Disintegration Schemes of Radioactive Substances. III. I^{131}

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The disintegration of the 8-day isotope of iodine, I^{131}, has been studied by spectrometer and coincidence techniques. The beta-ray spectrum is simple and has an end point of 0.595 ± 0.01 Mev. Each beta-ray is accompanied by two cascade gamma-rays of energies 367 ± 7 and 80 ± 1 kev, respectively. Both gamma-rays are partially internally converted, the 367-kev gamma-ray in the K shell in 0.8 percent of the disintegrations.

INTRODUCTION

THE 7.8-day activity of iodine, widely used for biological tracer work, has been assigned to I^{131}, although a slight possibility exists that it should instead be assigned to I^{129}.[1] This isotope emits beta-rays of moderate energy and also gamma-rays,[1,2] but not x-rays.[3] We have investigated the radiations from this isotope with a view to analyzing the possible modes of disintegration and to discovering the energy levels of the product nucleus Xe^{131}. The accurate understanding of the modes of disintegration is necessary for the calculation of radiation dosages in biological work in which this isotope is of therapeutic value.[4]

PROCEDURES

The coincidence amplifiers and magnetic lens beta- and gamma-ray spectrometer used in this work have been previously described.[5,6] Platinum screen cathode gamma-ray counters were used rather than copper cathode counters because of their considerably higher efficiency in detecting low energy gamma-rays. High efficiency of gamma-ray counters for low energy gamma-radiation is of course an advantage, but it is partially offset by the increased probability that a single quantum from the source causes a coincidence by means of secondary quanta produced by the photoelectric and Compton effects. The number of such spurious coincidences depends of course upon the relative geometry of source and counters. In the usual arrangement with source between counters, the number of spurious coincidences is a minimum with respect to geometry, but still appreciable, and no consistent results on the efficiencies of the counters could be obtained while the source was in this position. This experience is in direct contrast to that of Dunworth,[7] who found that Compton coincidences were negligible with the source between brass cathode counters, which are considerably less efficient for low energy quanta.

In order to eliminate these spurious coincidences, $\frac{5}{8}$ inch of lead was inserted between the two platinum counters used in gamma-gamma-

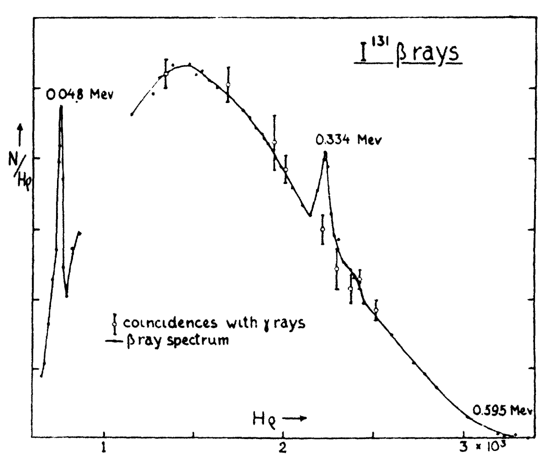

FIG. 1. Beta-ray spectrum and beta-gamma-coincidence spectrum of I^{131}.

* These results were reported at the meeting of the American Physical Society, Princeton, New Jersey, December, 1941.

[1] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 775 (1938); G. T. Seaborg, J. J. Livingood, and J. W. Kennedy, Phys. Rev. **55**, 794 (1939).

[2] G. F. Tape, Phys. Rev. **56**, 965 (1939).

[3] J. G. Hamilton and M. H. Soley, Am. J. Physiol. **127**, 557 (1939).

[4] S. Hertz, A. Roberts, J. H. Means, and R. D. Evans, Trans. Am. Assn. Study of Goiter, p. 260 (1939).

[5] M. Deutsch, Phys. Rev. **59**, 684A (1941).

[6] A. Roberts, J. R. Downing, and M. Deutsch, Phys. Rev. **60**, 544 (1941).

[7] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).

coincidence measurements, and the source placed symmetrically above the lead so that it could see each counter, and so that scattered quanta would have to penetrate the lead to cause a coincidence. In addition, we have also used a $\frac{1}{4}$-inch lead screen with a small hole over which the source is mounted in beta-gamma-coincidence experiments. These precautions were most essential when quantitative readings on counter efficiency were to be taken.

PREPARATION OF SOURCES

When tellurium is bombarded by deuterons, at least three other radioactive isotopes of iodine besides I^{131} are formed, with half-periods from 25 minutes to 13 days. Rather than work with such a mixture of isotopes, we utilized the fact demonstrated by Livingood and Seaborg,[1] that I^{131} is the decay product of the 25-minute and 1.2-day isomers of Te^{131}. Accordingly, tellurium was bombarded with deuterons in the M.I.T. cyclotron, and the radioactive iodine formed during the bombardment was separated out. The target solution was then permitted to stand for three to five days, the optimum time for growth of maximum I^{131} activity, and the iodine again separated. This gave for all practical purposes a source of radioactively pure I^{131}. It has been shown that another iodine activity grows from tellurium,[8] but this activity is far too weak to interfere with the experiment. The iodine was usually converted to AgI, since it was found that thin sources of NaI, in the presence of air, lost iodine to neighboring surfaces.

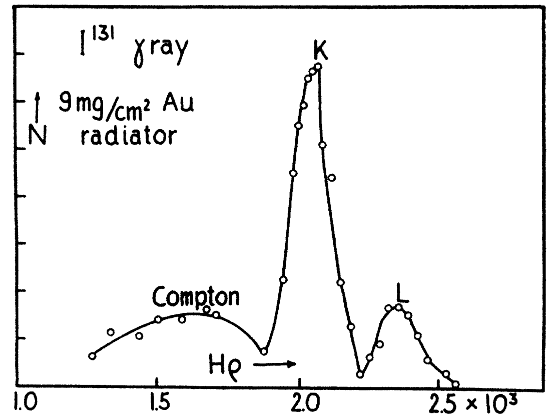

FIG. 4. Secondary electron spectrum excited by the gamma-rays of I^{131}. The photoelectron lines and Compton group are due to a 367-kev gamma-ray.

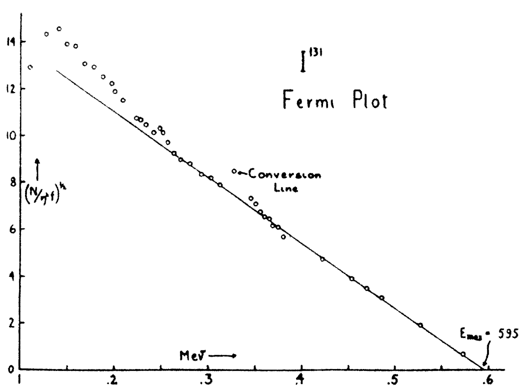

FIG. 2. Fermi plot of the beta-ray spectrum of I^{131}. The departure from linearity may be instrumental in origin.

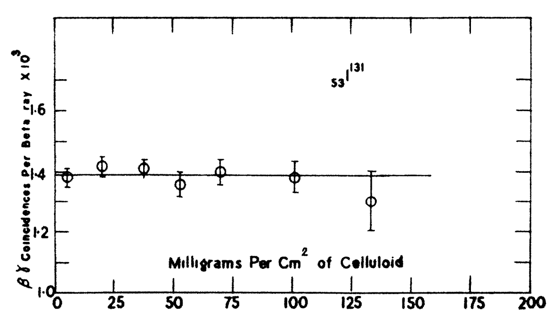

FIG. 3. The number of beta-gamma-coincidences for beta-ray in I^{131} as a function of the amount of absorber in front of the beta-counter. The independence of the rate of the absorber thickness shows the spectrum to be simple.

RESULTS

The beta-ray spectrum taken with the magnetic lens spectrometer is shown in Fig. 1. The maximum energy, both by inspection, and by extrapolation of the Fermi plot (Fig. 2), is 0.595 ± 0.01 Mev. Within the experimental error, the Fermi plot is a straight line over the range investigated. The shape of the beta-ray spectrum and of the beta-gamma-coincidence spectrum taken on the spectrometer are seen from Fig. 1 to be identical. The absorption of beta-gamma-coincidences (Fig. 3) shows the number of beta-gamma-coincidences per beta-ray to be independent of absorber thickness. All of these observations point to the conclusion that the beta-ray spectrum consists of a single transition. This is verified by integration of the beta-ray

[8] A. Roberts and J. W. Irvine, Jr., Phys. Rev. 59, 936A (1941).

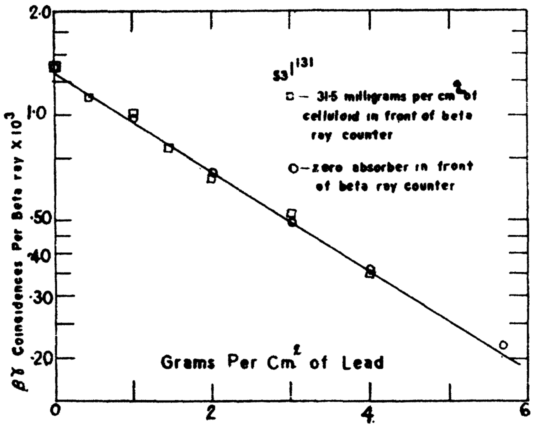

FIG. 5. The absorption of beta-gamma-coincidences by lead absorbers in front of the gamma-ray counter, for two different values of absorber in front of the beta-ray counter. The identity of the two curves shows the identity of the gamma-radiation coinciding with the two sets of beta-rays detected. The absorption of the coinciding gamma-rays is consistent with that of a 367-kev gamma-ray.

spectrum, according to the method[6] previously described.

The secondary electron spectrum of the gamma-rays (Fig. 4) shows a single gamma-ray of energy 367 ± 7 kev. The energy of this line is accurately determined, since it appears not only as Compton and photoelectron secondaries, but also in internal conversion (Fig. 1). No other gamma-ray of energy greater than 180 kev is observed.

The beta-ray spectrum of Fig. 1 shows two peaks due to internal conversion, one due to the 367-kev gamma-ray, and one due to a gamma-ray of 80 kev. The softer conversion line was found with a special thin window counter after the presence of the gamma-ray and its energy had been determined by means of measurements on gamma-gamma-coincidences, as described below.

ABSORPTION OF BETA-GAMMA-COINCIDENCES

To investigate these radiations further, the number of beta-gamma-coincidences was measured with various thicknesses of lead absorber before the gamma-ray counter. The absorption of the beta-gamma-coincidences is identical with the absorption of the gamma-rays alone (Fig. 5) and is not altered by the interposition of a celluloid absorber in front of the beta-counter. We can therefore conclude that all the gamma-rays are associated with the entire beta-ray spectrum since they each coincide equally. Also, over 90 percent of the observed gamma-ray counts are absorbed with the absorption coefficient consistent with a 367-kev gamma-ray, and so we conclude that the efficiency of the gamma-ray counter for detecting the 80-kev gamma-ray is quite low.

ABSORPTION OF GAMMA-GAMMA-COINCIDENCES

On considering the 80-kev gamma-ray which was first revealed by the presence of gamma-gamma-coincidences, we see that the main possibilities are that it is either in cascade with the 367-kev gamma-ray, or that it occurs in an alternate transition in cascade with other gamma-rays which are either too soft or too low in intensity to be observed in the secondary electron spectrum. Orbital electron capture is ruled out by the fact that ${}_{53}I^{131}$ grows from ${}_{52}Te^{131}$.

In investigating complex gamma-radiation by coincidence methods the following considerations are of interest. An absorption curve of the gamma-radiation in a single counter will in principle show the several components of the gamma-radiation. If the efficiency of the counter for detecting gamma-rays of equal abundance should vary considerably among the gamma-rays, the apparent intensity of the gamma-rays for which the counter has the lowest efficiency will be small—perhaps small enough so that the radiation will be entirely missed. Quantitatively, if there are two cascade gamma-rays for which the efficiency of the counter is ϵ_1 and ϵ_2, respectively, N disintegrations per minute will produce $N(\epsilon_1+\epsilon_2)$ counts per minute in the gamma-ray counter. If a thickness of absorber in which the absorption coefficients of the gamma-rays are μ_1 and μ_2, respectively, is interposed, the number of counts per minute will be $N(\epsilon_1 e^{-\mu_1 t}+\epsilon_2{}^{-\mu_2 t})$. Thus, while a sum of two exponential absorptions is to be expected, the abundance of each is weighted by the efficiency of the counter for detecting the appropriate radiation.

When coincidences of two cascade gamma-rays are measured, the number of gamma-gamma-coincidences per minute will be $N_{\gamma\gamma}=N(\epsilon_1\epsilon_2'+\epsilon_2\epsilon_1')$, where primed efficiencies are indicated for the second counter. If the counters are of identical construction and differ in efficiency only by a geometrical factor, so that $\epsilon'=k\epsilon$, then

$N_{\gamma\gamma}=2Nk\epsilon_1\epsilon_2$.[9] Now if an absorber is placed in front of one gamma-ray counter only, the number of gamma-gamma-coincidences will be $N_{\gamma\gamma} = N(k\epsilon_1\epsilon_2 e^{-\mu_2 t} + k\epsilon_2\epsilon_1 e^{-\mu_1 t}) = kN\epsilon_1\epsilon_2(e^{-\mu_1 t} + e^{-\mu_2 t})$.[10] This represents a combination of the same exponentials as the absorption in a single counter, but the two gamma-rays are now equally weighted, and the less efficient gamma-ray more easily observed. Curve 3 in Fig. 6 shows the absorption of gamma-gamma-coincidences with lead absorber in front of one counter. The harder component has the absorption characteristic of the 367-kev gamma-ray, and the other gamma-ray is much softer.

In view of this evidence, we may assume that the 80-kev gamma-ray is in cascade with the 367-kev gamma-ray.

In addition, measurements may be taken with absorber in front of both counters, the absorbers always being of equal thickness. In this case the number of coincidences is $N_{\gamma\gamma}=N(k\epsilon_1 e^{-\mu_1 t}\epsilon_2 e^{-\mu_2 t} + k\epsilon_2 e^{-\mu_2 t}\epsilon_1 e^{-\mu_1 t}) = 2kN\epsilon_1\epsilon_2 \exp-(\mu_1+\mu_2)t$.[11] Here a single absorption should be observed, with an apparent absorption coefficient $\mu_1+\mu_2$. Figure 6 shows the absorption curve obtained with lead. It is notable that a simple exponential absorption is expected only in the case when there are only two different gamma-rays present.

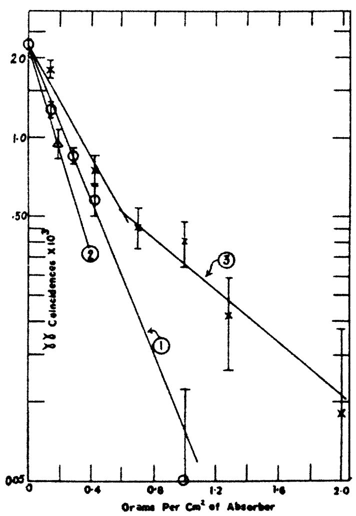

FIG. 6. Curve 3. Absorption of gamma-gamma-coincidences by lead absorber in front of one counter. Curve 1. Absorption of gamma-gamma-coincidences by lead absorber in front of both counters. Curve 2. Absorption of gamma-gamma-coincidences by tin absorbers in front of both counters.

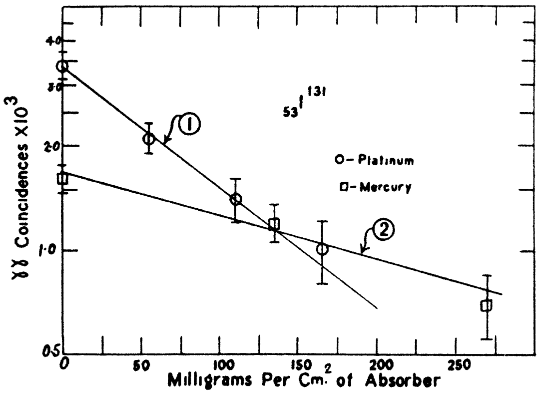

FIG. 7. Critical absorption of gamma-gamma-coincidences. Curve 1. Pt absorbers. Curve 2. Hg absorbers. Curves taken with different source strengths.

From these two experiments the absorption coefficient of the soft radiation in lead is obtained. However, there are two energies of gamma-radiation for which lead has the observed absorption coefficient of 4 cm² g⁻¹ one on either side of the K-absorption limit. Absorption in tin (Fig. 6) showed the energy to be below the K edge of lead, and then critical absorption in Pt and Hg (Fig. 7) gave the energy as 80 ± 1 kev. After this determination of the energy, confirmation of the value was found in the discovery of the conversion line to be expected for such low energy radiation, as shown in Fig. 1. It is of interest that Mitchell, Langer, and McDaniel[12] found a 70-kev gamma-ray in cascade with a considerably harder gamma-ray in Sb^{124} by similar methods (although not by critical absorption).

EFFICIENCY MEASUREMENTS

The exact efficiency of a gamma-ray counter for each of the two gamma-rays can be found by measuring both beta-gamma- and gamma-

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[9] For n cascade γ-rays the number is

$$N_{\gamma\gamma}=kN\sum_{i,j}^{n}\epsilon_i\epsilon_j \quad (i\neq j).$$

[10] For n cascade γ-rays,

$$N_{\gamma\gamma}=kN\sum_{i,j}^{n}\epsilon_i\epsilon_j(e^{-\mu_i t}+e^{-\mu_j t}) \quad (i<j).$$

[11] For n cascade γ-rays the number is

$$N_{\gamma\gamma}=2kN\sum_{i,j}^{n}\epsilon_i\epsilon_j e^{-(\mu_i+\mu_j)t} \quad (i<j).$$

[12] A. C. G. Mitchell, L. M. Langer, and P. W. McDaniel, Phys. Rev. 57, 1107 (1940).

gamma-coincidences with the same geometry of source and gamma-ray counter, as was pointed out by Dunworth.[7] The number of beta-gamma-coincidences per beta-ray recorded is $N\beta_\gamma/N\epsilon_\beta=\epsilon_1+\epsilon_2$ and the number of gamma-gamma-ray coincidences per gamma-ray recorded in a gamma-ray counter substituted for the beta-ray counter is $N_{\gamma\gamma}/Nk(\epsilon_1+\epsilon_2)=2\epsilon_1\epsilon_2/(\epsilon_1+\epsilon_2)$, provided that the counters differ in efficiency by only a geometrical factor. In the geometry in which this experiment was performed, the number of beta-gamma-coincidences per beta-ray was 0.82×10^{-3}, and the number of gamma-gamma-coincidences per gamma-ray 0.145×10^{-3}. On solving the equations for ϵ_1 and ϵ_2, the values $\epsilon_1=0.74\times10^{-3}$ and $\epsilon_2=0.07\times10^{-3}$ are obtained. Assigning the higher efficiency to the 367-kev gamma-ray, we see that the observation that about 90 percent of the gamma-ray counts are due to it is predicted by the efficiencies.

DIRECT MEASUREMENT OF INTERNAL CONVERSION COEFFICIENT

By mounting a source in the spectrometer on a thin window, it is possible to observe coincidences between beta-rays emitted through this window and conversion electrons focused by the spectrometer (and of course between conversion electrons emitted through the window and beta-rays focused by the spectrometer, as well as between beta- and gamma-rays (Fig. 8)). The number of conversion electron-beta-coincidences per recorded beta-ray permits a direct calculation of the fraction of disintegrations giving conversion electrons, provided the transmission of the spectrometer is known. Thus, in the case of the 332-kev conversion electrons produced in the K shell by the 367-kev gamma-ray, we observe 6.5×10^{-5} electron-beta-coincidence (above the background) per recorded beta-ray. The transmission of the spectrometer is 0.8 percent, and thus the number of K conversion electrons per disintegration is $6.5\times10^{-5}/0.008$ or 8×10^{-3} electron per disintegration. The L conversion line is not resolved, and is estimated not to exceed 25 percent of the K conversion. The assumptions involved in this calculation are that the beta-counter counting the focused conversion electrons is 100 percent efficient, and that the conversion electrons are equally likely to accompany any observed beta-ray. The latter assumption is true because the beta-ray spectrum has been shown to be simple; furthermore, interposing absorber to cut out part of the beta-ray spectrum does not change the electron-beta-coincidence rate per beta-ray recorded.

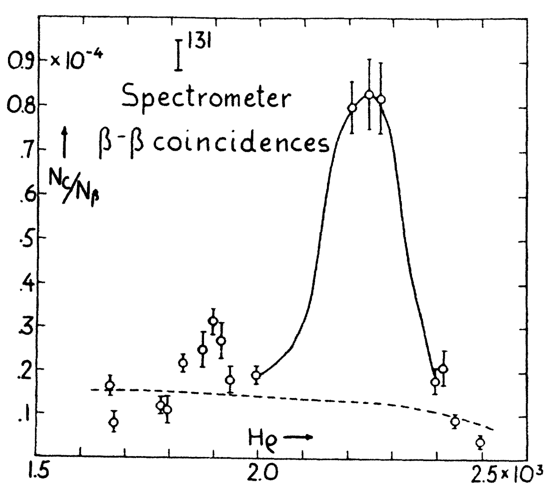

FIG. 8. Conversion electron-beta-ray coincidences as taken on the spectrometer.

The observed conversion coefficient is verified by a calculation of the height of the conversion line relative to the continuous spectrum (Fig. 1) if we take into account the line width and the shape of the spectrum. It is worth noting how much more prominent the conversion line appears in the electron-beta-coincidence spectrum (Fig. 8) than in the continuum (Fig. 1).

The observed peak in the electron-beta-coincidence spectrum at 250 kev may perhaps be due to another gamma-ray giving perhaps a fifth as many conversion electrons as the 367-kev line. No other evidence for such a gamma-ray has been found, and the line may be due to some secondary effect, as for example coincidences between beta-rays and Comptons focused by the spectrometer. The low coincidence rate necessary in electron-beta-coincidence measurements, where the individual counting rates are perhaps 60 and 20,000 counts per minute, respectively, makes the taking of accurate data somewhat difficult.

MATRIX ELEMENT

Using the universal time constant of the Fermi theory $\tau=3000$ sec. as given by Gronblom,[13] and

[13] B. O. Gronblom, Phys. Rev. 56, 508 (1939).

the values of the $F(Z, \eta)$ transition probability function given by Evans,[14] we find for the matrix element for the $I^{131}-Xe^{131}$ transition the value $G^2=5\times10^{-4}$.

The small value of the matrix element probably means that the beta-ray transition is forbidden. The beta-transitions to the ground state and the lower excited state are not present to as much as 2 percent of the main transition, so that they must be even more highly forbidden.

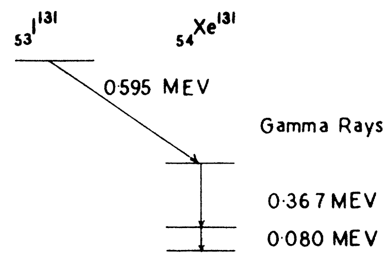

FIG. 9. Probable disintegration scheme of I^{131}. The order of emission of the gamma-rays is not known.

ELECTRIC MULTIPOLE ORDER OF THE GAMMA-RAYS

No theory of internal conversion probabilities for the region of $Z\sim53$ has been given, so far as we are aware. However, by "interpolating" between the theory for low Z[15] and the theory for high Z[16] which predict, respectively, dependence on Z^3 and Z^4, we find that the observed internal conversion coefficient of 0.008 for the 367-kev gamma-ray is probably characteristic of the emission of either electric dipole or quadrupole radiation. Our measurements on the low energy conversion electrons of the 80-kev gamma-ray were not sufficiently accurate to determine the conversion coefficient, mainly because of uncertainty as to the transmission of the counter window. In this case, however, the lifetime of the excited state against emission of an 80-kev gamma-ray would be so long for any but quadrupole[17] or dipole radiation that no coincidences with this gamma-ray would have been observed.

In order to account for the absence of 447 $(=367+80)$ kev radiation, it is necessary to suppose that there is an angular momentum difference between ground and 447-kev levels of at least two units if the states have opposite parity. If the assignment of the activity to mass number 131 is correct, then since the angular momentum of the ground state of Xe^{131} is $\frac{3}{2}$, the angular momentum of the 447-kev level must be at least 7/2. The angular momentum of the ground state of I^{131} is then at least 9/2.

Since I^{131} grows from the 25-minute Te^{131}, a consideration of the angular momentum of the ground state of this nucleus is of interest, especially since this nucleus possesses an isomeric state of half-period 1.2 days, 177 kev above the ground state, estimated by Helmholz to have an angular momentum differing from that of the ground state by at least three units.[18] It is not known whether the 25-minute period is accompanied by gamma-radiation. If it should be found not to emit gamma-radiation, then it will be reasonably certain that of the two isomeric states it is the ground state of Te^{131} which has the higher angular momentum.

The mode of disintegration of I^{131} (Fig. 9) seems sufficiently well-established to make it a useful substance for the calibration of the efficiency of gamma-ray counters.

ACKNOWLEDGMENTS

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[14] R. D. Evans, "Introduction to the atomic nucleus," M.I.T. course notes.

[15] S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939).

[16] H. R. Hulme, N. F. Mott, F. Oppenheimer, H. M. Taylor, Proc. Roy. Soc. **A155**, 315 (1936).

[17] I. S. Lowen, Phys. Rev. **59**, 835 (1941).

[18] A. C. Helmholz, Phys. Rev. **60**, 415 (1941).