

value is less than the observed value.⁴ More exact calculations, using Coulomb wave functions for the incident and deflected proton, would be of considerable interest in order to see if such a calculation gives a smaller cross section.

Since the cross section is not expected to vary rapidly with nuclear charge or mass, we have not continued the

⁴ W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, New York, 1954), third edition, p. 267.

study of other elements. Likewise, we have not tried alpha bombardment because of the weak beams available, despite the probable reduction in background. The most promising extension would seem to be to higher proton energy. The predicted cross section is rising rapidly and in the region of 2 to 3 Mev might be expected to have a value some 10 times its value at 1.5 Mev. The pair-production effect then might be measurable.

Gamma Rays from $C^{13} + d$ and the Excited States of $C^{14}\dagger$

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 (Received October 16, 1957)

A study has been made of the γ rays produced by the bombardment of C^{13} by deuterons from the Brookhaven Van de Graaff accelerator with particular emphasis on the γ rays from the $C^{13}(d,p)C^{14}$ reaction. A search for C^{14} γ -emitting states and cascade transitions was made using a three-crystal pair spectrometer and other scintillation counters at bombarding energies up to 3.7 Mev. From coincidence measurements the assignment of the 0.81-Mev γ ray to the cascade transition between the C^{14} 6.89- and 6.09-Mev states was confirmed and a previously unreported 0.62-Mev γ ray was assigned to the cascade transition between the C^{14} 7.35- and 6.72-Mev states. Limits on the lifetimes of the C^{14} 6.89-

6.72-, and 6.1-Mev states were obtained from measurements of the Doppler shifts of the 0.81-, 6.72-, and 6.1-Mev γ rays. The lifetime limit for the 6.72-Mev state was combined with previous measurements to give a most probable assignment of $J^\pi=3^-$ for the 6.72-Mev state, while the lifetime limit for the C^{14} 6.89-Mev state showed that the 0.81-Mev transition is chiefly dipole. Measurements of the angular correlation of the 0.81- and 6.1-Mev γ rays were interpreted by expressing the angular correlation functions in terms of the relative populations of the magnetic substates of the C^{14} 6.89-Mev state. By this method an assignment of $J=0$ was obtained for the C^{14} 6.89-Mev state.

I. INTRODUCTION

IN spite of its anomalously long lifetime, the C^{14} nucleus is difficult to reach by any of the more usual nuclear reactions with the exception of $C^{13}(d,p)C^{14}$. In fact, all that is known about the excited states of C^{14} has been determined from observations on the proton groups and γ rays following the bombardment of C^{13} by deuterons. Proton groups have been observed^{1,2} corresponding to bound C^{14} states at 6.091, 6.589, 6.723, 6.894, and 7.346 Mev in addition to eight other states above the neutron binding energy (8.169 Mev). Angular distributions have been obtained²⁻⁴ for the proton groups corresponding to these bound states and have been analyzed by the stripping theory of Butler to determine the parities and a range of possible J values for the states. Gamma-rays of 0.81, 6.1, and 6.7 Mev observed⁵ following the bombardment of C^{13} enriched targets by deuterons have been assigned to the transi-

tion between the C^{14} 6.89- and 6.09-Mev states and to the ground-state transitions of the C^{14} 6.09- and 6.72-Mev states. These γ transitions have not been studied in great detail. The present investigation was undertaken, then, to gain additional information concerning the excited states of C^{14} from a study of the γ rays following the bombardment of C^{13} by deuterons.

Using relatively high efficiency techniques, a search was made for γ rays originating from C^{14} states which might have escaped previous detection. Coincidence measurements were used to assign cascade γ rays—one of which was observed in the coincidence measurements only. Doppler-shift measurements were made on several γ rays and the lifetime limits obtained from these measurements gave information concerning the γ -emitting states involved. In one case, angular correlation measurements were used to give a definite spin assignment to a C^{14} state.

II. GAMMA-RAY SPECTRUM FROM $C+d$

All the observations on the γ rays following the bombardment of carbon by deuterons reported herein were carried out by using a 100- $\mu\text{g}/\text{cm}^2$ target of C^{13} , 70% enriched, cracked onto a 0.004-in. gold backing. The deuterons were accelerated by the Brookhaven 4-Mev research Van de Graaff accelerator. Detection of the γ rays was accomplished using two 2 by 2 in.

[†] Work performed under the auspices of the U. S. Atomic Energy Commission.

* Guest Physicist at Brookhaven National Laboratory on a fellowship of Deutsche Forschungsgemeinschaft.

¹ Sperduto, Buechner, Bockelman, and Browne, *Phys. Rev.* **96**, 1316 (1954).

² McGruer, Warburton, and Bender, *Phys. Rev.* **100**, 235 (1955).

³ R. E. Benenson, *Phys. Rev.* **90**, 420 (1953).

⁴ F. A. El Bedewi, *Proc. Phys. Soc. (London)* **A69**, 221 (1956).

⁵ F. Ajzenberg and T. Lauritsen, *Revs. Modern Phys.* **27**, 77 (1955).

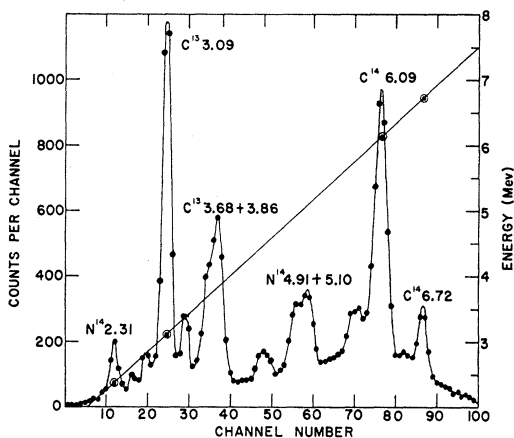


FIG. 1. Three-crystal pair spectrum of the γ rays from the $C^{12,13}+d$ reactions at $E_d=2.9$ Mev. Several of the more prominent pair peaks, all corresponding to ground-state transitions, have been labeled by the residual nucleus and the energy level to which they belong.

NaI(Tl) crystals and one 2 by $1\frac{1}{2}$ in. NaI(Tl) crystal mounted on Dumont type 6292 photomultipliers and one 3 by 3 in. NaI(Tl) crystal mounted on a Dumont type 6364 photomultiplier. The γ -ray spectra were displayed on a RIDL 100-channel pulse-height analyzer.

A. Three-Crystal Pair Spectrometer Studies

As the first step in the study of C^{14} γ -ray transitions, a survey of the high-energy γ rays following deuteron bombardment of the 70% C^{13} target was made using a three-crystal pair spectrometer. The design of the spectrometer and the associated circuitry was essentially the same as described by Alburger and Toppel,⁶ while the resolution and efficiency of a similar spectrometer

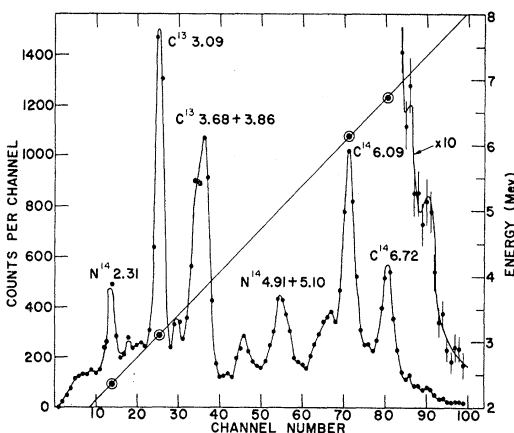


FIG. 2. Three-crystal pair spectrum of the γ rays from the $C^{12,13}+d$ reactions at $E_d=3.7$ Mev. The points above channel 84 have been multiplied by 10. Several of the more prominent pair peaks, all corresponding to ground-state transitions, have been labeled by the residual nucleus and the energy level to which they belong.

⁶ D. E. Alburger and B. J. Toppel, Phys. Rev. **100**, 1357 (1955).

has been described by Bent and Kruse.⁷ Several spectra were taken at 0° to the beam at deuteron bombarding energies of 2.9 and 3.7 Mev. The second energy represents a compromise between reliable Van de Graaff operation and the desire for excitation of as high-lying nuclear levels as possible. Two pair spectra taken with an integrated charge of 3500 microcoulombs are shown in Figs. 1 and 2. The γ rays observed at the two energies are listed in Table I. The reactions believed responsible for these γ rays are

$$C^{12}(d,n)N^{13}, \quad Q = -0.28 \text{ Mev};$$

$$C^{12}(d,p)C^{13}, \quad Q = 2.72 \text{ Mev};$$

$$C^{13}(d,n)N^{14}, \quad Q = 5.31 \text{ Mev};$$

$$C^{13}(d,p)C^{14}, \quad Q = 5.94 \text{ Mev};$$

$$C^{13}(d,\alpha)B^{11}, \quad Q = 5.16 \text{ Mev}.$$

All the γ rays listed except the one at 2.61 Mev have been previously reported, and the assignments, except for those noted, follow previous work.

A definite assignment for the 2.61-Mev γ ray cannot be made. It has been tentatively assigned to a N^{14} cascade or to an unknown contaminant. Two possibilities are a transition between the N^{14} 4.91- and 2.31-Mev states⁸ and a transition between the N^{14} 7.72- and 5.10-Mev states. The 4.46-Mev γ ray is assigned to the ground-state transition from the B^{11} 4.46-Mev level.

TABLE I. Assignments of the γ rays from bombardment of a $100\text{-}\mu\text{g}/\text{cm}^2$ 70% C^{13} target with 2.9- and 3.7-Mev deuterons.

Energy (Mev)		Assignment
$E_d=2.9$ Mev	$E_d=3.7$ Mev	
(2.31) ^a	(2.31)	N^{14} (2.31 \rightarrow gnd)
2.62	2.60	N^{14} (cascade) or contaminant ^b
2.81	2.81	N^{14} (5.10 \rightarrow 2.31) ^c
(3.09)	(3.09)	C^{13} (3.09 \rightarrow gnd)
3.40	3.38	N^{14} (5.69 \rightarrow 2.31)
3.75	3.75	C^{13} (3.68 and 3.86 unresolved \rightarrow gnd)
4.46	4.45	B^{11} (4.46 \rightarrow gnd) ^d
5.03	5.05	N^{14} (5.10 and 4.91 unresolved \rightarrow gnd)
5.75	5.75	N^{14} (5.69 and 5.83 (?) unresolved \rightarrow gnd)
(6.09)	(6.09)	C^{14} (6.09 \rightarrow gnd)
6.47 (?)	6.46 (?)	N^{14} (6.46 \rightarrow gnd)
(6.72)	(6.72)	C^{14} (6.72 \rightarrow gnd)
	7.12 (?)	N^{14} (7.03 \rightarrow gnd)
	7.35	C^{14} (7.35 \rightarrow gnd) and/or N^{14} (7.40 \rightarrow gnd) ^e

^a The γ rays in parentheses were used for energy calibration. For these calibration peaks the Doppler-shift correction was estimated from previous γ -ray energy measurements and from the Doppler-shift measurements reported in the present paper.

^b Previously unobserved.

^c Observed from the $C^{13}(p,\gamma)N^{14}$ reaction by Woodbury, Day, and Tollestrup, Phys. Rev. **92**, 1199 (1953). Previously unobserved from $C^{13}(d,n)N^{14}$.

^d See reference 11. Note added in proof.—Ranken, Bonner, McCrary, and Rabson, Phys. Rev. **109**, 917 (1958) have recently performed experiments which definitely establish this assignment.

^e Assigned to the N^{14} 7.40-Mev level (see reference 11) before the C^{14} 7.35-Mev level was reported (see reference 2).

⁷ R. D. Bent and T. H. Kruse, Phys. Rev. **108**, 802 (1957).

⁸ The N^{14} 2.31-Mev state is 0^+ and the N^{14} 4.91-Mev state has usually been considered to be 0^- . However, Broude, Green, Singh, and Willmott [Phil. Mag. **2**, 1006 (1957)] have recently reported a γ transition between the N^{14} 8.70-Mev 0^- state and the N^{14} 4.91-Mev state indicating that the 4.91-Mev state does not have zero spin.

The absence of the γ ray corresponding to the ground-state transition from the 2.14-Mev first-excited state of B^{11} is not surprising since the relative efficiency of the pair spectrometer for 4.46- and 2.14-Mev γ rays is 3.7:1 and it is estimated that a 2.14-Mev γ ray would not have been observed if it had an intensity equal to or less than that of the 4.46-Mev γ ray. Moreover, the relative populations of the B^{11} 4.46- and 2.14-Mev levels following bombardment of C^{13} by 14.8-Mev deuterons has been measured⁹ to be $\approx 3.5:1$ and it is quite possible that the 4.46-Mev level is populated more strongly than the 2.14-Mev level at deuteron energies below 3 Mev also.

The 7.35-Mev γ ray is assigned to either C^{14} or N^{14} or both. Of these two assignments, C^{14} is the more likely since C^{14} has a level at 7.346 ± 0.02 Mev² while N^{14} has a level at 7.40 ± 0.02 Mev,¹⁰ and thus the energy of the 7.35-Mev γ ray (reported as 7.34 ± 0.04 Mev by Bent, Bonner, and Sippel¹¹ at an average deuteron energy of 3.7 Mev) is in better agreement with the C^{14} level, especially if it is short-lived enough to have a Doppler shift (maximum shift ~ 40 kev).

B. Coincidence Measurements

The γ -ray spectrum of $C^{13} + d$ in the energy region below that obtainable by the three-crystal pair spectrometer was obtained with a single 2-in. NaI(Tl) crystal at a number of different bombarding energies and at various angles to the deuteron beam.

The γ rays observed are listed in Table II. The assignments of these γ rays—which have been observed earlier—follow previous work.⁵ The singles spectrum between 0.55 and 0.95 Mev obtained at $E_d = 2.9$ Mev is shown in Fig. 3. The 0.73-, 0.81-, and 0.87-Mev γ rays were previously observed by Mackin, Mills, and Mims¹² who made the assignments given in Table II for these three γ rays on the basis of threshold considerations and precise energy measurements. In the present experiment the full-energy-loss peaks corresponding to these γ rays were not completely resolved in the singles spectrum; however, by subtracting the background it was possible to separate three peaks of the proper shape at the expected energy positions. It

TABLE II. Low-energy γ rays from bombardment of a 100- $\mu\text{g}/\text{cm}^2$ 70% C^{13} target with 2- to 3.7-Mev deuterons.

Energy (Mev)	Assignment
0.17	C^{13} (3.86→3.68)
0.51	$C^{12}(d,n)N^{13}(\beta^+)C^{13}$
0.73	N^{14} (5.83→5.10)
0.81	C^{14} (6.89→6.09)
0.87	O^{17} (0.87→gnd)
1.64	N^{14} (3.95→2.31)

⁹ McGruer, Warburton, and Bender (unpublished).
¹⁰ E. J. Burge and D. J. Prowse, Phil. Mag. 1, 912 (1956).
¹¹ Bent, Bonner, and Sippel, Phys. Rev. 98, 1237 (1955).
¹² Mackin, Mills, and Mims, Phys. Rev. 98, 43 (1955).

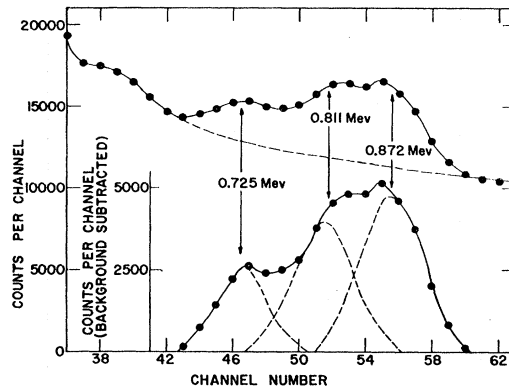


Fig. 3. Upper curve: Single-crystal spectrum of the γ rays from the $C^{12,13} + d$ reactions for E_γ between 0.55 and 0.95 Mev. The spectrum was taken with $E_d = 2.9$ Mev and at 0° to the beam. Lower curve: The singles spectrum decomposed into three peaks of the indicated energies.

was established that the 0.87-Mev γ ray was due to a target contaminant by observing that it persisted when the beam was striking the gold backing of the target so that the γ rays from $C + d$ had completely disappeared. The yield of the three γ -ray lines was roughly checked as a function of bombarding deuteron energy from 2 to 3.7 Mev. The yields of the 0.73- and 0.81-Mev γ rays were observed to increase uniformly with increasing deuteron energy while the yield of the 0.87-Mev γ ray had a slight minimum at $E_d = 2.9$ Mev. For this reason later studies of the 0.81-Mev γ ray were made at $E_d = 2.9$ Mev in order to keep the error due to the overlapping 0.87-Mev line at a minimum.

To confirm the assignments of the 0.73- and 0.81-Mev γ rays and to search for additional low-energy γ rays too weak to appear in the singles spectrum, coincidence experiments were performed with the 3-in. crystal placed at 0° to the beam and one 2-in. crystal placed at 90° to the beam. Both crystals were approximately 0.5 in. from the target and the average beam intensity was 0.02 μa . The singles spectrum obtained in the 3-in. crystal at $E_d = 3.7$ Mev is shown in Fig. 4.

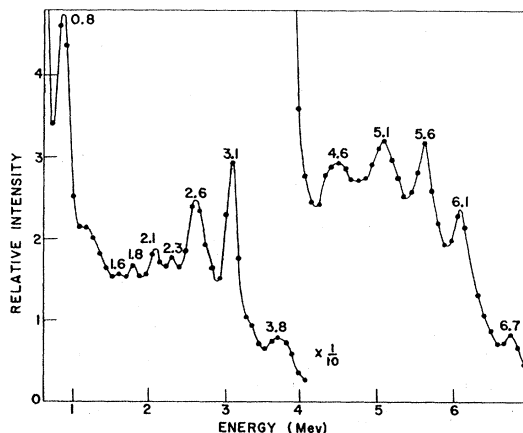


Fig. 4. Single-crystal spectrum of the γ rays from the $C^{12,13} + d$ reactions at $E_d = 3.7$ Mev.

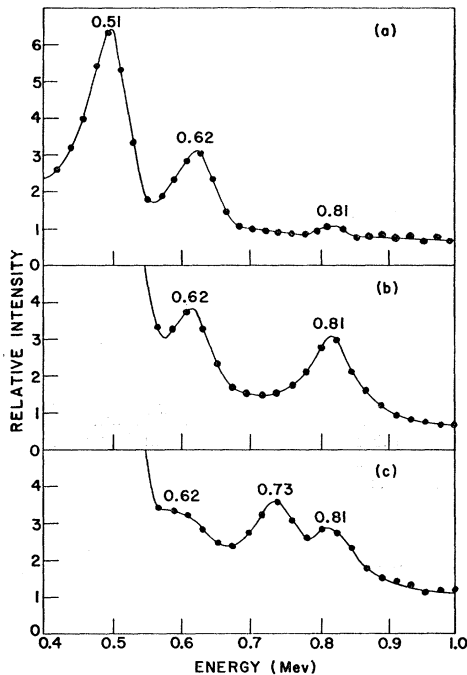


FIG. 5. Coincidence spectrum of the γ rays from the $C^{12,13}+d$ reactions at $E_d=3.7$ Mev. (a) Spectrum in coincidence with 6.4- to 7.0-Mev radiation, (b) spectrum in coincidence with 5.8- to 6.4-Mev radiation, (c) spectrum in coincidence with 4.3- to 4.9-Mev radiation.

These pulses went to a single-channel pulse-height analyzer which could be adjusted to count any portion of the total γ -ray spectrum. The analyzer window was adjusted by self-gating the spectrum from the 3-in. crystal with the pulses from the analyzer window and displaying the "self-gated" pulses with the 100-channel analyzer.

Initially, the low-energy spectrum displayed by the 2-in. crystal in coincidence with the 6.7-Mev peak of Fig. 4 was recorded at $E_d=3.7$ Mev with an analyzer window width of 0.5 Mev. This spectrum is shown in Fig. 5(a). The spectrum recorded by the 3-in. crystal for a single γ ray with an energy within the range of present interest (5-8 Mev) consists of one-quantum-escape and two-quantum-escape peaks of approximately equal intensity and a relatively weaker full-energy-loss peak, while the spectrum is approximately constant from zero energy to the low-energy side of the two-quantum-escape peak. Thus the spectrum of Fig. 5(a) contains possible contributions from any γ rays with energies higher than 6.7 Mev and in particular from the 7.1- and 7.4-Mev γ rays (see Fig. 2). In Fig. 5(b) is shown the spectrum in coincidence with the 6.1-Mev peak (which consists mainly of the 6.1-Mev γ -ray full-energy-loss peak) and in Fig. 5(c) is shown the spectra in coincidence with the 4.6-Mev peak (which consists mainly of contributions from the one-quantum-escape peaks of the 4.9- and 5.1-Mev γ rays and the two-quantum-escape peak of the 5.7-Mev γ ray).

From the variation of the intensity of the 0.73- and 0.81-Mev γ rays with window setting shown in Fig. 5, it was concluded that the 0.73-Mev γ ray was in coincidence with a γ ray with energy between 4.0 and 5.5 Mev while the 0.81-Mev γ ray was in coincidence with a γ ray with energy between 5.5 and 6.5 Mev. By narrowing the window and taking several spectra in coincidence with various portions of the spectra between 4.0 and 6.5 Mev, it was established that the 0.73-Mev γ ray was in coincidence with the N^{14} 5.1-Mev γ ray while the 0.81-Mev γ ray was in coincidence with the C^{14} 6.1-Mev γ ray, thus confirming the previous assignments¹² of these γ rays. Energy measurements were obtained for the 0.73-Mev and 0.81-Mev γ rays from the coincidence spectra taken at $E_d=2.9$ Mev. The coincidence spectra were calibrated with the Cs^{137} 0.662-Mev γ ray and the Co^{60} 1.172- and 1.332-Mev γ rays. These measurements gave 0.731 ± 0.008 and 0.813 ± 0.008 Mev in good agreement with the values of 0.729 ± 0.003 and 0.811 ± 0.003 Mev obtained by Mackin *et al.*¹³ at $E_d=2.6$ Mev.

In taking the various coincidence spectra described above, a search was made for γ rays with energies less than 2 Mev which were in coincidence with the γ rays between 4.5 and 7.5 Mev shown in Fig. 2. The γ ray at 0.62 Mev shown in Fig. 5 was the only one observed in these measurements which had not been seen in the singles spectra. An upper limit of 0.25 of the intensity of the 0.62-Mev γ ray is set for any other γ ray with energy between 0.7 and 2.0 Mev in coincidence with a γ ray with an energy between 4.5 and 7.5 Mev. In Figs. 5(b) and 5(c) the spectra are shown rising to the 0.51-Mev full-energy-loss peak which is shown in Fig. 5(a). This annihilation radiation arose partly from random coincidences, but for the most part from coincidences in which an high-energy γ ray was stopped in the 3-in. crystal by pair production with the subsequent capture of one of the associated annihilation radiations in the 2-in. display crystal. The intensity of this radiation was reduced greatly by placing a lead absorber between the crystals; however, it was still of sufficient intensity to obscure any γ rays with energies between 0.4 and 0.6 Mev, and to give considerable background below 0.4 Mev. An upper limit of twice the intensity of the 0.62-Mev γ ray is set for any γ ray with energy less than 0.4 Mev and in coincidence with γ rays between 4.5 and 7.5 Mev. Energy measurements of the 0.62-Mev γ ray were made from the coincidence spectra at $E_d=3.7$ Mev using Cs^{137} , Co^{60} , and the annihilation radiation for calibration. These measurements gave 0.621 ± 0.007 Mev. This energy is in agreement with three possible cascades, N^{14} 7.03 \rightarrow 6.46 Mev, C^{14} 6.72 \rightarrow 6.09 Mev, and C^{14} 7.34 \rightarrow 6.72 Mev. Of these, the C^{14} 6.72 \rightarrow 6.09-Mev cascade is eliminated by the coincidence measurements shown in Fig. 5 where it is seen from the relative yields of Figs. 5(a) and 5(b) that the 0.62-Mev γ ray is in coincidence with a γ ray of higher energy than 6.1 Mev. The 0.62-Mev γ ray was

not observed in a later coincidence measurement with the window centered at 7.4 Mev, thus showing that the 0.62-Mev γ ray is in coincidence with a γ ray with energy between 6.1 and 7.1 Mev. Additional coincidence spectra were taken with a window width of 0.6 Mev, first with the window centered at 6.4-Mev and second with the window centered at 7.0 Mev. The yield of the 0.62-Mev γ ray for these two measurements was the same within the estimated error, indicating strongly that the 0.62-Mev γ ray is in coincidence with the 6.7-Mev γ ray and not with the 6.46-Mev γ ray. As a further check the procedure was reversed. The window was centered at 0.62 Mev and the spectrum in coincidence with the pulses from the single-channel pulse-height analyzer was recorded. This coincidence spectrum is shown in the lower curve of Fig. 6. The singles spectrum shown in the upper curve was used for energy calibration. The coincidence spectrum of Fig. 6 contains contributions from the 5.1- and 6.1-Mev γ rays due to coincidences with the Compton distributions of the 0.73- and 0.81-Mev γ rays. It also contains contributions from quantum-escape peaks due to overlap of the analyzer window with the tail of the 0.51-Mev full-energy-loss peak. However, the appearance of the 6.7-Mev full-energy-loss peak confirms the assignment of the 0.62-Mev γ ray to the C^{14} 7.35 \rightarrow 6.72 Mev transition. Using the most accurate energy determination for the C^{14} 6.7-Mev level of 6.723 ± 0.015 Mev,¹ this assignment gives a best value for the level excitation of $6.723 + 0.621 = 7.344 \pm 0.017$ Mev, in excellent agreement with the value of 7.346 ± 0.02 Mev obtained by McGruer *et al.*²

The relative yield of the 0.62-Mev γ ray was determined from coincidence measurements to be approximately 12:4:1 at deuteron energies of 3.7, 2.9, and 2.6 Mev, respectively. The relative yield of the 7.35-Mev γ ray, which was not observed at $E_d = 2.9$ Mev, was determined from the three-crystal pair-spectrometer measurement to be $>2:1$ at deuteron energies of 3.7 and 2.9, respectively. These measurements are consistent with the assignment of the 7.35-Mev γ ray to C^{14} but do not rule out the assignment to N^{14} .

Since neither the 7.35-Mev γ ray nor the 0.62-Mev γ ray was observed in the singles spectrum, the branching ratio of the decay of the 7.35-Mev state was not measured. In any case, only a limit for the branching ratio could have been obtained since the 7.35-Mev γ ray could have originated, wholly or in part, from the N^{14} 7.4-Mev state. However, a rough comparison of the various measurements reported herein does indicate that the ratio of the C^{14} 7.35-Mev γ ray to the C^{14} 0.62-Mev γ ray is of the order of unity or less.

Both the 6.72- and 7.35-Mev states of C^{14} are known from the stripping analysis³ of the $C^{13}(d,p)C^{14}$ reaction to be $J^\pi = 1^-, 2^-,$ or 3^- . Therefore the 0.62-Mev transition has the lowest possible multiplicities $M1$ and/or $E2$, while—for an assumed assignment of 1^- to the 7.35-Mev state—the 7.35-Mev $1^- \rightarrow 0^+$ ground-

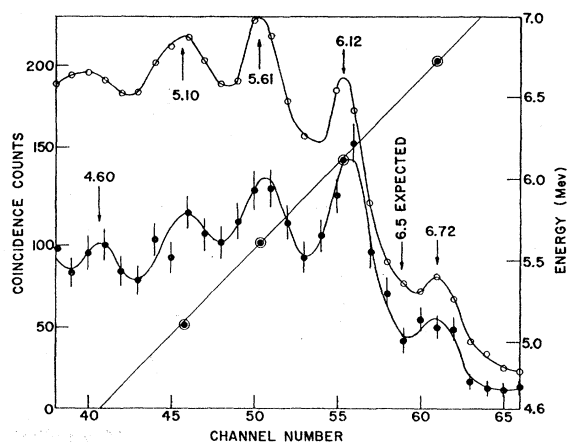


Fig. 6. Upper curve: Single-crystal spectrum of the γ rays from the $C^{13} + d$ reactions at $E_d = 3.7$ Mev. Lower curve: spectrum in coincidence with 0.57- to 0.67-Mev radiation.

state transition would be $E1$. The Weisskopf estimate presents mean lives of 1.3×10^{-13} and 1.6×10^{-9} sec for 0.62-Mev $M1$ and $E2$ transitions, respectively, and 2.6×10^{-18} sec for a 7.35-Mev $E1$ transition. The available evidence¹³ for radiative transitions in light nuclei indicates that it would be possible, but highly unlikely, for the $E1$ transition to be retarded and/or the $M1$ transition to be enhanced sufficiently for the 0.62-Mev transition to compete with the 7.35-Mev transition to the extent observed if the 7.35-Mev state were 1^- . A 0.62-Mev $E2$ transition would certainly not be expected to compete with a 7.35-Mev $E1$ transition. Therefore, it is concluded that the 7.35-Mev state is most likely 2^- or 3^- and not 1^- .

III. DOPPLER SHIFT MEASUREMENTS

The method of measuring γ -transition lifetimes by observations of the Doppler shift due to the center-of-mass motion of the γ -emitting nucleus has been well-described in the literature.^{14,15} This method can be used to measure γ -transition lifetimes in the range $5 \times 10^{-12} \gtrsim \tau \gtrsim 10^{-14}$ sec, but usually requires accurate measurements and stringent precautions against systematic errors. In many cases it is relatively easy, however, to set a lifetime limit by observing the existence or absence of a Doppler shift. Thus a relatively rough measurement of the Doppler shift of a γ ray emitted following a nuclear reaction can often give information useful in describing the nuclear states involved.

A. C^{14} 0.81-Mev Gamma Ray

Doppler Shift Measurement

The Doppler shift of the 0.81-Mev γ ray was measured at $E_d = 2.9$ Mev by using a 2-in. right cylinder of

¹³ D. H. Wilkinson, *Phil. Mag.* **1**, 127 (1956).

¹⁴ R. G. Thomas and T. Lauritsen, *Phys. Rev.* **88**, 969 (1952).

¹⁵ Devons, Manning, and Bunbury, *Proc. Phys. Soc. (London)* **A68**, 18 (1955).

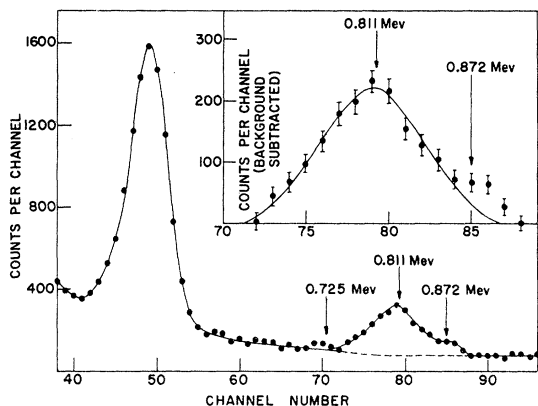


FIG. 7. Spectra in coincidence with 5.3- to 6.3-Mev radiation. The peak at channel 49 is the full-energy-loss peak of annihilation radiation. The insert shows the assumed shape and position of the 0.81-Mev γ -ray full-energy-loss peak.

NaI(Tl) mounted on a Dumont type 6292 photomultiplier which was picked for its non-rate-dependent gain. The crystal was irradiated with its center 13 cm from the target, at 0° to the beam and at a backward angle of 120° . In order to obtain a well-resolved full-energy-loss peak from the 0.81-Mev γ ray, the spectra from the 2-in. crystal were recorded in coincidence with the 6.1-Mev γ ray. The 3-in. crystal was placed at 90° and 2 cm from the target. The pulses from the 3-in. crystal went to the single-channel pulse-height analyzer which was set to pass the full-energy-loss and one-quantum-escape peaks of the 6.1-Mev γ ray. A typical spectrum obtained at 0° to the beam is shown in Fig. 7. The annihilation radiation peak and the unresolved 0.73- and 0.87-Mev peaks shown in Fig. 7 are due to chance coincidences. The 0.51-Mev peak was used to monitor the gain and to correct for shift of the gain with time and position. The counting rate used represented a compromise between the uncertainties introduced by time-dependent drifts of the gain and by chance coincidences.¹⁶ Six spectra were taken alternately at 0° and 120° . For each spectrum the pulse heights of the 0.51-Mev and 0.81-Mev γ rays were determined by plotting the peaks on large-scale graph paper. The pulse heights of the 0.81-Mev peaks were then corrected for gain shifts using the 0.51-Mev peaks for reference. Four determinations of the Doppler shift were obtained by comparing the corrected pulse height of each measurement (except the first and last) with the average of the two flanking measurements. The measured shift was $1.2 \pm 0.3\%$, where the uncertainty is the standard deviation.

¹⁶ In the course of this experiment a measurement was made of the Doppler shift of the O^{17} 0.87-Mev peak. No Doppler shift was detected within the accuracy of the measurement, in agreement with the measurement of $(2.5 \pm 1) \times 10^{-10}$ sec obtained by J. Thirion and V. L. Telegdi [Phys. Rev. **92**, 1253 (1953)] for the mean life of the O^{17} 0.87-Mev state. The contamination of the 0.81-Mev peak by the 0.87-Mev peak—which is due to chance coincidences—would be expected, then, to cause a small systematic decrease in the measured Doppler shift of the 0.81-Mev peak.

The Doppler shift calculated from the kinematics of the reaction—assuming a lifetime short compared to the stopping time of the C^{14*} nuclei and a center-of-mass distribution of the protons symmetric about 90° —was 1.1%.¹⁷ Therefore, it is concluded that the mean life of the C^{14} 6.89-Mev state is short compared to 3×10^{-13} sec, this time being a conservative upper limit for the mean stopping time of the C^{14*} nucleus for the target material and kinematics involved.

Discussion

The 6.09-Mev level is the only C^{14} excited state with known spin and parity. The stripping analysis²⁻⁴ of the $C^{13}(d,p)C^{14}$ reaction gives 0^- or 1^- , while the presence of the γ transition to the 0^+ ground state rules out a spin assignment of zero; therefore, the C^{14} 6.09-Mev state is assigned 1^- . The stripping analysis of the C^{14} 6.89-Mev state gives $J^\pi = 0^\pm, 1^\pm$, or 2^+ ,¹⁸ so that the 0.81-Mev γ transition between the 6.89- and 6.09-Mev states has the possible multipolarities $E1, M1, E2, M2$, or $E3$. We can use the limit on the lifetime to show that the transition is predominantly dipole in character. The Weisskopf estimate gives 1.9×10^{-15} and 5.9×10^{-14} sec for the mean lives of assumed $E1$ and $M1$ transitions, respectively. Either is consistent with the upper limit on the mean life of 3×10^{-13} sec. The Weisskopf estimate gives 4.7×10^{-10} sec for the mean life of an assumed $E2$ transition. The available evidence¹³ for light nuclei indicates that the Weisskopf lifetime estimate for $E2$ transitions is a rather good lower limit, at least within a factor of Z^2 (i.e., collective contributions of all the protons). Both $M2$ and $E3$ transitions—for which the Weisskopf estimates are 1.4×10^{-8} and 1.7×10^{-4} sec, respectively—are clearly inconsistent with the lifetime limit and it is concluded that the 0.81-Mev transition is predominantly dipole. For the purpose of later discussion we adopt the conservative limit $\delta^2 \leq 0.1$, where δ^2 is the ratio of the intensity of quadrupole radiation to that of dipole radiation.

B. The 6.1-Mev and 6.72-Mev Gamma Rays

Doppler Shift Measurement

It was observed by Bent *et al.*¹¹ that energy measurements⁵ of the C^{14} 6.72-Mev γ ray were in better agree-

¹⁷ For a lifetime short compared to the stopping time, the Doppler shift is smallest when the protons are emitted at 0° , and largest when they are emitted at 180° . For stripping reactions the protons are usually peaked in the forward direction and therefore it seems likely that the Doppler shift for the reaction considered was less than 1.1%. A Doppler shift of 0.9% (one standard deviation below the measured value) would correspond to a center-of-mass angle between the mean direction of emission of the protons and the beam of approximately 70° .

¹⁸ McGruer, Warburton, and Bender [Phys. Rev. **100**, 235 (1955)] reported that the angular distribution of the $C^{13}(d,p)C^{14}$ reaction to the 6.89-Mev state corresponded to $l_n = 1$ (in which case $J^\pi = 0^+, 1^+$, or 2^+). Preliminary results by one of the present authors (E.K.W.) indicate that the angular distribution obtained by McGruer *et al.* is fitted by $l_n = 0$ (in which case $J^\pi = 0^-$ or 1^-) at least as well as by $l_n = 1$. In the present investigation we shall assume that the stripping analysis gives $J^\pi = 0^\pm, 1^\pm$, or 2^+ .

ment with the measured energy position¹ of the C^{14} 6.72-Mev state if no Doppler-shift correction was applied to the γ -ray energy measurements. Although this comparison is not definite because the expected Doppler shift is comparable to the uncertainties of the measurements, it could indicate that the 6.72-Mev state is relatively long-lived ($\tau > 3 \times 10^{-13}$ sec). In order to confirm this possibility, a measurement of the Doppler shift of the 6.72-Mev γ ray was undertaken.

From Fig. 4 it can be seen that the 6.72-Mev full-energy-loss peak is relatively weak and is rather poorly resolved. Thus it would be expected that a Doppler-shift measurement of the 6.72-Mev γ ray obtained from measurements on the full-energy-loss peak would be influenced by neighboring γ -ray peaks, in particular the 6.1-Mev full-energy-loss peak. For this reason the Doppler shift of the 6.72-Mev γ ray was obtained by two methods. First, the Doppler shifts of the 6.1-Mev and 6.72-Mev γ rays were measured from singles spectrum and second, the relative shift of the two γ rays was measured using the three-crystal pair spectrometer. Both measurements were made with $E_d = 2.9$ Mev.

Singles Spectrum Measurements.—The γ rays were detected with the 3-in. crystal. The crystal was irradiated with its center 20 cm from the target, at 0° to the deuteron beam and at a backward angle of 120° . The γ -ray spectra were recorded using the 100-channel analyzer with a bias of 400 channels. The full-energy-loss-peaks of the 6.1-Mev and 6.72-Mev γ rays were investigated separately. A typical spectrum for the 6.72-Mev full-energy-loss peak is shown in Fig. 8. To obtain the pulse height of this peak the background was subtracted and the centroid of the remaining peak was found numerically. This method was possible because of the large number of experimental points in each peak. For the 6.1-Mev peak this method could not be used since the peak contained contributions from the quantum-escape peaks of the 6.72-Mev γ ray. Instead, the pulse height was located by drawing the peak on large-scale graph paper. The gain of the system was monitored and corrections were made for shifting of the gain with time and position in a manner similar to that used by Wilkinson¹⁹ in his measurement of the Doppler shift of the first excited state of B^{11} . For both peaks, eight spectra were taken alternately at 0° and 120° to the beam. From these data the Doppler shifts of the 6.1-Mev and 6.7-Mev γ rays were determined to be 35 ± 5 and 3 ± 6 kev, which compare to Doppler shifts between 0° and 90° of 23 ± 3 and 2 ± 4 kev, respectively. The relative shift of the γ lines is then 21 ± 5 kev. The uncertainties are the standard deviations and do not include the possible error due to overlapping of the peaks.

Three-Crystal Pair Spectrometer Measurement.—The pair spectra were taken in the same manner as described in Sec. II and were similar to the spectrum shown in

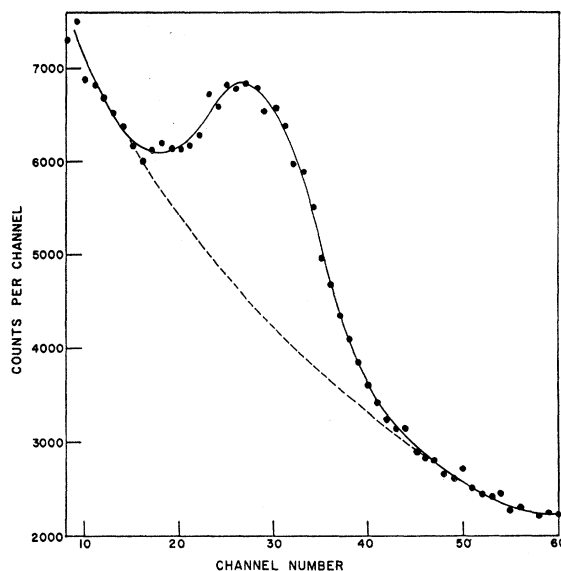


FIG. 8. The full-energy-loss peak of the C^{14} 6.72-Mev γ ray taken at $E_d = 2.9$ Mev with a bias of 400 channels.

Fig. 1. Four spectra taken at 90° to the beam were alternated with the four taken at 0° to the beam. For each spectrum in the sequence of eight, the pulse heights of the 6.1-Mev and 6.7-Mev two-escape peaks were determined by drawing the peaks on large-scale graph paper. By using this method it was believed possible to locate a peak to within one-tenth channel (≈ 6 kev); however, the uncertainty quoted for the final result did not depend on this but on the standard deviation of the measurement. The measured relative shift of the two peaks was 29 ± 6 kev. This measurement agrees within the experimental uncertainties with the relative shift of 21 ± 5 kev obtained from the singles spectra and it is concluded that the error due to overlapping of the peaks can be neglected.

Discussion

For the 6.1-Mev γ ray the calculated shift between 0° and 120° , including a correction for the finite counter size, and assuming an isotropic distribution of the C^{14*} nuclei in the center-of-mass system is 68 kev. Since the 6.1-Mev γ -ray transition is known to be $E1$ and the Weisskopf estimate presents 4.6×10^{-18} sec for the mean life of this transition, it can be assumed that the mean life is short compared to 3×10^{-13} sec. Therefore, the difference in the calculated and observed Doppler shift is assumed to represent a nonisotropic distribution for the C^{14*} nuclei in the center-of-mass system.²⁰ The measured Doppler shift of 35 ± 5 kev corresponds then to a nonisotropic distribution in which the mean

²⁰ A nonisotropic proton-gamma angular correlation could also cause the Doppler shift to be degraded; however, the stripping reaction to this level proceeds by capture of a neutron with zero angular momentum and thus, aside from compound nuclear effects, the proton-gamma angular correlation is expected to be isotropic.

¹⁹ D. H. Wilkinson, Phys. Rev. **105**, 666 (1957).

center-of-mass angle of the protons is approximately 35° . This distribution is quite consistent with that expected for a stripping reaction with $l_n=0$.

The smallest possible Doppler shift between 0° and 90° for the 6.72-Mev γ ray with $E_d=2.9$ Mev and a lifetime short compared to the stopping time of the C^{14} nuclei occurs when all the protons are emitted at 0° to the beam and is calculated from the kinematics of the reaction to be 22 kev while the measured Doppler shift was 2 ± 4 kev. Therefore, it is concluded that the measured Doppler shift is consistent only with a lifetime for the 6.72-Mev state large compared to the stopping time of the C^{14} nuclei, and is given by $\tau > 3 \times 10^{-13}$ sec.

The 6.72-Mev level is known from stripping results to be $J^\pi=1^-, 2^-,$ or 3^- . Since the C^{14} ground state is $J^\pi=0^+$, the corresponding γ -ray transitions are $E1, M2,$ and $E3$. The Weisskopf estimate gives 3.4×10^{-18} sec, 3.6×10^{-13} sec, and 6.6×10^{-11} sec, respectively for the mean lives of these three transitions. Therefore, the Doppler-shift measurement is consistent with $J^\pi=2^-$ or 3^- , but eliminates the possibility $J^\pi=1^-$. Bent *et al.*²¹ have measured the relative external and internal pair peak heights of the 6.1- and 6.72-Mev lines following the $C^{13}(d,p)C^{14}$ reaction. They found, for an assumed $E1$ transition for the 6.1-Mev γ ray, the possible multipolarities for the 6.72-Mev γ ray are, in order of preference, $E2, E1, M1, E3, M2,$ and $E4$, the last two being slightly outside the experimental errors. $E2, M1,$ and $E4$ are ruled out by the assignment of negative parity to the 6.72-Mev state, while $E1$ is ruled out by the lifetime limit; therefore their measurement indicates a preference for the assignment $J^\pi=3^-$ (corresponding to $E3$) over the assignment 2^- (corresponding to $M2$).²²

IV. ANGULAR CORRELATION OF THE 0.81-MEV AND 6.1-MEV GAMMA-RAYS

A. Introduction

General Considerations

It would often be advantageous to obtain theoretical expressions for the angular correlation of two successive γ rays involved in reactions of the type $X(h_1, h_2)Y^*(\gamma_1\gamma_2)Y$, in which h_1 and h_2 are heavy particles (with or without intrinsic spin) carrying arbitrary orbital angular momentum and where γ_1 is emitted from a level Y^* . The particle h_2 is not observed. Such reactions are quite prolific in photons and so are usually easy to study. A reaction of this type is formally described as a quadrupole cascade and, in principle, exact expressions for the angular correlations of γ_1 and γ_2 can be derived. However, the derivation requires, in general, rather complete knowledge of the properties

of the various states and particles involved in the formation of Y^* and such information is usually not available. It is possible, however, to express the angular correlations of γ_1 and γ_2 in terms of the relative populations of the magnetic substates of Y^* without requiring any knowledge of the mechanism of formation of Y^* , and as will be illustrated by the results of the present investigation, such a description can be useful in the interpretation of angular correlation and angular distribution measurements on γ_1 and γ_2 .

We consider a double cascade in which a nucleus goes from a state Y^* with spin J_1 through a state with spin J_2 to a final state Y with spin J_3 by the successive emission of two γ rays (γ_1 and γ_2) with multipolarities 2^{L_1} and 2^{L_2} respectively. We choose the beam (direction of h_1) as the quantization axis and define the direction of emission of γ_i by the Euler angles $\alpha_i\beta_i0$ where β_i is the polar angle between the direction of emission of γ_i and the quantization axis and $\alpha_2-\alpha_1=\phi$ is the dihedral angle between the γ_1-h_1 and γ_2-h_1 planes. The angular distribution of γ_1 with respect to the beam is²³

$$W_1(\beta_1) = \sum_{m_1 m_2} P(m_1) (J_2 L_1 m_2 M_1 | J_1 m_1)^2 F_{L_1}^{M_1}(\beta_1), \quad (1)$$

where $P(m_1)$ is the relative population of the m_1 -substate of the state with spin J_1 , $(J_2 L_1 m_2 M_1 | J_1 m_1)$ is a Clebsch-Gordan coefficient, and $M_1 = m_1 - m_2$. The $F_L^M(\beta)$ give the characteristic directional distribution of the radiation emitted in a transition between magnetic substates with $\Delta m = M$ and have been tabulated²⁴ through $L=5$.

The angular distribution of γ_2 with respect to the beam is given by

$$W_2(\beta_2) = \sum_{m_1 m_2 m_3} P(m_1) (J_2 L_1 m_2 M_1 | J_1 m_1)^2 \times (J_3 L_2 m_3 M_2 | J_2 m_2)^2 F_{L_2}^{M_2}(\beta_2), \quad (2)$$

and the angular correlation of γ_1 and γ_2 is given by

$$W(\beta_1\beta_2\phi) = \sum_{m_1 m_2 m_2' m_3} P(m_1) (J_2 L_1 m_2 M_1 | J_1 m_1) \times (J_2 L_1 m_2' M_1' | J_1 m_1) (J_3 L_2 m_3 M_2 | J_2 m_2) \times (J_3 L_2 m_3 M_2' | J_2 m_2') F_{L_1 L_1}^{M_1 M_1'}(\alpha_1 \beta_1) \times F_{L_2 L_2}^{M_2 M_2'}(\alpha_2 \beta_2), \quad (3)$$

where $M_1' = m_1 - m_2'$, $M_2' = m_2' - m_3$. The directional distribution functions,

$$F_{LL}^{MM'}(\alpha\beta) = \sum_{\mu\mu'} D_{M\mu}^L(\alpha\beta) D_{M'\mu'}^L(\alpha\beta) F_{L\mu}^\mu(0), \quad (4)$$

are expanded in terms of the $F_{LL}^{MM}(00) = F_L^M(0)$ which vanish unless $M = M' = \pm 1$. The expansion

²¹ Bent, Bonner, McCrary, and Ranken, Phys. Rev. **100**, 771 (1955).

²² This preference strengthens the argument (see Sec. II) that the 7.35-Mev state is not 1^- , since the lowest possible multipolarity allowed for the 0.62-Mev transition would be $E2$ if the 6.72-Mev state were 3^- and the 7.25-Mev state were 1^- .

²³ The angular distribution and correlation functions given here follow most closely the formalism given in the general treatment of double cascade theory of K. Alder, Helv. Phys. Acta **25**, 235 (1952).

²⁴ W. R. Arnold, Phys. Rev. **80**, 34 (1950).

coefficients $D_{M,\mu}^L(\alpha\beta)$ are the (M,μ) th elements of the rotation matrix of dimension $2L+1$. The $F_{LL}^{MM'}(\alpha\beta)$ have been evaluated from Eq. (4) for dipole and quadrupole radiation by using the rotation matrices of dimension 3 and 5 respectively and are given in Table III.

The problem treated here is identical formally to the angular correlation of the last two γ rays in a triple cascade transition in which the first transition defines the quantization axis and, in fact, Eqs. (2) and (3) can be obtained quite easily from the relations given by Biedenharn, Arfken, and Rose²⁵ for triple cascade transitions by suitable changes of sum convention and by introducing the populations of the first intermediate state explicitly.

C¹³(d,p)C^{14*}(γ₁γ₂)C¹⁴ Correlation

Measurements of the angular distributions of the proton groups from the C¹³(d,p)C¹⁴ reaction^{2,18} show that the spin of the C¹⁴ 6.89-Mev state is ≤ 2. The absence of a γ transition to the 0⁺ ground state suggests a zero-spin assignment for the 6.89-Mev state. However, a spin assignment of 1 or 2 cannot be ruled out on the basis of present experimental evidence. To decide between a spin assignment of zero and an assignment of 1 or 2, measurements of the angular correlation of the 0.81- and 6.1-Mev γ rays were carried out.

The double cascade under consideration is that of a state with spin J going through the intermediate 6.1-Mev state with spin 1 to a final state of spin 0 by the successive emission of two dipole γ rays.²⁶ For $J=0$ no propagation vector is defined and the correlation function, which is therefore independent of the mode of excitation of the 6.89-Mev state and the beam direction, is given by $W'(\theta)=1+\cos^2\theta$,²⁷ where θ is the angle subtended by the directions of emission of γ₁ and γ₂. For $J>0$ the angular correlation is a rather complicated function of the $\alpha_i\beta_i$ and has, in general, a different dependence on the $P(m_1)$ for each choice of geometry.

For special choices of geometry the angular dependence of the correlation reduces to a dependence on θ alone, thereby greatly reducing the difficulty of evaluating Eq. (3) and of interpreting the experimental measurements.

The following correlation functions were measured:
 $W(\beta_1,90,0)\equiv W'(90-\beta_1)$.—For this correlation $\theta=90-\beta_1$ and the correlation function evaluated from

²⁵ Biedenharn, Arfken, and Rose, Phys. Rev. **83**, 586 (1951).
²⁶ The Doppler-shift measurement (see Sec. III) showed that the 0.81-Mev γ transition was predominantly dipole. To simplify the following discussion it is assumed that it is strictly dipole in character. However, admixtures of quadrupole radiation have been considered and it has been found that the results presented herein are valid as long as $\delta^2 \leq 0.1$.
²⁷ L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953). We designate correlations which are a function of θ only by W' .

TABLE III. The directional distribution function $F_{LL}^{MM'}(\alpha\beta)$ for pure dipole and quadrupole radiation. The values of $F_{LL}^{MM'}(\alpha\beta)$ not listed are given by the symmetry relations $F_{LL}^{L,M,M'} = (-)^{M'-M} F_{LL}^{-M,-M'}$ and $F_{LL}^{L,M,M'} = F_{LL}^{L,M',M}$.

L	M	M'	$F_{LL}^{MM'}(\alpha\beta)$
1	-1	-1	$\frac{1}{2}(1+\cos^2\beta)$
		0	$(1/\sqrt{2})e^{i\alpha}\cos\beta\sin\beta$
		1	$\frac{1}{2}e^{2i\alpha}\sin^2\beta$
1	0	0	$\sin^2\beta$
		-2	$\frac{1}{2}(1-\cos^4\beta)$
		-1	$-e^{i\alpha}\cos^3\beta\sin\beta$
2	-2	-2	$-\frac{1}{2}(\sqrt{6})e^{2i\alpha}\cos^3\beta\sin^2\beta$
		-1	$-e^{3i\alpha}\sin^3\beta\cos\beta$
		0	$-\frac{1}{2}e^{4i\alpha}\sin^4\beta$
2	-1	-1	$\frac{1}{2}(\cos^2 2\beta + \cos^2\beta)$
		0	$\frac{1}{4}(\sqrt{6})e^{i\alpha}\sin 2\beta\cos 2\beta$
		1	$\frac{1}{2}e^{2i\alpha}\sin^2\beta(4\cos^2\beta-1)$
2	0	0	$\frac{3}{4}\sin^2 2\beta$
		0	

Eq. (3) is

$$W'(\theta) = 1 - P(0)\cos^2\theta \quad \text{for } J=1, \quad (5)$$

and

$$W'(\theta) = 1 + \frac{11P(0)+6P(1)-3}{6-4P(0)}\cos^2\theta \quad \text{for } J=2. \quad (6)$$

The $P(0)$, etc., are the populations of the $m_1=0$, etc., substates of the 6.89-Mev state with spin J_1 . It is assumed that $P(m_1)=P(-m_1)$ (i.e., no polarization) and the populations are normalized to $P(0)+2P(1)=1$ for $J=1$, and $P(0)+2P(1)+2P(2)=1$ for $J=2$. The normalization conditions have been used to eliminate $P(1)$ from Eq. (5) and $P(2)$ from Eq. (6).

$W(90,90,\phi)\equiv W'(\phi)$.—For this correlation $\theta=\phi=\alpha_2-\alpha_1$ and the correlation function is

$$W'(\theta) = 1 - P(0)\cos^2\theta \quad \text{for } J=1, \quad (7)$$

and

$$W'(\theta) = 1 + \frac{2P(0)}{3+5P(0)+6P(1)}\cos^2\theta \quad \text{for } J=2. \quad (8)$$

$W(0,\beta_2,0)\equiv W'(\beta_2)$.—For this correlation $\theta=\beta_2$, and the correlation function is

$$W'(\theta) = 1 + [2P(0)-1]\cos^2\theta \quad \text{for } J=1, \quad (9)$$

and

$$W'(\theta) = 1 + \frac{3-2P(0)-12P(1)}{3-2P(0)}\cos^2\theta \quad \text{for } J=2. \quad (10)$$

Angular distribution of γ₁.—For $J=0$, no propagation vector is defined and γ₁ is emitted isotropically with respect to the deuteron beam. For $J>0$ the angular distribution of γ₁ with respect to the beam is

$$W_1(\beta_1) = 1 + \frac{3P(0)-1}{3-P(0)}\cos^2\beta_1 \quad \text{for } J=1, \quad (11)$$

and

$$W_1(\beta_1) = 1 + \frac{3-6P(0)-9P(1)}{3+2P(0)+3P(1)}\cos^2\beta_1 \quad \text{for } J=2. \quad (12)$$

It is seen that all of these correlations are of the form $1 + A \cos^2\theta$, where A is the anisotropy (coincidence rate at 180° divided by that at 90° , minus 1). Thus it is only necessary to measure the coincidence rate at two angles in order to determine the correlations uniquely.

B. Angular Correlation Measurements

$W(\beta_1, 90, 0)$ Correlation

The 3-in. crystal (counter 2) was mounted horizontally with its axis at 90° to the beam direction and its front face 10 cm from the target. A 6-cm thick lead collimator with a 4-cm diameter aperture was mounted on the face of the crystal. A 2-in. crystal (counter 1) was rotated about the target in the horizontal plane with its face 12 cm from the target. The pulses from counter 2 were sent to the single-channel pulse-height analyzer which was adjusted to count the full-energy-loss and one-quantum escape peaks of the 6.1-Mev γ ray. The spectrum from counter 1—which was in coincidence with the pulses accepted by the single-channel analyzer window—was recorded by the 100-channel analyzer. The coincidence spectra in these, and the following, measurements were taken at $E_d = 2.9$ Mev and were similar to the spectrum of Fig. 7. For each measurement the coincidence spectrum, the counts accumulated by the single-channel analyzer, and the integrated beam current were recorded. The time of observation for a measurement varied from 30–90 minutes. The relative coincidence rate was obtained for each measurement from the number of counts in the 0.81-Mev full-energy-loss peak obtained by subtracting the background and the small contributions from the 0.87- and 0.73-Mev full-energy-loss peaks. Measurements were taken alternately with counter 1 at 0° and at 90° to the beam direction. In all, four determinations of the anisotropy were obtained from six measurements of the coincidence rate, each determination being made from a comparison of one of the measurements (except the first and last) with the average of the two flanking measurements. The chance correction averaged about 5% of the true rate. The anisotropy, after a finite solid angle correction²⁸ of 7%, was 0.97 ± 0.10 .

$W(90, 90, \phi)$ Correlation

The 3-in. crystal (counter 2) was placed exactly as in the $W(\beta_1, 90, 0)$ correlation measurement and the 2-in. crystal (counter 1) was rotated in a vertical plane perpendicular to the beam direction with its face 10 cm from the target. Six measurements of the coincidence rate were made, three made with the angle between the counters equal to 180° being alternated with three made with the angle equal to 90° . The data were analyzed exactly as in the $W(\beta_1, 90, 0)$ measurement. The solid-angle correction was also 7%, while

²⁸ M. E. Rose, Phys. Rev. **91**, 610 (1953).

the chance correction for the six measurements averaged about 25% of the true rate. The anisotropy was 1.1 ± 0.3 .

$W(0, \beta_2, 0)$ Correlation

For this correlation measurement two 2-in. crystals were used. Counter 1 was placed with its axis coincident with the beam direction and its face 12 cm from the target. Counter 2 was rotated about the target in the horizontal plane with its face 12 cm from the target. Measurements were taken with counter 2 at 90° and at 30° to the beam direction, the latter angle being the angle of closest approach of the two crystals. Again, the data were taken and analyzed exactly as in the $W(\beta_1, 90, 0)$ correlation measurements with the exception that the anisotropy was obtained by extrapolating from the measurements made with the angles between the two counters at 30° and 90° with an associated increase in the error of the determination. The solid-angle correction was 9% while the chance correction for the six measurements averaged about 10% of the true rate. The anisotropy was 0.9 ± 0.2 .

Angular Distribution of the 0.81-Mev γ Ray

The angular distribution of the 0.81-Mev γ ray (γ_1) relative to the deuteron beam was obtained from singles spectra measured with a 2-in. crystal rotated between 0° and 90° to the beam. Five singles spectra taken at 0° to the beam were alternated with five singles spectra taken at 90° . The 10 spectra were all similar to that shown in Fig. 3. The 0.87-Mev γ ray arises from the $\frac{1}{2}^+$, 0.87-Mev O^{17} state⁵ and thus the angular distribution of this γ ray relative to the deuteron beam must be isotropic. This fact was used in the analysis of the 10 singles spectra. The background was first subtracted and the peaks resolved as shown in Fig. 3. The relative yield of the 0.81-Mev γ ray at 0° and 90° was then obtained in the following ways: from the area under the 0.81-Mev peak, from the relative areas under the 0.81-Mev and 0.87-Mev peaks, from the relative peak heights of the 0.81-Mev and 0.87-Mev peaks, and from the sum of the areas under the 0.81- and 0.87-Mev peaks. From these measurements the anisotropy of the distribution was -0.01 ± 0.07 .²⁹

C. Discussion

The three angular-correlation measurements and the angular-distribution measurement are all consistent with an assignment of $J=0$ to the C^{14} 6.89-Mev level.

²⁹ The decomposition of the spectrum (Fig. 3) was aided by a careful analysis of the spectra of individual γ rays obtained from radioactive sources. The uncertainty of the anisotropy measurement arose largely from the separation of the 0.81- and 0.87-Mev peaks. The error due to background subtraction was relatively small since the relative size of the 0.81- and 0.87-Mev peaks was rather insensitive to the assumed background. From the same spectra the anisotropy of the 0.73-Mev γ ray was measured to be 0.02 ± 0.10 . The uncertainty of this measurement was larger than for the 0.81-Mev γ -ray distribution largely because it was more sensitive to the assumed background.

For equal populations of the magnetic substates of the 6.89-Mev level the correlation functions of Eqs. (5) through (10) reduce to those given by double cascade theory²⁷ and are $W'(\theta) = 1 - \frac{1}{3} \cos^2\theta$ for $J=1$ and $W'(\theta) = 1 + (1/13) \cos^2\theta$ for $J=2$. These correlation functions are quite different from that expected for $J=0$ and also from the experimental measurements. It would be surprising if the relative population of the magnetic substates for either $J=1$ or 2 deviated sufficiently from uniformity in such a way as to bring the correlations for $J=1$ or 2 into agreement with all three correlation measurements, especially since the angular distribution of γ_1 was isotropic within the uncertainty of the measurement in agreement with that expected for equal populations of the substates. And, in fact, it can be shown from Eqs. (5) through (12) that a spin assignment of 1 or 2 to the 6.89-Mev state is inconsistent with the measurements described above.

The physically realizable limits for $P(m)$ with the normalization chosen are $0 \leq P(0) \leq 1$ and $0 \leq 2P(1) \leq 1$, so that for $J=1$ the maximum positive anisotropy allowed by Eqs. (5) and (7) for the $W(\beta_1, 90, 0)$ and $W(90, 90, \phi)$ correlations is zero. The experimentally determined anisotropies for these correlations were 0.97 ± 0.10 and 1.1 ± 0.3 , respectively; thus, both measurements are clearly inconsistent with an assignment of $J=1$ to the 6.89-Mev state. The measured anisotropy for the $W(0, \beta_2, 0)$ correlation was 0.9 ± 0.2 which corresponds to a population [see Eq. (9)] of $1.12 \geq P(0) \geq 0.72$, while the angular distribution of the 0.81-Mev γ ray, for which the anisotropy was measured to be -0.01 ± 0.07 gives [see Eq. (11)] $0.40 \geq P(0) \geq 0.26$. Since these two measurements are not consistent with the same value of $P(0)$ for an assumed assignment of $J=1$, they combine to give a third independent determination showing that the spin of the 6.89-Mev state is not 1.

The measurements of the angular correlations and the angular distribution define limits on the possible populations for the magnetic substates for any assumed spin assignment to the 6.89-Mev state. In Fig. 9 the populations for $J=2$ possible within the experimental uncertainties for three of the four measurements are illustrated graphically. These values were obtained from Eqs. (6), (10), and (12). From Fig. 9 it is seen that the three measurements are just consistent within the uncertainty of the measurements with a spin assignment of 2 for the 6.89-Mev state, provided that $P(0) \simeq 0.57$ and $P(1) \simeq 0$ so that these measurements are not conclusive. However, the anisotropy for the $W(90, 90, \phi)$ correlation is limited to a maximum positive value [see Eq. (8)] of 0.25 which occurs for $P(0) = 1$, while the measured anisotropy was 1.1 ± 0.3 so that no populations within the triangular region of Fig. 9 are allowed by the measurement of $W(90, 90, \phi)$, and the $W(90, 90, \phi)$ correlation measurement is inconsistent with a spin assignment of 2 for the 6.89-Mev state.

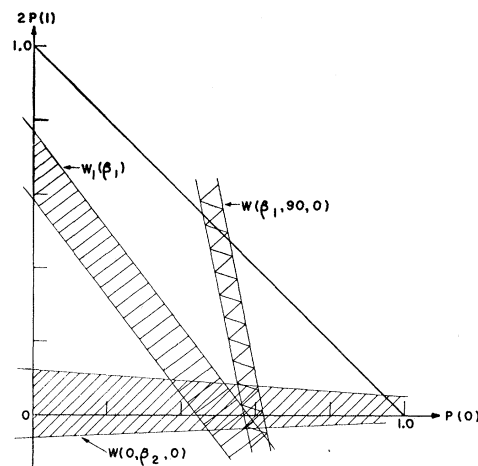


FIG. 9. The limits imposed on the relative populations of the magnetic substates of the 6.89-Mev state for an assumed spin of 2. The hatched areas show the values of $P(0)$ and $2P(1)$ consistent within the quoted uncertainties for the measured $W(0, \beta_2, 0)$ and $W(\beta_1, 90, 0)$ correlations and the angular distribution $W_1(\beta_1)$ of the 0.81-Mev γ ray (γ_1).

Since the stripping results gave possible spin assignments of 0, 1, or 2 for the C^{14} 6.89-Mev level and the angular correlation measurements just described are inconsistent with the assignments $J=1$ and 2 it is concluded that the C^{14} 6.89-Mev level has spin zero.

V. SUMMARY AND CONCLUSIONS

A weak transition was observed between the C^{14} 7.35- and 6.72-Mev states. The ratio of the intensity of a 7.35-Mev ground-state transition to this cascade transition was estimated to be of the order of unity or less. The observation of the 7.35→6.72-Mev state transition indicates that the 7.35-Mev state is not 1^- .

Lifetime limits were obtained from measurements of the Doppler shifts of several γ rays. These limits provided ancillary information useful in the description of several C^{14} states. In the case of the 6.72-Mev state, the lifetime limit helped in assigning a most probable spin and parity of 3^- . In the case of the 6.89 Mev state, the lifetime limit indicated that the 0.81-Mev transition to the 6.1-Mev state was predominantly dipole. The latter information was essential to a conclusive interpretation of the angular-correlation measurements made on the 0.81- and 6.1-Mev γ rays.

The angular-correlation measurements on the 0.81- and 6.1-Mev γ rays gave a unique assignment of $J=0$ for the 6.89-Mev state. It is clear that the interpretation of these measurements depended to a large extent on the large anisotropy of the angular correlation for $J=0$. Because of this large anisotropy it was possible to eliminate the possibility of $J=1$ and $J=2$ with rather rough measurements. In general, there is more than one unknown spin or multipolarity involved in a γ cascade following a nuclear reaction. Moreover, the anisotropies

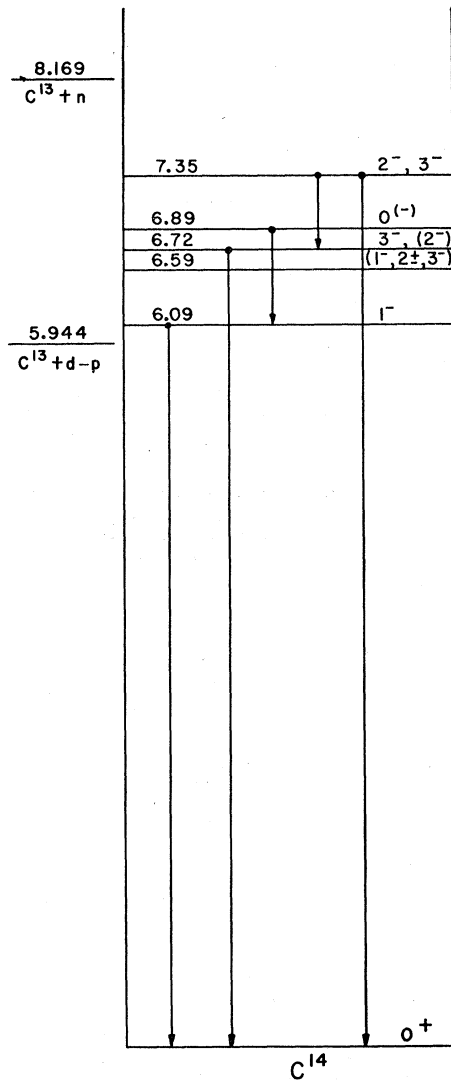


FIG. 10. The presently available information for the bound excited states and γ transitions of C^{14} . Less likely or uncertain assignments are enclosed in parentheses.

expected for different spin assignments are not usually so widely different as in the present case. For these reasons the method, used here, of comparing experi-

mental angular correlations to the correlations consistent with the allowed population distributions of the initial state is probably of rather limited use.³⁰

The results of this—and previous—investigations on the bound excited states and γ rays of C^{14} are summarized in Fig. 10.³¹ It is seen that the state at 6.59 Mev is the only bound state for which no γ rays have been assigned. The lack of observation of γ rays from this level is not surprising since the cross section for the formation of this level by $C^{13}+d$ at $E_d=14.8$ Mev was measured² to be less than one-tenth of the cross section for the formation of any of the other bound levels.

VI. ACKNOWLEDGMENTS

The authors are indebted to Mr. O. Kistner for the loan of a 3-in. NaI(Tl) crystal and are grateful to Dr. E. N. Hatch, Dr. D. H. Wilkinson, and Dr. B. H. Flowers for discussions concerning these experiments.

³⁰ A search through the literature revealed only one example [the $F^{19}(p,\alpha)O^{16}(\gamma_1\gamma_2)O^{16}$ reaction through the O^{16} 10.98-Mev state (see reference 7)] of an angular correlation measurement of the type considered here for which this method could be useful in assigning a nuclear spin value.

³¹ Note added in proof.—Figure 10 has been modified in proof to include recently available information. F. Gabbard and T. W. Bonner (to be published) have used the neutron threshold method to obtain an accurate energy value of 7.469 ± 0.005 Mev for the N^{14} 7.4-Mev level, while Ranken, Bonner, McCrary, and Rabson, Phys. Rev. **109**, 917 (1958) used a magnetic lens spectrometer to obtain an energy value (Doppler-corrected) of 7.323 ± 0.025 Mev for the 7.3-Mev γ ray observed following the bombardment of C^{13} with 4.5-Mev deuterons. Therefore the 7.3-Mev γ ray is definitely assigned to the C^{14} 7.35-Mev level ground-state transition. We therefore conclude from the present experiment that the C^{14} 7.35-Mev level decays by emission of 0.62-Mev and 7.35-Mev γ rays of approximately (within a factor of five) equal intensity.

In addition, the spin and parity assignments given in Fig. 10 include a recent reinvestigation by Warburton, Baranger, and McGruer (unpublished) of the stripping results (see reference 18). The Butler stripping radius assumed by McGruer *et al.* for the $l_n=0$ theoretical angular distribution was too large. If a radius is used which is consistent with the best-fitting radius for $l_n=0$ angular distributions obtained under similar kinematical conditions [see for example E. K. Warburton and J. N. McGruer, Phys. Rev. **105**, 639 (1957)], then it is found that the angular distribution for the 6.89-Mev level is fitted by $l_n=0$ better than by $l_n=1$. Therefore the parity of the 6.89-Mev level is most likely negative. Similarly the angular distribution for the 6.59-Mev level is fitted by $l_n=2$ or a mixture of $l_n=1$ and 3, which gives $J^\pi=1^-, 2^-, 3^-$, or 2^+ , respectively. Neither fit for the 6.59-Mev level is particularly good so that only a tentative assignment of $J^\pi=1^-, 2^\pm, 3^-$ is made for this level.