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Application of scintillation counters to beta ray spectroscopy

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UMI®
APPLICATION OF SCINTILLATION COUNTERS
TO BETA RAY SPECTROSCOPY

by

James P. Palmer

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of
The Requirements for the Degree of

DOCTOR OF PHILOSOPHY

Major Subject: Physics

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1950
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I. INTRODUCTION

The scintillation counter is rapidly proving to be a powerful tool in nuclear studies. Many phosphors, both organic, such as anthracene and stilbene, and ionic, such as KI(Tl) and NaI(Tl), have been studied in regard to their usefulness as emitters of light when struck by nuclear particles. This is the first prerequisite of a phosphor, it must scintillate. Different experiments require this scintillation to have special properties. If one is merely interested in detecting nuclear radiations, then the requirement on the phosphor is less stringent. It must merely respond to the particular radiation. In the study of decay schemes, by coincidence counting, a phosphor must emit its light scintillations promptly if one wishes to determine whether two particles are in true coincidence. Stilbene is an excellent phosphor for this work since its light is emitted in a time of the order of $2 \times 10^{-9}$ seconds. Anthracene is only slightly poorer, its light being emitted in a time of the order of $2 \times 10^{-8}$ seconds. In some phosphors such as CdS(Ag) the light emission extends over a period of $10^{-4}$ microseconds and, therefore, one is limited in the maximum counting rate at which he can operate the detector. This, in general, may not be important. One may also, as in this
work, be interested in linearity of the crystal for the actual measurement of particle energies. That is, are the number of light quanta emitted actually proportional to the energy of the impinging electron? This has been shown to be the case for anthracene (1).

The work to be described here sets three requirements for the phosphor. First, it must respond linearly to the energy of an incoming beta particle; second, it must scintillate promptly; and third, the quanta emitted must be sufficient in number, even for low energy electrons, that they will produce measurable pulses well above the noise background in a photomultiplier. Anthracene was chosen as a particularly attractive phosphor for this work.

The application of the scintillation counter to beta ray spectroscopy fulfills the need for an instrument capable of analyzing very weak beta activities. The ability to judge the energy of the beta particles, even if not with the precision attained by magnetic spectrometers, is then of value and cases of this type have been investigated in the present work. Furthermore, the counter's speed of response makes it an extremely useful tool for coincidence experiments.

In Chapter III a description of the experimental apparatus for measuring and recording pulses of a particular height as well as coincidences between two pulses will be
given. Chapter IV will be concerned with an analysis of the response of the system to mono-energetic electrons. How this response distorts an observed beta spectrum and what corrections to the observed spectrum are appropriate will also be discussed. Chapter V deals with the experimental results of determining beta ray end-point energies and the distribution in energy of beta rays.
II. REVIEW OF THE LITERATURE

The detection of alpha particles by scintillation counters dates back to Crookes and Rutherford.

Blau and Dreyfus (2), using a ZnS screen and a photomultiplier, showed that the photomultiplier current varied directly with the proximity of the alpha particle emitter. Coltman and Marshall (3) and Kallman (4) showed that individual light flashes could be detected. When Kallman showed that naphthalene was transparent to its own radiations and could be used to detect gamma and beta rays, many workers in this country entered the field. Deutsch (5) confirmed and extended Kallman's observations and Bell (6) showed that anthracene gave larger light flashes than naphthalene.

Shortly thereafter many laboratories started investigating the properties of scintillation counters. In the summer of 1948 a conference was held at the University of Rochester to discuss and compare scintillation and crystal counters. In June, 1949, a year later, a second conference (7) was held at Oak Ridge, devoted only to scintillation counters. Here Morton discussed the limitations of phototubes and the prospects of improving them. P. R. Bell showed for the first time that anthracene when used with the RCA-C-7132 photomultiplier (later RCA 5819) could be used
effectively for beta ray spectroscopy. It was primarily his results which inspired this work.

Until this day Bell's work in beta spectra has remained for the most part a standard for other investigators (8, 9).

A great deal of work has gone into finding good phosphors, phosphors which will convert a large fraction of the energy of the impinging particle into light energy. Kallman (10) calls this the "physical efficiency" of the phosphor. The radiated light should be largely concentrated in the region of maximum spectral sensitivity of the phototube as well as emitted in a short time. The emitting phosphor should not absorb its own radiation. This property Kallman refers to as the "technical efficiency".

Perhaps Kallman, above all others, has investigated the important properties of scintillation phosphors such as emission time, transparency, energy yield for various radiations, the wave length of the emitted light, and conversion efficiencies (8).

Hofstadter (11) has demonstrated the importance of NaI(Tl) for studying gamma ray energies. Since the crystal responds to gamma radiation by the photoelectric and pair processes, it gives good sharp gamma ray lines.

It is very difficult to survey the literature and determine which phosphor is the best for beta work but since
anthracene is known to respond admirably to beta rays this phosphor was chosen for this work.

Further reference to the literature will be given in the body of this report.
III. EXPERIMENTAL APPARATUS

A. System Block Diagram

The system has been designed as a scintillation-coincidence spectrometer. Since both channels are identical with the exception of the delay circuit for varying the coincidence resolving time, it is appropriate to follow the chain of events, in a general way, through only one channel. Referring to Fig. 1, scintillation counter one in its response to an energetic beta particle puts out a pulse to the pre-amplifier. The pre-amplifier shapes this pulse to the desired shape for operation of the differential discriminator. The main linear amplifier further amplifies the pulse and feeds it to the differential discriminator which consists of all the boxes in the top channel to the top scaler in Fig. 1. In the differential discriminator the pulse is either accepted or rejected in accordance with whether or not its height is between $E$ and $E + \delta E$, $E$ and $\delta E$ being previously selected by the operator. In practice $\delta E$ is chosen much smaller than the width of the transmission curve. If this pulse is between $E$ and $E + \delta E$, the pulse is sent on to the top scaler, which completes the necessary components for the scintillation spectrometer.
In coincidence counting a pulse in the lower channel, Fig. 1, opens a gate of chosen width (0.533 microseconds in the work here described). If in the upper channel, Fig. 1, a pulse appears in this gate a coincidence impulse is delivered from a coincidence tube to the coincidence scaler.

Either channel may be operated independently of the other if desired.

The instrument has negligible drift over six-hour periods after the initial warm up.

In Fig. 2 is shown a photograph of the equipment.

B. Phototube and Crystal

Crystals of anthracene which have been used in this work have either been prepared in this laboratory from the melt by the furnace gradient technique (8) or have been purchased from Harshaw Chemical Company. At first the Harshaw crystals were inferior to our own but with continued work on their part, it was found that their most recent ones are somewhat superior. As time goes on Harshaw will undoubtedly supply better and better crystals.

The crystal is cemented with Canada balsam to a one-inch by two-inch diameter specimen of highly polished lucite, chosen for its optical clarity and "light-piping"
Fig. 1. Block diagram of coincidence scintillation spectrometer.
Fig. 2. Photograph of the instrument
ability. This assembly was next cemented to the active surface of a 5819 phototube, again with Canada balsam. Of course, the best mechanical fit of the lucite to the tube was first made by recessing the end of this short lucite rod to accept the curved surface of the photomultiplier.

The 5819 photomultiplier was chosen for this work since it has a large photocathode with end-window construction. Furthermore, its photocathode is a more efficient quantum converter, of the order of 3-8% depending on the tube, than other phototubes on the commercial market. Its signal to noise ratio is also greater.

With these improvements, the photomultiplier is far from perfect. Its photocathode is not uniform and hence its conversion efficiency will be different for different areas on the tube. This means that a determination of the energy of a mono-energetic beta ray will differ from one area of the tube to the next. In an effort to avoid collimation of the light striking the photo-surface, which in practice would diminish its intensity without very special precautions, it is found experimentally that one can collect light over an area equal to the effective area of the photo-surface with little of the difficulty mentioned above by using the short lucite rod, mentioned above, to separate the scintillating crystal from the photocathode. This is quite important for it decreases the line-width of the
instrument when comparison is made with the case when a large crystal is cemented to the tube. The resolution is also improved somewhat by staggering the dynode potentials from the photocathode to the anode in the ratios 2, 1, 1, 1, ---. The phototubes were operated at 920 volts in these experiments. Each dynode is at a higher potential than the preceding one. The electrons converted from photons at the photocathode are multiplied by secondary emission at each succeeding dynode stage. Thus, if a tube has "n" stages, each with a multiplication ratio $r$, the gain of the tube is $r^n$. Actually the number of secondary electrons produced at each dynode surface will vary in a statistical manner, so that pulses obtained from single electrons are not all of the same height. This has been studied by Morton and Mitchell (12) and by Hoyt (13). It is shown in Chapter IV that the electron multiplying surfaces of the phototube will increase the relative variance in pulse height by a factor $\frac{r}{r - 1}$. This being the case one must make the relative variance in the number of electrons entering the multiplier as small as possible. If it is necessary to use these commercial tubes, which are good, but of course can always be improved, then it is especially important to get the very best crystals obtainable and collect as much of the light scintillations as possible. In this work both tubes and
crystals have been as carefully selected as possible. One hardly can emphasize enough the dominant role played by the crystal in destroying the resolution. As soon as one is faced with the problem in actual practice and observes the difference in detail of an internal conversion line, such as the 630 kev. line in Cs$^{137}$, caused by two anthracene crystals which look a great deal alike in physical appearance, he will never again doubt that excellent crystals are required. It should be emphasized and re-emphasized that anthracene crystals to be really good must be as transparent to their own radiation as possible, contain no fissures, be single crystals, and as free from impurities as possible. Anthracene, when used with a photomultiplier as a scintillation counter, responds to beta rays over 15 kev. with 100% efficiency ($1_f$). It responds to gamma rays chiefly through the Compton process.

In many experiments it is desirable to count particles of low energy. Noise in the photomultiplier will determine this low energy limit. Morton (12) has shown that at room temperature in a good tube (type 1P21) there will be about 1,000 pulses per second that are larger than the average for one photoelectron, and only two noise pulses per second larger than eight electrons. With the 5819 the number of noise pulses is somewhat greater, but the
greater light collection which results from the larger photocathode will in many cases more than compensate for this increase of noise. Of course, cooling the multiplier can greatly reduce the noise background — Morton (12) observed a factor of 100 decrease at dry-ice temperature. Coincidence counting with two multipliers viewing the same source will also reduce the noise background. This work was done at room temperature and at the energies investigated, background noise was so low as to be completely negligible.

The phototube is sensitive to magnetic fields, even to weak fields like that of the earth. For this reason a mu-metal shield surrounds the phototube. It was demonstrated that without magnetic shielding different orientations in the earth’s field can change the counting rate by as much as 25%.

The phototube-lucite-crystal assembly is wrapped carefully with a 1/4 mil. foil of highly reflecting aluminum, as shown in Fig. 3, to increase the light collection efficiency. The whole system is shielded against extraneous light with a cardboard tube to reduce scattering. The top of this tube contains a small, thin, 1/32-inch walled aluminum mandrel, 1/2 inch in diameter. This mandrel centers and supports a cylindrical lucite source holder
Fig. 3. Phototube and crystal assembly and geometry for taking beta spectra without coincidence.
one inch long with a 3/64-inch wall. The source is de-
posited on a thin film which covers, like the covering on
a drum, one end of this holder. For the coincidence ex-
periment a flat lucite wafer whose center had been removed
to accommodate the source, served as the source holder.

G. Pre-amplifier and Amplifier

Fig. 4 is a schematic diagram of the pre-amplifier
which serves to couple the photomultiplier pulses, after
suitable pulse shaping, to the main amplifier.

The anode of the photomultiplier, with a recovery time
of 800 microseconds, is coupled by a cathode follower to
a highly degenerative amplifier whose output couples into
a delay line differentiator. This permits a square pulse
to be formed of 0.8 microseconds duration and whose height
is proportional to the charge accumulated on the anode of
the multiplier. This charge is proportional, on the average,
to the energy of the impinging beta ray. The pre-amplifier
has an overall gain of unity.

Fig. 5 is an electrical schematic of the power supply
for the pre-amplifier.

The main amplifier used here is a Bell-Jordan linear
amplifier which has been modified at its input so that it
will pass the square pulse formed by the pre-amplifier with
little distortion. The Bell-Jordan smears the rise time
Fig. 4. Electrical schematic of the phototube and pre-amplifier
Fig. 5. Electrical schematic of pre-amplifier power supply
of the pulse from 0.2 to 0.3 microseconds. This amplifier has been used so frequently that no further comments are necessary.

D. Differential-Coincidence Discriminator

In Fig. 6 is shown an electrical schematic of a typical differential discriminator channel. Tube $V_1$ has its bias chosen by the 50K precision potentiometer to fire on pulses greater than some preset voltage $E$. Tubes $V_1$, $V_2$, and $V_3$ comprise this trigger circuit, called No. 1 discriminator. If a pulse greater than $E$ arrives at the grid of $V_1$, a square pulse of fixed height is delivered to the grid of $V_4$. This pulse is delayed by a 0.4 microseconds delay line differentiator before being fed to the grid of $V_5$. The remainder of the circuit from $V_5$ through $V_9$ shapes the pulse to the desired shape for actuating the anti-coincidence circuit, tubes $V_{10}$ and $V_{11}$. Tube $V_{16}$ of No. 2 discriminator has its bias adjusted to fire at a voltage level of $E + 5E$, $5E$ being determined by the battery shown in Fig. 6. The rest of the circuit from $V_{18}$ through $V_{24}$ is a pulse shaping circuit, as before. No delay exists in this channel. From the pulse diagrams shown on Fig. 6, it is clear that $V_9$ and $V_{24}$ will simultaneously deliver pulses to the anti-coincidence circuit if the input pulse is greater than $E$ and $E + 5E$. Discriminator No. 1 has
Fig. 6. Electrical schematic showing a typical differential pulse height discriminator
its pulse delayed and shaped so that this pulse appears inside the pulse delivered from No. 2 discriminator. With this the case, a pulse is delivered from V_{10} which is of the wrong polarity to fire V_{12}. Hence, anti-coincidence has been accomplished and this pulse is rejected. On the other hand, if the input pulse is greater than E but less than \( E + \delta E \), only No. 1 discriminator will deliver a pulse to the anti-coincidence circuit whose output is then of the correct polarity to fire V_{12}. Further pulse shaping occurs from V_{12} through V_{15} before a pulse is delivered to channel A scaler. Channel B differential discriminator is identical to the one described above with one exception. The preferred input circuit shown in Fig. 7 replaces discriminators No. 1 and No. 2. It was found that when one was counting small pulses in the presence of large ones at rates in excess of 50,000 per second, biases on V_1 and V_2 were changed slightly by grid current drawn. This is not to be tolerated in precise work since the actual height measured for a pulse will be in error. For this reason all beta spectra presented in this report were taken using the preferred input.

Fig. 8 shows a typical calibration curve of a discriminator. Fig. 9 is an electrical schematic of the power supply for the differential coincidence discriminators.
Fig. 7. Electrical schematic of a preferred differential discriminator input.
Figure 8. Discriminator Calibration curve.
Fig. 9. Electrical schematic of the differential coincidence discriminator power supply
Pulses from the differential discriminator channels, A and B, feed into the coincidence circuit shown in Fig. 10. Tubes $V_{25}$, $V_{27}$, $V_{28}$, and $V_{29}$ comprise a gate circuit for varying the coincidence resolving time. Tube $V_{30}$ is the coincidence tube. Tubes $V_{31}$, $V_{32}$, $V_{33}$, and $V_{34}$ shape the coincidence pulse for actuating the coincidence scaler.

E. Scalers

The scalers used with this equipment are of two types. The unit used to count coincidence pulses is a commercial scale of 64 sold by Industrial Development Laboratory. The other two units, see Fig. 11, are identical and are similar to the Higginbotham scaler with the exception of the first four scaling units. These units are cathode follower coupled to increase their speed of response. These scalers require no adjustment and have a two pulse resolving time of 0.2 microseconds.

Fig. 12 is an electrical schematic of the power supply for the 0.2 microsecond scalers.
Fig. 10. Electrical schematic of the coincidence circuit
Fig. 11. Electrical schematic of the 0.2 microsecond scaler
Fig. 12. Electrical schematic of the power supply for the 0.2 microsecond scalers
IV. THEORETICAL CONSIDERATIONS

A. Response to Mono-energetic Electrons

Suppose an electron of energy $E$ strikes the anthracene crystal and is completely absorbed. The energy of the electron will be converted, in part, into $n'$ light quanta. The relative variance in the number of quanta $n'$ we shall designate as $X_{n'}^2$. This quantity is probably no greater than

$$\frac{1}{n'} = \frac{1}{ME},$$

(1)

where $M$ is a constant of proportionality and $E$ is the beta ray energy. An average fraction $f$ of these quanta will be focussed by the light reflector onto the photomultiplier cathode and some of them converted to photoelectrons. Designate the relative variance in optically focussed quanta for $n'$ into the optical system by $X_{n'}^2$. Let the average gain of the photocathode be designated as $r_K$, which is the number of photoelectrons ejected for one quantum into the photocathode. This quantity is certainly less than one, of the order of 0.03-0.08. Let the relative variance in photoelectrons produced by the quanta impinging on the photocathode be $X_K^2$. Assume all of the ejected photoelectrons are collected at the first dynode and that in the multiplying process no secondary electrons are lost. Let the average gain by secondary emission of the dynodes be designated by $r$ and the
relative variance due to the multiplication process by \( X_T^2 \).

Since at each stage in the process all events are independent, the relative variance in the output electrons will be given by the sum of the component parts. That is:

Relative variance in the output =
\[
X_x^2 + X_F^2 + X_K^2 + X_T^2 .
\]  

(2)

Now consider a single dynode in the photomultiplier and let \( p_n \) be the probability that \( n \) electrons will be ejected for one impinging electron. Since we will be interested in the aggregate contribution of the various parts of the photomultiplier system to the statistical variation of the system, this problem can best be treated with the use of the generating function appropriate to a dynode stage -- all stages being regarded identical in the present analysis. The generating function for the process of observation is defined by the series

\[
f(x) = p_0 + p_1 x + p_2 x^2 + \cdots + p_n x^n + \cdots
\]

(3)

The generating function possesses the following properties:

\[
f(0) = p_0 ;
\]  

(4)

\[
f(1) = 1 .
\]  

(5)
The mean value \( \bar{n} \) of a series of observations; namely,
\[
\bar{n} = \frac{\sum np_n}{n},
\]
(6)
is readily found to satisfy the relation
\[
\frac{d\bar{n}}{dx} \bigg|_{x=1}.
\]
(7)
Similarly, the variance of \( n \), defined by the relation
\[
\frac{(n - \bar{n})^2}{n} \sum (n - \bar{n})^2 p_n,
\]
(8)
is found to be related to the generating function by the equation
\[
\frac{(n - \bar{n})^2}{dx^2} = \frac{df(x)}{dx} \bigg|_{x=1} + \bar{n} - \bar{n}^2.
\]
(9)
For a single dynode structure with one electron impinging
\[
\bar{n} = r = \frac{\sum np_n}{n} = \frac{df(x)}{dx} \bigg|_{x=1}.
\]
(10)
Now if two electrons impinge on the dynode the probability that \( n \) electrons will be ejected is the sum
\[
p_n p_0 + p_{n-1} p_1 + \cdots + p_0 p_n,
\]
which is the coefficient of \( x^n \) in the expansion of \( f^2(x) \).
Similarly, the probability that \( n \) electrons will be ejected for \( \overline{m} \) impinging will be the coefficient of \( x^n \) in
\[
f^m(x) \equiv F(x).
\]
(11)
The relative spread in the number \( n \) will now be given by
\[ \frac{\hat{N}^2 - \bar{N}^2}{\bar{N}^2} = \left( \frac{\mathrm{d}^2 f(x)}{\mathrm{d}x^2} \right)_{x=1} + \left( \frac{\mathrm{d}f(x)}{\mathrm{d}x} \right)_{x=1} - 1, \]

\[ = \frac{1}{\bar{N}} \frac{\hat{N}^2 - \bar{N}^2}{\bar{N}^2} = \frac{1}{\bar{N}} \bar{x}^2. \quad (12) \]

Equation (12) states that the relative variance in the number of electrons out for \( \bar{N} \) in is equal to the relative variance in the number of electrons out for one in reduced by a factor of \( \frac{1}{\bar{N}} \).

If \( \bar{N} \) represents the average value of the number of photoelectrons \( n \) from the photocathode then it follows, since the photomultiplier has ten stages, that

\[ x_f^2 = \frac{1}{\bar{N}} x^2 + \frac{1}{r \bar{N}} x^2 + \cdots + \frac{1}{r^9 \bar{N}} x^2, \]

\[ = \frac{1}{\bar{N}} x^2 \left( 1 + \frac{1}{r} + \frac{1}{r^2} + \cdots + \frac{1}{r^9} \right), \]

\[ = \frac{1}{\bar{N}} x^2 \frac{r}{r - 1}. \quad (13) \]

By the same reasoning

\[ x_k^2 = \frac{1}{r^n \bar{N}} x_k^2, \quad (14) \]

\[ x_f^2 = \frac{1}{r^\bar{N}} x_f^2, \quad (14a) \]

where \( x_k \) represents the relative variance in photoelectrons for one quantum hitting the photocathode. \( x_f \) represents the relative variance in quanta impinging on the photocathode for one quantum emitted by the crystal. But

\[ \bar{N} = r_k \bar{n} \bar{f}. \quad (15) \]

Substituting (13), (14), and (15) in (2) we have
Relative variance in the output

\[ x^2 = x^2 + \frac{1}{n'} x^2 + \frac{1}{n'} x^2 + \frac{1}{r K T n'} \frac{r}{r - 1} x^2. \tag{16} \]

The relative variance \( x^2 \) can be replaced by \( \frac{1}{n'} \). Using equation (1), we find by substitution into (11) that the relative variance in the output

\[ \frac{1}{M E} \left( 1 + x^2 + \frac{1}{r} x^2 + \frac{1}{r K T} \frac{r}{r - 1} x^2 \right). \tag{17} \]

We see from equation (17), subject to the condition that \( x^2 \) is given by (1), that the fractional deviation in output electrons from the photomultiplier is proportional to the inverse square root of \( E \). Consequently, the deviation is proportional to the square root of \( E \), a vital result in determining transmission corrections.

By observing the conversion line in Cs137, Fig. 17, and remembering the above result, we note that the line shape can be approximated closely by a Gaussian function of the form

\[ T(V, E_0) = \frac{A}{(KE_0/\pi)^{1/2}} \times \frac{(V - E_0/K)^2}{KE_0}. \tag{18} \]

where \( A, K, \) and \( k \) are proportionality constants. The coefficient of the exponential must be chosen so that

*Note that \( \sqrt{KE_0} \) is the half-width of the line down 1/e from the maximum.
\[
\int_{0}^{\infty} T(V, E_0) \, dv = 1, \tag{19}
\]

since \(T(V, E_0)\) really represents the probability that a count be observed at voltage \(V\) when the beta particle has energy \(E_0\). The total probability must be unity, assuming a pulse of some height is registered for every electron which strikes the crystal.

Hopkins (1) has measured the line width for this type of instrument and his observations seem to agree with the theory. He has also found that the response of the scintillation counter is proportional to the energy of the impinging electron plus a small constant such that a straight line fits the experimental data very well. This line, however, intersects the energy abscissae at 20 kev., as mentioned above, instead of going through the origin. At energies of interest in this work 20 kev. is so small compared with the energies investigated that it has been ignored.

B. Transmission Corrections

1. In the body of a beta spectrum

In the body of the spectrum of a beta emitter the observed counting rate will be given by the integral equation
\[ N_e(V) = \int_0^U T(V,E_0) n_t(E_0) dE_0, \]
\[ = \int_0^\infty T(V,E_0) n_t(E_0) dE_0. \]  \hspace{1cm} (20)

where \( U \) represents the end point of \( n_t(E_0) \), the true distribution.

The question we ask is: Given \( N_e(V) \) from the experimental data, and knowing \( T(V,E_0) \), what is the true distribution \( n_t(E_0) \)?

Equation (20) can be approximated in the following manner. Make a Taylor's expansion of \( n_t(E_0) \) about the energy \( E \) corresponding to the voltage \( V \), the unit in which \( E \) is measured; that is,

\[ E = kV \]  \hspace{1cm} (21)

We get:

\[ N_e(V) \approx n_t(E) \int_0^\infty T(V,E_0) dE_0 + n'_t(E) \int_0^\infty (E_0 - E) T(V,E_0) dE_0 \]
\[ + \frac{1}{2} n''_t(E) \int_0^\infty (E_0 - E)^2 T(V,E_0) dE_0 \]  \hspace{1cm} (22)

where

\[ M_n = \int_0^\infty e^{-\frac{(V-E_0/k)^2}{KE_0}} (E_0-kV)^n dE_0 \]  \hspace{1cm} (23)

From integrals given by de Haan (15), we find
\[ M_0 = kA \]  \hspace{1cm} (24) \\
\[ M_1 = \frac{k^2 A}{2v} R^2 \]  \hspace{1cm} (25) \\
\[ M_2 = \frac{k^3 A R^2}{2} \left( 1 + 3/2 \frac{R^2}{v^2} \right) \sim \frac{k^3 A R^2}{2} \]  \hspace{1cm} (26) \\

where
\[ R^2 = L V, \]  \hspace{1cm} (27) \\
L being a constant. \( R \) represents the half-width of the line at 1/e of the maximum when the line centers at voltage \( V \). 

Proceeding with these results for the Gaussian \( V \)-dependence we have
\[ N_e(V) = k A n_t(E) + \frac{k^2 A R^2}{2v} n_t'(E) + \frac{k^3 A R^2}{4} n_t''(E) \]  \hspace{1cm} (28) \\

Differentiating this equation twice with respect to \( V \) and neglecting derivatives beyond the second we have three equations with the three unknowns \( n_t(E), n_t'(E), \) and \( n_t''(E) \). Solving for \( n_t(E) \), we get
\[ n_t(E) = \frac{1}{kA} \left[ N_e(V) - \frac{R^2}{2v^2} v N_e'(V) - \frac{1}{4} \frac{R^2}{v^2} v^2 N_e''(V) \right] \]  \hspace{1cm} (29) \\
Equation (29) is used to correct the body of the spectrum. 

The two derivatives are calculated by the 7-multiplier (16) method of least squares. Thus, if the measurements are \( Y_3, Y_2, Y_1, Y_0, Y_1, Y_2, Y_3 \), then
\[ y_0' = \frac{1}{252\text{(interval)}} \left( 22y_{-3} - 67y_{-2} - 58y_{-1} + 58y_1 + 67y_2 - 22y_3 \right) \]  
(30)

\[ y_0'' = \frac{1}{42\text{(interval)}^2} \left( 5y_{-3} - 3y_{-1} - 4y_0 - 3y_1 + 5y_3 \right) \]  
(31)

The constant interval between consecutive points is measured in the same units as the line half-width. This 7-multiplier method involves fitting the best cubic to the observed data.

2. Near the end-point of a beta spectrum

Near the end-point of a beta spectrum theory predicts a parabolic shape for the form of the spectrum and, hence, we can write for the true distribution

\[ n_t(E_0) = n_t(E) \left( \frac{E_0 - U}{E - U} \right)^2 \quad \text{for} \quad E_0 < U, \]
\[ = 0 \quad \text{for} \quad E_0 > U, \]  
(32)

where \( U \) is the energy end-point of the spectrum.

For convenience, to avoid troublesome integrals, we will replace our Gaussian distribution with the best-fitting triangle which, of course, can be characterized by the width \( R \) of the triangle at half-maximum. Using these facts a solution of the integral equation (20) leads to the end-point correction shown in the accompanying graph, Fig. 13.
Fig. 13. End-point correction to beta spectra
C. Coincidence Spectrometry

A good review of the methods involved in coincidence counting is given in the doctoral thesis of W. W. Pratt (17). We will here write down only the vital equations necessary for the Rb$^{86}$ data discussed in Chapter V.

Referring to Fig. 14, the observed coincidence rate $N_0$ is related to the true coincidence rate $N_0$ and the accidental rate $N_a$ by the equation

$$N_0 = N_a + N_0 \quad (33)$$

The accidental rate is determined by the rates $N(\beta)$ and $N(\gamma)$ in the separate channels and the resolving time $T$ of the coincidence circuit by the equation

$$N_a = N(\beta) N(\gamma) T. \quad (34)$$

Normally a factor two multiplies the right-hand side of this equation, but in this case the construction of the coincidence system voids this factor. The reason for this is that the coincidence system used here orders the separate rates $N(\beta)$ and $N(\gamma)$. That is, a count must appear in the gamma channel before a beta channel count can be recorded in coincidence.

From these two equations $N_0$ can be determined if $T$, $N(\beta)$, $N(\gamma)$, and $N_0$ are known.
Fig. 14. Geometry and block diagram for coincidence counting
Using two separate sources in which there are no true coincidences, the observed rate equals the accidental rate and thus $T$ can be determined experimentally. A series of determinations led to an average value for $T$ of 0.533 microseconds. The resolving time $T$ was made longer than would have been preferred since coincidence was made after the pulses had been amplified and run through the discriminators. The rise time of the Bell-Jordan amplifiers (0.2-0.3 microseconds) sets a lower limit on the resolving time and one was chosen greater than this rise time for obvious reasons.
V. BETA SPECTRA

A. $^{32}$P

This isotope is well known. It is a pure beta emitter of the allowed type and has been investigated by Lawson (18) and many others. It is of interest here as a check on the methods of scintillation beta ray spectroscopy.

Approximately 0.5 mg./cm$^2$ source thickness of this isotope was mounted on a 50 microgr./cm$^2$ formvar film, which was then mounted in a holder near the anthracene crystal. Fig. 3 illustrates the geometry used in the experiment. The counting rate was then measured as a function of the pulse height selector setting using a single differential discriminator. The data collected in this way are shown by solid dots in Fig. 15. The body of the experimental data was then corrected for the poor resolution of the instrument as was discussed under "Transmission Corrections" in Chapter IV. A Kurie plot was made from these corrected data as is shown by open circles in Fig. 16. This determination leads to an endpoint of 1.9 mev. It will be noticed that the abscissa scale is in terms of the total energy expressed in units of the rest mass of the electron (0.5108 mev.). A con-
Fig. 15. Experimental spectrum of $^{32}P$ showing transmission corrections to the data (open circles)
Fig. 16. Kurie plot of $^{32}$P showing transmission corrections (solid circles)
version from pulse height in volts to total energy units was made by using the internal conversion line in Cs\(^{137}\) (630 kev.) as a calibration of the voltage scale. The geometry in this calibration determination was the same as above. The Cs\(^{137}\) isotope was mounted on a 1/4 mil. aluminum foil. Using 1.9 mev. as the end-point one is now able to make the end-point correction to the experimental data as was discussed under "Transmission Corrections" in Chapter IV. The effect on the Kurie plot caused by these corrections is shown by solid circles. This leads to a corrected end-point of 1.86 mev. This determination agrees with the value of J. L. Lawson (18) of 1.72 mev. to 8%.

Now \(^{32}\)P is known to be an allowed spectrum and therefore all points on the Kurie graph should have fallen on a straight line. The statistics in this experiment were excellent and the equipment itself could not have introduced such a large departure from a straight line at lower energies. A slight error in the calibration determination would have left this effect unchanged. One possible explanation, since this effect is seen in other spectra in this investigation, is that electrons are scattered out of the crystal before they have lost all of their energy and hence one would observe a preponderance of low energy pulses. With this the case one could correct for the anomaly by knowing the true response at low energies when the
apparatus is used to analyze mono-energetic electrons. This in effect would amount to transmission corrections other than the ones used here. The effect only occurs in the lower energy region of a spectrum and should not affect the bulk of the spectrum below the end-point. The end-point determination would likewise remain unaltered. P. R. Bell (8) has also observed this anomaly. It is of interest at this point, that the K and L conversion lines of Te\textsuperscript{125}, 78 kev. and 105 kev., respectively, were not resolved.

B. Cs\textsuperscript{137}

The radiations from this isotope are well known (19, 20). Less than 1 mg./cm\textsuperscript{2} of this material in thickness was deposited on a \textfrac{1}{4} mil. aluminum foil and placed near the crystal so that the geometry in this experiment was approximately 50\. The counting rate was then measured as a function of the pulse height in volts with and without a lucite absorber to stop the beta rays. The results are plotted in Fig. 17, the two curves being labeled "gamma only" and "gross counts", respectively. The difference between these two curves is plotted as the "gamma-subtracted" curve and clearly shows the beta ray and the conversion line. The fact that the line is known to have an energy of 630 kev. establishes a calibration point in energy
Fig. 17. Experimental beta and gamma spectrum of Cs$^{137}$ showing the conversion line.
units on the pulse height setting in volts. The background count was measured and found to have a completely negligible effect on the spectra as is the case in all spectra presented here.

Ca$^{137}$ is known to have an internally converted gamma ray (630 kev.), a lower beta ray (maximum energy 0.51 mev.), and a higher beta ray (maximum energy 1.2 mev.) (19). The higher beta ray is about 4% in abundance. Knowing the presence of these beta rays and their approximate energies allows one to deduct the contribution of the higher component to the lower beta-ray spectrum. This was done after the body of the lower spectrum was corrected, as discussed under "Transmission Corrections" in Chapter IV, for the poor resolution of the instrument. Next a Kurie plot was made. This is shown in Fig. 18. The best straight line through the open points gives a provisional end-point of 2.05 m$^2$e units. Using this provisional end-point, an end-point correction to the data was made and the effect of these corrections on the Kurie plot is shown as solid points. A smooth curve drawn through the corrected points leads to an end-point of 0.5 mev. for the lower beta rays. This value is within 2% of the known value of 0.51 mev. (19, 20). This beta ray, according to P. R. Bell (7), is first forbidden. A first forbidden correction could
Fig. 18. Kurie plot of Cs$^{137}$ showing transmission corrections (solid circles)
have been applied but has not been since it is felt the result would not be definitive. Our data agrees well with that of P. R. Bell's scintillation work. It is interesting that the Kurie plot of Bell's is much like our own with perhaps slightly greater curvature in spite of the fact that our transmission corrections to the data involve both first and second derivative corrections. It is understood that Bell uses only the second derivative correction (16). Furthermore, he has applied this second derivative correction quite near the end-point where it is clear that it cannot be applied legitimately since the spread in the transmission curve near the end-point overlaps the end-point as is seen from the magnitude of these end-point corrections in Fig. 18. Furthermore, one should point out that although the second derivative correction is larger near the end-point, it is not sufficiently large to mask completely the first derivative correction. This is particularly the case when one proceeds down into the body of the spectrum where the two corrections are of the same sign and of the same order of magnitude.

G. Rb$^{86}$

Fig. 14 illustrates the geometry used in this experiment as well as a block diagram of the arrangement of the necessary apparatus for coincidence counting. A fairly
thick source (15 to 20 mg./cm²) of Rb₂CO₃ was prepared and placed on a thin nylon film. A thick source was used to increase the number of bona fide coincidence counts to a reasonable number so that one could get fairly decent statistics without counting for an unreasonable length of time. Still the source was thin enough that the lower beta-ray spectrum measured would not be appreciably affected. However, it was found necessary to count for intervals of thirty minutes nevertheless. ³⁷Rb⁸⁶ decays to ³⁸Sr⁸⁶ by two alternative paths. According to Zaffarano, Kern and Mitchell (21), 20% of the transitions go by way of a beta decay (0.716 mev.) to an excited state in ³⁸Sr⁸⁶ which is followed by gamma radiation (1.081 mev.), in coincidence, to the ground state of ³⁸Sr⁸⁶. A beta transition (1.822 mev.) directly to the ground state of ³⁸Sr⁸⁶ accounts for 80% of the transitions.

In Fig. 14 the detector labeled "beta-counter" recorded counts caused by both beta rays and also, but to a smaller degree, since the anthracene crystal was only thick enough to stop the lower beta ray, some gamma counts when the gamma ray is converted in the crystal by the Compton process, the process most probable. The detector labeled "gamma counter" counted only gamma rays since the 1/2 cm. absorber of aluminum was sufficient to absorb all beta radiation. Here a thick anthracene crystal was used to
increase the probability for detecting gamma rays. If now one uses a differential discriminator in the beta channel to select all pulses as to heights and if all these ordered pulses are run into a coincidence circuit into the other side of which is fed all of the gamma-ray pulses above noise, the coincidence impulses out will dictate the number of ordered beta rays in the inner spectrum of a particular energy determined by the differential discriminator setting. Since one must allow for the chance coincidences due to the finite resolving time of the coincidence circuit, it was necessary to record the gross counting rates in both the beta channel and gamma channel. This is clearly pointed out under "Coincidence Spectrometry" in Chapter IV.

The coincidence resolving time was determined both before and after the experiment by using two random sources in which there were no true coincidences. (See Chapter IV, "Coincidence Spectrometry.") The average value of the coincidence resolving time from eighteen determinations was 0.533 microseconds.

The number of true coincidence counts at a particular pulse height setting were determined for several pulse height settings to obtain the beta spectrum shown in Fig. 19. Here each point represents the average of four determinations, each of 30 minutes duration. The spectrum was taken four times.
Fig. 19. Experimental spectrum of the inner beta ray in Rb$^{86}$. These data were taken by coincidence counting. Also shown is the Ca$^{137}$ conversion line.
The experiment terminated with a calibration of the pulse height scale using again the Cs$^{137}$ conversion line shown also in Fig. 19.

After correcting the spectrum in the usual way, described previously, a Kurie plot was made and the results are shown in Fig. 20. No end-point corrections have been made since it is felt that the data could be in error by as much as 15%. The Kurie plot gives an end-point of 674 kev. for the lower beta ray. This is to be compared with 716 kev. determined by Zaffarano, Kern, and Mitchell (21).

D. Sr$^{90}$

The beta spectrum of Sr$^{90}$ was taken for comparison with the spectrum of Rb$^{86}$, since it is known that Sr$^{90}$ is first forbidden with an end-point of 537 kev. (22, 23). The Rb$^{86}$ source was removed and a 5 mg./cm$^2$ source of Sr$^{90}$, chemically separated from its Y$^{90}$ daughter, was put in its place. The 1/2 cm aluminum absorber used in the Rb$^{86}$ experiment was removed. Of course, no coincidence counting was done in this experiment; only the beta spectrum was taken. The experimental data are shown in Fig. 21. After calibrating with Cs$^{137}$ and applying the usual corrections for the poor resolution of the instrument a Kurie plot, shown in Fig. 22, was made. This leads to an end-point of the Sr$^{90}$ beta ray of 562 kev. in excellent agreement with
Fig. 20. Kurie plot of Rb$^{86}$.
Fig. 21. Experimental spectrum of Sr-90

Counts per min. x 10^{-3}

Counts

40

80

Pulse Height (Volts)

630 keV
Fig. 22. Kurie plot of Sr\textsuperscript{90} showing transmission corrections (solid circles)
published values (22, 23). The Kurie plot has only a slight curvature, which may or may not be significant. In this experiment, as in all experiments except the Rb$^{86}$ coincidence experiment, the statistics are excellent and the equipment presented no drift problems since the data were taken in less than one hour. It is interesting that the Y$^{90}$ daughter component could be detected to have an energy of approximately 2.3 mev. although the intensity of this component was about a thousand times lower than its Sr$^{90}$ parent.

From a comparison of the Rb$^{86}$ and Sr$^{90}$ spectra and their Kurie plots one might speculate that Rb$^{86}$ has the same forbiddenness which Sr$^{90}$ has; namely, first forbidden. This is merely a speculation, however, since we have no detailed experimental proof because of resolution difficulties previously mentioned. All we really know is that for the greater portion of the line shape we are not too far in error. End-point determinations of known beta emitters confirm this. Whether a tail in the transmission on the lower energy side exists, and the data indicate something of this kind is true, is not proven. Furthermore, if such a tail exists, its shape as a function of energy is also unknown. Small departures from the transmission used in this work, such as a small tail at lower energies coupled with the possibility that the line width may not go quite with the square root of energy, makes the determina-
tion of forbiddenness in beta spectra strictly question-
able. P. R. Bell (8, 9) seems to put great faith in his
calculations but these determinations have been unproven.

E. Ag$^{112}$ and Ag$^{113}$

Stable CdNO$_3$ was bombarded at the synchrotron by
x-rays whose spectrum extended to nominally 70 mev. The
silver activities produced by the gamma-proton reaction
were then separated chemically from the Cd constituent and
prepared on a 1 mil. aluminum foil. In the sample were
present some long life activities below 1 mev. and two
higher energy beta groups attributable to Ag$^{112}$ and Ag$^{113}$.
The sample was quite weak but this gave no problem stati-
tically when using the scintillation spectrometer parti-
cularly since the source was laid directly on the 1/4 mil.
aluminum light reflector covering the surface of the
crystal (1 1/2" dia. by 1/2" thick). With the discriminator
set to count all pulses above 3.2 mev. a half-life deter-
mination, Fig. 23 was made. This result gave 3.2 hours for
the Ag$^{112}$ beta activity. At the end of this time most of
the higher component had died and the discriminator was
next set to count pulses above 700 kev. A half-life deter-
mination, Fig. 23, yielded 5.3 hours for the Ag$^{113}$ beta ray.
These half-life determinations agreed with determinations
made by M. L. Pool (24), R. B. Duffield and J. D. Knight (25),
Fig. 23. Half-life determinations of Ag\textsuperscript{112} and Ag\textsuperscript{113}
and J. A. Seiler (26) for Ag$^{112}$ and Ag$^{113}$.

A new sample, prepared as before, was made. In this experiment the composite spectrum for the higher beta ray and the lower one was taken. After five half-lives of the Ag$^{112}$ activity the spectrum was again taken for the lower beta ray. The spectrum in no case was extended below 700 kev. By correcting the Ag$^{113}$ data for decay Fig. 24, a subtraction was made and the two components separated. The spectra, now separated, were next corrected to a time 1/2 hour after the bombardment ceased, Fig. 25.

In the usual manner corrections were made and Kurie-graphs drawn, Figs. 26 and 27. The end-points found for the activities were 4.2 mev. and 2.0 mev., respectively, for Ag$^{112}$ and Ag$^{113}$. These determinations have an accuracy of 8% or better. The energy of Ag$^{112}$ agrees closely with 3.6 mev. determined by J. H. Seiler (26) but does not agree with 2.2 mev. determined by M. L. Pool (24). Pool's determination came from a cloud chamber experiment while that of Seiler was determined by absorption. Why Pool's determination is so low is not known.

Assuming both spectra are allowed, a theoretical determination for the total normalized counting rate of each spectrum was made by the formula

$$N(W) = \frac{W}{\alpha} F(\alpha, 4\beta) (W-U_o)^2$$  \hspace{1cm} (35)
Fig. 24. Experimental spectra of the Ag activities
Fig. 25. Separated spectra of Ag$^{112}$ and Ag$^{113}$
Fig. 26. Kurie plot of Ag$^{112}$.
Fig. 27. Kurie plot of Ag¹¹³
where $N(W)$ = number of counts per unit energy interval

$W$ = total energy in MoC$^2$ units

$\alpha$ = momentum in MoC units

$U_o$ = end-point of the beta spectra

Using the slopes of the Kurie-graphs, Figs. 26 and 27, the total unnormalized counting rates were determined. The relative yield is related to the relative cross sections for the reactions by the equation

$$\frac{R_{113}}{R_{112}} = \frac{n_{113}}{n_{112}} \frac{\sigma_{113}}{\sigma_{112}} \frac{1-e^{-\lambda_{113}t_0}}{1-e^{-\lambda_{112}t_0}}$$

where

$t_o$ = time of the bombardment = 3.5 hours

$\frac{R_{113}}{R_{112}}$ = relative yield of Ag$^{113}$ to Ag$^{112}$ = 2.2

$\frac{n_{113}}{n_{112}}$ = ratio of abundance of Cd$^{114}$ to Cd$^{113}$ = 2.28

$\lambda_{113} = 0.131$ hr$^{-1}$

$\lambda_{112} = 0.217$ hr$^{-1}$

Substitution yields

$$\frac{\sigma_{113}}{\sigma_{112}} = 1.5$$
VI. CONCLUSIONS

Ca$^{137}$, Sr$^{90}$, and P$^{32}$ have end-points which are well
known. In obtaining spectra, it is demonstrated that
end-point determinations can be made with this instrument
to an accuracy of something like 8% or better. With data
equally as good statistically, end-point determinations for
Ag$^{113}$ and Ag$^{112}$ have been found to be 2.0 mev. and 4.2 mev.,
respectively. The end-point for the inner beta spectrum
of Rb$^{86}$ has been found to be 674 kev. within about 15%.
Without further proof one can only speculate that Rb$^{86}$ has
the same degree of forbiddenness as Sr$^{90}$, namely, first
forbidden.

At a maximum x-ray energy of nominally 70 mev., using
the synchrotron to produce Ag$^{112}$ and Ag$^{113}$ from Cd$^{113}$ and
Cd$^{114}$ respectively, by a gamma-proton reaction, the ratio
of the cross section of Ag$^{113}$ to Ag$^{112}$ is found to be 1.5.
The relative yield of Ag$^{113}$ to Ag$^{112}$ equals 2.2.

It is probable that corrections for forbiddenness can
be made to the beta spectra data from this instrument.
This can be done with certainty only when a detailed know-
ledge of the transmission is known.

Because the instrument can investigate activities
about 1000 times as weak as those needed for magnetic
vegetate near easterly is at 1. in thel.
- from as Home State College. of course being able to in-
- many activities which can be produced by the 70 mev. synchro-
- spectrometers. It promises to be a very useful tool for

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VII. RECOMMENDATIONS

As is always the case in experimental work, a piece of work is never really finished. Always items arise which could have improved the experimental work. Most important of these is a thorough examination of the transmission of the instrument for mono-energetic electrons. Questions which arise are: What is the shape of the transmission distribution for values of energy far removed from the center where most of the contribution is found and, in particular, what is the detailed structure in the low energy region? Does the half-width of the line really vary precisely as the square root of the particle energy? Such questions as these can be studied in detail and work on these matters is continuing. These are vital points for the determination of structure in a beta spectrum and to a lesser degree in the end-point determinations. One would like also to improve the resolution, and, if possible, to such a point that corrections are hardly necessary. Of course the answer to this is simply better crystals, better optics, better phototubes — particularly photocathode. By better crystals is meant crystals with higher "technical efficiencies" and "physical efficiencies". A photo-surface with a conversion efficiency of approximately 5% is rather
good but percentage wise, extremely poor. Work should be, and will be carried on in these directions at this laboratory.

Again it would be very nice to use multi-channel discriminators so that the whole spectrum could be taken in a single run. This is excellent for short life activities which the synchrotron can produce nicely. In pulse height discrimination it again would be better to take a coarse slice out of the pulse distribution, amplify this slice and perform differential discrimination on the amplified section. This means the setting of the discriminator window could be made small easily. If $\delta E$ represents the coarse window selection and $G$ = the gain of the amplified section, the effective value of the window would be $\delta E/G$. Circuit difficulties are simpler when attacked in this manner. More work should go into developing circuits of higher stability so that drifts for longer periods than six hours and from turn-on to turn-off of the equipment would be so small that the instrument could be used as an absolute calibration in itself without the attendant difficulties associated with calibration from such sources as Cs$^{137}$ every time an experiment is performed. Often for source geometrical reasons Cs$^{137}$ calibration at the end of an experiment can be difficult.
Furthermore, better amplifiers than the Bell-Jordan Amplifier are needed for very fast counting. I would recommend an amplifier with 10 mc. response, a gain of 1000 with coarse and fine gain adjustments similar to those found on the Bell-Jordan Amplifier.

When one remembers that the gain of the 5819 photomultiplier tube varies as the seventh power of its high voltage supply, he realizes extreme stability in this respect is necessary if we are to have the instrument a standard in itself.
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