Disintegration of I^{124} and I^{123}†

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The disintegration of I^{124}(4.2 days) has been studied with the help of magnetic spectrometers and scintillation spectrometers. The disintegration occurs 71% by electron capture and 29% by positron emission. Three positron groups were found having end-point energies of 2130 (46.0%), 1531 (46.4%), and 786 (7.5%) kev. The most energetic positron group has a shape characteristic of $\Delta I=\pm 2$, yes. Positron-gamma coincidence experiments show that this group goes to the ground state. Gamma rays of energies 2700, 2300, 2100, 1700, 1520, 1350, 723, and 603 kev together with annihilation radiation and Te K x-rays have been found and the relative intensities measured. A disintegration scheme, consistent with the levels of Te^{124} as determined from the decay of Sb^{124}, has been established. No beta rays were found, showing that a transition to Xe^{124} is highly improbable. The former work on I^{123}(13.5 hours) has been substantiated and, in addition, it seems highly unlikely that any positrons are emitted from I^{123}.

A. IODINE-124

I. Introduction

THE characteristics of the distintegration of I^{124} were studied by Mitchell, Mei, Maienschein, and Peacock[1] (hereafter referred to as I). They produced I^{124} by bombarding unseparated antimony with alpha particles from the cyclotron. Thus, their sample contained I^{126} and I^{123} as well as I^{124}. They measured, in a magnetic spectrometer, the distribution of positrons from I^{124} together with internal conversion electrons and also photoelectrons produced by gamma rays in a radiator. The disintegration of I^{124} gives the states of Te^{124} which are also produced by the decay of Sb^{124}. The results of the investigation showed gamma rays at 0.603, 0.73, 1.72, and ~1.95 Mev, which are essentually the strong lines seen in the decay of Sb^{124}. The positron distribution showed three groups with end-point energies of 2.20, 1.50, and 0.67 Mev, the highest energy group having a unique first forbidden shape. It was evident that the difference between the end points of the two higher energy groups agreed with neither the 0.603-Mev nor the 0.723-Mev gamma-ray energy. The scheme was drawn up showing the most energetic group going to the first excited state of Te^{124}.

In the interim, Stevenson and Deutsch[2] looked for coincidences between the high-energy positron group and gamma rays and could find none, implying that the highest energy group goes to the ground state. Marquez and Perlman[3] found that I^{124} decayed 70% by electron capture and 30% by positron emission. Recently, the spin[4] of I^{124} has been found to be 2. This makes it appear very likely that the highest energy positron group goes to the ground state. Finally, some difficulties were found in standardizing I^{124} for therapeutic[5] use on the basis of the scheme given in I. In view of these facts, and since techniques in nuclear spectroscopy have improved greatly in the last ten years, it was decided to reinvestigate the disintegration of I^{124}.

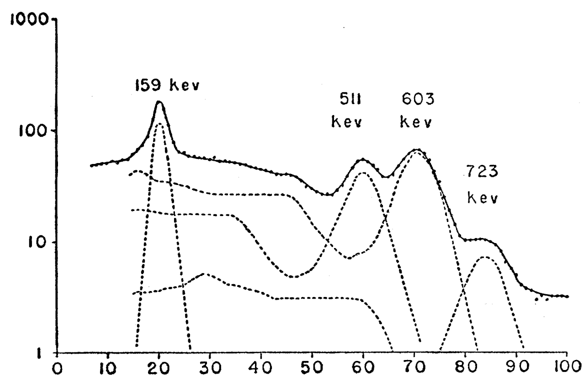

FIG. 1. Scintillation spectrogram of gamma rays of I^{124} (low-energy region).

II. Preparation of Sources; Apparatus

Sources were prepared by bombarding separated Sb^{121} (99.4%), as the metal, with 23 Mev alpha particles in the cyclotron. A small amount of KI, of known iodide concentration, was added to the active material to act as carrier. Nitric acid was added to the mixture and the iodine was distilled over. The iodine was purified by repeated oxidations and reductions and extractions into CCl_4 and water. The purified source was layed down as NaI on thin Zapon and covered by a thin (<10 microgram/cm^2) Zapon film.

The particle spectrum—positrons and internal conversion electrons—was measured with the help of a magnetic lens spectrometer and a 180° magnetic spectrometer. The gamma-ray measurements were carried out with a scintillation spectrometer using either a 1-in. by 1-in. or a 3-in. by 3-in. NaI(Tl) crystal. The scintillation spectrometer had previously been calibrated for

† Supported by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] Mitchell, Mei, Maienschein, and Peacock, Phys. Rev. **76**, 1450 (1949).
[2] D. T. Stevenson and M. Deutsch, Phys. Rev. **83**, 1202 (1951).
[3] L. Marquez and I. Perlman, Phys. Rev. **78**, 189 (1950).
[4] Garvin, Green, and Lipworth, Bull. Am. Phys. Soc. Ser. II, **2**, 383 (1957).
[5] Newbery, Dyson, Francois, and Mallard, Phys. in Med. and Biol. **2**, 72 (1957).

intensity. Beta-gamma coincidences were investigated with the help of a fast-slow coincidence scintillation spectrometer.

In all experiments, there was some I^{123} present from $Sb^{121}(\alpha,2n)I^{123}$. This was present in the early part of the various runs but died out with a half-life of 13.5 hours. Intensity measurements were made on I^{124} after the I^{123} had disappeared. The half-life of I^{124} was found to be 4.2±0.2 days.

III. Results

(a) Gamma-Ray Spectrum

The gamma-ray spectrum was measured using a scintillation spectrometer in conjunction with a 100-channel analyzer. In some instances a 1-in.×1-in. NaI(Tl) crystal was used which had been calibrated for intensity work. For the high-energy gamma rays, a 3-in.×3-in. crystal was used. A copper plate, 0.254 cm thick, was placed directly below and in contact with the source so that annihilation radiation would be produced at a standard position. Corrections to the gamma-ray intensities were made for absorption in the copper. The intensity of the K x-rays was measured without the copper and was checked by an intercomparison with Cs^{137}. The results are shown in Figs. 1 and 2 and the gamma-ray energies and intensities are given in Table I. The error in the intensities is approximately 10 %.

TABLE I. I^{124} energies and relative intensities of gamma rays.

Energy (kev)	Relative intensity	Probability per disintegration
27 (x-ray)	0.88	...
511	0.46	0.29
603	1.00	0.62
723	0.17	0.11
1350 {1326	0.03	0.02 {...
1350 {1361		0.01[a]
1350 {1370		0.01
1520	0.03	0.02
1700	0.13	0.08
2100	0.02	0.01
2300	0.02	0.01
2700	0.01	0.01
	K capture	0.64
	L capture	0.07
	Total electron capture	0.72

[a] From gamma-gamma coincidences.

The relative intensities were determined in tems of the line at 603 kev. In order to determine the number of electron captures, the flouroescent yield correction was applied to the K x-ray intensity of tellurium. In addition, the contribution from L capture was also taken into account. Using the calculations of Brysk and Rose[6] or of Zweifel,[7] the value of L/K for $Z=52$ is 0.115. Thus

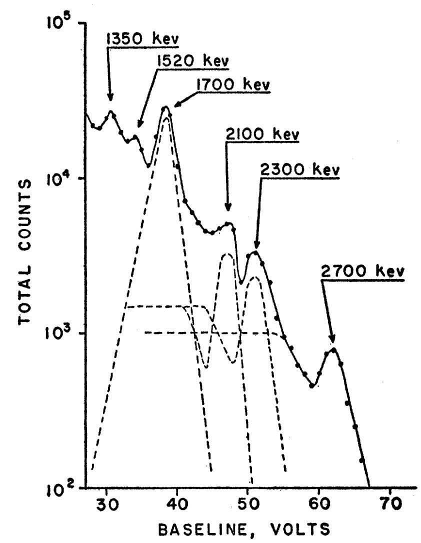

FIG. 2. Scintillation spectrogram of gamma rays of I^{124} (high-energy region).

on the basis of $I_{603}=1.000$, the K capture is 1.030, the L capture 0.119, and the total electron capture 1.149. The probability of positron emission is given by the intensity of annihilation radiation, 0.46. The probabilities per distintegration are thus given in column 3 of Table I. It will be seen at once that the disintegration of I^{124} takes place 71% by electron capture and 29% by positron emission, in agreement with Marqez and Perlman.[3]

The energies of the lines given in Table I agree with those of the stronger lines seen in the decay of Sb^{124}. The latest and most definitive work on Sb^{124} appears to be that of Zolotavin, Grigoryev, and Abroyan.[8] A comparison between their scheme and that derived from the present work on I^{124} will be given below, but it is interesting to note here that the high-energy lines beyond that at 1700 kev have been found.

(b) The Positron Spectrum and Conversion Lines

The particle spectrum (positrons and internal conversion electrons) of the active material was measured in a magnetic lens spectrometer without a baffle to separate positrons from electrons and also in a 180° spectrometer. The spectrum was measured shortly after bombardment, in which case the internal conversion lines for the gamma ray of I^{123} at 159 kev were seen, and again later after the I^{123} had died out. Numerous experiments both on the positrons and the intensity of annihilation radiation showed that the contribution of positrons from I^{123}, if any, is negligible. Figure 3 shows the particle distribution for a source of I^{124}. In addition to the particle distribution, a K line and an L line will

[6] H Brysk and M. E. Rose, Oak Ridge National Laboratory Report ORNL-1830, 1955 (unpublished).

[7] P. F. Zweifel, *Proceedings of the Rehovoth Conference on Nuclear Structure*, edited by H. J. Lipkin (North Holland Publishing Company, Amsterdam, 1958), p. 300.

[8] Zolotavin, Grigoryev, and Abroyan, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **20**, 289 (1956) [Columbia Tech. Transl. **20**, 271 (1956)].

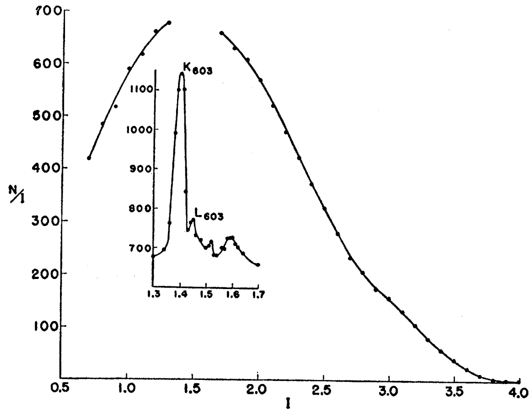

FIG. 3. Positrons and internal conversion electrons from I^{124}.

be seen for the gamma ray at 603 kev and internal conversion lines for the gamma-ray complex at 646, 713, and 723 kev.

A Fermi analysis was made of the positron distribution. A typical analysis of the data from the 180° spectrometer is shown in Fig. 4. The highest energy group (end point 2130 kev) does not have an allowed shape and can be fitted with a unique first forbidden ($\Delta I=2$, yes) shape. This group goes to the ground state of Te^{124}, as will be shown in the beta-gamma coincidence experiments, and the assignment is therefore consistent with the spin and parity (2^-) of the ground state of I^{124}. Two lower energy groups were found as is shown in Fig. 4 and Table II. The results obtained using the magnetic lens spectrometer are substantially in agreement with the values shown in Table II. Owing to the many subtractions involved, it is difficult to say whether there are any positrons emitted to the higher energy states of Te^{124}. If such transitions exist they must be of very low abundance.

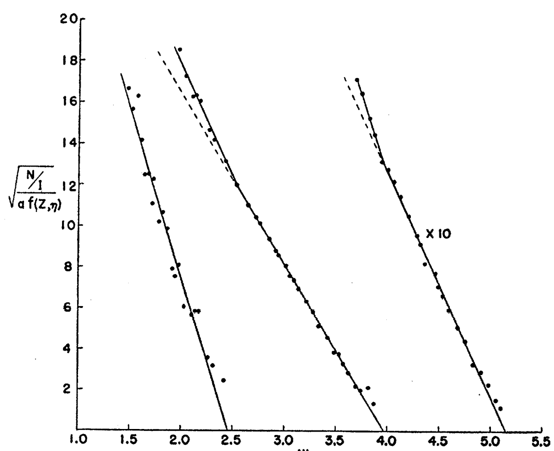

FIG. 4. Fermi analysis of positron distribution from I^{124}. For the highest energy group $a=C_1^{(2)}$ (first forbidden unique); otherwise $a=1$.

On reversing the field of the 180° spectrometer, in order to look at the electron emission, only the internal conversion lines for the 159-kev gamma ray of I^{123} and the 603-kev line of I^{124} were found. There appeared to be no beta-ray emission connected with the 4.2-day disintegration of I^{124}. This is of interest since I^{124} lies between the two stable isotopes Te^{124} and Xe^{124}.

The internal conversion lines shown in Fig. 3 are the K and L lines for the gamma ray at 603 kev and lines for the 658, 713, 723 kev complex. Since the internal conversion coefficient for the gamma ray of energy 603 kev has been measured by Langer, Lazar, and Moffat[9] and by Zolotavin *et al.*,[8] from the decay of Sb^{124}, no attempt was made to make a precision determination of this quantity. A rough check was obtained by measuring the ratio of the number of electrons under the K line to the total number of positrons and comparing this with the ratio of the intensities of the 603-kev gamma-ray line and that of annihilation radiation. The result for the internal conversion coefficient is $\alpha_K=7\times10^{-3}$ which is to be compared with $\alpha_K=3.4\times10^{-3}$ by Langer, Lazar, and Moffat and $\alpha_K=4.26\times10^{-3}$ by Zolotavin *et al.*[8] It is to be remembered that the method

TABLE II. Distribution of positrons from I^{124}.

Energy (kev)	Relative abundance (percent)	Probability per disintegration	log ft	$\log[(W_0^2-1)ft]$
2130±20	46.0	0.131	7.61	9.11
1531±30	46.4	0.133	7.34	···
786±50	7.5	0.022	7.68	···

used here depends on measuring the intensities of two gamma rays with a scintillation counter and is not expected to yield as accurate results as the more direct method used by the other authors.

(c) *Positron-Gamma Coincidence Experiments*

In order to test the disintegration scheme further, positron-gamma coincidence experiments were carried out using two single-channel scintillation spectrometers, one equipped with a NaI(Tl) crystal and the other with an anthracene crystal. Coincident pulses were observed with the help of a fast-slow coincidence circuit. The channel measuring gamma rays was set on the gamma ray at 603 kev and that observing positrons (anthracene crystal) was set at various places, some beyond 1530 kev and some between 800 kev and 1530 kev (second group). The results are shown in Fig. 5 in which the number of coincidences per recorded gamma ray is plotted against the dial setting of the scintillation spectrometer measuring positrons. The number of coincidences beyond the end point of the second group is very small and can be accounted for as being the result of coincidences between

[9] Langer, Lazar, and Moffat, Phys. Rev. 91, 338 (1953).

Compton electrons of gamma rays in coincidence with the 603-kev gamma ray. This experiment shows that the most energetic positron group goes to the ground state and that the next most energetic group goes to the state at 603 kev.

(*d*) *Gamma-Gamma Coincidence Experiments*

No systematic study of gamma-gamma coincidences has been made in this report on I^{124}. It is felt that the angular correlation work of Lindqvist and Marklund[10] on the gamma rays of Te^{124} produced from the disintegration of Sb^{124} gives the essentials of the decay scheme. The gamma ray at 1520 kev was first seen in the decay of Sb^{124} by Dzhelepow and Zhukovsky[11] and also seen in I^{124} by Girgis and van Lieshout.[12] Since neither of the above was able to fit this gamma ray into the scheme in an unambiguous fashion, the present authors performed certain gamma-gamma coincidence experiments involving this line.

From energy considerations alone it appeared not to be possible to fit this line into existing energy states. A group of experiments was arranged to perform positron-gamma coincidences between the line at 1520 kev, observed in a 3-in.×3-in. crystal, and positrons, observed with an anthracene crystal, using a relatively broad energy region. No positron-gamma coincidences were observed, indicating that the line at 1520 kev arises from electron capture and comes from a state high in the disintegration scheme.

Gamma-gamma coincidences were measured with the help of a 3-in.×3-in. crystal for the high-energy lines, in order to resolve the 1520-kev line from the much stronger 1700-kev line, and a $1\frac{1}{2}$-in.×$1\frac{1}{2}$-in. crystal for the low-energy lines. Since the line at 723 kev is not in coincidence with that at 1700 kev, one channel was set on the peak of that line and the other channel was swept through the high-energy region. The results are shown in Fig. 6(A) in which it will be seen that the line at 723 kev is in coincidence with that at 1520 kev and also with one at ~1300–1400 kev. The latter is to be expected from the decay scheme of Zolotavin *et al.*[8] The fixed channel was then set on the line at 603 kev and the same high-energy region was investigated. It was found that there are coincidences between the line at 603 kev and that at 1520 kev [Fig. 6(B)], although they are somewhat swamped by the large coincidence rate between the 603-kev and the 1700-kev lines. It would, therefore, appear that the line at 1520 kev ends on the level at 1326 kev, from which the line at 723 kev follows, and comes from a level at approximately 2850 kev. Any direct transition from this high state to the ground state would probably not be resolved from the line at 2700 kev seen in the scintillation spectrum. From a comparison of the coincidence data and the singles spectrum, it is estimated that the line labeled as 1361 in the decay scheme is approximately the same intensity as that labeled 1370. The energy values are taken from Zolotavin *et al.*

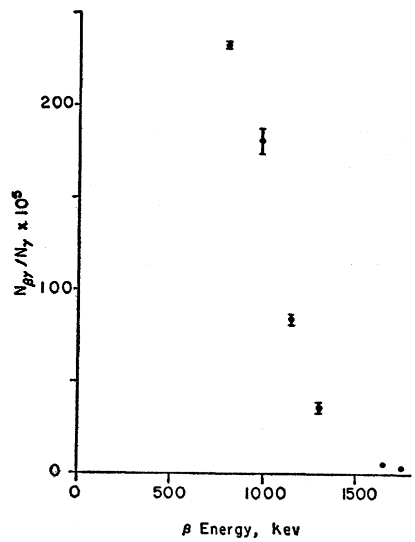

FIG. 5. Positron-gamma coincidences in I^{124}.

IV. Disintegration Scheme

The main features of the levels of Te^{124} are reasonably well known from work on Sb^{124}. Some of the many weak gamma rays, recently discovered, have added complications to the level system[11] which are difficult to substantiate with present-day techniques. The main problem here is to see whether the results obtained from the

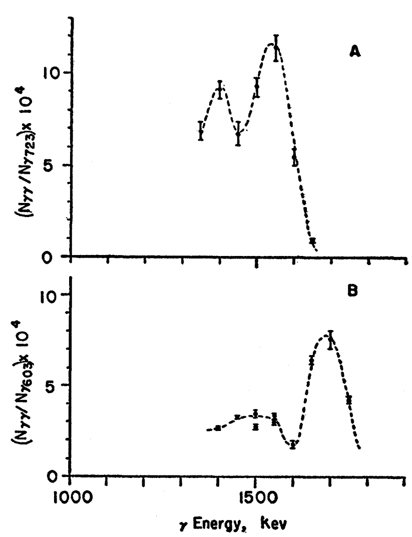

FIG. 6. Gamma-gamma coincidences (A) between the 723-kev and the 1520-kev line; (B) between the 603-kev and the 1520–1700-kev group.

[10] T. Lindqvist and I. Marklund, Nuclear Phys. 4, 189 (1957).
[11] B. S. Dzhelepow and N. N. Zhukovsky, Nuclear Phys. 6, 655 (1958).
[12] R. K. Girgis and R. van Lieshout, reported in Strominger, Hollander, and Seaborg, Revs. Modern Phys. **30**, 585 (1958).

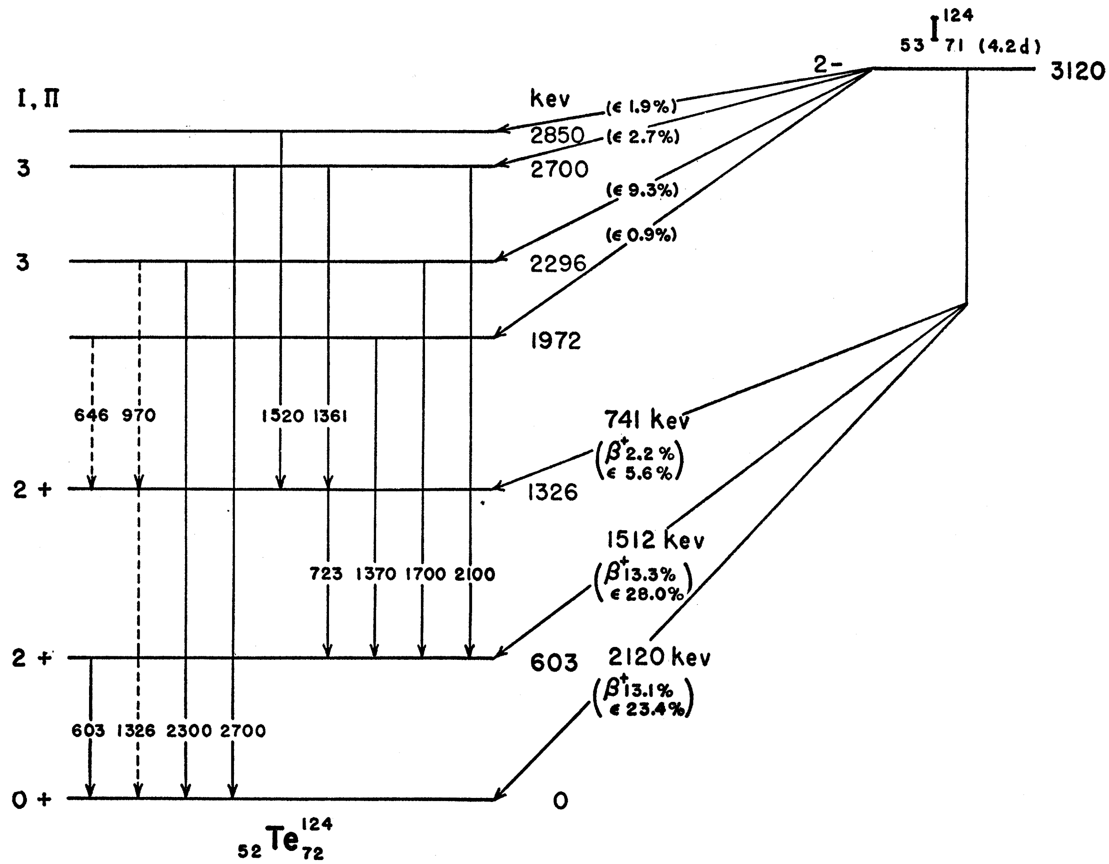

FIG. 7. Disintegration scheme of I^{124}.

disintegration of I^{124} agree with the levels of Te^{124} as determined from the disintegration of Sb^{124}. It is, of course, not to be expected that the relative intensities of the various gamma rays will agree.

Figure 7 shows the agreement of the results obtained herein with the scheme of Zolotavin *et al.*[8] It is to be noted that the most energetic positron group goes to the ground state and that the end-point energies of the next two groups are consistent with the energies of the levels of Te^{124}. The gamma rays seen in this experiment are shown by solid lines. Lines, unresolved in these experiments but resolved by Zolotavin *et al.*, are shown by dotted lines. The spin assignments are based on the work of Lindqvist and Marklund and of Zolotavin *et al.* The line at 1520 kev is placed in the scheme as a result of the coincidence experiments mentioned above. The values of the probability of electron capture and positron emission (given in percent of total disintegrations) from I^{124} to the various levels of Te^{124} were calculated with the help of Table I and Table II, and are shown, for each of the levels involved, in Table III along with the values of log *ft*.

TABLE III. Values of probability of electron capture and positron emission together with values of log *ft* for I^{124}.[a]

Level (kev)	Electron capture plus positron emission (percent)	log *ft*	$\log[(W_0^2-1)ft]$
2850	1.9	6.6	
2700	2.7	6.8	
2296	9.3	6.9	
1972	0.9	8.2	
1326	7.8	7.7	
603	41.3	7.3	
0	36.5	7.6	9.1

[a] For the lowest three levels the figures given are for $\epsilon+\beta^+$, and $f=f_+ + f_K$; otherwise figures are ϵ only and $f=f_K$.

B. IODINE-123

Since I^{123} is produced by the $Sb^{121}(\alpha,2n)I^{123}$ reaction, the properties of this isotope were reinvestigated here with substantially the same results found in I.

The only radiation associated with the decay of I^{123} (13.5 hr) is a gamma ray of energy 159 kev. Positrons were looked for by measuring the positron spectrum of the I^{123}–I^{124} mixture as a function of time and also by measuring the relative intensity of the gamma ray at 159 kev to annihilation radiation of the mixture. Both the annihilation radiation and the positron spectrum decayed with the half-life characteristic of I^{124}. The evidence tends to support the supposition that I^{123} decays entirely by electron capture.

The energy of the gamma ray determined from the internal conversion spectrum and from the photoline as seen in the scintillation spectrum is 159 ± 1 kev. The $K/(L+M)$ ratio for the internal conversion line is 6.6 ± 0.1.

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Spacings of Nuclear Energy Levels

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A simple statistical model is suggested in terms of which the general features of the level spacing distribution can be understood, and which involves no special assumptions, other than that of Porter and Thomas.

INTRODUCTION

In the last few years a wealth of experimental data concerning the widths and spacings of nuclear energy levels has become available.[1] The initial theoretical response to the information these data supplied was directed toward the understanding of the statistical properties of the neutron widths[2]; and these investigations culminated in the very successful paper of Porter and Thomas.[3] These authors inferred a normal distribution for the reduced neutron width from the plausible assumption of a highly complex, rapidly varying wave function for compound nuclear states, which is not highly correlated with the wave functions of nearby states. In their paper, Porter and Thomas show their inference to be strongly supported by experimental evidence.

With the success of this simple approach in mind, Wigner suggested an analogous examination of the distribution of level spacings.[4] In particular, he pointed out that the distribution of spacings between adjacent eigenvalues of matrices whose elements were randomly chosen would show a deficiency of small spacings, contrary to the expectation if the eigenvalues themselves were uncorrelated. This so-called "level repulsion" effect has been observed experimentally in connection with the nuclear resonance levels.[5] Blumberg and Porter[6] demonstrated the level repulsion effect numerically by diagonalizing random matrices of fairly large order, all of whose elements had the same normal distribution. Rosenzweig[7] made more accurate numerical calculations of the same type and obtained a rather detailed histogram which agreed very well with the distribution of spacings originally suggested by Wigner [see Eq. (8)], except for the largest spacings.†

It is the purpose of this paper to indicate a simple statistical model in terms of which the general features of the level spacing distribution can be understood, and which involves no special assumptions, other than that of Porter and Thomas.[3]

THEORY

Nuclear resonance states are determined by the zeros of the function[8]

$$f_c(E)\equiv\int_S\left(\frac{\partial X}{\partial n}+b_cX\right)\Phi_c dS, \tag{1}$$

where E is the energy of the bombarding particle; X is a solution of the Hamiltonian equation $HX=EX$ inside the nuclear surface, S, in configuration space; n is the outward normal to S; Φ_c is a channel wave function in channel c; and b_c is a real arbitrary constant. By appropriate choice of b_c the level shift may be made

* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

[1] J. A. Harvey and D. J. Hughes, Phys. Rev. **109**, 471 (1958), and references cited therein.

[2] J. A. Harvey and D. J. Hughes, Phys. Rev. **99**, 1032 (1955), and references cited therein.

[3] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).

[4] E. P. Wigner, Oak Ridge National Laboratory Report ORNL-2309, November 1, 1956 (unpublished), p. 67.

[5] J. A. Harvey, Phys. Rev. **98**, 1162 (1955); I. I. Gurevich and M. I. Pevsner, J. Exptl. Theoret. Phys. U.S.S.R. **31**, 162 (1956) [translation: Soviet Phys. JETP **4**, 278 (1957)]; also Nuclear Phys. **2**, 575 (1957).

[6] S. Blumberg and C. E. Porter, Phys. Rev. **110**, 786 (1958).

[7] N. Rosenzweig, Phys. Rev. Letters **1**, 24 (1958).

† *Note added in proof.*—Professor E. P. Wigner kindly pointed out to the author that this slight disagreement arose from a failure to account for the dependence of the local average level spacing on the eigenvalue (energy). When Rosenzweig's results were corrected for this effect the disagreement for large spacings was removed.

[8] R. G. Sachs, *Nuclear Theory* (Addison-Wesley Publishing Company, Inc., Cambridge, 1955), p. 291.