with

with.

several valuable suggestions.

(b) Finally, it seems at least possible that the ionizing source might decay quadratically as would be the case in reaction (14) if the removal of the metastables were controlled by this process. The metastables themselves would then disappear according to the simple recombination equation and we would have to write

$$
q = \beta M^2 = \beta (1/M_0 + \beta t)^{-2}, \tag{25}
$$

where again  $\beta$  is the probability that a metastable atom is colliding with another one causing the reaction (14) and  $M_0$  is the initial concentration of such excited atoms. with another one causing the reaction (14)<br>the initial concentration of such excited<br>iq. (17) then has the solution  $X = C_1 M^{\frac{1}{4}(1+d)}$ ,<br>where  $d^2 = 1 + 4\alpha/\beta$ , so that<br> $n = \frac{\beta M}{2\alpha} \times \frac{(1-d)M^d - (1+d)C}{M^d - C}$ , (26)

For  $\delta=0$  Eq. (17) then has the solution  $X=C_1M^{\frac{1}{2}(1+d)}$  $+C_2M^{-\frac{1}{2}(1-d)}$ , where  $d^2=1+4\alpha/\beta$ , so that

$$
n = \frac{\beta M}{2\alpha} \times \frac{(1-d)M^d - (1+d)C}{M^d - C},
$$
 (20)

PHYSICAL REVIEW VOLUME 84, NUMBER 2 OCTOBER 15, 1951

# The Decay of  $Sr^{87m}$ ,  $Y^{87m}$ , and  $Y^{87}$

LLOYD G. MANN\* AND PETER AXEL Physics Department, University of Illinois, Urbana, Illinoist (Received June 29, 1951)

The 2.8-hr Sr<sup>87</sup>, 80-hr Y<sup>87</sup>, and 14-hr Y<sup>87</sup><sup>m</sup> activities were studied by means of a thin lens beta-ray spectrometer, Geiger counters in coincidence, and scintillation crystals.

The following results were obtained: (a)  $Sr^{87m}: t_1 = 2.80 \pm 0.05$  hr,  $\gamma$ -energy=390 $\pm 2$  kev, K conversion coefficient=0.24 $\pm$ 0.05, ratio of K to  $(L+M)$  conversion=6.9 $\pm$ 0.4; (b) Y<sup>87</sup>:  $t<sub>i</sub>$ =80.0 $\pm$ 1 hr, decays to  $Sr<sup>87</sup>$ , more than 99 percent K-capture, 0.7-Mev positrons, gamma-ray follows K-capture with energy =485 $\pm$ 3 kev and K conversion coefficient=3.2 $\pm$ 0.7 $\times$ 10<sup>-3</sup>; (c) Y<sup>87m</sup>: t<sub>1</sub>=14 $\pm$ 1 hr,  $\gamma$ -energy=384 $\pm$ 3 kev (definitely different from Sr<sup>87m</sup>  $\gamma$ -ray), K conversion coefficient=0.24 $\pm$ 0.07.

These data are compared with the predictions of the theories of internal conversion, nuclear isomeric transitions, shell structure, and beta-decay. The results are incorporated in a decay scheme and probable spin and parity assignments are given. Conventional beta-decay theory and the spin orbit coupling shell model give incompatible assignments to the energy levels in Ys'.

## INTRODUCTION

HIS experiment was undertaken to establish the  $\bf{l}$ decay scheme for  $Y^{87m}$ ,  $Y^{87}$ , and  $Sr^{87m}$ . From the data that were available in the literature, it seemed likely that a careful investigation of these activities would produce convincing checks of several different theories. These earlier data will be given below in the course of the presentation of our own data. The results of our investigation improved the precision of the known data and removed several inaccuracies and inconsistencies which had existed.

The data which were obtained are summarized in the decay scheme shown in Fig. 1.The description and discussion of the results will be presented in three sections: (1) general experimental findings for each

activity, (2) measurement and interpretation of conversion coefficients, and  $(3)$  comparison of results with the theory of nuclear isomers, the spin orbit shell. structure model, and beta-decay theory.

 $C = M_0^{\frac{2\alpha n_0 - (1-d)\beta M_0}{\alpha}}$ 

This relation consists of elementary functions only and should, for any specific case, be relatively easy to analyze. But since this case probably is of minor importance a more detailed discussion shall be dispensed

In general, however, it can be seen that in some cases finite solutions of more complex recombination problems do exist and it is felt that these rather than numerical integrations or more crude approximations should be used whenever possible. The writer wishes to thank Professor L.B. Loeb, who initiated this analysis, for his advice and guidance and also Professor S. C. Brown for

 $\frac{2\alpha n_0-(1+d)\beta M_0}{h}$ 

The incompatibility between the assignments in  $Y^{87}$ from beta-decay theory and shell structure does not depend on detailed quantitative analysis. The discrepancy is gross and would require either drastic changes in the decay scheme or a modification of one of these theories.

#### SOURCE PREPARATION

 $Y^{87}$  and  $Y^{87m}$  were produced by cyclotron bombardments of ordinary metallic strontium and chemically pure strontium nitrate with protons or deuterons.<sup>1</sup> The bombardments using 10-Mev deuterons were most satisfactory, since they produced less of the unwanted

<sup>\*</sup>Now at Stanford University, Stanford, California. Some of the material of this paper was included in the doctorate thesis of L.G.M.

t This research was supported by the joint program of the ONR and ABC.

The authors are indebted to the cyclotron staffs at the University of Chicago and the University of Washington in St. Louis for these bombardments.



FIG. 1. Decay scheme of  $Y^{87m}$ ,  $Y^{87}$ , and  $Sr^{87m}$ . Of the two possible assignments given for the states of  $Y^{87}$ , those taken from the shell model theory seem more reasonable.

100-day  $Y^{88}$ . A 400-microampere hour bombardment gave about two millicuries of 80-hr  $Y^{87}$  activity. In the course of a short bombardment about equal numbers of radioactive  $Y^{87}$  nuclei are formed in the 14-hr and 80-hr states.

The radioactive Y was separated from the bombarded Sr by precipitating  $Y(OH)$ <sub>3</sub> from an HCl solution of Y



FIG. 2. Conversion electron spectrum of the 390-kev gammaray. The dotted curves indicate the contributions due to the  $K$ and  $(L+M)$  electrons.

and Sr using NH<sub>4</sub>OH. The effectiveness of this procedure was tested by checking the radioactivity resulting from both an  $Y^{90}$ -Sr<sup>90</sup> separation and an  $Y^{87}$ -Sr<sup>87</sup> separation. These tests showed that the separation was good to 2 percent after a single precipitation and 0.1 percent after a repetition.

Additional chemical purification was necessary to produce sufficiently thin beta-spectrometer sources. Considerable inactive iron was found (presumably introduced in the preparation of cyclotron targets). The iron was separated from the yttrium by performing the hydroxide precipitation of  $Y(OH)_3$  in an oxalic acid solution.<sup>2</sup>

### 2.80-hr  $Sr^{87m}$

The present investigation of  $Sr^{87m}$  was undertaken mainly to obtain an accurate experimental value for the internal conversion coefficient. In addition, the reports of other investigators were in general confirmed, discrepancies were removed and the experimental determinations were made with improved precision.

This 2.8-hr activity was first reported by Stewart, Lawson, and Cork<sup>3</sup> and confirmed by Stewart<sup>4</sup>; its isomeric nature and isotopic assignment were shown by Lawson, and Cork<sup>s</sup> and confirmed by Stewart<sup>4</sup>; its<br>isomeric nature and isotopic assignment were shown by<br>DuBridge and Marshall.<sup>5,6</sup> In addition, Helmholz,<sup>7</sup> Robertson, Scott, and Pool,<sup>8</sup> the present authors,<sup>9</sup> and Hyde and O'Kelley" have published data on this activity.

Investigation of this Sr isomer, separated chemically from its 80-hr Yparent, showed the presence of Sr Auger electrons,  $K$  and  $L$  conversion electrons, x-rays, and gamma-rays. Each electron line was observed to decay with the proper half-life in the spectrometer. This double thin lens spectrometer was adjusted to have a resolution of 2.8 percent and a transmission of about 1 percent. For a precise energy determination the Sr conversion electrons were studied in equilibrium with the 80-hr  $Y^{87}$  parent source. The gamma-ray energy was determined as  $390\pm 2$  kev using the 624-kev conversion electrons of Cs<sup>137</sup> as a standard.<sup>11</sup> Other reported determinations of this energy are 386 kev' reported determinations of this energy are 386 kev<br>and  $394 \pm 4$  kev.<sup>10</sup> The K to  $L+M$  ratio was determine by analyzing the partially resolved conversion line as shown in Fig. 2 and comparing peak heights. The shape of the upper energy edge of the  $K$  conversion line was obtained from the  $K$  conversion line of the 88-kev gamma-ray of Ag<sup>109</sup> which is completely resolvable from the  $L$  line in our spectrometer. The value of the  $K$  to  $L+M$  ratio was determined as 6.9 $\pm$ 0.4; two other

- <sup>3</sup> Stewart, Lawson, and Cork, Phys. Rev. 52, 901 (1937).<br><sup>4</sup> D. W. Stewart, Phys. Rev. 56, 629 (1939).
- 
- L. A. DuBridge and J. Marshall, Phys. Rev. 56, <sup>706</sup> (1939). ' L. A. DuBridge and J. Marshall, Phys. Rev. 58, <sup>7</sup> (1940). '
- 
- <sup>7</sup> A. C. Helmholz, Phys. Rev. 60, 415 (1941).
- 
- 
- <sup>8</sup> Robertson, Scott, and Pool, Phys. Rev. **78**, 318 (1950).<br><sup>9</sup> L. G. Mann and P. Axel, Phys. Rev. 80, 759 (1950).<br><sup>10</sup> E. K. Hyde and G. D. O'Kelley, Phys. Rev. 82, 944 (1951).<br><sup>11</sup> L. M. Langer and R. D. Moffat, Phys. R
- 

<sup>&</sup>lt;sup>2</sup> M. J. Glaubman developed the purification procedure and performed much of the necessary chemical work

reported values are 6 to  $7^7$  and  $7.2.^{10}$  A search was also made for the conversion electrons of the 485-kev gamma-ray, but these were not present in a  $Sr^{87m}$  source. The half-life was measured with Geiger counters which detected the x-rays, gamma-rays, conversion electrons, and the electron x-ray coincidences as shown in Fig. 3. The x-ray was differentiated from the gamma-ray by using absorbers. In addition, the electrons from one source were followed with a Geiger counter for 9 halflives. The value of the half-life is  $2.80 \pm 0.05$  hr in excellent agreement with  $2.75 \pm 0.1$  hr<sup>6</sup> and  $2.80 \pm 0.03$ excellent agreement with  $2.75 \pm 0.1$  hr<sup>6</sup> and  $2.80 \pm 0.03$ <br>hr.<sup>10</sup> The total conversion coefficient was measured as  $0.28 \pm 0.06$  by a coincidence method which depends on the decay scheme and which will be discussed below. This value is more precise than the previously measured value of 0.15.' As would be expected from Fig. <sup>1</sup> neither  $x-x$  nor  $x-y$  coincidences were observed. The relative intensities of the x-ray, gamma-ray, and conversion electrons are determined in the course of the measurement of the conversion coefficient. The absence of any other radiations is also indicated by these intensity measurements.

## 80-hr Y<sup>87</sup>

The detailed investigation of the 80-hr transition showed the presence of a gamma-ray of 485 kev which had not been reported in this activity. The 80-hr activity was first reported by Stewart<sup>4</sup> (82 $\pm$ 4 hr) and assigned to  $Y^{87}$  by DuBridge and Marshall<sup>6</sup> (80 $\pm$ 3) hr. X-rays, gamma-rays, and  $x-\gamma$  coincidences as well as the  $Sr^{87m}$  daughter electrons and  $e-x$  coincidences were followed for more than three half-lives giving a value  $80.0\pm1$  hrs. Representative curves are shown in Fig. 4 and Fig. 8. (In these and other gamma-ray or x-ray curves, it was necessary to subtract the contribution of the 100-day  $Y^{88}$  which was present.)

The evidences that the 485-kev gamma-ray was associated with the 80-hr activity are given by the following data:

1. The  $x-\gamma$  coincidence rate observed with an 80-hr half-life in Fig, 4 is not present in either the 2.8-hr or 14-hr activities.

2. An 80-hr Y source which had the Sr removed at zero time showed the initial gamma-ray activity (presumably the 485-kev gamma-ray) and the growth of the 2.8-hr Sr activity (the 390-kev gamma-ray). The data taken on a NaI scintillation detector are shown in Fig. 5.

3. The 469-kev  $K$  conversion electrons were observed in the spectrometer with an  $Y^{87}$  source and were not observed with a  $Sr^{87m}$  source. This conversion peak was followed for 200 hours and decayed with an 80-hr half-life.

4. The 469-kev  $K$  conversion electrons were observed in the spectrometer and exhibited a 14-hr growth in a freshly bombarded V source, followed by an 80-hr decay, as shown in Fig. 6.



FIG. 3. Decay curves of 2.8-hr  $Sr^{87m}$ . A. Electrons  $\times T_0$ . B.  $e^- - x$  coincidences  $\times$  64. C. X-rays. D. Gamma-rays counted in x-ray counter.

These data seem conclusive despite the fact DuBridge and Marshall<sup>6</sup> saw no 80-hr gamma-rays and that Robertson, Scott, and Pool<sup>8</sup> associate this gamma-ray with the 2.8-hr daughter activity. Hyde and O'Kelley<sup>10</sup> did not see the conversion electrons but indicate that they may have overlooked them, considering the low intensity.

The ratio of the 469-kev electrons to the 374-kev electrons of the Sr daughter is  $1/(59\pm2)$  as measured by peak counting rates. This gives a value of the  $K$ 



Fro. 4. Decay curves of photons in 80 hr Y<sup>87</sup>. A. X-rays.<br>B. Gamma-rays. C. X- $\gamma$  coincidences X640. A 100-day Y<sup>88</sup> background has been subtracted.



FIG. 5. Growth of 2.8-hr Sr<sup>87</sup><sup>m</sup> gamma-rays into an 80-hr Y<sup>87</sup> source. Detector: 3-cm NaI scintillation crystal. A. Equilibrium activity extrapolated to zero time. B. Observed gamma-ray counting rate. C. Difference curve  $(A - B)$ , showing a half-life of  $2.6\pm0.3$  hr.

conversion coefficient of the 485-kev gamma-ray as of  $3.2\pm0.7\times10^{-3}$ . It should be noted that most of the error in the determination of this conversion coefficient is directly related to the error in the 390-kev gamma-ray conversion coefficient.

In addition to the radiations already mentioned a very weak positron spectrum (less than 1 percent of total conversion electrons) of maximum energy about 0.7 Mev was observed in the spectrometer. Because of the small intensity it was necessary to use a thick source and the resultant spectrum was very poor. These positrons were too weak to be followed for more than two half-lives but they seemed to decay with an 80-hour half-life. These positrons were also reported by Robertson, Scott, and Pool.<sup>8</sup>

#### 14-hr  $Y^{87m}$

In the course of this investigation, the genetic relationship between the 14-hr and 80-hr Y was definitely established.<sup>9</sup> The energy of the transition was measured and the fact that it differed from the  $Sr^{87m}$ gamma-ray by only 6 kev was confirmed. This fact was<br>first reported by Hyde and O'Kelley.<sup>10</sup> The interna. first reported by Hyde and O'Kelley.<sup>10</sup> The interna conversion coefficient was also measured with respect to the  $Sr^{87m}$  conversion coefficient and found to be about the same.

The 14-hr activity was first reported by Stewart<sup>4</sup>  $(14\pm2)$  hr) and later correctly assigned by DuBridge and Marshall<sup>6</sup> as an excited isomeric level in  $Y^{87}$ , genetically related to the 80-hr ground state. However, DuBridge and Marshall reported a 0.5-Mev gamma-ray and some conversion electrons. Their difIiculty in establishing the character of the radiations more precisely was due to the masking effect of the 2.8-hr Sr which quickly grows into the 80-hr ground state. Robertson, Scott, and Pool<sup>8</sup> postulated the decay of the 14-hr state to the isomeric level of Sr. In an earlier report,<sup>9</sup> we also postulated some branching directly to  $Sr^{87m}$  because of our inability to resolve the two electron lines, 6 kev apart (1 percent in momentum). All of the aforementioned investigators had used cyclotron bombarded Sr and had, therefore, both the 14-hr and 80-hr activities to contend with.

The most conclusive proof of the existence of a 14-hr gamma-ray activity of only slightly less energy 14-hr gamma-ray activity of only slightly less energy than the 2.8-hr Sr was given by Hyde and O'Kelley.<sup>10</sup> They used 100-Mev protons to bombard niobium and produced 94-minute Zr<sup>87</sup>. This positron emitter decays predominantly to the 14-hr Y state. With this source it was possible to observe the 14-hr conversion line, follow its decay and also to watch the growth of 80-hr activity together with its 2.8-hr Sr daughter. In addition, Hyde and O'Kelley used a spectrometer of higher resolution and were able to resolve both lines even when they were present in equal intensity.

The present investigation with the beta-ray spectrometer and with Geiger counter coincidence experiments established the following facts, before the14-hr and 2.8-hr electron lines were resolved:

i. The 80-hr activity grew from the 14-hr activity as indicated by the  $469$ -kev  $K$  conversion line shown in Fig. 6.

2. There were 14-hr x-rays, gamma-rays, electrons, and electron-x-ray coincidences as shown in Figs. 7 and 8.

3. There were no detectable 14-hr x-gamma coincidences.

While these data would have been consistent with the branching postulated earlier, $\frac{1}{2}$  there were two contradictory indications: (1) It was impossible to observe the predicted 2.8-hr growth in a 14-hr  $Y^{87m}$ source after Sr had been separated. (2) The efficiency for detecting the 14-hr x-ray in a krypton-filled Geiger counter was greater by a factor of 4 than that of the 2.8-hr x-ray. (This indicated that the 14-hr x-rays were Y x-rays, since these are detected much more efficiently by krypton. )

To resolve the two gamma-rays, a fresh  $Y^{87}$ ,  $Y^{87m}$ source was obtained and studied in the spectrometer as soon as possible after the Sr had been chemically removed. While two conversion lines could not be resolved, it was obvious that the low energy side of the line was decaying with about a 14-hr half-life and that the high energy side of the line grew in rapidly (presumably 2.8-hr) and then grew more slowly. The line shape was followed in detail for about 2 half-lives (7

days). The 80-hr activity was then corrected back in time using the 14-hr growth relation of the type shown in Fig. 6. The extrapolated 80-hr activity was subtracted from the unresolved lines and an electron line was found about 6 kev below the  $Sr^{87m}$  line. It was planned to repeat this procedure, but before this was done, the report of Hyde and O'Kelley<sup>10</sup> became available.

Our resultant value for the 14-hr gamma-ray transition is  $384\pm3$  kev. This value is to be compared with 389 kev reported by Hyde and O'Kelley. (It should be noted that the calibration of the spectrometer used by Hyde and O'Kelley is evidently 4 or 5 kev above the one used in this investigation. ) Our subtraction process is not precise enough to determine a  $K/L$  ratio for comparison with the value of 8.3 given by Hyde and O'Kelley.

In order to determine the contribution of the 14-hr activities to the various Geiger counter rates shown in Figs. 7 and 8 it was necessary to take into account the growth of the 80-hr activity (as indicated by the dotted curves, which were patterned after Fig. 6). This subtraction necessarily limited the precision of the 14-hr measurements; our value for the lifetime is  $14\pm 1$  hr. The 14-hr activity measurements were further complicated by the existence of some of the 2- or 3-hr x-ray and gamma-ray activities in Y resulting from deutero<br>bombardment of  $Sr<sup>3,4,6</sup>$  However, these short-live bombardment of Sr.<sup>3,4,6</sup> However, these short-live activities disappeared completely and ceased to influence the experiments after about 15 hr.

In addition to the observed x-rays, gamma-rays, and electrons which will be discussed more quantitatively below, a 14-hr positron spectrum of maximum energy about 1.1 Mev was observed. However, Hyde and  $O'K$ elley<sup>10</sup> convincingly assign these positrons to 14.6-hr  $Y^{86}$  which would have been produced in our strontium target by either a  $(d,2n)$  or a  $(p,n)$  reaction.

No other 14-hr activities were found. Specifically, we did not see any trace of the high energy conversion did not see any trace of the high energy conversion<br>electrons reported by Hyde and O'Kelley.<sup>10</sup> While we may have overlooked the low intensity conversion electrons they reported in the region above 1 Mev, we would have seen any appreciable gamma-ray intensity in our observations with Geiger counters (from curves such as shown in Fig. 7). In addition, if these gammarays followed the 384-kev gamma-rays, appreciable 14-hour  $x-\gamma$  coincidences would have resulted. Neither our data nor any reasonable decay scheme consistent with our data give any indication of high energy gamma-rays originating in 14-hr  $Y^{87m}$ .

#### LIMITS ON UNREPORTED BRANCHINGS

The 2.8-hr Sr isomer was investigated carefully to show that the conversion electrons of the 485-kev gamma-ray were not present. In addition, the coincidence data indicated a one-to-one correspondence between x-rays and electrons to within 5 percent. No experiments would have detected weak branches of

other gamma-rays emanating from this state, if they gave many fewer conversion electrons than the 485-kev gamma-ray from 80-hr Y.

Specihcally, the data gave no evidence for low energy excited levels which would be necessary to explain gamma-rays following beta-decay in Rb<sup>87</sup>. It is particularly difficult to explain the existence of these gammarays in a Rb<sup>87</sup> source, since any state to which Rb<sup>87</sup> (spin 3/2) decays would probably also be reached from the 2.8-hr Sr state. It is energetically possible for the 2.8-hr Sr state to decay by K-capture to  $Rb^{87}$ . However, the energy available is so low that this mode of decay would be less than 1 percent and undetectable in our experiments.

The 80-hr  $Y^{87}$  activity was studied in sufficient detail to rule out the presence of any conversion lines except those due to the 485 kev and the 390 kev from the Sr daughter. The 390-kev transition was observed to grow into the. 80-hr source. A very careful search was made for conversion electrons of very low energy in a combined 14-hr, 80-hr, 2.8-hr source; however, only Auger electrons were observed.

A freshly prepared 80-hr Y source was followed using a NaI scintillation detector to observe the relative proportion of the 485-kev gamma-ray and the growing 390-kev gamma-ray. Using the measured conversion



FIG. 6. Fourteen-hr growth and 80-hr decay of the conversion<br>electrons from the 485-kev gamma-ray. Electrons viewed in spectrometer. A. Equilibrium activity extrapolated back to zero time. B. Observed counting rate. C. Difference curve  $(A - B)$  showing the 14-hr half-life. Curves B and C are extrapolated back to the time of bombardment.



FIG. 7. (a) Decay of gamma-rays from Y<sup>87</sup> source. The 2.8-hr Sr<sup>87m</sup> daughter was in equilibrium. A. Actual total counting rate. B. Long-lived activity extrapolated back to zero time, after the 100-day  $\gamma$ -ray background was subtracted. C. Known 14-hr growth of 80-hr activity. D. Difference curve showing 14-hr (384-kev) γ-ray contributio<br>to total counting rate. (b) Decay of x-rays and x-γ coincidences in Y<sup>87</sup> source. A. X-γ coincidences×640. B. x-rays. C. Long-lived x-ray activity extrapolated to zero time after the 100-day background was subtracted. D. Known 14-hr growth of 80-hr x-ray activity. E. Difference curve showing 14-hr x-ray contribution.

coefficient (which is based only on the one-to-one correspondence between  $K$ -capture in 80-hr Y and growth of 2.8-hr Sr) it was possible to show the equality of intensity to within about 8 percent. Except for this equality there can be no direct experimental evidence that the 390-kev state was formed through the 875-kev state. However, the multipolarity of the 485-kev gamma-ray and shell structure both add support to this contention. None of the experimental data would have indicated a small  $K$ -capture branch directly to the ground state. However, if the other aspects of the decay scheme are accepted, the equality of the intensities of the 390-kev and 485-kev gamma-rays put a i0 percent upper limit on this branch. Of course, it is improbable for the 80-hr Y to have appreciable branching to both members of the Sr<sup>87</sup> isomeric pair.

The positrons associated with the 80-hr Y were placed in the decay scheme mainly on the basis of the extremely high  $K$ -capture to positron ratio. This ratio would not be theoretically understandable if both  $K$ capture and positrons led to the same state. Although the decay scheme shown is not consistent with betadecay theory, at least it is consistent with expected  $K$ capture to positron ratios.

The only aspect of the 14-hr decay which has not yet been mentioned is its possible branching directly to the ground state of Sr. Although e-x coincidence data were taken, the relative inefficiency of the detector for Sr x-rays prevents our reducing the upper limit of this

direct  $K$ -capture branch below about 15 percent. However, even if all the positrons we observed were due to  $Y^{87m}$ , instead of  $Y^{86}$ , they would have represented only a 1 percent branch. The theoretical prediction for the  $K$ -capture branch is about the same value. Furthermore, Hyde and O'Kelley<sup>10</sup> report that the positrons were less than O.i percent of the electrons; this corresponds to a branching of less than 0.025 percent.

#### INTERNAL CONVERSION COEFFICIENTS

A measurement of the internal conversion coefficient is essentially a determination of the branching ratio between conversion electrons and gamma-rays. Thus, it is necessary to know the absolute detection efficiency for the  $K$  conversion electrons or for the  $K$  x-rays which accompany them and for the gamma-rays. While it is usually very difficult to determine the absolute detection efficiency, in the case of some simple known decay schemes coincidence experiments can be used to obtain efficiencies directly. The efficiencies determined in this manner include all of the corrections which are usually very difficult to determine, such as the absorption of the radiation by the source or any absorbers, the solid angle factor, the absolute efficiency of detector for a radiation which reaches it and most of the possible electronic idiosyncracies of the recording circuits.

In these experiments the electrons were detected by a commercial bubble side window Geiger counter whose window thickness was about 2.5 mg/cm<sup>2</sup> (counter 1).



I'io. 8. Decay of elecric. o. Decay of elec-<br>trons and  $e^- - x$  coincidences in  $Y^{87m}$  source. A. Observed coincidence counting rate $\times$ 64. B. Known growth of 80-hr coincidences from 14-hr activity. C. Difference curve {A—B), showing 14-hr (13.6-hr) contribution to coincidences (X64). A', B', and C' are similar curves for electron counting rate  $\times\frac{1}{10}$ .

The x-rays were detected in a Geiger counter (counter 2) filled to a pressure of 30 cm of krypton with about 5 percent methylal used as a quencher. A 5-mil gold cathode Geiger counter (counter 3) was used to detect the gamma-rays. It was 6lled to a pressure 10 cm of argon with 5 percent ethyl alcohol as a quencher.

The coincidences were recorded by an electronic circuit with a resolving time which was varied between 0.2 and 0.4 microsecond. The circuits were capable of introducing artificial delays up to 1 microsecond. This delay feature was used to insure the fact that the resolving time was large enough to count the coincidences with an efficiency of 100 percent. In addition, several of the experiments were done with a fourchannel delay unit<sup>12</sup> which provided a continuous check on the constancy of the relative delay between two Geiger counters.

The procedure by which the conversion coefficients were derived from data such as shown in Figs. 3 to 8 and the corrections which had to be made are most easily seen by examining the equations which are given below. These equations give the expected counting rate,  $n$ , for a source having a rate of decaying nuclei,  $N$ , assuming the decay scheme shown in Fig. 1. The following symbols and values will be used:

 $\alpha = N_e/N_i$  total conversion coefficient,

 $k = N_K/(N_L + N_M + \cdots)$  = ratio of K shell conversion to all other conversion  $(=6.9)$ ,

 $r =$ ratio of K-capture to L capture (=10.1),<sup>13</sup>

observed in this experiment.

 $E=$  detector efficiency including solid angle factor; subscripts are used to differentiate between counters and radiations. The gamma-rays were counted in either counter 2 or counter 3;  $\gamma$  will be used for the 390-kev and  $\gamma'$  for the 485-kev gamma-ray.

For the 2.8-hr Sr<sup>87</sup><sup>m</sup> Source

$$
n_{x} = N \frac{\alpha}{1 + \alpha} \frac{k}{1 + k} f E_{1x}
$$
\n
$$
n_{e} = N \frac{\alpha}{1 + \alpha} E_{2e}
$$
\n
$$
n_{\gamma} = N \frac{1}{1 + \alpha} E_{3\gamma}
$$
\n
$$
n_{ex} = N \frac{\alpha}{1 + \alpha} \frac{k}{1 + k} f E_{1x} E_{2e} \text{ (coincidences)}.
$$

## For the 80-hr Y<sup>87</sup> Source in Equilibrium with the  $Sr^{87m}$

(For this equilibrium there are 1.04 Sr decays for each Y decay.)

$$
n_x = N_Y[(r/1+r) + 1.04(\alpha/1+\alpha)(k/1+k)]fE_{1x}
$$
  
\n
$$
n_e = N_Y1.04(\alpha/1+\alpha)E_{2e}
$$
  
\n
$$
n_{\gamma+\gamma'} = N_Y[E_{3\gamma'} + 1.04(1/1+\alpha)E_{3\gamma}]
$$

$$
n_{ex} = N_Y 1.04(\alpha/1+\alpha)(k/1+k) f_{1x} E_{1x}
$$

 $f =$  fluorescence yield = fraction of  $K$  shell holes which

<sup>&</sup>lt;sup>12</sup> The authors are indebted to Dr. Sherman Frankel for the use of this multichannel unit, which he designed, built, and tested. 's M. E. Rose and J. L. Jackson, Phys. Rev. 76, 1540 (1950).

<sup>&</sup>lt;sup>14</sup> A. H. Compton and S. K. Allison, X-Rays in Theory and Experiment (D. Van Nostrand Company, Inc., New York, 1935).<br><sup>15</sup> H. S. W. Massey and E. H. S. Burhop, Proc. Roy. Soc. (London) **A153**, 661 (1936).

This set of eight experimental counting rates  $n$ , can be used to determine the 7 unknowns. In addition, several of these unknowns can be checked independent of these radioactive sources. The ratio of the efficiencies of the 485- to the 390-kev gamma-ray (i.e.,  $E_{3\gamma}/E_{3\gamma}$ ) for the gold cathode Geiger counter was determined from the curves given by Saurrer<sup>16</sup> as 0.93. Both  $E_{2e}$ and  $E_{1x}$  were checked by using an In<sup>114</sup> source. Thus only four unknowns, the two source strengths, the gamma-ray efficiency, and the conversion coefficient, were determined by using experimental values for the observed counting rates in these eight equations. The internal consistency of these values confirms both the decay scheme and the reliability of the procedure.

The internal conversion coefficient can be obtained directly from any of the following ratios:

$$
\frac{(n_e/n_x)_{\rm Sr}}{(n_e/n_x)_{\rm Y}} = \frac{(n_{ex}/n_x)_{\rm Sr}}{(n_{ex}/n_x)_{\rm Y}} = \frac{1}{1.04} \frac{r}{r+1} \frac{\alpha+1}{\alpha} \frac{k+1}{k} + 1
$$
  
= 1.52 + 0.524(1/\alpha), (1)

$$
\frac{(n_x/n_\gamma)_{\rm Sr}}{(n_x/n_\gamma)_{\rm Y}} = \frac{0.81\alpha^2 + 1.72\alpha}{1.81\alpha + 0.90}.
$$
 (2)

TABLE I. Total conversion coefficient  $\alpha$ .

Source No.			
Coincidences			
Singles	A 31	0.23	

In this experiment, we concentrated our efforts on determining  $\alpha$  from Eq. (1), since it is independent of the position of the 485-kev gamma-ray and of any branching which might occur from the 80-hr state directly to the 2.8-hr state.

Physically, this equation represents a comparison of the number of x-rays of a  $Sr^{87m}$  source with those of an equilibrium 80-hr  $Y^{87}-2.8$ -hr Sr $^{87m}$  source. It was not until we had taken data on several sources that the decay scheme was established so that Eq. (2) could be used to determine  $\alpha$ . The results obtained from 3 different sources, using Eq. (1) are given in Table I.

If, in addition, a correction is made for the coincidences due to the 485-kev conversion electrons (2.8 percent), the values of  $\alpha$  from coincidences become 0.34 and 0.26. The final value is  $\alpha=0.28\pm0.06$ , obtained by weighting source 2 a little heavily because of the superiority of the data. When this value is corrected for the  $K$  to  $L$  ratio, the  $K$  conversion coefficient is found to be  $0.24 \pm 0.05$ . The error indicated is twice the average deviation and represents a reasonable estimate of the accuracy based on a detailed study of the curves from individual determinations.

The conversion coefficient of the 485-kev gamma-ray can be calculated from the determined coefficient of the 390-kev gamma-ray by using the measured conversion electron ratio of 1/59 and the decay scheme in Fig. 1.

The resultant value of  $\alpha$  is 3.5 $\pm$ 0.7 $\times$ 10<sup>-3</sup>. The critical aspect of the decay scheme for this calculation is the assertion that a negligible fraction of the 2.8-hr state is formed from the 80-hr Y state without going through the 875-kev state in Sr<sup>87</sup>.

These conversion coefficients are compared with the values obtained from theory in Table II.The theoretical values were taken from the following sources:

1. The  $K$  conversion coefficients are taken from the tables of relativistically calculated  $K$  conversion coefficients, which have been privately circulated by Rose,  $et~al.^{17}$ 

2. The electric  $K$  to  $L$  ratios are obtained from the nrelativistic calculations of Hebb and Nelson.<sup>18</sup> nonrelativistic calculations of Hebb and Nelson.

3. The magnetic  $K$  to  $L$  ratios were determined from a pproximate values given by Tralli and Lowen.<sup>19</sup> the approximate values given by Tralli and Lowen.

In the case of each of the gamma-rays, the experimental error is so large that two different assignments are possible. However, since the experimental determination of the 485-kev gamma-ray conversion coefficient is based on that of the 390-kev gamma-ray, the errors in these two values are correlated. Thus, if the multipolarity of the 390-kev gamma-ray is Magnetic 4, the 485-kev gamma-ray is Magnetic 1; if the 390-kev were E5, the 485-kev would be E2.

The experimental value of the  $K/(L+M)$  ratio for the 390-kev gamma-ray does not agree with either theoretical value. However, this is the rule for isomers rather than the exception and the calculations of the theoretical values are only approximate. Despite this disagreement, the  $K/(L+M)$  ratio can be used, since a smooth empirical curve can be drawn for other M4 transitions (if empirical K to  $L+M$  ratios are plotted as a function of  $Z^2/E$ ) and our value fits on this curve. Furthermore, Sunyar and Goldhaber<sup>20</sup> have shown that experimental values of K to L (or K to  $L+M$ ) ratios are consistently lower than the theoretical values. Their empirical curves predict a  $E4 K$ -to-L ratio of less than 4; for E5 the value would be less than 2. Thus, from our experimental results the multipolarities of the 390 kev and 485-kev gamma-rays can be assigned as M4 and  $M1$ , respectively.

The internal conversion coefficient of the 384-kev gamma-ray in 14-hr  $Y^{87m}$  can be measured with the same technique. In this case it is simplest to determine this coefficient by comparing  $Y^{87m}$  and  $Sr^{87m}$ . Figure 1 can be used to give the following equations:

$$
\frac{(n_e/n_\gamma)_{14-\text{hr } \text{Y}}}{(n_e/n_\gamma)_{2.8-\text{hr } \text{Sr}}} = \frac{\alpha_\text{Y}^{s\gamma_m}}{\alpha_\text{S}r^{s\gamma_m}},\tag{3}
$$

$$
\frac{(n_x/n_y)_{\mathbf{Y}^{87m}}}{(n_x/n_y)_{\mathbf{Y}^{87m}}} = \frac{[\alpha E_{2x}k/(k+1)]_{\mathbf{Y}^{87m}}}{[\mathbf{X}^T_{2x}k/(k+1)]_{\mathbf{Y}^{87m}}}. \tag{4}
$$

$$
(n_x/n_y)_{\text{Sr}}^{s_{7m}} \quad [\alpha E_{2x}k/(k+1)]_{\text{Sr}}^{s_{7m}}
$$

<sup>17</sup> Rose, Goertzel, Spinrad, Harr, and Strong, report privately circulated. '

- 
- <sup>18</sup> M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).<br><sup>19</sup> N. Tralli and I. S. Lowen, Phys. Rev. **76**, 1541 (1949).<br><sup>20</sup> A. W. Sunyar and M. Goldhaber, Phys. Rev. 83, 216 (1951)

and M, Goldhaber and A, W. Sunyar, Phys. Rev.  $83,906$  (1951).

 $\equiv$ 

<sup>&</sup>lt;sup>16</sup> H. Saurrer, Helv. Phys. Acta 24, 381 (1950).

Experimental				Theoretical		
Gamma- energy	Conversion ratio $K/(L+M)$	Total conversion $N_{\rm e}/N_{\rm Y}$	conversion $N_{eK}/N_{\gamma}$	K conversion <sup>a</sup>		$K/L$ ratiob. <sup><math>\circ</math></sup>
$390 \text{ keV}$	$6.9 + 0.4$	0.28	$0.24 + 0.05$	E4 0.079 E5 0.274	M4 0.194 M5 0.640	E5 5.6 M4 8.3
485 key		$3.5 \times 10^{-3}$	$3.2 + 0.7 \times 10^{-3}$	$E1 \; 1.12 \times 10^{-3}$ $E2 \, 3.76 \times 10^{-3}$	$M12.64\times10^{-3}$ $M2\,8.6\times10^{-3}$	

TABLE II. Experimental and theoretical conversion coefficients.

<sup>a</sup> See reference 17. b See reference 18. b See reference 19.

Since the energies of the radiations are almost the same, the efficiency factors do not appear in Eq. (3). However, in Eq.  $(4)$ , the x-ray efficiencies differ by a factor of 4, since krypton absorbs Y x-rays, but not Sr  $x$ -rays, in the K shell. This relative efficiency was determined experimentally by comparing the e-x coincidence rates per detected electron for the 14-hr and 2.8-hr activities.

The required data were taken before the 384-kev transition in  $Y^{87m}$  was identified. They, therefore, are not as precise as they could be using this comparison technique. The results give the ratio as  $1.0 \pm 0.2$ ; a ratio of 1 would establish the equality of the multipolarities of the 384-kev and 390-kev transitions. The  $K$  to  $L$  ratio of 8.3, reported by Hyde and O'Kelley for the 384-kev transition, while somewhat higher than the expected value, would also indicate an  $M4$  assignment.

#### ISOMERIC THEORY

The theory of isomers can be used to calculate the expected gamma-ray lifetimes for radiations of different multipolarities once the energies are known. The gamma-ray lifetime can also be obtained from the experimental data by correcting the experimental lifetime for any other competitive mode of decay. If internal conversion is the only competitive process, the correction factor is  $(1+\alpha)$ , where  $\alpha$  is the experimentally determined total internal conversion coefficient. For 2.8-hr Sr this factor is 1.28; within our precision a correction factor of 1.28 is also suitable for 14-hr Y.

These corrected experimental values indicate a forbiddenness of  $l=5$  according to the classification of biddenness of *l*=5 according to the classification c<br>Axel and Dancoff.<sup>21</sup> This assignment would be consisten with a multipolarity of either  $M4$  or  $E5$ . However, an improved formula for gamma-ray lifetime has been suggested by Weisskopf.<sup>22</sup> This formula gives the gamma-ray half-lifetime for an electric k-pole transition as

$$
\tau \gamma^{k}(E) = \left(\frac{k}{k+1}\right) \left[1 \times 3 \times 5 \times \cdots \times (2k+1)\right]^{2}
$$

$$
\times \left(\frac{137}{W}\right)^{2k+1} \frac{1}{\rho^{2k}} 4.54 \times 10^{-22} \text{ sec,}
$$

<sup>21</sup> P. Axel and S. M. Dancoff, Phys. Rev. 76; 892 (1949).

~ V. Keisskopf and J. M. Blatt, to be published in <sup>a</sup> book on theoretical nuclear physics.

where  $\rho$ =nuclear radius/2.82 $\times$ 10<sup>-13</sup> and W=energy/  $mc<sup>2</sup>$ . According to Weisskopf a magnetic k-pole transition is less probable and leads to a lifetime which is  $180\rho^2$  times the electric lifetime. The predictions of these formulas are compared with the experimental value in Table III.<sup>†</sup>

In addition to the theoretical values, Table III lists the value obtained from the empirical formula for the the value obtained from the empirical formula for the  $M4$  group obtained by Sunyar and Goldhaber.<sup>20</sup> This formula contains a statistical factor dependent on the spin of the upper state of the isomer,  $I_i$ , and is

$$
\tau_{\gamma} = 1.0 \times 10^4 (2I_i + 1)/A^2 E_{\text{Mev}}^9.
$$

For  $Y^{87}$ ,  $I_i$  is taken equal to 9/2, following the predictions of the shell model.

Using the Keisskopf formulation, an assignment of M4 is the only acceptable one. While the agreement of the M4 assignment with the empirical formula of Sunyar and Goldhaber is excellent, their empirical analysis indicate that this lifetime might also be suitable for  $E4$ . However, the  $E4$  possibility is completely inconsistent with the conversion coefficient.

#### SHELL STRUCTURE

Using the shell model of Mayer, $2<sup>3</sup>$  predictions can be made of the spin and parity of odd  $A$  nuclei. For  $Sr^{87}$ with 49 neutrons, the prediction is for a  $g_{9/2}$  or  $p_{1/2}$ ground state and a second excited state of  $p_{3/2}$ . The measured<sup>24</sup> ground-state spin of  $9/2$  when used in conjunction with the shell model uniquely gives the order of the Sr<sup>87</sup> levels as  $g_{9/2}$ ,  $p_{1/2}$ , and  $p_{3/2}$ . For Y<sup>87</sup> with 39 protons, the shell model predicts either the  $p_{1/2}$ 

TABLE III. Gamma-ray lifetimes.

Energy	Experimental	Weisskopf and Blatt			Sunyar and Goldhaber
		M4	F.A	E5	M4
390	$1.29\times10^{4}$	$6.7 \times 10^{4}$	$6.6\times10$	$4.9\times10^{7}$	$1.3 \times 10^{4}$
384	$6.45 \times 10^{4}$	$7.5 \times 10^{4}$	$7.4\times10$	$5.6 \times 10^{7}$	$7.3 \times 10^{4}$

 $\ddagger$  *Note added in proof*: A revised lifetime estimate was published by V. F. Weisskopf in Phys. Rev. 83, 1073 (1951). This estimate would increase the lifetime of electric transitions by the factor  $(l+3/3)^2$ . It also changes the ratio of magnetic to electric lifetime to  $18\rho^2$ . These changes are small but strengthen the assignment

given. ~ M. G. Mayer, Phys. Rev. 78, 16 (1950).

<sup>24</sup> M. Heyden and H. Kopferman, Z. Physik 108, 232 (1938).

or  $g_{9/2}$  orbit. The spin for the neighboring Y<sup>89</sup> has been measured<sup>25</sup> as  $1/2$ . From this and the fact that the 80-hr ground state decays to the excited states in  $Sr^{87}$ , it seems reasonable to assign the levels in  $Y^{87}$  as  $p_{1/2}$ and  $g_{9/2}$ . However, it will be pointed out below that this assignment is inconsistent with beta-decay theory.

It is interesting to compare  $Sr^{87}$  and  $Y^{87}$ , since they are representable as a single neutron and a single proton, respectively, added to a core of 38 protons and 48 neutrons. This comparison seems more promising because the 6rst excited state in each nucleus occurs at close to the same excitation energy. However, a more careful examination indicates that the similarity in energy is probably accidental. One factor is that the positions of the  $p_{1/2}$  and  $g_{9/2}$  states are interchanged in the two nuclei. Furthermore, if the nuclear forces were identical, the coulomb repulsion would tend to make the odd proton (i.e., Y), more stable with the  $g_{9/2}$ configuration, whereas the  $p_{1/2}$  seems more probable. Of course, it is quite reasonable to expect the extra 10 neutrons to make an appreciable difference.

## BETA-DECAY THEORY

The calculations and published curves of Feenberg and Trigg<sup>26</sup> were used to compare the observed data with beta-decay theory. For a 700-kev positron, the ratio of  $K$ -capture to positron emission for allowed transitions is about 5.9. Since the observed ratio was about 300 it seemed likely that the main  $K$ -capture branch was going to a higher energy state. If the positrons are emitted during a transition from the 80-hr Y to the 390-kev Sr state, the main  $K$ -capture branch can lead to the 875-kev Sr state. The theoretical K-capture to positron ratio for this branch is 760. and these positrons would not have been noticed in the 700-kev positron group. The log $ft$  for the main  $K$ capture branch would then be 5.65 indicating an allowed transition.

However, all other comparisons with beta-decay theory are inconsistent with shell structure assignments. If the theoretical  $K$ -capture to positron ratio for allowed transitions is used, the maximum branching ratio from the 80-hr Y to the 390-kev Sr state is 2.5 percent. This leads to a logft of 7.6 indicating a forbidden transition. Similarly, if the maximum experimentally allowable branching of about 2 percent is assumed for  $K$ -capture plus positron branching from the 14-hr Y state to the ground state of Sr, the logft value is 7.4. This experimental limit for the 14-hr Y is based on the 1.1-Mev positron data taken with our cyclotron produced sources. According to Hyde and O'Kelley, the main fraction of the positrons we observed are associated with 14.6-hr  $Y^{86}$  and the upper limit on positron branching in  $Y^{87}$  is about 0.025 percent. This lower limit is supported by the fact that the positrons we observed were not of the proper energy to fit into our own decay scheme.

These beta-decay results could be consistent with the other results only if either:

(1) The level assignments in  $Y^{87}$  were changed to  $f_{5/2}$  and  $i_{13/2}$ . The small beta-branchings would then be expected, since the transitions would be second forbidden. This would be incompatible with Mayer's shell model.

(2) Some special selection rule were operative inhibiting both of the expected beta-branches.

The authors are deeply indebted to the group which built and tested the beta-ray spectrometer used in this experiment. Professor W. E. Meyerhof supervised the design and construction, and Dr. A. J. Saur and J. Ovadia gave invaluable assistance. Dr. Sherman Frankel helped with some of the design problems of the spectrograph and also designed some of the electronic equipment.

We are grateful to Dr. Ovadia and Mr. M. J. Glaubman for their assistance during the course of the experiments. We also wish to thank Professor F. Metzger for many helpful discussions and for his aid in carrying out the experiments with scintillation detectors.

<sup>&</sup>lt;sup>25</sup> M. F. Crawford and N. Olson, Phys. Rev. 76, 1528 (1949).<br><sup>26</sup> E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399<br>(1950). We wish to thank Mr. Trigg for sending us an enlarged set of the graphs contained in this article.