Coincidence Measurements. Part II. Internal Conversion

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It has been found that for a nucleus which has a simple β spectrum followed by internally converted γ radiation, the total absolute conversion coefficient can be obtained simply from single counting and coincidence rates. The method is independent of source strength, solid angle of counters, and counting efficiencies. The procedure could also be applied to processes involving complex β decay if the branching ratio were known.

The shape of the $\beta_{-e}-$ per observed β particle (i.e. $N_{\beta e}-/N_\beta$) curve is dependent upon the relative energies of the β particle and the conversion electrons as well as upon the conversion coefficient α.

The conversion coefficients have been obtained for gold[198], cesium[134], and iridium[192,194].

1. INTRODUCTION

INTERNAL conversion coefficients have been most commonly obtained from a beta spectrograph study of the radiation emitted by a radioactive source. If a γ ray is partially internally converted the monoenergetic conversion electrons appear as a peak superimposed on the smooth continuous β ray background. The conversion coefficient is then found by comparing the area under the excess peak with the total area under the continuous β curve. Clearly this method is difficult and generally does not lend itself to a high degree of accuracy. As an example, we may note the case of the 0.4-Mev line of gold[198]. Richardson[1] obtained a value of $\alpha=10$ percent, Plesset[2] 4 percent, and Siegbahn[3] $\sim$1 percent. In addition, the spectrograph method cannot be easily applied to short-lived materials or to those which require the accurate measurement of energies below a few hundred kilovolts.

2. THEORY

Let us consider the decay scheme indicated in Fig. 1. N is the number of disintegrations per second, γ_1 is converted with a conversion coefficient α, $\gamma_2\cdots\gamma_n$ represent non-converted γ rays, $\epsilon_1\cdots\epsilon_n$ are the respective efficiencies of the γ counter for the γ radiation, ϵ_β is the efficiency of the β counter, σ with the appropriate subscript is the fraction of the sphere subtended by the counter. It is assumed that the efficiency of the γ counter is $\ll 1$ (actually ~ 0.01).

Coincidences are then measured between the β rays and conversion electrons. The source is fixed to counter (2). This coincidence rate is:

$$N_{\beta e^-}=2\alpha N\sigma_{\beta_1}\epsilon_\beta\sigma_{\beta_2}\epsilon_\beta$$

(if $\alpha\sigma_\beta$ is small). The counting rate of counter (1) is:

$$N_{\beta 1}=(1+\alpha)N\sigma_{\beta_1}\epsilon_\beta.$$

We next consider the $\beta-\gamma$ coincidence rate by placing an aluminum plug in front of the window of counter (1). Counter (2) is left unchanged.

$$N_{\beta\gamma}=(1+\alpha)N\sigma_{\beta_2}\epsilon_\beta\sigma_\gamma\left[\frac{(1-\alpha)}{(1+\alpha)}\epsilon_1+\sum_2^n\epsilon_n\right]$$

and the γ counting rate is

$$N_\gamma=N\sigma_\gamma\left[(1-\alpha)\epsilon_1+\sum_2^n\epsilon_n\right].$$

The ratio is then found

$$R=N_{\beta e^-}/N_\beta/N_{\beta\gamma}/N_\gamma\cong 2\alpha/(1+\alpha)^2.$$

In the more exact case when α and σ_β may become large, a correction must be made in the β counting rate, N_β, and in the $\beta-\gamma$ rate, $N_{\beta\gamma}$, for the probability that both the β particle and the conversion electron will simultaneously pass through the same counter and be counted as a single particle. Considering this, the ratio becomes

$$R\cong 2\alpha/[1+\alpha-(\alpha/2)\sigma_{\beta_2}\epsilon_{\beta_2}]^2.$$

[1] J. Richardson, Phys. Rev. **55**, 609 (1939).
[2] E. H. Plesset, Phys. Rev. **62**, 181 (1942).
[3] K. Siegbahn, Proc. Roy. Soc. **189**, 527 (1947).

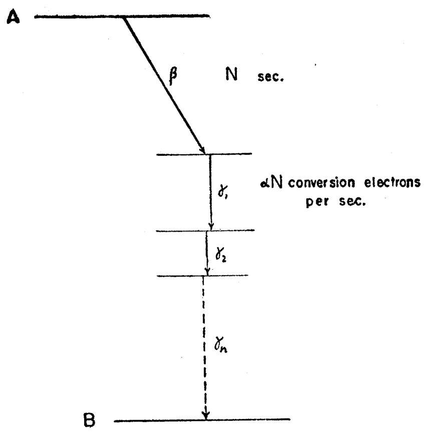

FIG. 1. Decay of nucleus A to nucleus B by the emission of a simple β spectrum. A series of γ rays, $\gamma_1\cdots\gamma_n$ follow the β decay. γ_1 is internally converted with a conversion coefficient α.

In practice $\sigma_{\beta_2}\sim 0.1$, $\epsilon_{\beta_2}\sim 1$, so that if this correction were neglected the error introduced for $\alpha=0.1$ would be ~ 0.5 percent. For $\alpha=1$ the error would be ~ 2.5 percent.

A similar equation can be found for a cascade level scheme in which several γ lines are partially converted. For example, if γ_1 is converted with a coefficient α_1 and γ_2 with a coefficient α_2, then

$$R=N_{\beta e^-}/N_\beta/N_{\beta\gamma}/N_\gamma$$
$$=2[\alpha_1+\alpha_2+\alpha_1\alpha_2]/[1+\alpha_1+\alpha_2]^2.$$

The total conversion coefficient $\alpha=\alpha_1+\alpha_2$. If $\alpha_1\alpha_2$ is small $R\simeq 2\alpha/(1+\alpha)^2$. Even if α_1 and α_2 were as large as 0.1 the error introduced would be ~ 5 percent. Thus, in cases where R is small the total absolute conversion coefficient can be obtained. If R is large so that $\alpha_1\alpha_2$ could not be ignored, it would be necessary to know the relative values of α_1 and α_2.

All the previous calculations are for nuclei which have a simple β spectrum. Similar equations can be obtained for complex spectra, however, to obtain α it is necessary to know the branching ratio. We have, therefore, limited our experimental work to nuclei which apparently have simple β spectra.*

* Note added in proof: It is assumed throughout that both the conversion electrons and gamma rays are emitted without a preferred direction with respect to the beta particle.

Shape of $N_{\beta e^-}/N_\beta$ Curves as a Function of Absorber Thickness

The previous equations for α have been developed on the assumption that the γ rays and conversion electrons are not partially absorbed. In practice each must pass through a small thickness of material to enter the counter. It is, therefore, necessary to obtain the ratio $N_{\beta e^-}/N_\beta$ as a function of absorber thickness and extrapolate to zero absorber thickness. Such extrapolation ordinarily would introduce little error if counters with thin windows were used.

We again consider the decay indicated in Fig. 1. An absorber will be placed between the source and counter (1). Let F_1 and F_2 be the fraction of β particles and conversion electrons, respectively, which are transmitted by the absorber. Then

$$N_{\beta e^-}=(F_1+F_2)\alpha N\sigma_{\beta_1}\sigma_{\beta_2}\epsilon_\beta{}^2,$$

and

$$N_{\beta 1}=(F_1+\alpha F_2)N\sigma_{\beta_1}\epsilon_\beta,$$

$$N_{\beta e^-}/N_\beta=(F_1+F_2)\alpha\sigma_{\beta_2}\epsilon_\beta/(F_1+\alpha F_2).$$

Initially (with 0 absorber), F_1 and $F_2=1$.

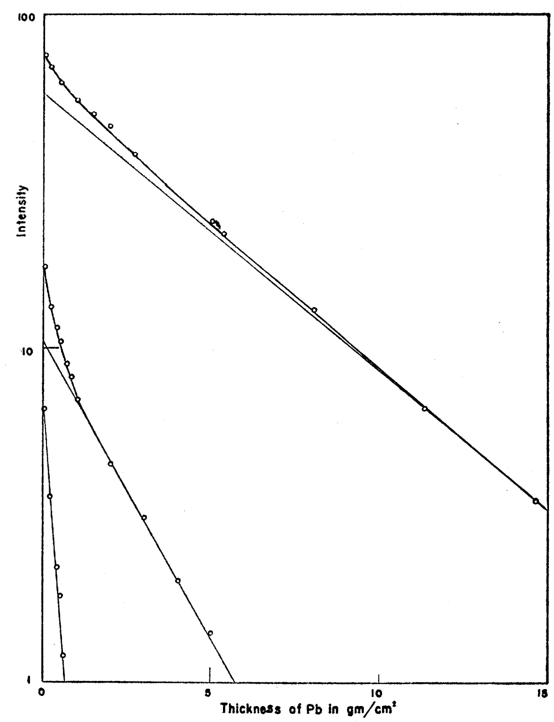

FIG. 2. Absorption curve of the γ rays of Au^{198}. Only the first portion is shown. This clearly is complex and can be broken into three components.

The initial and final values of the curve can be calculated. These are shown in Table I. That is, if F_1 is always equal to F_2, $N_{\beta e^-}/N_\beta$ as a function of absorber thickness will be a constant. If $F_1 > F_2$ (a) for small α the final value of this ratio will be $\approx\frac{1}{2}$ the initial value; (b) for large α the ratio again approaches a constant value. For $F_1 < F_2$ (i.e., essentially if the β energy is less than the e^- energy) the ratio will rise as the thickness is increased. This rise for small α will be $\sim 1/2\alpha$. Similar conditions exist if more than one γ ray is converted. The α then becomes the total conversion coefficient.

3. EXPERIMENTAL

In practice we have used the metal type self-quenching counters with an end window of $\frac{1}{2}$ mil Cellophane. A small brass disk was fitted into the end of counter (2). The sample was placed in a $\frac{1}{8}$-inch hole drilled through the center of the disk. This was found to be necessary to eliminate spurious coincidences resulting from the scattering of electrons from one counter to the other. In order to determine the conversion coefficient of a given nucleus it was found convenient to determine the ratio $N_{\beta e^-}/N_\beta$ as a function of absorber thickness. In all cases it was necessary to correct the β_{-e^-} coincidences for $\beta-\gamma$ and $\gamma-\gamma$ coincidences. For most nuclei studied this was a very small correction in the region of interest. The value of $N_{\beta e^-}/N_\beta$ extrapolated to zero absorber thickness was then used in calculating α. We have investigated the effect of various absorbers, namely Cellophane and aluminum, on the extrapolated value and have found no difference.

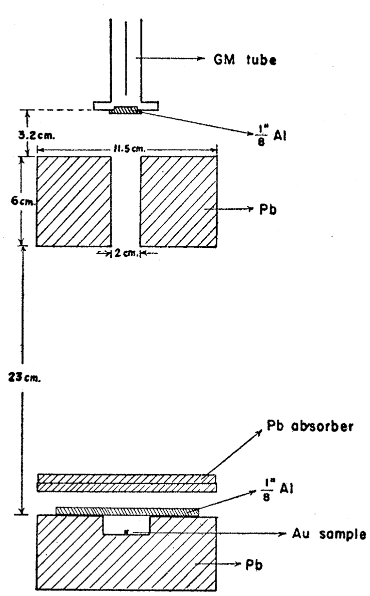

FIG. 3. The geometric arrangement used in obtaining the absorption curve for the γ radiation of Au^{198}. The source strength was $\sim$50 millicuries.

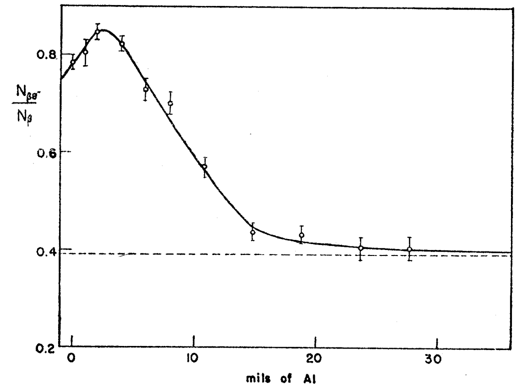

FIG. 4. The β conversion electron coincidence rate per observed β particle as a function of absorber thickness for Au^{198}. The extrapolated value is used to calculate α. The dotted line is the limit which the curve should approach if the e^- energy is less than the upper limit of the β energy.

Gold^{198}

Gold^{198} decays with a period of 2.7 days by the emission of a single β ray of energy 0.92 Mev. Siegbahn[3] has studied the γ radiation by observing the secondary electrons ejected from various radiators. He has found only one γ line of energy 0.401 Mev. However, Richardson,[1] from a cloud-chamber study, has observed three lines, namely, 70, 280, and 440 kev. Sizoo and Eijkman[4] have reported lines at 73, 250, and 410 kev and one of 2.5 Mev. Clark[5] in 1942 has reported $\gamma-\gamma$ coincidences from gold. It has been suggested by Siegbahn that these discrepancies may be due to other activities induced by deuterons on gold and that Au^{198} really emits only a simple β spectrum followed by a single γ ray.

It is mentioned by Siegbahn[3] that Norling[6] could not obtain any $\gamma-\gamma$ coincidence effects. This apparently is due to the extremely low efficiency of his lead counters for the lower energy

[4] G. J. Sizoo and C. Eijkman, Physica **6**, 332 (1939).
[5] A. Clark, Phys. Rev. **61**, 242 (1942).
[6] F. Norling, Ark. Mat. Astr. Fys. **A**, 27 (1941).

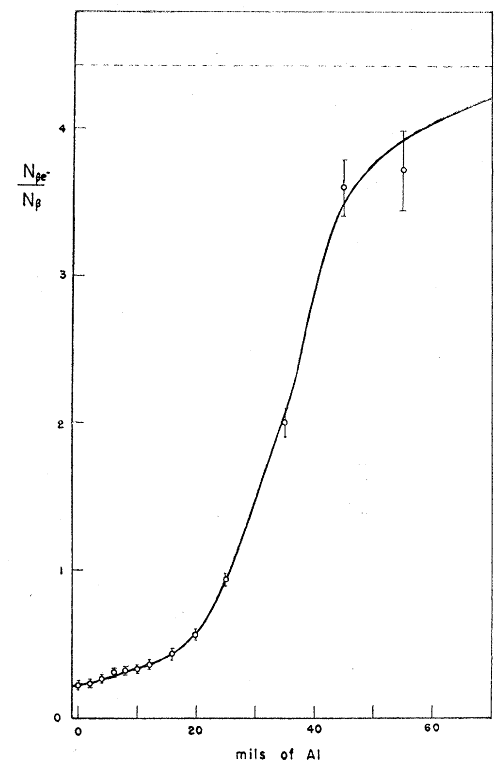

FIG. 5. Coincidence curve for Cs^{134}. The rise indicates that the e^- energy exceeds the maximum β energy. The calculated limit is indicated by the dotted line.

γ radiation, since in making an absorption measurement of the γ radiation he points out: "The curve reveals the peculiar fact that the weak component is not counted at all, . . . On the other hand the soft component could be observed with a brass tube of 1 mm wall. . . . It ought to be possible to observe $\gamma-\gamma$ coincidences by the use of such counting tubes of brass, but the intensities of the available sources were too small for this purpose."

We have obtained γ ray absorption curves of strong ($\sim$50 millicuries) gold samples produced by slow neutron bombardment at Oak Ridge. As Siegbahn suggests, this sort of sample should be most free from other activities. Our absorption curve is found to be complex in the low energy region and can be broken into two components of energies $\sim$300 and 70 kev (Fig. 2) in addition to the 0.4-Mev line. The geometric arrangement is indicated in Fig. 3.

It is not possible to estimate the relative intensities, since apparently the efficiencies of the counters vary over a wide range in this energy region. This is clear from the fact that different types of counters give considerably different relative values, yet all the curves show the complexity.

We have also looked for and found $\gamma-\gamma$ coincidences both from the sources prepared by slow neutron capture and by deuteron bombardment on the Michigan cyclotron. It would therefore seem that the gold β radiation is followed by two γ rays emitted in cascade.

It has previously been mentioned that the 0.4-Mev line following the disintegration of gold is partially converted. From the β spectral distribution as obtained with a cloud chamber, Richardson[1] has found a value of $\sim$10 percent while Plesset,[2] from a β spectrograph study (using the photographic method) gives 4 percent and Siegbahn[3] $\sim$1 percent.

Using the coincidence method described above we have obtained the "β_{-e^-} per observed β" curve indicated in Fig. 4. From the extrapolated rate and the $\beta-\gamma$ per observed γ rate we obtain a value $\alpha=4.70$ percent. It may be noted that the curve in Fig. 4 decreases with an increase in absorber thickness and seems to approach the theoretical value indicated by the dotted line.

The peculiar maximum in the $N_{\beta e^-}/N_\beta$ curve is apparently a real effect and presumably is due to the difference in absorption of the continuous β spectrum and the monoenergetic conversion electrons. In fact, a slope of zero should occur when $F_1/F_2=(dF_1/dx)/(dF_2/dx)$. Also the value of the absorber thickness at which the ratio is equal to the ratio at zero thickness should occur

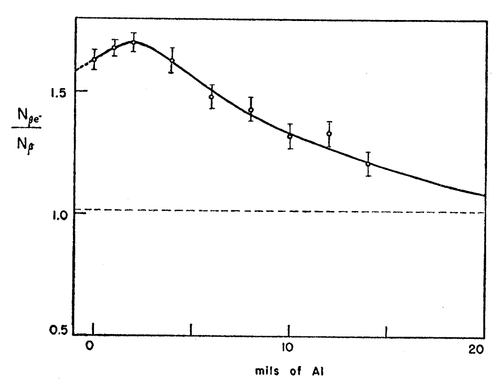

FIG. 6. Iridium coincidence curve. The e^- energy is less than the β energy.

TABLE I. Calculated initial and final values of $N_{\beta e^-}/N_\beta$.

	Initial value	Final value
$F_1 = F_2$	$(2\alpha/1+\alpha)\sigma_{\beta_2}\epsilon_\beta$	$(2\alpha/1+\alpha)\sigma_{\beta_2}\epsilon_\beta$
$F_1 > F_2$	$(2\alpha/1+\alpha)\sigma_{\beta_2}\epsilon_\beta$	$\alpha\sigma_{\beta_2}\epsilon_\beta$
$F_1 < F_2$	$(2\alpha/1+\alpha)\sigma_{\beta_2}\epsilon_\beta$	$\sigma_{\beta_2}\epsilon_\beta$

when $F_1 = F_2$. That is, the absorption curves would cross at this point. It would be of interest to check these points by comparing the absorption of 0.4-Mev monoenergetic electrons (from a Van de Graaff machine) with the total absorption curve of gold.

Cesium^{134}

In a similar manner, measurements were carried out on a sample of Cs^{134} produced by slow neutron bombardment in the Oak Ridge pile. It might be noted that the specific activities of most such samples are very large so that only an extremely small amount need be used as a counting source. The $N_{\beta e^-}/N_\beta$ curve as a function of absorber thickness is shown in Fig. 5. This curve clearly rises as should be expected if at least one of the converted lines has an energy greater than the upper energy limit of the continuous β spectrum. The total absolute conversion coefficient as calculated from the ratio $(N_{\beta e^-}/N_\beta)/(N_{\beta\gamma}/N_\gamma)$ is 2.51 percent. From this we see that the curve should rise to a value ~ 23 times the initial value. This final value is indicated by the dotted line in Fig. 5.

Relatively little is known concerning the γ radiation of Cs^{134}, however, once the relative conversion coefficients are found (if more than one line is converted), the absolute values of each can be obtained by using our absolute value for the total conversion coefficient.

TABLE II. Experimentally determined conversion coefficients.

Isotope	γ Energy	$\frac{N_{\beta e^-}}{N_\beta}$	$\frac{N_{\beta\gamma}}{N_\gamma}$	α
Au^{198}	0.401	0.0116	0.136	0.0470 ± 0.0024
Cs^{134}		0.00344	0.0719	0.0251 ± 0.0015
$Ir^{192, 194}$		0.0247	0.0714	0.286 ± 0.014

Iridium$^{192, 194}$

Strong samples of iridium$^{192, 194}$ (60 days) were obtained by slow neutron bombardment. No special difficulties were encountered in obtaining the total conversion coefficient. The value obtained for α is 28.6 percent. The $N_{\beta e^-}/N_\beta$ curve is shown in Fig. 6. It clearly drops and seems to approach the predicted value (dotted line). The fact that it drops indicates that none of the conversion electrons have energies greater than the maximum energy of the β particles.

4. DISCUSSION

It would seem that by the coincidence methods herein described much data could be quickly and easily obtained concerning internally converted γ radiation. Unfortunately, we are at present limited to γ rays which follow a simple β spectrum. The same methods could be applied to disintegrations involving complex β spectra if the branching ratio were known. We have not as yet attempted to apply these procedures to disintegrations involving positron emission. Difficulties would probably arise from the annihilation radiation if the experimental procedure were not changed somewhat.

5. SUMMARY OF EXPERIMENTAL RESULTS

The experimental data on gold, cesium, and iridium are indicated in Table II.