Gamma Rays in the Decay of Barium-131*

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Gamma rays accompanying electron capture of 12-day Ba^{131} have been studied with a single-channel scintillation spectrometer. Relative intensities have been determined for the 122 kev (198), 214 kev (148), 372 kev (100), 496 kev plus satellite (360), and 620 kev (33) gamma-rays. Three new gamma-rays have been found at 823±20 kev (2.0), 917±15 kev (7.2), and 1032±15 kev (11.0), and their assignment to Ba^{131} has been established by chemical purification and half-life. Crystal summing studies confirm that the 620-kev transition is the crossover of the 122-kev and 496-kev transitions, and no significant crystal summing is found for the three new gamma rays. A peak corresponding to 83 kev was observed, which appears to be due to two or more unresolved gamma rays, but no gamma ray could be found at 100 kev. The decay of Ba^{131} was followed for 9 half-lives using an end-window beta proportional counter, and the half-life was found to be 11.52±0.08 days. The ratio of the pile neutron cross section for the activation of Ba^{131} to that of long-lived Ba^{133} has been found to be $\sigma(Ba^{130})/\sigma(Ba^{132}) = 1.2\pm0.3$.

INTRODUCTION

THE complex gamma-ray spectrum associated with the electron capture of 12-day Ba^{131} has been studied by a number of investigators.[1–11] Table I summarizes the results of previous investigations and present findings. The gamma rays at 122, 214, 244, 372, 496, and 620 kev are well established.

Weak high-energy radiations were found in a rough scintillation spectrometer survey of a radiobarium source, which came to hand in connection with other studies. It seemed of interest to study these hard gamma rays, whose assignment had been in question. Because of its inherent stability, our automatic recording scintillation spectrometer is well suited to the study of low-intensity radiation.

EXPERIMENTAL

Forty-four grams of reagent grade barium nitrate were irradiated in the Oak Ridge reactor for 28 days. The irradiated sample was dissolved in water and the cesium daughter of Ba^{131} was separated by a chemical procedure involving the addition of sodium and cesium carriers and the precipitation of barium chloride with a concentrated (5:1) hydrochloric acid diethyl ether mixture. The precipitate was evaporated almost to dryness three times to expel NO_3^- and C^{14} as $C^{14}O_2$. La^{+++} and Fe^{+++} carriers were added to an aqueous solution of the barium chloride, and ferric hydroxide and lanthanum hydroxide were precipitated with excess ammonium hydroxide. The scavenging was repeated three times. Barium was precipitated in final form as barium chromate in acetate-buffered acetic acid by the addition of potassium chromate. The barium chromate was filtered, washed, dried, and mounted.

Isomeric states of Ba^{133} (39 hr) and Ba^{135} (29 hr) were allowed to decay before proceeding with the scintillation spectrometry. Pulse-height spectra from chemically purified and unpurified sources were identical.

An automatic-recording single-channel scintillation spectrometer was used for all experiments reported here. The detector consisted of a cylinder of NaI(Tl) $1\frac{1}{2}$ in. diameter×1 in. high mounted on a DuMont 6292 photomultiplier tube, with a mixture of MgO and $Mg(ClO_4)_2$ for diffuse reflector. It was encased in a tight-fitting thin brass can and sealed with Scotch electrical tape. The detector was operated in a graded shield.

RESULTS

Typical pulse-height spectra with source distances of 7.6 cm and 1.8 cm are shown in Fig. 1. Peaks corresponding to gamma rays of 122, 214, 372, 496, and 620 kev are clearly resolved. The 244-kev gamma ray is not clearly seen, but its presence can be inferred from the skewness of the 214-kev peak. No peaks are seen at 196 or 585 kev. A peak appears at approximately 83 kev, but none was fund at 100 kev. Under closer examination, the 83-kev peak appears considerably broader than the 87-kev line of Cd^{109}: this peak is probably due to two or more unresolved gamma rays. The 496-kev peak was consistently broader than the full energy peak in the pulse-height spectrum of the Be^7 gamma ray (478 kev), indicating the existence of an unresolved satellite.

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TABLE I. Gamma rays associated with the decay of Ba^{131}. A summary of energy measurements to date, along with present findings.

Experimental technique	Gamma-ray energy (kev) γ_1	γ_2	γ_3	γ_4	γ_5	γ_6	γ_7	γ_8	γ_9	γ_{10}	γ_{11}	γ_{12}	γ_{13}	γ_{14}	Reference
Pb and Al abs.						240		420	500					1200	1
Pb and Al abs.						220			500					1700	2
Thin-lens spectr.									496						3
Siegbahn spectr.				122		206		372	494						4
Thin-lens spectr.						214									5
Mag.-lens spectr.				122	196	213	241	371	497						6
Abs. and coinc. abs.				160					460			830		1200	7
Solenoidal spectr.	43	65	108	122		214	241	370	494						8
Perm. mag. spectr.	55	79	92	124		216	239	374	488	585	620				9
				133			249		497						
Scint. spectr.			100	122		220		370	500		620		900[a]	1020[a]	10
Scint. spectr.				122		214	c	372	496		618			1090[b]	11
Scint. spectr.			83[d]	122		214	c	372	496[d]		620	823	917	1032	Present work

[a] The authors were not able to say that these gamma rays were not due to impurities.
[b] Reported but not definitely attributed to Ba^{131}.
[c] Observed but unresolved by the scintillation spectrometer.
[d] Unresolved complexity.

Above the 620-kev peak, the count does not go to background (0.005 counts/sec per $\frac{1}{2}$-volt channel). Two peaks can be clearly seen at 920 and 1030 kev, and the hint of a third at about 820. The latter becomes much more evident when the two higher energy spectra are "peeled off." The half-life of the high-energy spectrum over three half-lives was 11.8±1 days. Together with the specific barium chemistry, this rules out all contaminants but 12.8-day Ba^{140}, and its daughter, 40-hr La^{140}. Ba^{140} has no intense gamma rays above 540 kev.[12] La^{140} has a very intense gamma ray at 1.60 Mev,[13] but pulse-height spectra taken to 1.5 Mev failed to reveal any trace of radiation above 1.03 Mev. On the basis of known neutron activation cross sections,[1,14] the ratio of Ba^{140} to Ba^{131} activities for this irradiation is expected to be about 10^{-6}. We therefore conclude that the high-energy spectrum is not due to Ba—La^{140} and assign it to Ba^{131}.

The nature of the high-energy spectrum might lead one to suspect that it is due to "pile-up," an effect attributable to the finite resolving time of the spectrometer. This possibility was ruled out by observing the pile-up arising from an intense source of Cs^{137}, and using the resolving time thus obtained to compute the pile-up due to lower energy gamma rays of Ba^{131}. This effect contributes no more than 0.2% to the data count for the spectrum at 7.6 cm (Fig. 1). In addition, the highest energy peak shows normal width; and, as has been pointed out above, the high-energy spectrum decays with the characteristic half-life of Ba^{131}. We therefore conclude that the spectrum represents new high-energy gamma rays of Ba^{131}, not reported with confidence before.

The results of several spectra were averaged to obtain the energies of the three hard gamma rays. Calibration was provided by the 1114-kev line of Zn^{65},[15] the 661-kev line of Cs^{137},[16] and the 496-kev line of Ba^{131}. The values obtained are 823±20 kev, 917±15 kev, and 1032±15 kev; the errors arise mainly from gain fluctuations, uncertainties in subtractions, and counting statistics.

Intensities of the 122 kev, 214 kev, 496 kev and satellite, 620 kev, and the three new gamma rays have been determined relative to the intensity of the 372-kev line, which is neither a cross-over transition, nor coincident with any other gamma rays, except possibly the weak one at 244 kev.[11] Full-energy peaks were isolated by subtracting the spectra of higher energy gamma rays from the gross spectrum, by a method similar to that used by McGowan.[17] Areas were obtained by assuming the peaks to be Gaussian, and measuring their heights and $(1/e)$ widths. McGowan's values for the ratio of full-energy area to total area were used, with calculated geometrical efficiencies obtained from the Mathematics Panel at Oak Ridge.[18] Intensities have been corrected for decay during the run, differential absorption in the counter window (0.010-in. brass and 0.030-in. aluminum), and pile-up. Background was negligible.

The need for a correction for constant channel width of the pulse-height analyzer over the single-channel pulse-height spectrum has been pointed out.[11] A method of determining such a correction experimentally was adopted in this investigation, based on the premise that the only factor which ultimately is capable of distorting the true differential pulse height spectrum and consequently the area under a peak is the relation of the window width of the analyzer to the width of the peak at the input to the analyzer. The necessary correction to the area of a peak of given width would be the ratio of the integral count to the areal count for a calibration

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peak. A number of calibration spectra were taken of the full energy peak of the 661-kev gamma ray of Cs^{137}. The analyzer window was held constant at $\frac{1}{2}$ volt, the window used for the Ba^{131} work, and the gain of the linear amplifier was varied, resulting in a number of peaks of different disperison. Peaks were produced whose $(1/e)$ width ranged from 1.65 volts to 7.1 volts, spanning the range of interest for this experiment. Within the counting accuracy, the areas agreed with each other and with integral counts of the peak. It was concluded that the pulse height spectrum is not distorted, and that areal counts need not be corrected for this effect.

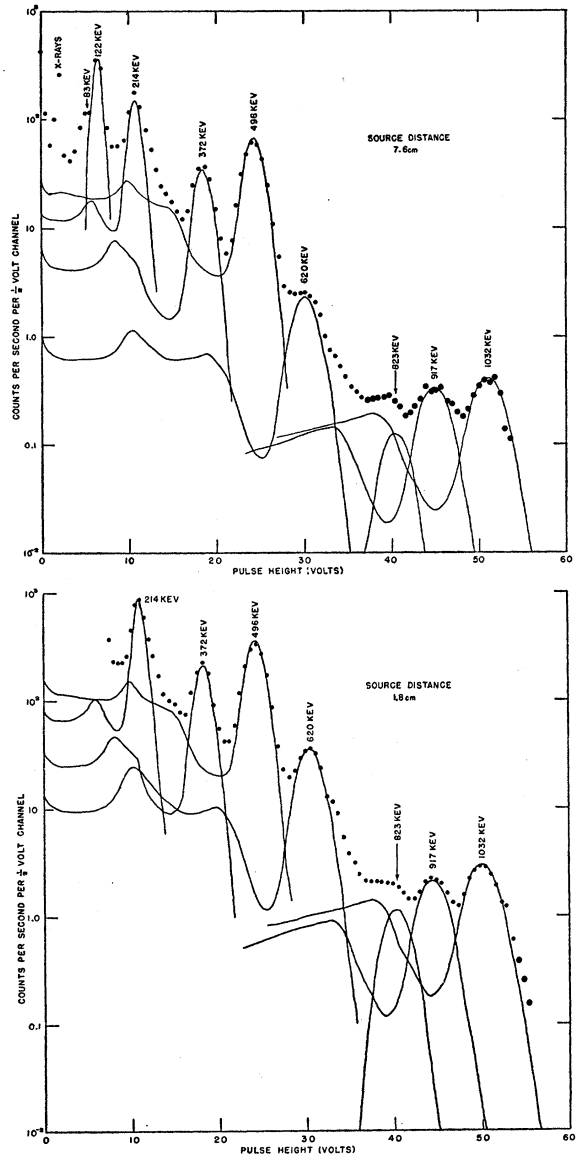

FIG. 1. Pulse-height spectra of Ba^{131} gamma rays at two source distances. The solid curves are the contributions of the various gamma rays to the gross spectrum. Statistical fluctuations are less than the diameter of the solid data points. For the spectrum at 1.8 cm, the source intensity was sufficient to jam the pulse-height analyzer below 7 volts.

TABLE II. Crystal summing study. Intensities of Ba^{131} gamma rays at two source distances (relative to 372-kev line as 100).

Transition energy (kev)	Relative intensity source distance 7.6 cm	1.8 cm
122	198±50	
214	148±30	144±30
372	100	100
496 and satellite	360±40	340±40
620	33±5	57±5
823	2.0±0.5	2.8±0.5
917	7.2±1	7.6±1
1032	11.0±1	14.0±1

The relative intensities obtained at the two source distances are displayed in Table II. The intensity of the 620-kev line is enhanced at the expense of the 496-kev line for small distances. This has been attributed to the summing of the 496-kev and 122-kev gamma rays.[11] The cross-over gamma ray at 620 kev is real, for its relative intensity is changed only by a factor two, and not by the pure summing factor of about ten. The measurements at 7.6 cm are free of summing effects, since the relative intensity of the 620-kev peak did not change as the distance was increased further. The intensities of the three hard gamma-rays relative to the 372-kev line did not change significantly with distance, which indicates that they are not cross-over transitions.

Our intensity measurements disagree with beta spectrometer results,[4,6,8] but are in essential agreement with the scintillation spectrometer work of Payne and Goodrich.[10] The latter also reported gamma-rays at 0.90 and 1.02 Mev with intensities 3% of the 496-kev line. Although Payne and Goodrich could not definitely assign them to Ba^{131}, they are undoubtedly the hard gamma rays which we have found at 917 and 1032 kev.

HALF-LIFE

The decay of Ba^{131} was followed through nine half-lives with a methane flow, 2π beta proportional counter with a 1.0 mg/cm^2 aluminized mylar end window. The half-life was found to be 11.52±0.08 days. The assigned error is twice the estimated standard error, as determined by a least squares fit.[19]

ACTIVATION CROSS-SECTIONS

We have determined the ratio of the neutron activation cross sections of Ba^{130} and Ba^{132}. The intensities of the gamma rays of Ba^{131} were measured in a fresh sample. After the Ba^{131} had decayed, the intensity of the 82-kev gamma ray of Ba^{133} was measured under identical conditions. Adopting the decay scheme of Lu, Kelly, and Wiedenbeck,[11] the sum of the intensities of the 496-kev, 372-kev, and 214-kev gamma rays, corrected for internal conversion, is an approximate measure of the Ba^{131} activity. To this approximation, the

[19] W. Edwards Deming, *Statistical Adjustment of Data* (John Wiley and Sons, Inc., New York, 1943), Chap. IX.

conversion of the 496- and 372-kev lines is negligible[4,8]; and the conversion coefficient of the 214-kev line is about 0.24.[8] Similarly, the intensity of the 82-kev gamma-ray of Ba^{133}, corrected for internal conversion, is a good measure of the Ba^{133} activity[20,21]; the internal conversion coefficient has been reported to be 3.5.[20] Corrected to the end of the pile irradiation, the activities of Ba^{131} and Ba^{133} are in the ratio 130:1. Assuming a uniform irradiation of 28 days, a 12.0-day half-life for Ba^{131}, and a 7.5-year half-life for Ba^{133},[22] the ratio of the pile activation cross sections is $\sigma(Ba^{130})/\sigma(Ba^{132})=1.2\pm0.3$.

If one recalculates for a 7.5-year half-life of Ba^{133}, one finds that the ratio obtained by Katcoff[1] is $\sim$2.2, while the ratio of the cross sections given in AECU-2040[14] is 0.002. Katcoff's results can be considered in fair agreement with the work reported here, but the Ba^{130} activation cross section appearing in AECU-2040 is certainly in error.

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Angular Distributions of Deuterons from (p,d) Reactions in Light Nuclei. I. Nitrogen*

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Deuterons have been observed from the reaction $N^{14}(p,d)N^{13}$, produced by 18.7-Mev protons. The deuterons were distinguished from proton background by a thin NaI crystal in a scintillation counter with the crystal thickness equal to the deuteron range. The angular distribution of deuterons to the N^{13} ground state was fitted by a curve calculated from Butler's theory for an angular momentum transfer $l_n=1$. This shows that the ground states of N^{13} and N^{14} have opposite parity and is consistent with the assignment of 1+ to the N^{14} ground state. The reduced width for the ground state transition is in qualitative agreement with that calculated using an independent-particle shell model with some indication that the N^{14} ground state is largely a D state. The transition $N^{14}(p,d)N^{13*}$ to the first excited state of N^{13} was not observed. The experimental upper limits on its cross section give upper limits of a few percent probability for admixtures of the configurations p^8s^2 and p^8sd in the N^{14} ground state. Application to the lifetime of C^{14} is discussed.

A. INTRODUCTION

A GREAT deal of recent work has been concerned with the angular distribution of the protons and neutrons from (d,p) and (d,n) stripping reactions.[1] Butler has shown[2] that the shape of the angular distribution in stripping determines the angular momentum carried into the target nucleus by the captured particle, e.g., by the captured neutron in a (d,p) reaction. By application of conservation laws for parity and angular momentum, this angular momentum transfer yields the change in parity and angular momentum between the initial state of the target nucleus and the final state of the residual nucleus. Thus if the parity and spin of the target nucleus are known, the parity and spin of the final state are determined (the latter with some ambiguity because of the vector addition rules for angular momentum). Because of this result, stripping reactions have given much information useful in nuclear spectroscopy.

With the present activity in the study of stripping reactions, it seems surprising at first that the inverse reactions (p,d) and (n,d) have been neglected, since, because of the reciprocity theorem,[3] Butler's results apply also to them. There is, however, an important practical reason for this neglect of "pick-up" reactions. The energy release Q in a ground-state stripping reaction is positive in almost every case, and is usually 5–10 Mev in magnitude. This means that low-energy deuterons will produce high-energy protons in a (d,p) reaction, while any background of scattered neutrons, etc., is at low energy and so is easily removed. For (p,d) reactions the situation is just the opposite; high-energy protons produce low-energy deuterons, which must be examined in the midst of a large background of protons of the same or higher energy. The interesting features of the angular distributions usually occur at small angles, where the background is particularly bad. Because of this difficulty, angular distributions from (p,d) reactions have been measured in a few favorable cases only.[4,5]

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