

Disintegration of Antimony-124

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The radiations from radio antimony of 60-day half-period Sb^{124} , have been investigated in a magnetic lens electron spectrometer. The beta-ray spectrum was found to have two components with maximum energies of 0.57 ± 0.02 and 2.24 ± 0.05 million electron volts. The gamma-ray spectrum has two lines, one at 0.60 ± 0.01 and the other at 1.69 ± 0.02 million electron volts. It appears quite certain that the 0.60 Mev gamma ray originates from Sb^{124} , and therefore these measurements substantiate the disintegration scheme proposed by Meyerhof and Scharff-Goldhaber [1],¹ with a small revision of energies.

I. Introduction

The radiations from 60-day Sb^{124} have been studied by several investigators. Hales and Jordan [2] used a 180° spectrometer to study the beta spectrum of Sb^{124} . They report the presence of two components with maximum energies at 0.74 ± 0.03 and 2.45 ± 0.07 million electron volts (Mev). Miller and Curtiss [3], using a thin magnetic lens spectrometer, also observed a complex beta spectrum, but with maximum energies at 0.53 and 2.25 Mev.

The most recent investigation of the beta and gamma radiations is that of Meyerhof and Scharff-Goldhaber [1] by means of coincidence-absorption measurements. They report the presence of two beta-ray components with energies of 0.7 and 2.4 Mev; and two gamma rays with energies of 0.6 and 1.7 Mev. A table summarizing the results of previous studies of Sb^{124} is also given by them.

Although the gamma radiations from Sb^{124} had been studied by a number of investigators prior to these authors, none of them had reported the presence of a 0.6-Mev gamma ray. With one exception, these prior reports indicated only one gamma ray with energy about 1.7 Mev. The present paper provides further information regarding the origin of this 0.6-Mev gamma ray.

The energies of the beta and gamma rays from Sb^{124} reported here were obtained by means of a spectrometer of the thin magnetic lens type. This instrument has been described by Miller and Curtiss [4]. The results obtained are in partial

agreement with those previously reported, but differ significantly in certain respects. It is believed that the type of spectrometer used by the authors provides a more accurate determination of the Sb^{124} beta and gamma energies than can be obtained from absorption measurements such as used by Meyerhof and Scharff-Goldhaber [1].

II. Preparation of Sources

To obtain radioactive Sb^{124} for these experiments, a sample of antimony, estimated to be about 99.95 percent pure, was prepared at the Bureau of Standards. This sample was then sealed in a quartz capsule and irradiated with slow neutrons in the Clinton Laboratories pile at Oak Ridge, Tenn. Since ordinary antimony consists of about 56 percent of Sb^{121} and 44 percent of Sb^{123} , the resultant reactions $\text{Sb}^{121}(n, \gamma)$ and $\text{Sb}^{123}(n, \gamma)$ produce a mixture of 2.8-day Sb^{122} and 60-day Sb^{124} [5].

The preparation of the beta and gamma sources used in the spectrometer has been described in detail in a previous paper [6]. The same methods were used here. To prepare a beta source, the antimony crystals were first ground up with a Carboloy mortar and pestle to a fine powder. A small amount of this powder was then deposited on a thin aluminum foil moistened with dilute glyptal solution.

The gamma source was prepared by placing a relatively large amount of the antimony crystals

¹ Figures in brackets indicate the literature references at the end of this paper.

in a brass capsule, the opening of which was then sealed with Apiezon-Q wax. A radiator of uranium foil (42 mg/cm^2) was used to convert the gamma rays from Sb^{124} into photoelectrons whose energy can be measured in the spectrometer.

III. Beta-Ray Spectrum of Sb^{124}

The beta-ray spectrum was determined about 6 months after receipt of the source from Oak Ridge. The amount of 2.8-day Sb^{122} left after this long an interval is completely negligible, so that the remaining activity must be due to 60-day Sb^{124} . Figure 1 shows the beta spectrum ob-

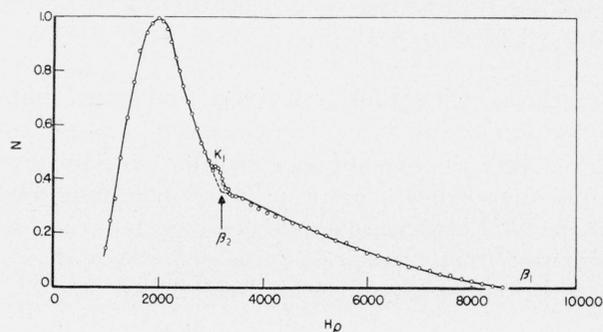


FIGURE 1. Primary beta-ray spectrum of Sb^{124} showing the intensity plotted against $H\rho$.

The K_1 peak is probably due to internal conversion electrons from a 0.60-Mev gamma ray.

tained. The quantity N represents the relative intensity over equal intervals of $H\rho$ (gauss-cm). It is equal to the number of counts per minute in the Geiger-Müller counter divided by $H\rho$.

The small K_1 peak in figure 1 is no doubt due to internal conversion electrons produced by a 0.60-Mev gamma ray, which will be discussed later. Omitting this peak, it is apparent that the beta spectrum of Sb^{124} consists of two components. The first part of the curve, up to the K_1 peak, is due to the sum of the two components; the remainder of the curve, beyond the K_1 peak, is due to the higher-energy component alone. Unfortunately, the K_1 peak occurs in the vicinity of the "break" in the beta distribution curve, which indicates the end point of the lower-energy beta-ray group. This tends to obscure the location of this end point.

If we neglect the K_1 peak, however, and extrapolate the parts of the curve immediately preceding and following this peak, their intersection is found

to lie at $H\rho = 3,175 \pm 75$ gauss-cm. This corresponds to an energy of 0.57 ± 0.02 Mev. The higher-energy component approaches the $H\rho$ axis asymptotically, which makes its end point difficult to determine by direct inspection of figure 1. For this purpose, therefore, we make use of the Fermi plot shown in figure 2, which will be discussed in detail in section IV.

The Fermi plot again indicates clearly the presence of two beta-ray components. The K_1 peak of figure 1 has been omitted from figure 2, as indicated by the central broken portion of the curve. The extrapolated portions of this Fermi plot give the end points of the two beta components as 0.58 ± 0.02 Mev (in agreement with the value obtained from fig. 1) and 2.24 ± 0.05 Mev. These values are lower than those obtained by Hales and Jordan (0.74 and 2.45 Mev) [2], but agree within the limits of experimental error with those of Miller and Curtiss (0.53 and 2.25 Mev) [3].

The Fermi plot of figure 2 does not result in straight line curves, in contrast with the results of Hales and Jordan [2]. This, however, is not necessarily significant because of the following considerations:

1. The relatively long half-life of Sb^{124} , together with the high maximum energy of its beta spectrum, indicates that the transitions involved are

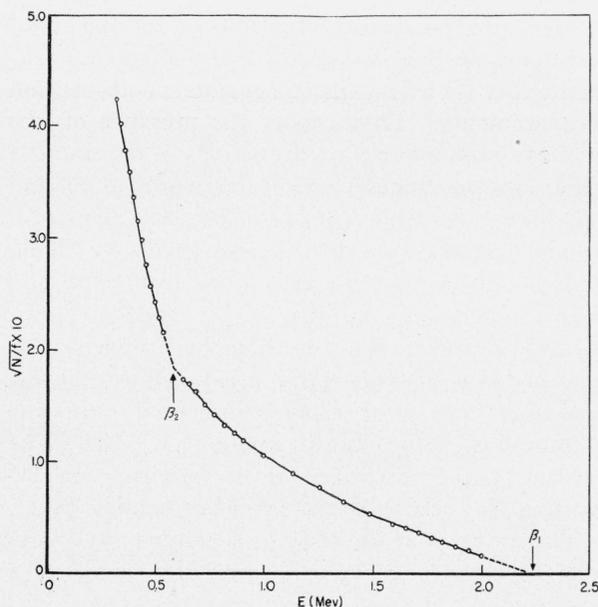


FIGURE 2. Fermi plot of the data shown in figure 1.

The original Fermi function $f(Z, \eta)$ was evaluated for $Z=51$, not the usual Kurie approximation.

highly forbidden. The simple Fermi theory does not strictly apply to transitions of this type.

2. At the time the beta spectrum was measured the source had decayed through three half-lives, and its specific activity had decreased to about one-eighth of its initial value. Consequently, it was necessary to prepare a beta source whose thickness was such that the amount of absorption and scattering within the source cannot be considered entirely negligible. This would tend to distort the observed beta-ray spectrum.

IV. Fermi Plot

The plot in figure 2 is based on Fermi's theory of beta-ray emission. Starting with eq 42 of Fermi's paper [7], let

$$K = g^2 \frac{256\pi^4}{[\Gamma(3+2S)]^2} \frac{m^5 c^4}{h^7} \left(\frac{4\pi m c \rho}{h} \right)^{2S} \quad (1)$$

$$Q^*_{mn} = \int v_m^* u_n d\tau. \quad (2)$$

(See Fermi's paper for meaning of these expressions). For a given isotope, K is constant.

Also, we have

$$\sqrt{1+\eta_0^2} = E_0 + 1, \quad (3)$$

where

η_0 = maximum momentum of beta rays emitted in units of $m_0 c$

E_0 = maximum energy of beta rays in units of $m_0 c^2$

m_0 = rest mass of electron,

and similarly,

$$\sqrt{1+\eta^2} = E + 1, \quad (4)$$

where η and E now refer to any electron with momentum η (equal to $H\rho/1700$) and energy E .

Substituting eq 1, 2, 3, and 4 in Fermi's eq 42, the probability of emission of an electron with momentum between η and $\eta+d\eta$ can be written

$$P(\eta)d\eta = K|Q^*|^2[\eta^2 F(Z,\eta)](E_0 - E)^2 d\eta, \quad (5)$$

where

$$F(Z,\eta) = \eta^{2S} e^{\pi y} |\Gamma(1+S+iy)|^2 \quad (6)$$

$$S = \sqrt{1 - \left(\frac{Z}{137}\right)^2} - 1 \quad (7)$$

$$y = \frac{Z}{137} \frac{\sqrt{1+\eta^2}}{\eta}, \quad (8)$$

and Z is the atomic number.

According to Fermi, in case of "allowed" transitions the matrix element Q^* in eq 5 is of the order of unity. For "forbidden" transitions, however, $|Q^*|^2 \ll 1$. In the latter case Q^* probably depends on the momentum η of the emitted beta rays, although the exact nature of this dependence is not known. As a first approximation, we shall assume that $|Q^*|^2$ is constant for all values of η .

Since the quantity N in the beta spectrum of figure 1 is proportional to $P(\eta)$, we have from eq 5

$$N\Delta\eta \propto f(Z,\eta)(E_0 - E)^2 \Delta\eta, \quad (9)$$

where

$$f(Z,\eta) = \eta^2 F(Z,\eta). \quad (10)$$

Taking all the $\Delta\eta$'s equal throughout the beta spectrum, eq 9 may be written

$$\sqrt{\frac{N}{f}} \propto (E_0 - E), \quad (11)$$

so that if we plot $\sqrt{N/f}$ against E , a straight-line curve is obtained which intersects the E axis at $E=E_0$. This graphical representation of the beta-ray spectrum is commonly referred to as a Fermi plot or Kurie plot, and is frequently used to determine the end point of a beta distribution.

It should be remembered, however, that eq 11 is based on the assumption that Q^* is independent of E , i. e.,

$$|Q^*|^2 = \text{constant}. \quad (12)$$

For allowed transitions, eq 12 is true—at least approximately. For forbidden transitions, however, this is not necessarily true, and the Fermi plot need not be a straight line. Figure 2 indicates, therefore, that the beta transitions involved in the decay of Sb^{124} are of the forbidden type.

The Fermi function $f(Z,\eta)$ used in making a Fermi plot is given by eq 10 and 6. The factor $F(Z,\eta)$ in eq 5 is due to the effect of nuclear attraction upon the emitted beta ray. When $\eta \gg Z/137$, $F(Z,\eta) \rightarrow 1$. That is, for relatively high beta energies and light atoms, the effect of nuclear attraction is small.

A nonrelativistic treatment of the theory of beta emission leads to the expression

$$F(Z,\eta) = \frac{2\pi y}{1 - e^{-2\pi y}}, \quad (13)$$

whereas the more rigorous relativistic theory leads to eq 6. According to Kurie, et al. [8], eq 13 is adequate for all light elements up to about

copper, $Z=29$ (provided η is not $\gg 1$). Hence in this case the Fermi function can be written

$$\left. \begin{aligned} f(Z,\eta) &= \eta^2 \frac{2\pi y}{1 - e^{-2\pi y}} \\ y &= \frac{Z}{137} \frac{\sqrt{1 + \eta^2}}{\eta} \end{aligned} \right\} \quad (14)$$

For Sb^{124} , however, $Z=51$ and $\eta_0 \cong 5$, so that the relativistic expression (eq 6) should be used. The Fermi function is then

$$f(Z,\eta) = \eta^{2+2S} e^{\pi y} |\Gamma(1+S+iy)|^2. \quad (15)$$

As $Z=51$, we have from eq 7

$$\left. \begin{aligned} \frac{Z}{137} &= 0.37 \\ S &= -0.072 \\ 2+2S &= 1.86 \\ 1+S &= 0.93 \end{aligned} \right\} \quad (16)$$

The numerical values of $Z/137$ and S in eq 16 have been rounded off to two significant figures. Substituting from eq 16 in eq 15, we have for Sb^{124}

$$\left. \begin{aligned} f(\eta) &= \eta^{1.86} e^{\pi y} |\Gamma(0.93 + iy)|^2 \\ y &= 0.37 \frac{\sqrt{1 + \eta^2}}{\eta} \end{aligned} \right\} \quad (17)$$

This is the function that was used in the Fermi plot of figure 2. The values of $f(Z,\eta)$ obtained from eq 17 have been compared with the corresponding values given by the nonrelativistic expression (eq 14). It is worth noting that the differences between the two values vary from 0 to 15 percent for the range of momenta considered here.

V. Gamma-Ray Spectrum of Sb^{124}

The gamma-ray spectrum is shown in figure 3. The distribution represented by the full curve was obtained about 1 month after receipt of the source from Oak Ridge. The activity of the 2.8-day Sb^{122} had by this time decayed to less than 10^{-3} of its initial amount, and was therefore negligible in comparison with the 60-day Sb^{124} activity. The continuous distributions in figure 3 are due to Compton recoil electrons, mostly from the

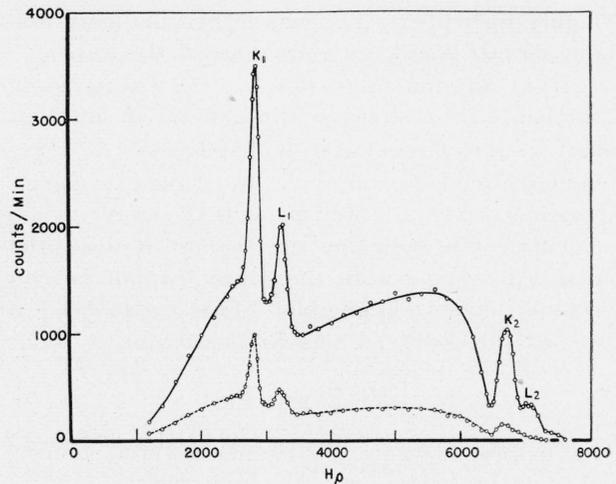


FIGURE 3. Secondary electron spectrum produced by Sb^{124} gamma rays in uranium.

K_1 , L_1 are photoconversion lines of a 0.60-Mev gamma ray; K_2 , L_2 are due to a 1.69-Mev gamma ray. Solid curve is gamma spectrum determined 1 month after preparation of source; broken curve is spectrum obtained 5 months after preparation of source.

brass capsule containing the source; whereas the superimposed peaks are due to photoelectrons from the uranium radiator (the photoelectrons from the brass capsule are relatively negligible in number)[6].

Figure 3 clearly shows the presence of two gamma rays, with energies of 0.60 ± 0.01 Mev and 1.69 ± 0.02 Mev. These values are obtained by adding the K -binding energy of uranium (0.115 Mev) to the energies of the K -photoelectrons ejected from the uranium radiator by the two gamma rays, as indicated by the location of the K_1 and K_2 peaks. A further check may be obtained by adding the L -binding energy of uranium (0.022 Mev) to the L -photoelectron energies indicated by the L_1 and L_2 peaks.

The energy of the 1.69 Mev gamma ray agrees within the limits of experimental error with the value 1.70 ± 0.02 Mev previously reported by Kruger and Ogle [9], and with the value 1.72 ± 0.03 Mev reported by Rall and Wilkinson [10]. The 0.60 Mev gamma ray, however, has not been reported by any investigators prior to Meyerhof and Scharff-Goldhaber [1]. This is difficult to understand, since figure 3 indicates that it is even more intense than the 1.69 Mev gamma ray.

Rall and Wilkinson [10] also reported a 0.61 Mev gamma ray, which they tentatively assigned to 30-day $\text{Te}^{122,124}$. Both half-life and identity of these isotopes were indicated as being in doubt.

This suggested the possibility that the 0.60 Mev gamma ray observed by us might be identical with the 0.61 Mev gamma ray reported by Rall and Wilkinson. To check this possibility, a chemical separation was made of the antimony and tellurium in the source about 9 months after its receipt from Oak Ridge. The separated tellurium fraction was then found to emit no detectable radiation. Had there been a 30-day $\text{Te}^{122,124}$ gamma activity in the tellurium fraction, it would almost certainly have been observed. We may safely conclude, therefore, that the 0.60 Mev gamma ray shown in figure 3 is not due to such an activity.

To determine whether the relative intensity of the 1.69 and 0.60 Mev gamma rays remains constant with time, the gamma spectrum was redetermined 4 months (2 half-lives of Sb^{124}) after its first measurement. This second determination is shown by the broken curve in figure 3. Comparison of the two curves indicates that the two gamma rays decay approximately at the same rate and are therefore probably both due to 60-day Sb^{124} .

VI. Disintegration Scheme of Sb^{124}

Meyerhof and Scharff-Goldhaber [1] have recently determined the relative intensities of the beta and gamma rays of Sb^{124} by means of Geiger-counter coincidences. Their measurements indicate that the relative intensity of the two beta-ray components is approximately 1:1, and that of the two gamma rays is about 2:1. According to them, the hard beta ray is followed by one gamma ray, whereas the soft beta ray is followed by two gamma rays. We were unable to make any accurate determination of relative beta- and gamma-ray intensities from our measurements. In general, however, the relative intensities indicated in figures 1 and 3 tend to agree with the values reported by these authors.

The beta- and gamma-ray energies determined by us, combined with the above data of Meyerhof and Scharff-Goldhaber, can be fitted into the disintegration scheme shown in figure 4. This is identical with the decay scheme proposed by them, except for the more accurate determination of the energies involved.

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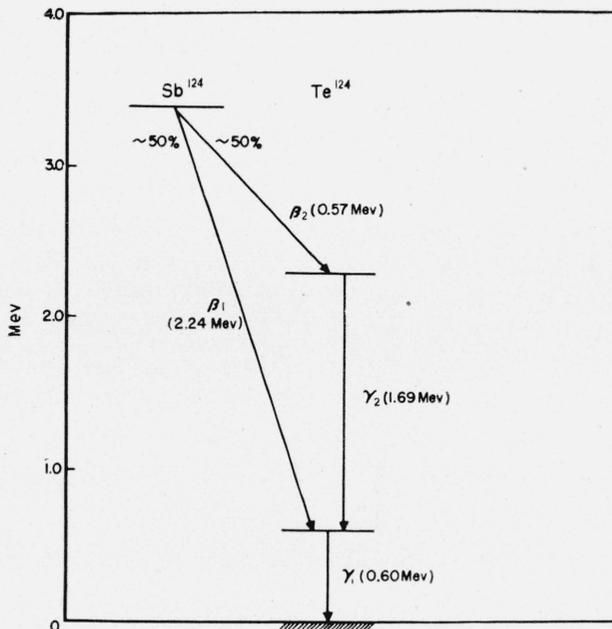


FIGURE 4. Suggested disintegration scheme for Sb^{124} based on the observed beta- and gamma-ray spectra and on the coincidence measurements reported by Meyerhof and Scharff-Goldhaber [1].

Beta transitions indicated above include energy equivalent of the electron mass.

Standards for the numerical evaluation of the original Fermi function for $Z=51$; and also A. Schwebel for the chemical separation of antimony and tellurium in the source.

VII. References

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