Radiation from Excited States of Carbon-13†

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A scintillation spectrometer and a magnetic lens spectrometer have been used to study gamma rays from excited states of C^{13} at 3.84 and 3.68 Mev, produced in the reactions $C^{12}(d,p)C^{13*}$ and $B^{10}(\alpha,p)C^{13*}$. Lines have been measured at 169.5±0.4 kev, 3.844±0.015 Mev, and 3.69±0.02 Mev. The last one was shown to exhibit a Doppler shift, suggesting a lifetime less than 3×10^{-13} second. The 170-kev line was shown to result from a transition between the 3.84- and 3.68-Mev levels, indicating a level energy 3.675 ±0.015 Mev. Its internal conversion coefficient was found (assuming an isotropic distribution) to be $(1.4\pm0.3)\times10^{-4}$, consistent with electric dipole radiation. Lack of knowledge of the angular distribution adds to the uncertainty and makes it impossible to rule out magnetic dipole radiation. Relative line intensities showed that the upper level decayed through the 3.68-Mev level with a probability 0.24±0.05. Transitions to the 3.1-Mev level were not detected and were concluded to have intensities less than 3% of the ground-state transitions.

INTRODUCTION

THE excitation energies, spins, and parities of the lower levels of C^{13} now seem well established.[1] This paper reports an investigation of the radiation from the second and third excited states at 3.684±0.015 and 3.844±0.015 Mev,[2] respectively. The results consist of new values of the gamma-ray energies, an internal conversion coefficient, and information on the radiative lifetimes.

A 3-Mev electrostatic accelerator was used as a source of deuterons and singly-charged helium ions to produce the C^{13} gamma rays in the reactions $C^{12}(d,p)C^{13*}$ and $B^{10}(\alpha,p)C^{13*}$. A 20-microgram/cm² natural boron target was prepared by cracking diborane gas on tantalum, and the carbon targets were graphite flakes or unbacked carbon foils prepared by cracking natural gas on tantalum and detaching the deposit. Efforts to prepare boron foil targets were unsuccessful. The complementary techniques of scintillation spectroscopy and magnetic lens spectroscopy were used to establish the presence of gamma lines at 3.69, 3.84, and 0.170 Mev, and to study certain of their properties.[3] The first two lines were interpreted as representing ground-state transitions from the known levels, and gamma-gamma coincidences were measured to confirm the assignment of the third line as a transition between the levels.

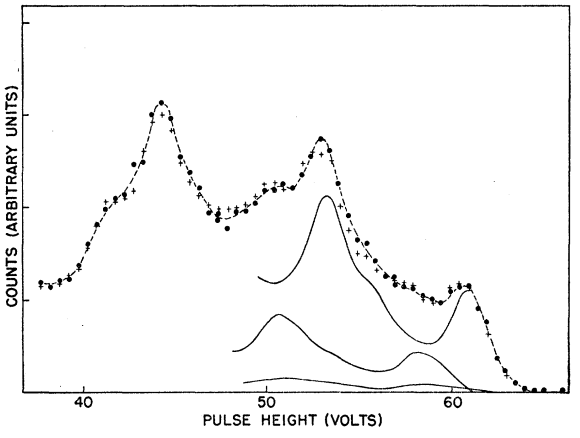

FIG. 1. Pulse-height spectrum in NaI(Tl) from $B^{10}(\alpha,p\gamma)C^{13}$. $E_\alpha=1.64$ Mev. Circles represent readings taken with counter in line with the beam; crosses correspond to 155 degrees. Solid curves represent component spectra from lines at 3.84 Mev and 3.68 Mev. The lowest curve is an estimate of the Doppler-shifted 3.68-Mev spectrum.

DOPPLER SHIFT AND RELATIVE LINE INTENSITIES

A study of Doppler effects was made on high-energy gamma rays from $B^{10}(\alpha,p)C^{13*}$ by comparing energies measured at 0° and 155° with respect to the incident He^+ beam.[4] The beam energy was set by electrostatic analysis to give maximum yield from a resonance at 1.64 Mev at which the 3.84-Mev level is produced with 22 times[5] the intensity of the 3.68-Mev level. A scintillation counter [NaI(Tl) cylinder, 1½ inches diameter, 1½ inches long; Dumont 6292 photomultiplier] was set up two inches from the target, first on a radius coincident with the He^+ beam direction, then at 155° removed from it. The scintillation spectrum was studied at each position with a ten-channel pulse-height analyzer and is shown in Fig. 1. Circles represent the 0° data and crosses, the 155° data. The spectrum from the 2.62-Mev line from ThD was used as a stability check. No shift between the spectra was detected, and it was concluded that the lifetime of the 3.85-Mev level is significantly longer than the stopping time in tantalum for the recoil carbon nuclei,[6] about 3×10^{-13} second.

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

[2] Sperduto, Buechner, Bockelman, and Browne, Phys. Rev. **96**, 1316 (1954).

[3] A preliminary account of part of this investigation was given by R. J. Mackin, Jr., Phys. Rev. **92**, 529(A) (1953).

[4] W. R. Mills and R. J. Mackin, Jr., Phys. Rev. **95**, 1206 (1954).

[5] Shire, Wormald, Lindsay-Jones, Lunden, and Stanley, Phil. Mag. **44**, 1197 (1953).

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

The experiment was repeated at a resonance (2.30 Mev) at which the 3.68-Mev line intensity was found to be about five times that of the 3.85-Mev line. Figure 2 shows the measured spectra.

The shape of the (unshifted) curve produced by decay of the 3.85-Mev level was taken to be that of Fig. 1, adjusted for the change in pulse amplification used. Subtracting it from each curve results in two spectra from a nominal 3.68-Mev transition, shifted by $1.3\pm0.2\%$ of the line energy. The theoretical maximum shift expected to result from net motion of the center of mass is 1.9%, but the experiment's poor geometry prevents drawing any quantitative conclusion as to the level's mean life. The expected mean life is two orders of magnitude below the stopping time.

The subtraction analysis of Fig. 2 suggested a shape for the scintillation spectrum produced by an individual line at 3.68-Mev. Using this shape together with an estimate of the intensity of the shifted 3.68 line obtained from proton-group intensities,[5] a rough analysis was made of the 0° curve in Fig. 1. After subtraction of the weak shifted spectrum (the lowest curve in Fig. 1), a more careful analysis of the remainder was made, which was based on identical 3.68-Mev and 3.85-Mev line shapes with a photopeak like that of the 3.68-Mev line measured at 2.3 Mev. Figure 1 shows the resultant curves as solid lines.

The intensity ratio of the gamma rays emitted at 0° was determined from the relative photopeak heights, corrected for the (empirical) relative photopeak efficiency,[7] and was found to be: (total 3.68)/3.85=0.46. By assuming the angular distributions corresponding to the accepted angular momenta involved,[5,8] and comparing with the proton group intensities, it was calculated that the 3.85-Mev level decays by cascade through the 3.68-Mev level with a probability 0.24 ± 0.05.

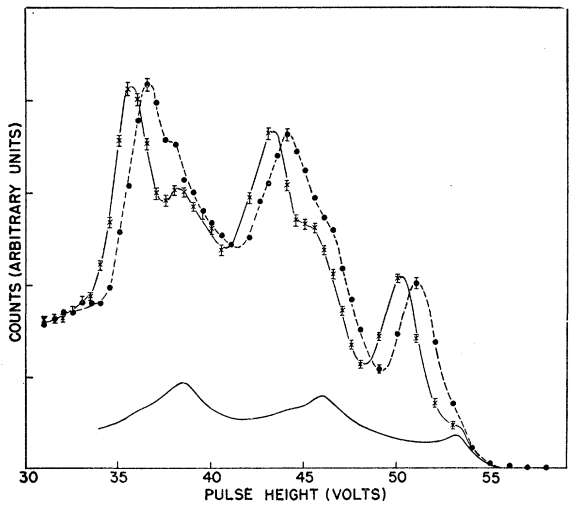

FIG. 2. Pulse-height spectrum in NaI(Tl) from $B^{10}(\alpha,p\gamma)C^{13}$. $E_\alpha=2.3$ Mev. Circles represent 0° readings; crosses, 155° readings. Lower solid curve is the "total unshifted spectrum."

[7] H. H. Woodbury, thesis, California Institute of Technology, 1953 (unpublished).
[8] A. G. Stanley, Phil. Mag. **45**, 430 (1954).

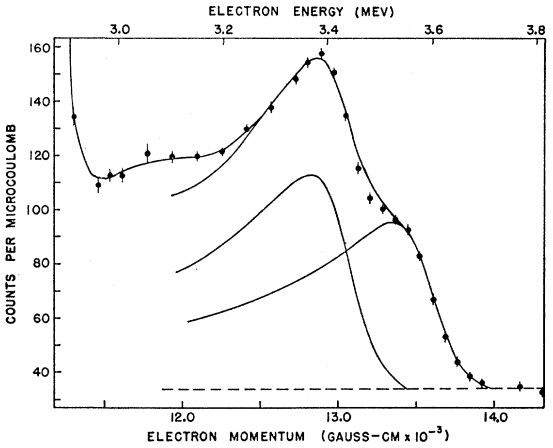

FIG. 3. Spectrum of Compton electrons produced by the 3.84- and 3.68-Mev gamma rays of $C^{12}(d,p\gamma)C^{13}$ in 40-mg/cm² graphite. The solid curves represent theoretical line shapes.

By using this branching probability in conjunction with the relative gamma-ray intensities calculated for the 2.3-Mev resonance and assuming isotropic emission, it was found that at $E_\alpha=2.3$ Mev the 3.68-Mev level is produced with 3.5 times the intensity of the 3.85-Mev level.

ENERGY MEASUREMENTS

The Compton electron spectrum from a 40-mg/cm² graphite target-converter at the source-point of a magnetic lens spectrometer was studied to determine the energies of the two high-energy lines from $C^{12}(d,p)C^{13*}$ at $E_d=2.4$ Mev. A trans-stilbene scintillation-counter detector was used to record focused electrons. The spectrum (Fig. 3) was compared with theoretical line shapes[4,6] derived from the Klein-Nishina formula by folding with the spectrometer window curve (a Gaussian of width 1.9% of momentum), a Landau energy loss distribution for the target, and (for the lower line) a Doppler broadening distribution. The source of the discrepancy below 12.5 kilogauss-centimeters is believed to be the 3.4-Mev line[6] from $C^{13}(d,n)N^{14*}$. An alternative analysis, which employed the observed spectrum produced by the (3.097 ± 0.005)-Mev line (not shown) as a pattern for resolving the components, showed the same discrepancy in this region. The two analyses produced compatible energy values for the two gamma lines: 3.844 ± 0.015 Mev and 3.69 ± 0.02 Mev.

No Doppler correction is indicated for the upper line, according to the results of the previous section. Taking into account the fraction of the lower line's intensity resulting from direct production of the level instead of cascade (0.75), its Doppler correction (for isotropic distributions) is 19 kev. This figure may be changed by as much as 50% by the angular distributions involved. The indicated value for the level energy is

3.67±0.03 Mev; a better value is given in the next section. The thick-target yields of 3.84- and 3.69-Mev γ rays at $E_d=2.4$ are, respectively, 4.3×10^{-6} γ rays/deuteron and 5.8×10^{-6} γ rays/deuteron, to about 15%.

LOW-ENERGY LINE

The energy of the gamma line connecting the two levels was measured in the magnetic lens spectrometer through study of the internal conversion line and the photoelectric peaks from silver, thorium, and uranium converters. An Amperex 200C end-window Geiger counter served as detector. The results appear in Fig. 4. The momentum scale was calibrated on the ThB "F" line,[9] and account was taken of the component of the earth's field along the spectrometer axis, the atomic binding energy of the converter,[10] and electron energy losses in the converter. The thickness of the silver converter was determined by the energy loss of the internal conversion electrons passing through it, corrected for the energy difference between these electrons and the silver K-shell photoelectrons by a semiempirical energy-loss formula.[6] The thicknesses of the uranium and thorium converters were, respectively, 2 and 4 mg/cm^2, many scattering lengths for electrons in the K-photopeaks. The peak positions were therefore located by their extrapolated leading edges,[11] which obviated the need for energy loss corrections. The transition energy values are given in Table I.

Consideration was given to a possible energy shift of the internal conversion electrons resulting from motion in the vacuum of a fraction of the excited C^{13} nuclei recoiling from the rear face of the target. Equality of the widths of the internal conversion peaks with and without the silver backing (2.3±0.15 and 2.4±0.15%) indicated that such effects were not significant. The indicated value for the energy of the lower level is 3.675±0.015 Mev.

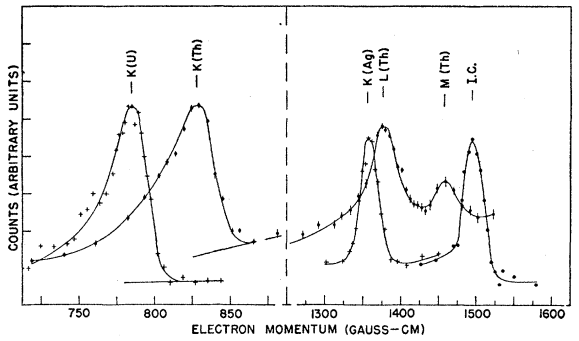

FIG. 4. Internal conversion line and photoelectric peaks produced by the 170-kev gamma rays from $C^{12}(d,p\gamma)C^{13}$ in silver, thorium, and uranium converters.

[9] G. Lindstrom, Phys. Rev. **83**, 465 (1951).
[10] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).
[11] Hornyak, Lauritsen, and Rasmussen, Phys. Rev. **76**, 731 (1949).

TABLE I. Measured energy values for the 170-kev transition.

Internal conversion	
Silver (K)	170.6±1.0 kev
Thorium (L)	170.1±1.0 kev
Thorium (K)	169.4±0.8 kev
Uranium (K)	168.5±0.5 kev
	168.7±0.5 kev
Average	169.5±0.4 kev

A measure of the internal conversion coefficient was found by comparing the area under the internal conversion line with the scintillation spectrum produced by the low-energy gamma rays in a 0.5-inch NaI(Tl) crystal in well-defined good geometry. The measurements were made in immediate succession without displacement of the target. It was necessary to correct for electron absorption in the 1.4 mg/cm^2 window of the detector counter[12] (10%) and for gamma-ray absorption in the spectrometer wall and the crystal covering. It was estimated[13] that 84% of the radiation incident upon the crystal produced pulses in the photo peak. The lens spectrometer solid angle was determined from the Compton spectrum of a calibrated Co^{60} source.

The internal conversion coefficient was found to be

$$Y_e/Y_\gamma=(1.4\pm0.3)\times10^{-4}.$$

This figure ignores angular distribution effects. It could thus be seriously in error, inasmuch as the conversion electron distribution, if anisotropic, differs sharply from the gamma-ray distribution at this low energy,[14] and inasmuch as the electrons were measured at about 20° and the gamma rays at 90° away from the deuteron beam direction.

The theoretical total conversion coefficients for the lower multipoles[15] are

$E1$	1.75×10^{-4}
$M1$	0.75×10^{-4}
$E2$	14.7×10^{-4}
$M2$	$7.\times10^{-4}$.

Agreement is obtained only with $E1$ (as expected from the accepted spins) although the qualification imposed by unknown angular distributions admits the possibility of $M1$.

A corroborative measurement was made of the internal conversion coefficient of the 238-kev line of ThB (of which the "F" line is the K-conversion peak). The resulting value was 0.68±0.13. The best known

[12] Feather, Kyles, and Pringle, Proc. Phys. Soc. (London) **A61**, 466 (1948).
[13] R. L. Heath and F. Schroeder, U. S. Atomic Energy Commission Report AECD IDO-16149, first rev., 1955 (unpublished).
[14] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).
[15] Rose, Goertzal, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951), and Rose, Goertzal, and Swift (privately circulated tables).

value, based on the intensity of the F-line relative to that of the parent beta spectrum, is 0.71.[16]

With the lens spectrometer and a thin carbon target, backed with a 20.2-mg/cm^2 thorium converter, a search was made for the gamma lines representing transitions to the 3.1-Mev level. The search was unsuccessful, and their intensities are estimated at less than three percent of those of the ground state transitions.

[16] D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) **63**, 223 (1950).

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The authors are indebted to T. Lauritsen and W. A. Fowler for many helpful discussions.

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Electron Capture and Alpha Decay of Np^{235}†

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The electron capture and alpha decay of Np^{235} have been investigated. An L/K capture ratio of 30 ± 2 is found from absolute L and K x-ray measurements. No gamma rays are observed in the electron capture decay of Np^{235}. An alpha branching ratio of $(3.5\pm0.4)\times10^{-5}$ was measured for this nuclide. The alpha particles have coincident 26- and 85-kev gamma rays with an intensity of 0.054 ± 0.005 gamma per alpha disintegration for the 85-kev gamma ray. The intensity of the 26-kev gamma ray is comparable, although somewhat more uncertain.

I. INTRODUCTION

THE nuclide Np^{235} was originally discovered as a product of deuteron irradiations of U^{235}.[1] In later work by James *et al.*,[2] a half-life of 410 days was measured for this nuclide, exhibiting predominantly electron capture decay with an L/K capture ratio of >9. A slight alpha branching (5×10^{-5}) was also reported with an alpha-particle energy of 5.06 Mev. The present report concerns a more detailed study of the decay of this nuclide using a scintillation crystal spectrometer and a proportional counter in connection with a multichannel pulse-height analyzer. Both gamma-gamma and alpha-gamma coincidence techniques were used in a study of the decay scheme.

II. EXPERIMENTAL METHODS AND RESULTS

The Np^{235} used in the present study was produced by the $(d,2n)$ reaction in a cyclotron bombardment of high-purity U^{235} (99.9+%) with 18-Mev deuterons. The target received a total of 2489 microampere hours of intermittent irradiation over a period of three months. After dissolution of the uranium foil in hydrochloric acid, the neptunium fraction was separated and purified by repeated hydroxide and fluoride precipitations with lanthanum carrier added, anion exchange column elutions using Dowex-1 resin[3] in a hydrochloric acid medium, and solvent extraction of the neptunium using thenoyltrifluoroacetone (TTA) dissolved in benzene.[4] The final purification was performed after waiting a sufficient length of time to insure the complete decay of shorter-lived neptunium isotopes (Np^{234}, Np^{236}) present in the original sample. An aliquot of the final preparation was followed for decay in a 4π counter as a check on radiochemical purity.

The relative intensities of the K and L x-rays resulting from the electron-capture decay of Np^{235} were measured using both a sodium iodide (Tl-activated) crystal and a xenon-filled proportional counter as detectors. The initial pulse, either from the phototube attached to the crystal or directly from the proportional counter was introduced into a 50-channel differential pulse-height analyzer, after being amplified in a preamplifier and linear amplifier. The 60-kev gamma ray of Am^{241} was used to calibrate sample geometry for the sodium iodide crystal. Knowing the dimensions of the proportional counter and the pressure of the xenon gas within the counter, it was possible to calculate the sample geometry and counting efficiencies for the L x-rays of the Np^{235} sample. The counting efficiencies of the L_α, L_β, and L_γ x-rays of uranium were 53.7%, 34.0%, and 23.5%, respectively. The calculated geometry and counting efficiencies were experimentally checked using the L x-rays of an Am^{241} standard.[5] The K and L x-ray spectra of Np^{235} are shown in Fig. 1. From the absence of

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] James, Florin, Hopkins, and Ghiorso, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV.

[2] James, Ghiorso, and Orth, Phys. Rev. **85**, 369 (1952).

[3] Dow Chemical Company, Midland, Michigan.

[4] E. K. Hyde, in *The Actinide Elements*, edited by G. T. Seaborg and J. J. Katz (McGraw-Hill Book Company, Inc., New York, 1954), Chap. 15, National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV.

[5] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952) and Phys. Rev. **87**, 1144 (1952).