

## Gamma Rays from the Deuteron Bombardment of Carbon-13<sup>†</sup>

R. J. MACKIN, JR.,\* W. B. MIMS,‡ AND W. R. MILLS, JR.

*Kellogg Radiation Laboratory, California Institute of Technology, Pasadena, California*

(Received December 30, 1954)

A magnetic lens spectrometer was used to investigate the gamma radiation produced by the deuteron bombardment of C<sup>13</sup> enriched carbon targets. A study of the Compton electron spectra from a thin Al converter at  $E_d=1.42$  Mev revealed the following gamma lines (energies in kev, not corrected for possible Doppler shift):  $4930\pm 40$ ,  $5130\pm 30$ ,  $5730\pm 30$ ,  $6120\pm 25$ . A weak line at  $3910\pm 50$  kev was established with the spectrum from a thick Al converter. At  $E_d=1.9$  Mev, additional lines appeared at  $6450\pm 50$ ,  $6730\pm 40$ , and  $811\pm 3$  kev. The last-named was identified by photoelectrons from a thorium converter. No new lines were discovered at  $E_d=2.7$  Mev. The well-known 6.12-Mev line is from C<sup>14\*</sup>, as are probably the 6.73- and 0.811-Mev lines. All others correspond to known levels of N<sup>14</sup>.

### I. INTRODUCTION

INFORMATION about the levels of N<sup>14</sup> below 7 Mev has been derived principally from investigations of neutron groups<sup>1</sup> and gamma rays<sup>2</sup> from C<sup>13</sup>+D<sup>2</sup>, and more recently from measurements on gamma rays<sup>3</sup> from C<sup>13</sup>+H<sup>1</sup>. In view of difficulties which have attended attempts to infer a consistent level scheme, it appeared worthwhile to re-examine certain controversial portions of the gamma-ray spectrum from the former reaction. Additional measurements were made at bombarding energies up to 2.7 Mev in a search for new gamma lines.

### II. EXPERIMENTAL PROCEDURE

A magnetic lens spectrometer<sup>2,4</sup> was used to study Compton electron spectra produced by gamma rays from C<sup>13</sup>+D<sup>2</sup>. A C<sup>13</sup> enriched, compressed soot target at the spectrometer focal point was bombarded by magnetically analyzed deuterons from a 3-Mev electrostatic accelerator. Because several closely-spaced gamma lines were anticipated, a 20-mg/cm<sup>2</sup> target was backed with an aluminum converter of minimum thickness consistent with adequate counting statistics, 21 mg/cm<sup>2</sup>. Additional runs were made with a 125-mg/cm<sup>2</sup> converter to detect any weak lines.

The spectrometer was operated with 1.9 percent resolution (Gaussian window) and an effective solid angle of 2.3 percent of a sphere. The momentum calibration for the Compton spectra was based on the ThD X-line at 9988.4 gauss-cm.<sup>5</sup> A helical baffle permitted separation of electrons and positrons. The detector of focused electrons was a magnetically compensated *trans*-stilbene scintillation counter<sup>6</sup> which

made possible energy discrimination against scattered electrons. The customary beam currents (0.5–1.5  $\mu$ a) provoked copious emission from the soot of thermal electrons or positive ions, depending on the target potential. In view of this difficulty, no attempt was made to measure beam current, and the detector count readings were normalized by means of a gamma-ray monitor. The monitor consisted of two thin-wall Geiger counters in coincidence, set along a line from the target perpendicular to the incident beam and separated from one another by  $\frac{1}{8}$ -inch Al so as to record only radiation above 1 Mev. The system was thus insensitive to the delayed (annihilation) radiation from N<sup>13</sup>, produced by C<sup>12</sup>(*d,n*)N<sup>13</sup>. Tests with natural sources confirmed that monitor and detector sensitivity were independent of the magnetic field. Measurements were made at bombarding energies of 1.42, 1.90, and 2.7 Mev.

A special search was made for gamma lines in the region 0.5 to 1.0 Mev with a carbon foil target<sup>7</sup> (about 0.3 mg/cm<sup>2</sup>), backed with a 20.2-mg/cm<sup>2</sup> thorium photoelectric converter.

### III. RESULTS AND ANALYSIS

Because internal conversion pairs contributed a significant background to the Compton electron spectrum, it was necessary to measure the positron spectrum. It is expected for light nuclei (and has been roughly verified in work at this laboratory)<sup>8</sup> that the electron and positron internal pair spectra will be practically identical. Accordingly, the difference of electron and positron curves was regarded as consisting entirely of Compton electrons. This spectrum is shown in Fig. 1, along with the positron data.

The spectrum above 20.5 kilogauss-centimeters (at  $E_d=1.42$  Mev) was analyzed by comparison with a theoretical Compton spectrum<sup>2</sup> which was derived from the Klein-Nishina formula. The theoretical curve included the effects of Doppler line broadening, electron energy losses in the converter, and the spectrometer

<sup>†</sup> Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

\* National Science Foundation Fellow, now at the Office of Naval Research, Washington 25, D. C.

‡ Commonwealth Fund Fellow, now at the Clarendon Laboratory, Oxford, England.

<sup>1</sup> R. E. Benenson, Phys. Rev. **90**, 420 (1953).

<sup>2</sup> R. G. Thomas and T. Lauritsen, Phys. Rev. **88**, 969 (1952).

<sup>3</sup> Woodbury, Day, and Tollestrup, Phys. Rev. **92**, 1199 (1953).

<sup>4</sup> Hornyak, Lauritsen, and Rasmussen, Phys. Rev. **76**, 731 (1949).

<sup>5</sup> W. Brown, Phys. Rev. **83**, 271 (1951).

<sup>6</sup> C. Wong, Phys. Rev. **95**, 761 (1954).

<sup>7</sup> J. D. Seagrave, Phys. Rev. **85**, 197 (1952); E. A. Milne, Phys. Rev. **93**, 762 (1954).

<sup>8</sup> Dougherty, Hornyak, Lauritsen, and Rasmussen, Phys. Rev. **74**, 712 (1948).

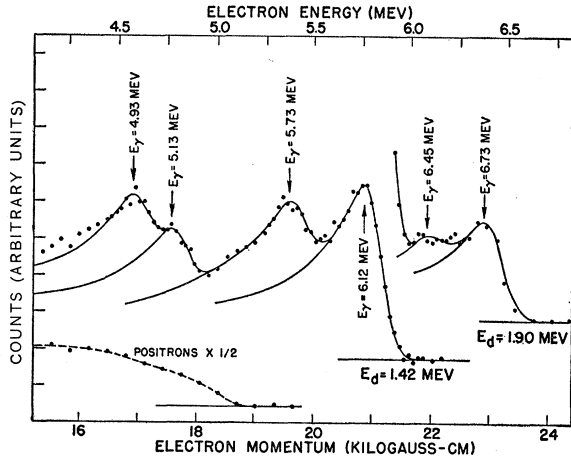


FIG. 1. Compton electrons from 20-mg/cm<sup>2</sup> C plus 21-mg/cm<sup>2</sup> Al converter.

resolution window.<sup>9</sup> In addition to the gamma-ray energy and yield determination, the theoretical line shape permitted extrapolation to lower momenta and made possible separation of the contributions from succeeding gamma lines. The solid curves in Fig. 1 were produced in this way and resulted in the line energies and relative yields given in Table I.

The discrepancy between points and curve below

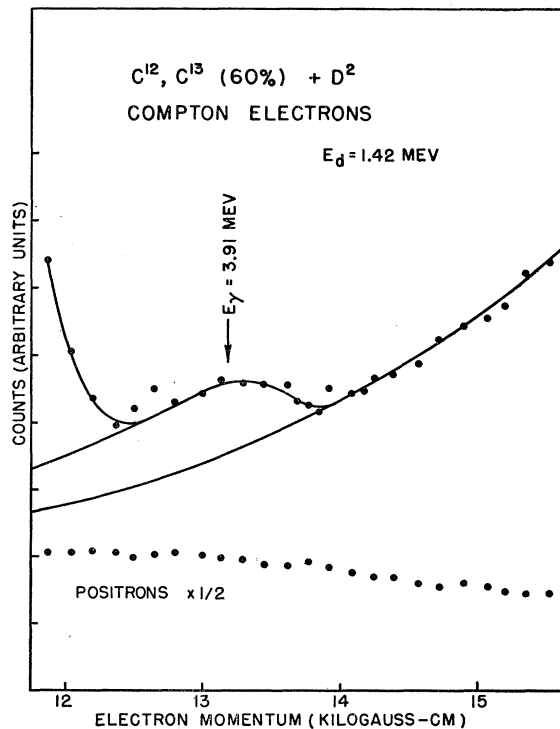


FIG. 2. Compton electrons from 20-mg/cm<sup>2</sup> C plus 125-mg/cm<sup>2</sup> Al converter.

<sup>9</sup> W. R. Mills, Jr., and R. J. Mackin, Jr., Phys. Rev. **95**, 1206 (1954).

16 kilogauss cm may reflect the presence of a gamma line at 4.5 Mev reported by the Rice Institute group<sup>10</sup> and assigned by them to the reaction  $C^{13}(d,\alpha)B^{11*}$ . It was not found possible to accentuate this line with a thicker converter.

Analysis of the data taken at  $E_d=1.9$  Mev, also shown in Fig. 1, involved the same procedure. The spectrum shows clearly the existence of a line at 6.73 Mev; the deviation from the calculated line shape below 22.5 kilogauss cm is taken to indicate the presence of a line whose energy is 6.45 Mev. The establishment of the 6.45-Mev line was dependent upon certainty that it was not a part of the 6.1-Mev line spectrum; the end point of the latter was carefully determined at  $E_d=1.4$  Mev.

Figure 2 shows a 3.91-Mev line (suspected earlier<sup>2</sup>) whose presence was confirmed by measurements with a 125-mg/cm<sup>2</sup> Al converter. The end point of the 3.4-Mev

TABLE I. Summary of results.

Gamma-ray energy (kev)	Maximum Doppler shift (kev)	Relative yield (Mev)	Assigned level (kev)	Reaction and reference
3910±50	19	0.14	N <sup>14</sup> 3945±15	N <sup>14</sup> (p,p')N <sup>14*a</sup>
4930±40	24	0.37	N <sup>14</sup> 4910±10	N <sup>14</sup> (p,p')N <sup>14*a</sup>
5130±30	25	0.34	N <sup>14</sup> 5104±10	N <sup>14</sup> (p,p')N <sup>14*a</sup>
5730±30	28	0.45	N <sup>14</sup> 5687±20	C <sup>13</sup> (d,nγ)N <sup>14</sup> (cascade) <sup>b</sup>
6120±25	30	1.00	C <sup>14</sup> 6095±15	C <sup>13</sup> (d,p)C <sup>14*c</sup>
6450±50	36		N <sup>14</sup> 6440±20	C <sup>13</sup> (p,γ)N <sup>14*d</sup>
6730±40	38		C <sup>14</sup> 6723±15	C <sup>13</sup> (d,p)C <sup>14*e</sup>
729±3	4		N <sup>14</sup> 5810±20	C <sup>13</sup> (p,γ)N <sup>14*d</sup>
811±3	4.5		C <sup>14</sup> 6894±15	C <sup>13</sup> (d,p)C <sup>14*e</sup>
869±3	5		O <sup>17</sup> 870.5±2	O <sup>16</sup> (d,p)O <sup>17*b</sup>

<sup>a</sup> Bockelman, Browne, Buechner, and Sperduto, Phys. Rev. **92**, 665 (1953).

<sup>b</sup> See reference 2.

<sup>c</sup> Sperduto, Holland, Van Patter, and Buechner, Phys. Rev. **80**, 769 (1950).

<sup>d</sup> See reference 3.

<sup>e</sup> Sperduto, Buechner, Bockelman, and Browne, Phys. Rev. **96**, 1316 (1954).

line immediately below was fixed by comparison with a theoretical line shape.

The photoelectron spectrum between 0.5 and 1.0 Mev (taken at  $E_d=2.6$  Mev) appears in Fig. 3. Three gamma lines at 729, 811, and 869 kev are indicated therein by their *K*-photopeaks. The expected *L*-peak of the 729-kev line is shown as a dashed curve. Correction for electron energy loss in the thorium converter<sup>4</sup> was checked by study of the Cs<sup>137</sup> gamma rays at  $661.60\pm 0.14$  kev<sup>11</sup> with the same converter. The 811-kev line was found to have an apparent threshold near  $E_d=1.9$  Mev.

All of the line energies are given in Table I, where also appear the relative yields (at  $E_d=1.45$  Mev) and the maximum possible Doppler shifts produced by center-of-mass motion. The actual line-shifts may differ

<sup>10</sup> Bent, Bonner, and Sippel, Phys. Rev. **95**, 649(A) (1954).

<sup>11</sup> Muller, Hoyt, Klein, and Du Mond, Phys. Rev. **88**, 775 (1952).

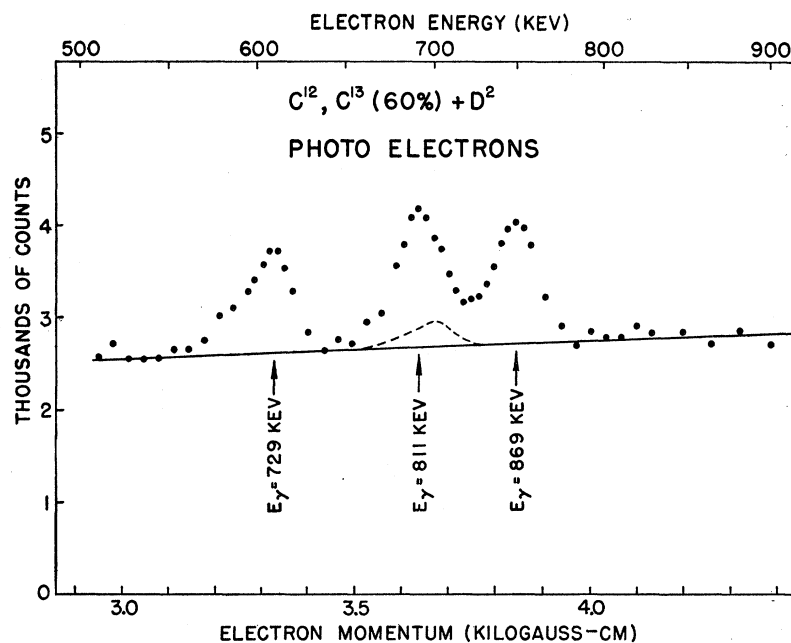


FIG. 3. Spectrum of photolines from 20.2-mg/cm<sup>2</sup> Th converter.

from these if the distribution of the motion of the emitting nuclei in the center-of-mass system is asymmetric about 90°, as in a stripping reaction, or if the mean life for emission is comparable with the nuclear stopping time. Other columns in the table suggest the most likely source of each line and give current values for the excitation energies of the assigned levels as reported by other investigators.

Measurements made at  $E_d = 2.7$  Mev revealed no further lines above 16 kilogauss-cm, although such lines are reported at  $E_d = 4$  Mev.<sup>10</sup>

#### IV. DISCUSSION

The 869-kev line at the end of Table I is attributed to the reaction  $O^{16}(dp)O^{17*}$ , presumably produced by bombardment of oxygen in the surface of the thorium converter. The line did not appear when a copper foil was interposed between target and converter. The line energy confirms the value given by Thomas and Lauritsen.<sup>2</sup>

The other assignments in the table are based on energy agreement with known levels together with threshold considerations. In particular, the yield of the 6.73-Mev line at  $E_d = 1.9$  Mev is believed to be too great to permit assignment to the 6.76-Mev level of  $B^{11}$ , since the bombarding energy is less than a hundred kev above threshold for the production of this level.

The assignment of the 3.91-Mev line to the 3.95-Mev level of  $N^{14}$  is consistent with the value given by Woodbury, *et al.*<sup>3</sup> for the branching ratio of that level. The assignment of the line as a cascade from the 6.23-Mev level to the 2.31-Mev level would appear to be excluded on the basis of the observation<sup>3</sup> that the former level decays only to the ground state.

The existence of a 5.82-Mev level in  $N^{14}$  is manifested only in the  $(p, \gamma)$  data<sup>3</sup> and in the assignment of the 729-kev line. For this assignment, a best level excitation value is  $5.104 + 0.729 = 5.833 \pm 0.013$  Mev. Our failure to observe a line of this energy sets its intensity as less than 0.2 that of the 5.73-Mev line and suggests a cascade branching ratio greater than earlier supposed.<sup>3</sup>

From observations on the gamma rays<sup>3</sup> in  $C^{13}(p, \gamma)N^{14}$  and elastically scattered protons<sup>12</sup> in  $C^{13}(p, p)C^{13}$ , it has been reasonably well established that the 8.06, 8.70, and 9.17-Mev levels in  $N^{14}$  have spins and parities  $1^-$ ,  $0^-$ , and  $2^-$ , respectively. Because of the large  $\gamma$ -width for the ground-state radiation from the 8.06-Mev level, and the absence of a transition to the 2.3-Mev level ( $J = 0^+$ ,  $T = 1$ ), it has been suggested<sup>13</sup> that the 8.06-Mev state has  $T = 1$ . Similarly, the 8.70-Mev level has been assigned  $T = 1$  on the basis of the large ground-state  $\gamma$ -ray width.<sup>14</sup>

Calculation shows that the analog states of the 6.1-, 6.7-, and 6.9-Mev levels in  $C^{14}$  should lie near 8.4, 9.0, and 9.2 Mev in  $N^{14}$ . It has been suggested, on the basis of the internal pair spectrum,<sup>2</sup> that the radiation from the 6.1-Mev level in  $C^{14}$  to the ground state is  $E1$  ( $M1$  and  $E2$  are not excluded), implying  $J = 1^-$  for this state. This assignment is borne out by the stripping analysis of the proton group from  $C^{13}(dp)C^{14*}$  (1). Thus, one might identify the 6.1-Mev level in  $C^{14}$  as the analog of the 8.06-Mev level in  $N^{14}$ . The absence of a ground-state transition from the 6.9-Mev state in  $C^{14}$  suggests  $J = 0$  for it, and a correspondence with the 8.70-Mev level in  $N^{14}$ . The apparent absence of levels between 8.06 and 8.70 Mev in  $N^{14}$  (except one

<sup>12</sup> E. A. Milne, *Phys. Rev.* **93**, 762 (1954).

<sup>13</sup> A. B. Clegg and D. H. Wilkinson, *Phil. Mag.* **44**, 1269 (1953).

<sup>14</sup> D. H. Wilkinson, *Phil. Mag.* **44**, 450 (1953).

with  $J=0$ ) then suggests that the analogs of the upper two  $C^{14}$  levels are inverted. A possible choice for the analog of the 6.73-Mev level is the level at 9.17 Mev whose large ground-state gamma-ray width is characteristic of electric dipole radiation, which would be inhibited if that level had  $T=0$ . Finally, it must be remembered that the analog level may thus far be

unobserved. If its reduced proton width is a large fraction of the so-called sum rule limit, the observed width may be of the order of Mev, and the level may be lost in the background.

The authors are indebted to C. C. Lauritsen and T. Lauritsen for enlightening discussions and valuable suggestions.

### Redetermination of the Disintegration Constant of $U^{238}$

A. F. KOVARIK AND N. I. ADAMS, JR.

*Sloane Physics Laboratory, Yale University, New Haven, Connecticut*

(Received December 30, 1954)

A recount of alpha particles from previously used specimens of natural uranium gives  $1.538 \times 10^{-10} \text{ yr}^{-1}$  for the disintegration constant of  $U^{238}$ .

IN 1932, the authors<sup>1</sup> made a new determination of the disintegration constant of uranium I ( $U^{238}$ ). Alpha particles from a thin layer of natural uranium in the form of the oxide  $U_3O_8$  were allowed to pass through a grid of known geometry into an ionization chamber. The electrical impulses there produced were amplified and applied to an electromechanical recording system. By this means the specific alpha activity of natural uranium was observed to be 1486 disintegrations per minute per milligram. In 1938, the apparatus was improved to give a better resolution of close counts, in connection with determination of thorium constants.<sup>2</sup> A subsequent recount made on some of the original uranium specimens gave the value 1501 disintegrations per minute per milligram.<sup>3</sup> Recently Fleming, Ghiorso, and Cunningham<sup>4</sup> have measured the specific alpha-activities of  $U^{234}$ ,  $U^{235}$ , and  $U^{238}$ . In their excellent paper these investigators discussed and evaluated previous work, not only on the isotopes in question, but on  $U^{238}$  as well. They also adopted "best values" for the various physical constants involved. Shortly before this, in connection with other work, the writers had constructed a new apparatus capable of much more rapid counting than the apparatus previously employed. It seemed of some interest, therefore, to make a fast recount on three of the original specimens, which were still intact. In all, approximately  $3 \times 10^6$  alpha particles were recorded, giving for the specific alpha-activity 1503 disintegrations per minute per milligram. This

agrees substantially with our 1941 value and with the recent "best value."<sup>4</sup>

Since the present accepted values of the isotopic atomic weights and of the other pertinent constants differ slightly from the values used in the earlier work, we must recalculate  $\lambda^{238}$ , the disintegration constant of  $U^{238}$ . The isotopic atomic weights are taken from Segre's book<sup>5</sup>; the other constants from Fleming *et al.*,<sup>4</sup> in whose paper references to the original sources may be found. It must be remembered that the atomic weights are based on the physical scale, on which the atomic weight of O<sup>16</sup> is 16.0000 and that of O (natural oxygen) is 16.0043. Using isotopic atomic weights and relative atomic abundances from the sources indicated above, we find that the atomic weight of U (natural uranium) is 238.103 and that the relative mass abundances of  $U^{238}$ ,  $U^{235}$ , and  $U^{234}$  are 0.99285, 0.00710, and 0.00005, respectively. Hence, since the alpha activities of the isotopes are in the ratios 1:0.046:1, the specific alpha activity of  $U^{238}$  is 739.9 disintegrations per minute per milligram. This corresponds to a number of  $U^{238}$  atoms equal to  $6.02501 \times 10^{20} / 238.125 = 2.53019 \times 10^{18}$ , since  $6.02501 \times 10^{23}$  is Avogadro's number (on the physical scale). Thus, finally,

$$\lambda^{238} = 1.538 \times 10^{-10} \text{ yr}^{-1},$$

with an estimated probable error of 0.2 percent. The corresponding half-life is  $4.507 \times 10^9$  years. It will be noted that these values differ from the earlier values<sup>1</sup> by only 0.4 percent, the effect of the increased value of the natural alpha activity being largely offset by the effect of the other small changes.

<sup>1</sup> A. F. Kovarik and N. I. Adams, Jr., *Phys. Rev.* **40**, 718 (1932).

<sup>2</sup> A. F. Kovarik and N. I. Adams, Jr., *Phys. Rev.* **54**, 413 (1938).

<sup>3</sup> A. F. Kovarik and N. I. Adams, Jr., *J. Appl. Phys.* **12**, 296 (1941).

<sup>4</sup> Fleming, Ghiorso, and Cunningham, *Phys. Rev.* **88**, 642 (1952).

<sup>5</sup> E. Segre, *Experimental Nuclear Physics* (McGraw-Hill Book Company, Inc., New York, 1953), Vol. 1, Part 5.