carriers is so much greater than that of the positive ions.

Whereas no measurements are available with which the present saturation curves in purified gases could be compared, a saturation curve at one atmos. of tank argon was taken for α-particles at right angles to the electric field. This can be compared with Helbig's[6] curve taken under similar conditions. While the argon used by us was supplied as 99.6 percent pure, Helbig's measurements were carried out in argon supplied as 99.5 percent pure. The two curves agree closely, both giving saturation at a field strength of about 18 volts/cm. It is interesting to note that this value is more than twice as great as the field strength required for saturation in purified argon at the same pressure when the paths of the α-particles are parallel to the field.

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The Angular Correlation of Successive Gamma-Rays*

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The angular dependence of gamma-gamma coincidences has been studied for Na^{24}, Cl^{38}, and Br^{82} in an attempt to evaluate the predicted angular correlations between successive quanta. The coincidence rates were found to be independent of the relative directions of the quanta to within a few percent except as the counters approached each other closely, where an increase was observed. This increase was attributed to scattered quanta and was measured by an independent method.

INTRODUCTION

It has been pointed out by Pryce and Dunworth[1] that correlations are to be expected between the directions of quanta emitted in cascade from an excited nucleus, due to the coupling between the emitted quantum and the anisotropic radiation field of the preceding quantum. D. R. Hamilton[2] has calculated these spatial correlations for two successive dipole or quadrupole transitions involving three radiation levels. Since this analysis shows that the correlations are determined by the multipole orders of the transitions and the spins of the levels involved, some knowledge of these important quantities should be gained from experimental determinations of the directional correlations.

Several workers,[1,2] using coincidence counting methods, have failed to observe any angular dependence in the rate of gamma-ray coincidences from such sources of successive quanta. Recently, however, Watase,[3] using Cl^{38} gamma-rays, reported a marked difference between the double coincidence rate from quantum pairs having directions differing by π radians and $\pi/2$ radians. No directional dependence in the coincidence rate was observed with Na^{24} gamma-rays.

The present paper describes angular coincidence experiments of somewhat greater completeness than have heretofore been reported. Several extraneous effects inherent in this kind of coincidence experiment are discussed and evaluated.

THE COINCIDENCE METHOD

If $W(\theta)$ represents the probability that two coincidence quanta 1 and 2 be emitted from a point source with a relative angle of θ between their directions, and if two counters of vanishingly

* This paper together with reference 5 is part of a thesis presented to the Graduate School of Yale University for the Degree of Doctor of Philosophy.

[1] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).

[2] D. R. Hamilton, Phys. Rev. **58**, 122 (1940).

[3] Y. Watase, Proc. Phys. Math. Soc. Japan **23**, 618 (1941).

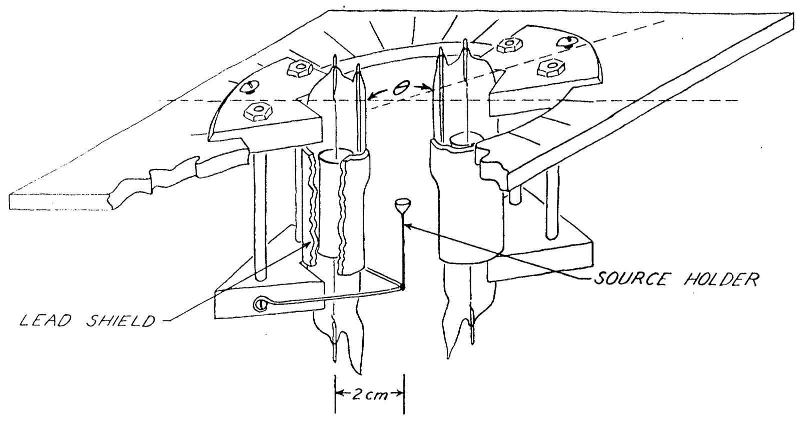

FIG. 1. Counter arrangement showing part of the circular track cut away. The track and counter cradles are brass. The source holder is aluminum foil. The counter to the right is movable from $\theta=60°$ to $\theta=300°$.

small aperture are symmetrically situated so as to intercept such quanta and are connected to an ideal amplifier, the number of true coincidence counts per single count in one counter is

$$C/N=[2\epsilon_1\epsilon_2/(\epsilon_1+\epsilon_2)]W(\theta), \tag{1}$$

where ϵ_1 and ϵ_2 are the net counter efficiencies (including counter aperture) for the respective quanta. In practice, the variation of quantum efficiency over the sensitive counter aperture and the imperfect spatial resolution of the counter set must be considered. The first effect is probably small,[4] while the second is easily calculated for a symmetrical arrangement of source and counters. Both effects may be included in the θ dependent term, and will tend to make the observed variation of C/N with θ somewhat less than the true variation given by $W(\theta)$. When correction has been made for the angular aperture of the counter set, the observed variation of C/N with θ gives $W(\theta)$ to within a multiplicative constant, since for a symmetrical counter arrangement $2\epsilon_1\epsilon_2/(\epsilon_1+\epsilon_2)$ is not a function of the angular settings of the counters. It is this function $W(\theta)$ which is related to the level spins and multipole orders in Hamilton's calculations.

In addition to any intrinsic angular dependence in C/N, the counters will themselves impose a variation of coincidence rate with θ, since they are composed of scattering material. Such coincidences are most probable for the penetration of both counters by a Compton electron originating at one of the counters. Fortunately, these coincidences are easily eliminated by surrounding the counters with absorbing shields. However, the less prevalent coincidences in which the recoil electron traverses one counter and the scattered quantum discharges the second counter cannot be avoided. The coincidence rate from this process may be represented as

$$N\varphi(\theta)\epsilon, \tag{2}$$

where N is the single counting rate, and $\varphi(\theta)$ a probability depending explicitly on θ and implicitly on the quantum energy. This factor $\varphi(\theta)$ could be calculated from the known angular spreads of recoil electrons and scattered quanta. However, it can be measured directly by using sources which emit no coincidence quanta. In such an experiment, the observed coincidence rate is

$$A'=A+N\varphi(\theta)\epsilon, \tag{3}$$

where A is the usual accidental coincidence rate $2\tau N^2$, τ being the resolving time of the coincidence set. It should be noted that in principle N will itself be a function of θ. Actually, this increase in N as the counters approach each other is quite negligible, being of the order of $N\varphi(\theta)\epsilon$.

A further variation of coincidence rate with θ may be due to beta-ray or gamma-ray pair production and subsequent two-quantum annihilation of the positrons in the source region. This is easily identified by the sharp maximum in the rate of coincidences at the in-line position of

[4] D. L. Jorgensen, Rev. Sci. Inst. 10, 34 (1939).

source and counters.[5] The elimination of absorbing material around the source makes this effect quite negligible.

If the angular resolving power of the coincidence apparatus is to be good, high single counting rates must be employed for a reasonable time of experiment. This is particularly true because of the low values of gamma-ray counter efficiencies. The angular aperture of the counter set shown in Fig. 1 was 0.75 steradian. The counters had copper cathodes and were filled with a mixture of 94 percent argon and 6 percent oxygen to a pressure of 9 cm of mercury.[6] One counter was kept fixed and the other moved on a circle about the source position. The cradles holding the counters were designed to keep scattering materials behind the sensitive counting volumes.

The coincidence amplifiers were of the usual three-stage resistance-capacity coupled design. The pre-amplifiers were a modified Neher-Pickering type.[7] Rossi pulse adding was employed with a grid-biased thyratron as coincidence discriminator. The smallest coupling condensers and resistors which permitted full saturation of the Rossi tubes were used throughout. The various grid and plate voltages were taken from glow-tube regulated voltage supplies backed with a constant voltage transformer. The high voltage counter supply used Street and Johnson[8] vacuum-tube regulation.

Single counting rates were measured with a hard-tube scale-of-sixteen[9] and self-quenching thyratron recording circuit. The single counting losses measured with "added sources"[10] and from the decay of sources of known half-life[11] were found to be in good agreement with the relation given by Alaoglu and Smith[12] in their analysis of single counting losses in a system composed of a counter, amplifier, scaling circuit, and recorder. By making use of their criteria, it was concluded that no counts were lost in the scaling circuit or recorder. For this case, the observed single counting rate is given by $N'=N/(1+N\sigma)$. For the amplifiers used, σ had a value of $8.0(10)^{-4}$ sec. This rather large resolving time could be associated only with the counter quenching circuit, and so it was probable that most of the single counting losses occurred at the counters themselves rather than in the amplifiers.

A study was made of the constancy of the coincidence resolving time τ, and it was found to increase with an increasing frequency of counter pulses arriving at the adding stage of the amplifier shown in Fig. 2. The effect was less pronounced as the adding resistance R was diminished, but the variation in τ could not be entirely eliminated.

In addition to the loss of single counts and the variation of coincidence resolving time with counting rate, there was an observed loss of true coincidence counts at high counting rates. This effect, unlike the other two, could not be evaluated quantitatively.

Notwithstanding the difficulties discussed, it was possible to determine the form of the angular correlation function $W(\theta)$ with considerable precision, by standardizing the single counting rates. The method which was used is outlined as

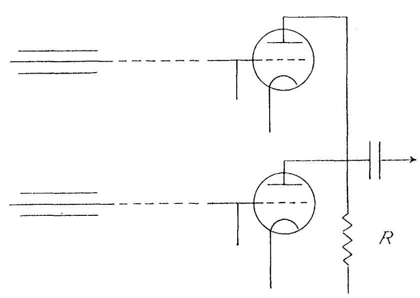

FIG. 2. Schematic circuit of the double coincidence amplifier showing the "adding resistance" R.

follows: The observed coincidence rate over a coincidence counting interval of t_2 seconds is

$$\bar{C}'+2\bar{\tau}\frac{1}{t_2}\int_0^{t_2} N_0{}^2 e^{-2\lambda t}dt$$

for a source of coincidences with a decay constant λ, where $\bar{\tau}$ is the coincidence resolving time observed with a very slowly decaying source emitting no true coincidences and of such

[5] R. Beringer and C. G. Montgomery, Phys. Rev. **61**, 222 (1942).
[6] C. G. Montgomery and D. D. Montgomery, J. Frank. Inst. **231**, 447–467, 509–545 (1941).
[7] T. H. Johnson, Rev. Sci. Inst. **9**, 218 (1938).
[8] Street and Johnson, J. Frank. Inst. **214**, 155 (1932).
[9] H. Lifschutz and J. L. Lawson, Rev. Sci. Inst. **9**, 83 (1938).
[10] Duffendack, Lifschutz, and Slawsky, Phys. Rev. **52**, 1231 (1937).
[11] A. Flammersfelt, Zeits. f. Physik **122**, 729 (1939).
[12] L. Alaoglu and N. M. Smith, Phys. Rev. **53**, 832 (1938).

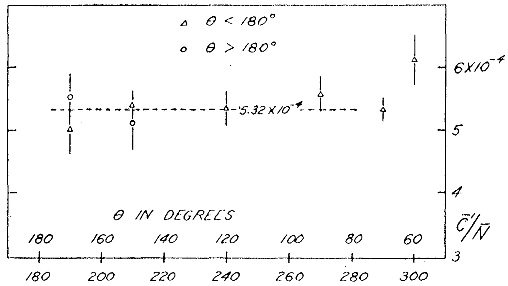

FIG. 3. The number of double coincidences per single count for Na^{24} gamma-rays as a function of the relative angle between the counters at the source.

strength as to produce

$$\bar{N}=\frac{1}{t_2}\int_0^{t_2} N_0 e^{-\lambda t}dt$$

single counts per second. The coincidence rate $\bar{C}'$ is then known, as is the ratio $\bar{C}'/\bar{N}$, which, if no true coincidence counts were lost, would be given by formula (1). When coincidences are lost in the counters and circuits, the factor $[2\epsilon_1\epsilon_2/(\epsilon_1+\epsilon_2)]W(\theta)$ in formula (1) must be multiplied by some complicated function of N containing various circuit constants as parameters. Since this loss term is a function only of the single counting rate for a given amplifier, it may be neglected in comparing observations for which the single counting rates are the same. For such a procedure, the variation of $\bar{C}'/\bar{N}$ with θ gives the form of $W(\theta)$ independently of the several kinds of counting losses.

In the early measurements, various values of $\bar{N}$ were used. These data showed systematic trends which could be attributed to the loss of coincidences at high counting rates. For the later measurements, a constant mean single counting rate was employed in the manner outlined. The coincidence resolving time $\bar{\tau}$ was determined frequently at this mean single counting rate using Ra gamma-rays as a source of accidental coincidences. A small, regular variation of $\bar{\tau}$ around a mean of about two microseconds was observed over periods of several weeks, and was attributed to slow changes in tube characteristics.

Na^{24}

The radiations from the 14.8-hour sodium activity have been widely studied and several level schemes have been advanced. The gamma-ray spectrometer experiments of Curran, Dee, and Strothers[13] and the coincidence experiments of Feather and Dunworth[14] indicate four gamma-ray levels with two modes of quantum dexcitation. Two quanta of 3.0 and 1.5 Mev are emitted in cascade in one mode, and two quanta of 2 Mev in the other mode. This second mode is of much lower intensity. The recent gamma-ray spectrometer experiments of J. Itoh[15] do not show the 2 Mev component, but give a three-level scheme, with two quanta of 2.8 and 1.4 Mev in cascade.

Sodium chloride sources of high specific activity were prepared by deuteron bombardment in the Yale cyclotron. Aged samples, free from Cl^{38} activity, were used for coincidence counting, prepared as pellets about 2 mm in diameter.

The variation of $\bar{C}'/\bar{N}$ with θ was measured twice without controlled singles rates and once with $\bar{N}=70$ sec.$^{-1}$. Each of the three runs showed no angular correlation in the range $90°\leqslant\theta\leqslant 270°$. Outside of this range, the coincidences rose rapidly as the counters approached each other. This rise was attributed to Compton coincidences. Figure 3 shows one set of data, and the results of the three runs are summarized in Table I. The mean $\bar{C}'/\bar{N}$ in Table I was calculated under the assumption of no correlations in the range $90°\leqslant\theta\leqslant 270°$. The agreement of the theoretical standard deviations in the mean $\bar{C}'/\bar{N}$, with the mean experimental deviations calculated from the number of coincidence counts used to determine each point, makes this assumption reasonable. One can say that angular correlations in $\bar{C}'/\bar{N}$ are less than 3 percent statistically. The curves indicate that correlations at any particular angular setting are less than 10 percent. The mean $\bar{C}'/\bar{N}$ is different in the three cases, being

TABLE I. Test for angular correlation.

Run	Mean $\bar{C}'/\bar{N}$ for $90°\leqslant\theta\leqslant 270°$	Standard deviation in mean $\bar{C}'/\bar{N}$	Mean experimental deviation in $\bar{C}'/\bar{N}$	R
1	$5.32(10)^{-4}$	$0.13(10)^{-4}$	$0.18(10)^{-4}$	10^4 ohm
2	4.64	0.06	0.21	10^5
3	5.20	0.12	0.22	$5(10)^4$

[13] Curran, Dee, and Strothers, Proc. Roy. Soc. **A174**, 546 (1940).
[14] N. Feather and J. V. Dunworth, Proc. Camb. Phil. Soc. **34**, 442 (1938).
[15] J. Itoh, Proc. Phys. Math. Soc. Japan **23**, 605 (1941).

higher for runs 1 and 3, where the "adding resistance" of the coincidence amplifier (see Fig. 2) was smallest.

Cl^{38}

The gamma-rays from 37.5-minute Cl^{38} have been observed by many workers, in particular by Curran, Dee, and Strothers[13] and J. Itoh.[15] Their experiments are in essential agreement, indicating two gamma-rays in cascade, with perhaps a third quantum of the same energy as the second of the cascade quanta. Itoh gives the energies as 2.2 and 1.6 Mev.

The Cl^{38} sources were prepared by deuteron bombardment of LiCl in the Yale cyclotron. Decay curves showed inappreciable contamination up to five hours after bombardment; beyond this time a small 15-hour activity, presumably from Na^{24}, was evident. In the coincidence experiments, chemically separated pellets of Cl^{38} in the form of AgCl were used and showed no contamination.

The results of the angular coincidence experiments are shown in Fig. 4. The mean single counting rate for each of these observations was 50 sec.$^{-1}$. As in the case of Na^{24}, no angular

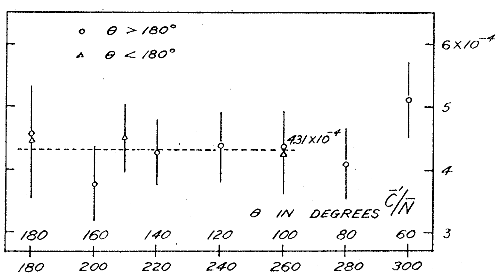

FIG. 4. The number of double coincidences per single count for Cl^{38} gamma-rays as a function of the relative angle between the counters at the source.

correlation was observed. The mean experimental deviation from the mean $\bar{C}'/\bar{N}$ was $0.17(10)^{-4}$ as compared with the theoretical standard deviation in the mean of $0.22(10)^{-4}$. Thus correlations are less than four percent statistically and less than about ten percent at any particular value of θ. This is in disagreement with the previous report which gave $(C/N)_{\theta=180°}/(C/N)_{\theta=90°}=0.85$ from coincidence counting at these two positions.

Br^{82}

Several recent studies of the gamma-rays from 36-hour Br^{82} have been reported.[16,17] Roberts, Downing, and Deutsch[16] give a level scheme with three gamma-rays of 0.547, 0.787, and 1.35 Mev in cascade. It was thought to be of some interest to see whether any angular correlations were detectable for this three-quantum process.

Br^{82} was produced by the bombardment of ethylene-dibromide with slow neutrons from the

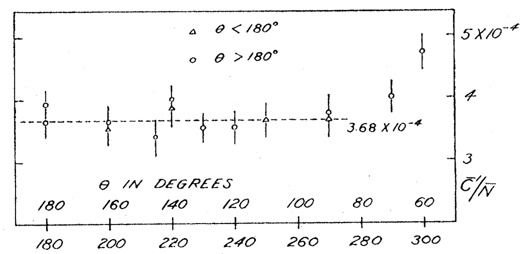

FIG. 5. The number of double coincidences per single count for Br^{82} gamma-rays as a function of the relative angle between the counters at the source.

Yale cyclotron. The active sample was concentrated by gravity separation in aqueous solution and the radio-bromine precipitated as AgBr, which was then aged for twenty-four hours before counting. The results of the angular coincidence experiments are shown in Fig. 5. The mean single counting rate for each of these observations was 70 sec.$^{-1}$. As before, no angular correlation is indicated in the range $90° \leqslant \theta \leqslant 270°$. In this range, the theoretical standard deviation from the arithmetic mean $\bar{C}'/\bar{N}$ of $3.68(10)^{-4}$ was $0.08(10)^{-4}$ and the mean observed deviation was $0.14(10)^{-4}$. Angular correlations are less than four percent statistically.

THE COMPTON COINCIDENCES

In each of the angular coincidence experiments, an increase in the coincidence rate was observed as the counters approached each other, as is to be expected for the Compton coincidences given by formula (2). This quantity $N\varphi(\theta)\epsilon$ was evaluated by means of formula (3), using Cu^{64} positron annihilation radiation as a source of uncorrelated quanta, which is valid in the region outside of the

[16] Roberts, Downing, and Deutsch, Phys. Rev. **60**, 544 (1941).
[17] J. Rotblat, Nature **148**, 371 (1941).

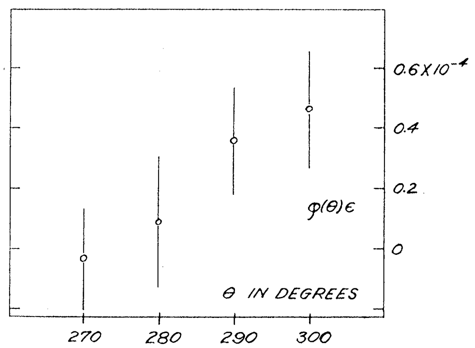

FIG. 6. The number of double coincidences per single count for Cu^{64} positron annihilation radiation as a function of the relative angle between the counters at the source. The coincidences are attributed to Compton scattering of the quanta.

allowed cone for two-quantum annihilation coincidences.[5]

The sources were prepared by bombarding copper foils with deuterons in the Yale cyclotron. The activated samples were then pressed into small pellets and wrapped in enough lead to stop all of the positrons. The results are shown in Fig. 6. The rise in the number of coincidences per single count from $\theta=270°$ to $\theta=300°$ is about $0.5(10)^{-4}$; slightly smaller than the contributions observed in Figs. 3, 4, and 5, but in accord with the low counter efficiency for the 0.5-Mev annihilation quanta.

THE COUNTER EFFICIENCY

Direct measurements of the gamma-ray counter efficiencies were not carried out. However, a comparison of the coincidence rates from the various sources was made, in which we used the generally accepted form of the change of efficiency with quantum energy for counters with copper cathodes as given by Dunworth and von Droste[1] and recently checked by Roberts, Downing, and Deutsch.[16] The coincidence rates under comparison were all taken at about the same mean single counting rate and with the same amplifiers. The scale of the efficiency curve was fixed with the measured $\bar{C}'/\bar{N}$ for Na^{24} of $5.2(10)^{-4}$, and by the four level scheme and the energies given by Curran, Dee, and Strothers.[13] The value of $\bar{C}'/\bar{N}$ calculated from this curve for Cl^{38} was $4.7(10)^{-4}$ for Itoh's level scheme,[15] which is in good agreement with the observed value of $4.3(10)^{-4}$. For Br^{82} the calculated value of $4.2(10)^{-4}$, obtained from the level scheme of Roberts, Downing, and Deutsch,[16] is in fair agreement with the observed value of $3.7(10)^{-4}$.

DISCUSSION

The lack of observed spatial correlations in the gamma-gamma coincidences from Cl^{38} and Na^{24} indicates that for these elements, this method of assigning level spins is not sufficiently sensitive under the limitations of present counting techniques. These negative results obtained do not constitute a check of the theory, since some of the calculated correlations are within the experimental errors. Further, nothing can be said as to which of these calculated correlations is most probable, since nothing is known of the level spins of these elements. The results of the experiments are therefore without satisfactory explanation, and a check of the theory must be made in a case in which the multipole orders and level spins can be independently assigned. Even if the theoretical predictions are verified, it is believed that considerable improvements must be made in counting methods, particularly as regards permissible resolving times, before the method of angular correlations can be used as a reliable means of investigating nuclear angular momenta.