

**Ba<sup>131m</sup>—A New Isomer\***

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A new isomer of Ba<sup>131</sup> with a half-life of 14.5 min has been produced by the reaction Cs<sup>133</sup>(*p,3n*)Ba<sup>131m</sup>. The mass assignment has been made by excitation functions and from cross bombardments using the reaction I<sup>127</sup>(Li<sup>7</sup>,*3n*)Ba<sup>131m</sup>. The isomer decays by emission of a 106±4 keV  $\gamma$  ray. The growth of the Ba<sup>131</sup> ground-state  $\gamma$  rays has been observed, consistent with a 14.5-min parent. No decay (<0.1%) has been observed directly to Cs<sup>131</sup>.

**I. INTRODUCTION**

IN recent work by Friedlander *et al.*<sup>1</sup> on nuclear charge dispersion in the high-energy fission of uranium, it was noted that the formation cross sections of Ba<sup>131</sup> were consistently low. A possible explanation for this could be an as-yet-undiscovered isomeric state. Further strength is added to this idea by the fact that measurable isomeric states exist for Ba<sup>137</sup> (2.6 min), Ba<sup>135</sup> (29 h), and Ba<sup>133</sup> (39 h) for transitions from the 11/2<sup>-</sup> to 3/2<sup>+</sup> states. Recently the existence of a 2.1-hour isomer of Ba<sup>129</sup> has been reported.<sup>2</sup> On this basis a search for an isomer of Ba<sup>131</sup> was undertaken.

**II. EXPERIMENTAL AND RESULTS**

"Spec-pure" cesium carbonate was irradiated in a thin aluminum capsule in the internal beam of the McGill proton synchrocyclotron. Initial experiments were performed at 30 MeV bombarding energy—roughly the optimum for the reaction Cs<sup>133</sup>(*p,3n*)Ba<sup>131</sup>. The carbon in the carbonate ion served to monitor the beam current by the well-known reaction C<sup>12</sup>(*p,pn*)C<sup>11</sup>.

In the first experiment the sample, after irradiation, was dissolved out of the aluminum tube with dilute sodium hydroxide (very little aluminum dissolved) and split into two parts. The first aliquot was taken immediately for  $\gamma$ -ray analysis by means of a 3-in.×3-in. NaI(Tl) crystal coupled to a 100-channel pulse-height analyzer. The  $\gamma$ -ray spectrum of the unseparated sample contained a prominent peak at 106 keV which decayed with a half-life of about 15 min, a peak at 510 keV corresponding to the annihilation radiation of C<sup>11</sup> which decayed with a half-life of 21 min, as well as the  $\gamma$ -ray peaks due to 12-day Ba<sup>131</sup>.

The second aliquot was subjected to a rigorous chemical purification in order to prove that the activity was that of a barium isotope. Barium, strontium, rubidium, cesium, aluminum, and magnesium carriers were added at appropriate intervals and the barium was repeatedly precipitated as chloride, chromate, and

then chloride. The solution was scavenged with ferric hydroxide. Barium carbonate and then barium nitrate were precipitated.

The 106-keV  $\gamma$  ray was still very prominent. It decayed with a half-life of 14.5 min, and the ratio of x-rays to 106-keV  $\gamma$  ray to the  $\gamma$  rays corresponding to the Ba<sup>131</sup> ground state was the same, corrected to identical times, as that in the unseparated sample. A typical spectrum is shown in Fig. 1.

The excitation function for this 106-keV  $\gamma$  ray was obtained by bombarding cesium carbonate at various energies, dissolving the target in dilute sodium hy-

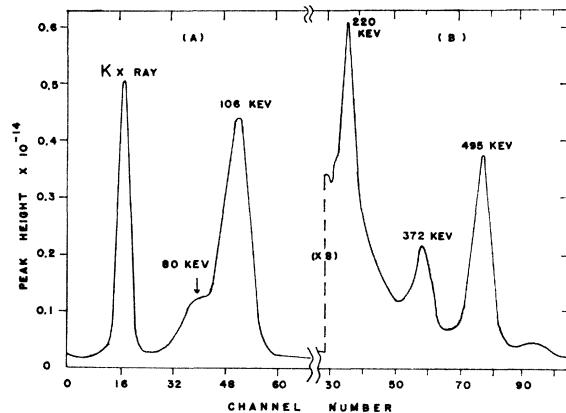


FIG. 1. Photon spectra of: (A) low-energy region showing peaks due to Ba<sup>131m</sup>, taken 64 min from the end of bombardment; (B) higher energy region, showing peaks due to Ba<sup>131g</sup>, taken 91 minutes from the end of bombardment.

droxide solution and comparing the intensities of the 106-keV  $\gamma$  ray of Ba<sup>131</sup> with the 510-keV annihilation radiation of the C<sup>11</sup> monitor. The cross sections for the reaction C<sup>12</sup>(*p,pn*)C<sup>11</sup> at various energies were taken from Crandall *et al.*<sup>3</sup> The results of these experiments are shown in Table I and the excitation functions for the isomeric and ground states of Ba<sup>131</sup> are given in Fig. 2. The 106-keV  $\gamma$  ray was assumed to be 100% abundant and its internal conversion coefficient  $\alpha_K$  to be 0.90.<sup>4</sup> The formation cross sections for the ground

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<sup>1</sup> G. Friedlander, L. Friedman, B. Gordon, and L. Yaffe, this issue [*Phys. Rev.* **129**, 1809 (1963)].

<sup>2</sup> E. Arbman and I. B. Haller, *Nucl. Phys.* **22**, 341 (1961).

<sup>3</sup> W. E. Crandall, G. P. Millburn, R. V. Pyle, and W. Birnbaum, *Phys. Rev.* **101**, 329 (1956).

<sup>4</sup> D. J. Horen, W. H. Kelly, and L. Yaffe, following paper, *Phys. Rev.* **129**, 1712 (1963).

TABLE I.  $Ba^{131m}$  and  $Ba^{131g}$  cross-section data.

Energy (MeV)	Duration (min)	$D^0_{C^{11}}$ ( $\times 10^{-7}$ )	$D^0_{Ba^{131m}}$ ( $\times 10^{-7}$ )	$D^0_{Ba^{131g}}$ ( $\times 10^{-7}$ )	$\sigma_{C^{11}}$ (b)	$\sigma_{Ba^{131m}}$ (b)	$\sigma_{m+g}$ (b)	$\sigma_{Ba^{131g}}$ (b)	$\sigma_m/\sigma_g$
20	6	...	0 <sup>a</sup>	...	0.15	...	...	...	...
25	6	24.5	108.2	1.46	0.52	0.83	1.17	0.33	2.5
30	15	59.5	397	58.3	0.75	1.90	2.45	0.55	3.5
35	6	28.0	164	20.0	0.80	1.71	2.16	0.45	3.8
40	5	38.1	105	12.5	0.87	0.85	1.08	0.23	3.6
45	5	55.6	68.0	8.24	0.87	0.38	0.50	0.12	3.2

<sup>a</sup> The 106-keV peak of  $Ba^{131m}$  was not detected at 20 MeV.

state were calculated using the 495-keV  $\gamma$  ray which was assumed to occur in 40% of the transitions.<sup>5</sup>

The excitation functions show definitely that the 106-keV activity is formed by a  $(p,3n)$  reaction on Cs and can only correspond to mass 131. The rigorous chemical purification proves that this is indeed an isotope of barium and thus must be  $Ba^{131m}$ .

The absolute cross-section values seem rather high. This may be due to dissociation of the cesium carbonate under the irradiation with consequent loss of  $C^{11}$  activity.

Further confirmation of this activity has been obtained using the HILAC at Berkeley. The 106-keV activity was produced by the reaction of carbon ions on Sn. This, however, due to the multi-isotopic nature of the tin and to the lack of beam energy discrimination used, produced many competing barium activities.  $Ba^{131m}$  was produced cleanly by the reaction  $I^{127}(Li^7,3n)Ba^{131}$  on a sodium iodide target shielded by 97.5 mg/cm<sup>2</sup> aluminum. This degraded the energy of the lithium ion beam below 38 MeV just above the Coulomb barrier for the above reaction, thus optimizing the formation of  $Ba^{131}$  and reducing to a minimum the amount of barium isotopes of lighter mass which would be formed. (It should be noted that both  $Ba^{130}$  and  $Ba^{132}$  are stable.)

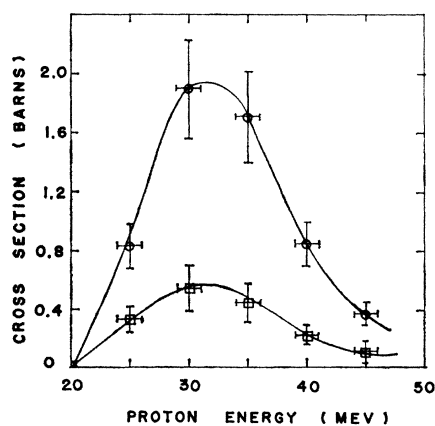


FIG. 2. Excitation functions for  $Ba^{131m}$  (●) and  $Ba^{131g}$  (□).

<sup>5</sup> *Nuclear Data Sheets*, National Academy of Sciences—National Research Council (U. S. Government Printing Office, Washington, D. C., 1961).

The sodium iodide target was dissolved in water, barium and cesium carriers added, and the mixture fumed with concentrated nitric acid to get rid of the iodine. The barium was then purified by repeated nitrate and chloride precipitations with appropriate additions of carrier.

The  $\gamma$  spectra obtained showed mainly the x-ray and 106-keV peaks. Both decayed with a 14.6-min half-life into a long-lived tail ascribable to  $Ba^{131g}$ . A peak at about 80 keV was first of all taken to be the escape peak of the 106-keV photon. Later work<sup>4</sup> showed that this was another  $\gamma$  transition in coincidence with the 106-keV peak. Other  $\gamma$  rays which were present in very much smaller intensity could all be ascribed to  $Ba^{131g}$ .

TABLE II. Cesium "milking" experiment.

Separation No.	$t^a$ (min)	$D^0$ <sup>b</sup> (c.p.m.)	$D^0/t$
1	14	1193	85.3
2	14	1106	79.0
3	13	1460	112.2
4	16	1750	109.3
5	1302	112 000	86.0
6	341	31 500	92.4

<sup>a</sup>  $t$  = time in minutes during which  $Cs^{131}$  was allowed to grow in.  
<sup>b</sup>  $D^0$  = counting rate of  $Cs^{131}$  extrapolated back to the time of separation, as calculated from the area under the x-ray peak, and corrected for chemical yield.

An attempt was made to determine whether any of the isomeric state decayed directly to  $Cs^{131}$ . Barium was rapidly separated from the cesium target and purified. Cesium fractions were successively removed and purified. The separation of cesium from barium was repeated four times at approximately 15-min intervals and twice more much later. The cesium samples, purified by cesium bismuth iodide and cesium platinum chloride precipitations<sup>6</sup> showed only the cesium x-ray peak due to the decay of  $Cs^{131}$  by electron capture. The data are given in Table II. The value of  $D^0$  in column 3 has been corrected for chemical yield and extrapolated back to the time of separation. The data shown in the last column are compatible with all the  $Cs^{131}$  being

<sup>6</sup> H. L. Finston and M. T. Kinsey, National Academy of Sciences—National Research Council Report NAS-IVS 3035, 1961 (unpublished).

formed by decay from the ground state of Ba<sup>131</sup>. The sensitivity of the experiment would suggest that less than 0.1% of the Ba<sup>131m</sup> proceeds by direct decay to Cs<sup>131</sup>.

An attempt was made to observe a growth in the ground state of Ba<sup>131</sup> due to decay of the isomeric state. A bombardment was made of a duration short compared to the half-life of the metastable state. The area of the 495-keV peak was then followed after a quick barium purification. From the classical growth and decay equations, one obtains the relationship

$$N_2^t = N_1^0 \frac{\lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t},$$

where the subscripts 1 and 2 refer to Ba<sup>131m</sup> and Ba<sup>131g</sup>, respectively,  $N^0$  is the number of atoms at time of separation, and  $N^t$  is the number of atoms at a time  $t$  later.

Since, in this case,  $e^{-\lambda_2 t} \approx 1$ , and  $\lambda_2 \ll \lambda_1$ , then

$$A_2^t = A_1^0 (\lambda_2 / \lambda_1) (1 - e^{-\lambda_1 t}) + A_2^0,$$

where  $A$  represents the counting rates of the species. Thus

$$(\lambda_2 / \lambda_1) A_1^0 e^{-\lambda_1 t} = [A_2^0 + (\lambda_2 / \lambda_1) A_1^0] - A_2^t.$$

The term in square brackets represents the flat portion of the growth curve and, in the experiment indicated, amounts to 7000 counts/min. The data are given in Table III. Column 1 gives the time elapsed between separation and measurement, column 2 shows the experimental observations, and column 3 gives the activity of the isomer itself in terms of the 12-day ground-state activity. This is plotted against time in Fig. 3 and gives a half-life of 13.7 min, by least squares analysis, in very satisfactory agreement with that obtained directly from observations of the 106-keV

TABLE III. Data for Ba<sup>131g</sup> growth.

$t$ (min)	$A_2^t$	$A_1^0 e^{-\lambda_1 t} =$ $7000 - A_2^t \times (1 - e^{-\lambda_1 t})$	$(\lambda_2 / \lambda_1) A_1^0$	$A_2^0$	$A_2^t$ - 1728
21	5154	1846	3322	1732	3426
26	5674	1325	3812	1862	3946
29	5819	1181	3958	1861	4091
32	5810	1190	4137	1673	4082
35	5617	1385	4286	1331	3889
40	6200	800	4498	1702	4472
43	6457	543	4604	1853	4729
48	6466	534	4745	1721	4738
54	6666	334	4880	1786	4938
59	6754	246	4965	1789	5026
95	6663	...	5224	1439	4935
114	6846	...	5258	1588	5118
124	7188	...	...	...	5460
150	6980	...	...	...	5252
180	7050	...	...	...	5322
210	6951	...	...	...	5222
267	7171	...	...	...	5443
269	6853	...	...	...	5125

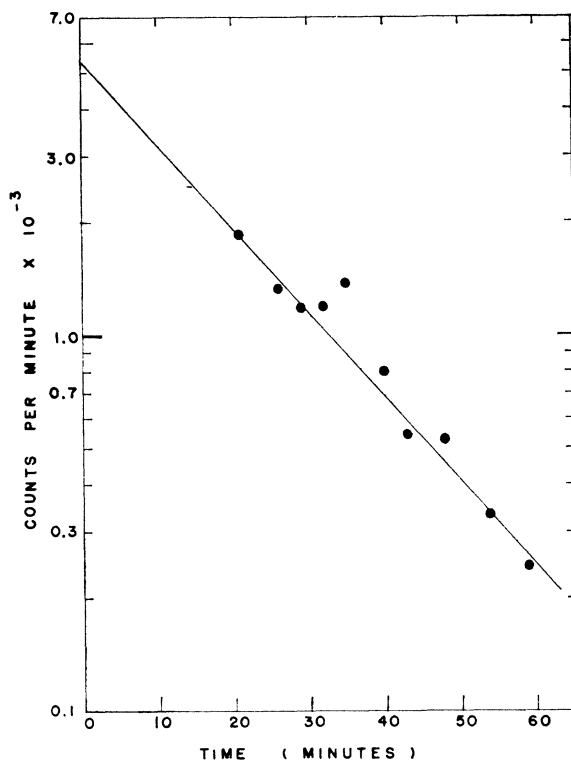


FIG. 3. Decay of Ba<sup>131m</sup> calculated from growth of the 495-keV peak of Ba<sup>131g</sup>.

$\gamma$  ray. Column 4 gives the contribution due to the isomer at various times  $t$ . (Since these are all in terms of the 495-keV  $\gamma$  ray from the ground state, the term  $\lambda_2 / \lambda_1$  will cancel.) The values in column 5,  $A_2^0$ , are found by subtracting those in column 4 from column 2. These have been averaged to give a value of 1728 counts/min, which is subtracted from the experimentally observed values in column 2 to give the data in

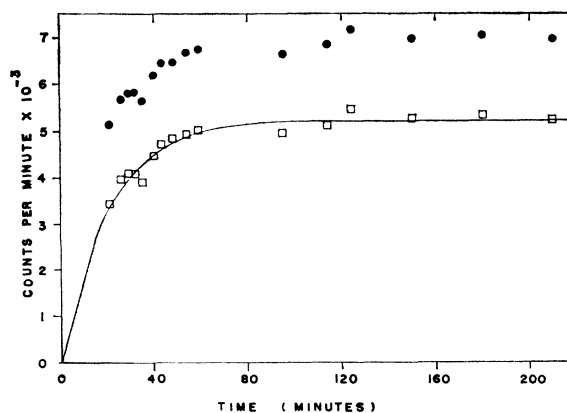


FIG. 4. Growth of the 495-keV peak of Ba<sup>131g</sup>. O Experimental points. □ Experimental points after Ba<sup>131m</sup>, directly formed, has been subtracted. — Theoretical growth curve of a 12-day daughter growing from a 15-min parent.

column 6 and from this the growth curve shown in Fig. 4. The original experimental points are shown as circles. The open squares give the growth curve derived as described, while the solid line is the theoretical growth curve for a 12-day activity growing from a 15-min parent.

$$\begin{aligned}(\lambda_2/\lambda_1)A_1^0 &= 5280 \text{ counts/min,} \\ A_1^0 &= 6.07 \times 10^6 \text{ counts/min,} \\ A_2^0 &= 1.72 \times 10^8 \text{ counts/min,} \\ \frac{\sigma_m}{\sigma_g} &= \frac{A_1^0 (1 - e^{-\lambda_2 T})}{A_2^0 (1 - e^{-\lambda_1 T})},\end{aligned}$$

where  $T$ , the length of bombardment, is 2 min.

$$\sigma_m/\sigma_g = 3.2.$$

This is in excellent agreement with the value of 3.5 at

a proton energy of 30 MeV obtained by direct measurement of the ratios of the 106- to 495-keV  $\gamma$  rays.

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### Characteristics of the Decay of $\text{Ba}^{131m}$ †

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Sources of  $\text{Ba}^{131m}$  were produced by bombarding sodium iodide with  $\text{Li}^7$  ions of about 38-MeV energy, and its half-life was redetermined as  $14.6 \pm 0.2$  min. Its decay was studied by means of scintillation techniques. The decay of  $\text{Ba}^{131m}$  proceeds by a  $(78 \pm 5)$ -keV  $E3$  transition ( $\alpha_K = 12.3 \pm 1.5$ ), followed by a  $(107 \pm 3)$ -keV  $M1 + E2$  transition ( $\alpha_K = 0.90 \pm 0.15$ ). Included is a discussion of possible spin and parity assignments for the levels involved in this decay. An auxiliary experiment showed that less than 1% of  $\text{La}^{131}$  decays lead to  $\text{Ba}^{131m}$ .

#### I. INTRODUCTION

RECENTLY, Tilbury and Yaffe discovered a 14-min isomer in  $\text{Ba}^{131}$  which decayed by emitting  $K$  x rays and a 106-keV photon.<sup>1</sup> On the basis of this half-life and energy, one would expect the isomeric transition to have multipolarity  $E3$  or  $M3$ . The ground-state spin and parity of  $\text{Ba}^{131}$  have been variously assigned  $\frac{1}{2}+$  and  $\frac{3}{2}+$ .<sup>2,3</sup> The  $\frac{1}{2}+$  assignment would require the spin and parity of the isomeric level to be  $\frac{7}{2}-$  for an  $E3$

transition and  $\frac{7}{2}+$  for an  $M3$  transition, whereas the  $\frac{3}{2}+$  assignment would require  $\frac{9}{2}-$  or  $\frac{9}{2}+$  for an  $E3$  or  $M3$  transition, respectively. To date, no isomeric states of these types have been found in other odd- $A$  nuclei in this region. Therefore, it was considered desirable to investigate further the decay of  $\text{Ba}^{131m}$  with the hope of determining the spin and parity of the isomeric level.

#### II. SOURCE PREPARATION AND APPARATUS

Sources of  $\text{Ba}^{131m}$  were produced by bombarding sodium iodide for approximately 15 min with about 38-MeV  $\text{Li}^7$  ions in the Berkeley Hilac (heavy-ion linear accelerator).

The sodium iodide was dissolved in water to which was added 10-mg Ba and 1-mg Cs carriers. Concentrated  $\text{HNO}_3$  was added and the solution boiled to fume off  $\text{I}_2$ . The barium was separated as  $\text{Ba}(\text{NO}_3)_2$  by the addition of fuming  $\text{HNO}_3$  to an ice-cold solution. Further purification consisted of two precipitations of  $\text{BaCl}_2$  by

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<sup>1</sup> R. S. Tilbury and L. Yaffe, preceding paper [Phys. Rev. 129, 1709 (1936)].

<sup>2</sup> *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences-National Research Council, Washington, D. C.); and references cited therein.

<sup>3</sup> C. B. Creager, C. W. Kocher, and A. C. G. Mitchell, Nucl. Phys. 14, 578 (1959).