available, no interpretation of those results can be made.

The results shown in Table II for the gyromagnetic ratios for intrinsic particle motion and collective motion suggest that the strong-coupling model is valid for Ta and Re at least. Note that the minus sign must be taken for g_K-g_R in the case of Ir^{191} to get a reasonable value of g_R. Independent checks on the values of δ_{12}^2 from angular distributions should be obtained before attaching too much significance to these results. It is worth noting that in most cases the K/L conversion coefficient data are consistent with the values of δ_{12}^2 presented here when the $M1$ coefficients are corrected for nuclear size. Also the values agree quite well with results of Fagg.[29]

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[38] De-Shalit, Huber, and Schneider, Helv. Phys. Acta **25**, 279 (1952).

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[40] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd (to be published).

It is worthwhile to comment that the agreement of measurements of $B(E2)$ between different laboratories and from conversion electron or gamma-ray observations is still no better than a factor of 2. It is noted that results for Ta differ from those of Stelson and McGowan[1] by about 20%, while the results on Re and Ir are in fair agreement with those of Huus[1] but vary as much as 50% from those of Bernstein and Lewis[28] and Fagg *et al.*[29] The Hg results are consistently lower than those of the Saclay group.[34] Some of the discrepancy can be traced to choice of conversion coefficients, but there appears to be a need for more accurate measurements, especially when one attempts to determine mixing ratios from the intensity ratio of crossover to cascade radiations.]

ACKNOWLEDGMENTS

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Bremsstrahlung and Pair Production in Condensed Media at High Energies

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The effect of multiple scattering on bremsstrahlung and pair production is considered. The probability of these processes decreases considerably at energies $\gtrsim 10^{13}$ ev.

The calculations are carried out with the aid of the density matrix. The formulas thus obtained yield the probability of pair production and bremsstrahlung for arbitrary electron and photon energies.

I. INTRODUCTION

At high energies, when the directions of the particles participating in pair production and bremsstrahlung almost coincide, large longitudinal distances begin to play an important role. Thus, if a photon of wavelength $\lambda\!\!\!^{-}$ is emitted during bremsstrahlung, a certain length $l \sim \lambda\!\!\!^{-}/(1-v/c)$ is found to be essential, v being the electron velocity. Landau and Pomeranchuk[1,2] have shown that multiple scattering over this length leads to a significant decrease of the probability of the aforementioned processes. An estimate of the cross sections for bremsstrahlung and pair production in the limiting case of ultra-high energies ($E \gg 10^{13}$ ev) is given in reference 2.

The intensity of emission of soft photons by electrons of arbitrary energy has been computed previously.[3] In that paper the classical formula for intensity of emission by an electron moving along a given trajectory was averaged over all possible trajectories. This procedure was carried out by means of the distribution function which was averaged over the positions of the atoms of the scattering medium and which satisfies the usual kinetic equation.

The aim of the present paper is the deduction of formulas for the probability of bremsstrahlung (formula 61) and pair production (formula 63) per unit path in a condensed medium for arbitrary photon and electron energies. This is done by connecting the transition probability with the density matrix and then using the equation for the density matrix averaged over the scattering atom coordinates deduced previously.[4,5] At low energies formulas (61) and (63) transform into the Bethe-Heitler formula[6]; in the limiting case of ultra-high energies the formulas confirm the estimation obtained in reference 2. At photon energies much lower than that of the electron, formula (61) changes into the expression obtained in reference 3. Finally, for very soft photons, when the deviation of the dielectric constant from unity is important, formula (56) of the present paper yields in the limiting case the same results as those of Ter-Mikaelyan.[7]

Formulas (61) and (63) can be used to construct a theory of shower production in condensed materials at energies exceeding 10^{13} ev.

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[6] B. Rossi and K. Greisen, Revs. Modern Phys. **13**, 240 (1941).

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