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Direction and Polarization Correlations of Successive Gamma-Rays*

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Experimental arrangements for the measurement of directional and direction-polarization correlations of successive gamma-rays have been set up and considerations given to instrumental corrections and procedures for the analysis of the data. Directional measurements have been made with Co^{60}, Rh^{106}, Sb^{124}, As^{76}, and Ir^{194}, and direction-polarization measurements with Co^{60}, Rh^{106} and ThC''. The results are discussed in the light of present information on the systematics of the spins of the excited states of even-even nuclei. The occurrence of mixtures of magnetic dipole and electric quadrupole radiation in directional correlation experiments is summarized.

I. INTRODUCTION

As part of a general program for the study of the radiations and the excited states of radioactive nuclei, measurements of directional and polarization correlations of successively emitted gamma-rays have been undertaken.

The theory of direction and direction-polarization correlations was initially investigated by Hamilton[1,2] for dipole and quadrupole radiation, and that for polarization-polarization correlations by Falkoff.[3] It was first demonstrated by Brady and Deutsch[4] and Metzger and Deutsch[5] that such measurements can be of practical aid in the determination of the character of gamma-rays and the spins and parities of nuclear levels. Further work along both experimental and theoretical lines has been rapid and extensive in recent years. A summary of developments can be found in the recent review articles of Deutsch[6] and Frauenfelder.[7]

Because of the nature of the decay process, angular correlation measurements have been particularly well suited to obtaining information on the excited states of even-even nuclei. Much of the data recently used in the establishment of systematics in the spins and parities of the excited states of such nuclei[8–11] has been derived from correlation experiments. In the work reported here, some emphasis has been given to neutron induced activities that have a relatively short half-life.

In general, angular correlation studies are complicated by the presence of four disturbing factors, in addition to instrumental effects, any of which may make a unique assignment of spins, and parities of levels, and the character of the gamma-rays difficult.

(1) Gamma-rays in cascade, other than those being studied, are often present, and unless some form of selection is employed, a composite correlation may be observed. In this investigation integral pulse-height discrimination with scintillation counters served to partially select one cascade in the presence of others.

(2) In some cases there are ambiguities in the interpretation of the measured correlation even if only pure radiation is considered.

(3) Atomic perturbations have been shown to be present in the directional correlation of Cd^{111},[12] and are suspected of influencing the correlation in other cases (Rh^{106}, Co^{60}).[13] Theoretical considerations of this effect have been carried out by Goertzel[14] and Alder.[15] Aside from their intrinsic interest and the possibility they offer for the determination of the magnetic moment of the intermediate nuclear state, such perturbations can make the interpretation of the measured directional correlation difficult. Even if the intermediate nuclear state has no measurable lifetime, the excited atom may have a magnetic moment large enough to perturb the correlation.[16]

(4) It appears that the transition probabilities for $M1$ and $E2$ radiation may be comparable. Thus, when one pair of adjacent levels being studied have spins which differ by 1 or 0 units of angular momentum and have the same parity, a directional correlation may be observed that represents a mixture, with interference, of those two kinds of radiation. Correlations involving such mixtures have been observed in a number of cases. Ling and Falkoff[17] and Lloyd[18] have computed the coefficients to be expected for mixtures of various radiations, and Zinnes[19] has computed the effect of mixtures in direction-polarization correlations.

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[1] D. R. Hamilton, Phys. Rev. **58**, 122 (1940).
[2] D. R. Hamilton, Phys. Rev. **74**, 782 (1948).
[3] D. L. Falkoff, Phys. Rev. **73**, 518 (1948).
[4] E. L. Brady and M. Deutsch, Phys. Rev. **78**, 558 (1950).
[5] F. Metzger and M. Deutsch, Phys. Rev. **78**, 551 (1950).
[6] M. Deutsch, Rep. Phys. Soc. Progr. Phys. **14**, 196 (1951).
[7] H. Frauenfelder, to be published in *The Annual Review of Nuclear Science*, Vol. II.
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[10] P. Staehelin and P. Preiswerk, Helv. Phys. Acta **24**, 623 (1951).
[11] G. Scharff-Goldhaber, Phys. Rev. **87**, 218 (1952).
[12] Aeppli, Bishop, Frauenfelder, Walter, and Zunti, Phys. Rev. **82**, 550 (1951).
[13] Aeppli, Frauenfelder, Heer, and Ruetschi, Phys. Rev. **87**, 379 (1952).
[14] G. Goertzel, Phys. Rev. **70**, 897 (1946).
[15] K. Alder, Phys. Rev. **84**, 369 (1951); **83**, 1266 (1951).
[16] H. Frauenfelder, Phys. Rev. **82**, 549 (1951).
[17] D. S. Ling and D. L. Falkoff, Phys. Rev. **76**, 1639 (1949).
[18] S. P. Lloyd, Phys. Rev. **83**, 716 (1951).
[19] I. Zinnes, Phys. Rev. **80**, 386 (1951).

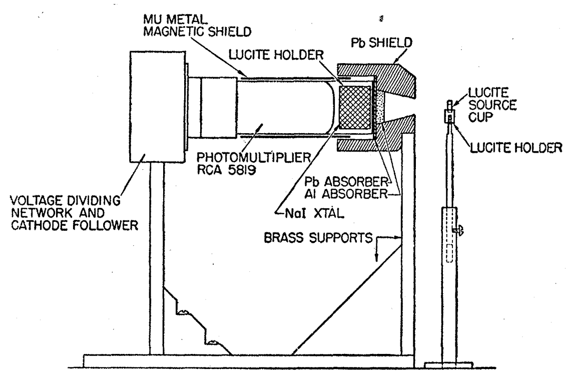

FIG. 1. The arrangement of the absorbers and the shields around the scintillation counter used in the measurement of directional correlations.

II. DIRECTIONAL CORRELATIONS

The determination of a directional correlation pattern involves the measurement of the coincidence counting rate as a function of the angle subtended at the source by two detectors. The general arrangement used in this investigation, is shown in Fig. 1. The Pb absorbers used in front of the scintillation counters to attenuate backscattered radiation were $\frac{1}{16}$ in. thick. The Al beta-absorbers were $\frac{1}{2}$ in. thick, and were used in addition to the Pb to prevent the counting of the energetic beta-rays that appear in some of the radioactive nuclei studied.

The electronic circuits involved were conventional and consisted of a cathode follower at the photomultiplier base, followed by a linear amplifier (5×10^{-8} sec rise time), pulse-height discriminator, and a blocking oscillator which generated a 1×10^{-7} sec pulse. One output of the blocking oscillator went to a univibrator which gave a pulse about 3×10^{-6} sec wide for operation of a scaler to record single events. Another output of the blocking oscillator went to a coincidence circuit employing a 6AS6 as a mixing tube. The coincidence resolving time was measured frequently in the course of the determination of a directional correlation. The counting rates used for the resolving time measurement were made the same as the ones involved with the source under investigation. The resolving time was about 1.2×10^{-7} sec.

The integral discriminator levels were kept sufficiently high so that the counters were insensitive to the Pb x-rays produced in the adjacent shields and absorbers. The "no bias" setting then was actually such that the discriminator levels were at about 0.1 Mev.

For the determination of the correlation function, measurements of the single and coincidence counts were made for fifteen minute periods at the variable angle θ, 90°, 270°, and $360°-\theta$. The sum of θ and $360°-\theta$ together with the sum of 90° and 270° were used in the calculation of $\epsilon(\theta)$.[20] An inverted order of taking data, i.e., 90°, θ, $360°-\theta$, 270°, was then followed. The coincidence counts were normalized by dividing by the product of the single counts. The method of normalization over-corrects for the decay of the source. However, due to the systematic method of taking data, as described above, this effect cancels to the first order and for the shortest activity studied (19 hours) was found to be negligible. The "fixed" counter was periodically rotated to a new position to insure that there was no scattering from adjacent room walls and equipment.

A function of the form $\epsilon(\theta)=a_2\cos^2\theta+a_4\cos^4\theta$ was fitted by a least squares procedure[21] to the experimental points. In a manner described in a previous publication[22] these coefficients were then adjusted for the finite detector resolution. The half-width at half-maximum δ_0 of the angular resolution curve was 12.0° in all cases except for Co^{60} where it was 10.5°, and for part of the work on Rh^{106}, where it was 6.6°. Values of $4\pi Bn/(2n+1)$ were determined by numerical integration and were, for example, in the 12.0° geometry: 1.0000, 0.9050, and 0.7653 for n equal to 0, 2, and 4.

In order to test the apparatus for the presence of effects, such as scattering which would artificially provide an anisotropy, the coincidence counting rate was measured as a function of θ using a source emitting a single gamma-ray. For this purpose a source of Be^7 known to be free of contaminants was used. The use of Cs^{137} and Fe^{59} was avoided inasmuch as these have been shown[23] to possess a small, but measurable, number of coincidence gamma-rays. With no bias and with $\frac{1}{16}$ in. of Pb in front of both counters, the coincidence counting rates at 90°, 130°, and 180° were found to be the same within the statistical accuracy. If

[20] In the notation of Brady and Deutsch (reference 4),

$$\epsilon(\theta)\equiv[N(\theta)-N(\pi/2)/N(\pi/2)]\times10^2.$$

[21] Least squares values for a_2 and a_4 may be obtained through the use of the following expressions: assuming each point weighted equally

$$a_2=\frac{[8]\sum_i\epsilon(\theta_i)\cos^2\theta_i-[6]\sum_i\epsilon(\theta_i)\cos^4\theta_i}{\Delta}\times10^{-2},$$

$$a_4=\frac{[4]\sum_i\epsilon(\theta_i)\cos^4\theta_i-[6]\sum_i\epsilon(\theta_i)\cos^2\theta_i}{\Delta}\times10^{-2},$$

where $[N]\equiv\sum_i\cos^N\theta_i$ and $\Delta\equiv[4][8]-[6]^2$.

The rms errors of these coefficients are given by:

$$\delta a_2=([8]/\Delta)^{\frac{1}{2}}\delta\epsilon\times10^{-2},$$

and

$$\delta a_4=([4]/\Delta)^{\frac{1}{2}}\delta\epsilon\times10^{-2}$$

where $\delta\epsilon$ is the rms error of each of the experimental points.

Often the error is desired on the quantity a_2+a_4 to facilitate comparison of the coefficients with the value of $\epsilon(180°)$. This error can be shown to be:

$$\delta(a_2+a_4)=\left(\frac{[4]+[8]-2[6]}{\Delta}\right)^{\frac{1}{2}}\delta\epsilon\times10^{-2}.$$

In the event that only a_2 is being considered, i.e., a_4 is taken as 0, the expressions for the least squares value and the error become

$$a_2=\frac{\sum_i\epsilon(\theta_i)\cos^2\theta_i}{[4]}\times10^{-2},$$

$$\delta a_2=(1/[4])^{\frac{1}{2}}\delta\epsilon\times10^{-2}.$$

[22] E. L. Church and J. J. Kraushaar, Phys. Rev. **88**, 419 (1952).
[23] F. R. Metzger, Phys. Rev. **85**, 727 (1952).

the upper limit of the experimental error were applied as a correction to the measured correlations, it would, for example, amount to a decrease of 2 percent in $\epsilon(180°)$ for As^{76}, where the effect was the largest. That is, the original value of 6.40 would be reduced to 6.27, which is still well within the indicated error.

As a further test of the experimental techniques, the directional correlation of the cascade gamma-rays of Co^{60} was measured. This correlation had been previously measured by Brady and Deutsch[1] and others and essentially agrees with that to be expected for a 4–2–0 (Q–Q) cascade. In the work here the anisotropy was measured at 120°, 140°, 160°, and 180°. By a least squares fit to the data and by adjustment for the finite detector size, values of 0.120 ± 0.052 and 0.043 ± 0.059 were obtained for a_2 and a_4, respectively. These are to be compared with the theoretical values of 0.1250 and 0.0417 for a cascade of the above type. The corresponding value of $\epsilon(180°)$ from the data is 0.163 ± 0.012, while the theoretical value is 0.1667. This experimental value cannot be said, however, to be incompatible with the value recently obtained by Aeppli *et al.*[12] of 0.148 ± 0.002. These authors indicate that the difference between their experimental value and the theoretical value may be attributable to a perturbation of the correlation by the electron shell. The directional correlation of Pr^{144} has also been measured by using the described apparatus and techniques and has been reported elsewhere.[24]

Rh^{106}

In Fig. 2, curve (a), is shown the measured directional correlation of the successive gamma-rays of Rh^{106}, using a ruthenium chloride source in solution and with no bias on the counters. Values of 2.245 ± 0.041 and 2.953 ± 0.047 were obtained for a_2 and a_4 by taking a weighted average of the corrected least squares values listed previously[22] for δ_0 equal 6.6° and 12.0°. The corresponding value of a_2+a_4 is 0.708 ± 0.011. Also shown in Fig. 2 are the experimental points, with their associated rms statistical errors, obtained in the 6.6° geometry.

To estimate the contribution from gamma-rays known to be present[25] other than those involved in the main cascade, 0.51–0.62 Mev, the gamma-ray spectrum was studied with a scintillation spectrometer. Lines at 0.51, 0.62, 0.87, 1.04, and 1.55 Mev were identified and their relative intensities determined to be 1.00, 0.53 ± 0.05, 0.02 ± 0.01, 0.07 ± 0.01, and 0.01 ± 0.01, respectively. These data are in essential agreement with those of Alburger.[25] Thus, if all the gamma-rays are considered to be counted in proportion to their intensity as well as to the efficiency for detection with a 3-cm NaI crystal, 84 percent of the coincidences will be due to the 0.51–0.62 Mev cascade.

The use of integral discrimination permitted the energy dependence of the directional correlation to be studied. By sufficiently raising the bias on one of the counters in two steps the value of $\epsilon(180°)$ was reduced by 16 percent and 34 percent, respectively, and the value of $\epsilon(130°)$ was reduced by 33 percent and 76 percent, respectively, from those measured with no bias on the counters. Through estimating the attenuation of the various cascade processes at the two higher bias settings and with a knowledge of the intensities and the counting efficiencies, small positive values were indicated for the anisotropy of the background coincidences.

The above work indicates that the background coincidences may be assumed to have approximately an isotropic angular distribution without introducing an appreciable error. If the coefficients of the main cascade are adjusted for this 16 percent isotropic background, new values of 2.673 and 3.515 are obtained for a_2 and a_4. Curve (b) in Fig. 2 shows this distribution.

Comparison of curve (b) and the theoretical 0–2–0 correlation function shows about a 12 percent difference. Both curves (a) and (b) indicate a larger anisotropy than those originally measured by Brady and Deutsch[4] and later by Steffen.[26] The recent measurements of Arfken *et al.*[27] are somewhat higher in the region of 150° to 180° than is shown in curve (b).

The 12 percent discrepancy between curve (b) and the theoretical distribution (c) may be due, as has already been suggested,[4,11,28,29] to a perturbation of the correlation by the electron shell. However, if such a perturbation is present, it could not be significantly

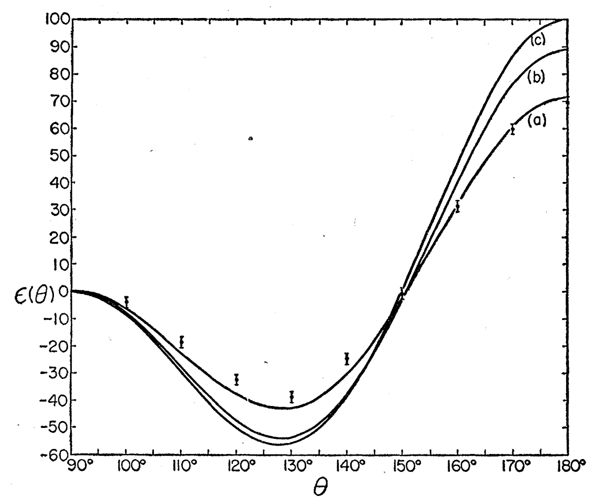

FIG. 2. The experimental points shown are those taken in the 6.6° geometry. Curve (a) represents the weighted averaged values of the 6.6° and 12.0° data corrected for the finite detector size. Curve (b) is curve (a) corrected for the estimated 16 percent isotropic background. The theoretical correlation function for a 0–2–0 (Q–Q) transition is shown in curve (c).

[24] D. E. Alburger and J. J. Kraushaar, Phys. Rev. **87**, 448 (1952).
[25] D. E. Alburger, Phys. Rev. **85**, 734 (1952) and Phys. Rev. **88**, 339 (1952).

[26] R. M. Steffen, Phys. Rev. **80**, 115 (1950).
[27] Arfken, Klema, and McGowan, Phys. Rev. **86**, 413 (1952).
[28] J. J. Kraushaar, Phys. Rev. **85**, 727 (1952).
[29] R. M. Steffen, Phys. Rev. **86**, 632 (1952).

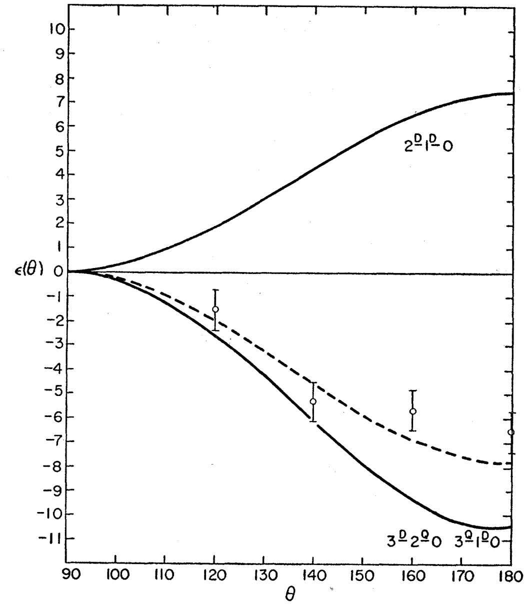

FIG. 3. The directional correlation of Sb^{124}. The dashed curve represents a least squares fit, corrected for the finite detector size, to the experimental points shown. The solid line curves are the theoretical distributions for pure dipole and quadrupole radiation that are nearest in agreement with the experimental curve.

altered by imbedding the sources in different media.[29] It may be that when the intermediate state has a short lifetime ($<5\times10^{-9}$ seconds[30] for Rh^{106}) it is more difficult to change the perturbation. It has been pointed out by Alder[15] and Frauenfelder[7] that if the electron shell is disturbing the correlation, the attenuation of the coefficients of the terms in an expansion in a Legendre series will be greater the higher the term. For a 0–2–0 transition, A_4/A_2 is 3.201; while for curve (b), A_4/A_2 is 3.546. This then provides additional evidence against the explanation of the discrepancy in terms of the influence of extra-nuclear fields.

Sb^{124}

The decay of 60-day antimony involves a complex beta- and gamma-ray spectrum. By far the most prominent gamma-ray cascade consists of the 1.70–0.603-Mev radiation. By the use of integral discrimination, the directional correlation of this cascade has been studied. It has been found previously by workers concerned with the β–γ directional correlation that the composite γ–γ coincidences showed no significant anisotropy.[31]‡

Sources of about 0.1 millicurie of Sb^{124} were prepared by neutron bombardment of metallic antimony in the Brookhaven reactor. The discriminator level of one of the counters was set so that the counter responded only to radiation appreciably above 0.7 Mev. The other counter was biased at about 0.1 Mev. The presence of adequate beta-ray absorbers precluded any contribution from the known β–γ correlation to the measured γ–γ anisotropy. The results of the measured γ–γ correlation are shown in Fig. 3. The dashed curve is a least squares fit of the points adjusted for the finite geometry of the apparatus. The value of a_2 thus obtained is -0.078 ± 0.009. Also indicated are the closest approaching computed curves, considering only pure dipole and quadrupole radiation and assuming that the ground state of Te^{124} has spin zero. The consideration of pure octupole radiation also provides no agreement with experiment.

The mixtures of dipole and quadrupole radiation in one of the transitions, which agree with the results, are shown in Table I. Recently Metzger[32] measured the internal conversion coefficient of the 0.603-Mev gamma-ray and found that it agreed with the theoretical value for electric quadrupole radiation. With the usual assumption concerning the spin and parity of the ground state of even-even nuclei, the first excited state, then, must have spin 2 and even parity. The fact that there is no parity change in that transition is also in agreement with Stump's[33] measurement of the β–γ direction-polarization correlation. Of the mixture possibilities listed, then, only 3–2–0 (D, Q–Q) is consistent with the above results.

TABLE I. Mixtures of dipole and quadrupole radiation in the first transition that are consistent with the directional correlation data for Sb^{124}.

Spins	α/β	Phase difference
3 —D,Q— 2 —Q— 0	36	0°
1 —D,Q— 1 —D— 0	7.8 or 0.115	180°
2 —D,Q— 1 —D— 0	5.6 or 4.8	0°

On the basis of the present experiments, the possibility that the correlation is being perturbed by the electron shell or perhaps by other causes cannot be dismissed. Inasmuch as the effect of such perturbations is to attenuate the coefficients, it is apparent that the original correlation must be negative and that a_2 must be equal to or greater than 0.078. Other than 3–2–0, only a 1–2–0 assignment would meet this requirement and still be consistent with present information. Of course the very weak intensity of the probable cross-over transition (~0.02 percent)[34] very much favors the 3–2–0 assignment.

[30] M. Deutsch and W. E. Wright, Phys. Rev. **77**, 139 (1950).
[31] S. L. Ridgeway, Phys. Rev. **78**, 821 (1950); J. R. Beyster and M. L. Wiedenbeck, Phys. Rev. **79**, 169 (1950); D. T. Stevenson and M. Deutsch, Phys. Rev. **83**, 1202 (1951).
‡ *Note added in proof:*—The γ–γ directional and polarization correlations of Sb^{124} have recently been measured by Kloepper, Lennox, and Wiedenbeck (Phys. Rev. **88**, 695 (1952)). Without using energy discrimination they found no anisotropy for the directional correlation.

[32] F. R. Metzger, Phys. Rev. **86**, 435 (1952).
[33] R. Stump, Phys. Rev. **86**, 249 (1952).
[34] V. Meyers and A. Wattenberg, Phys. Rev. **75**, 992 (1949).

If a mixture is present, there would be a strong argument in favor of having the parity of the 2.30-Mev level even. In principle, however, mixtures of electric dipole and magnetic quadrupole radiation could take place.

It is interesting to note that the directional correlation of Y^{88}, measured by Brady and Deutsch,[4] is similar to that shown in Fig. 1. One must assume, then, that the mixtures listed in Table I are also approximately applicable to that gamma-ray cascade process. In addition to the 2–1–0 (D, Q–D) assignment hitherto considered,[4,17] the other possibilities should also be viewed in the light of present information on the decay of Rb^{88} and Y^{88}. There does not appear at present to be any strong evidence favoring the 2–1–0 assignment over the 3–2–0. The importance of this assignment lies mainly in the fact that it may be one of the few remaining exceptions to the observation that the spin and parity of the first excited states of even-even nuclei are 2, even.[8,9] Further experiments on this nuclide are in preparation.

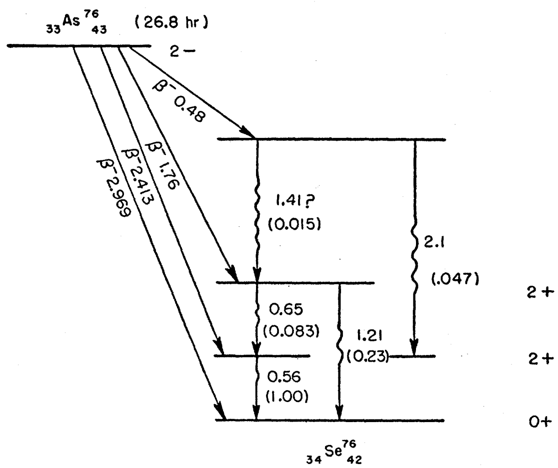

FIG. 4. The decay scheme of As^{76}. The quantities in parentheses are the relative gamma-ray intensities as indicated by Hubert.

As^{76}

The decay of As^{76} (27 hours) involves several cascade gamma-rays from the excited states of Se^{76}. In addition to the information summarized in *Nuclear Data*,[35] there has been some recent work on the beta-[36] and gamma-ray[37,38] spectra. Because of the discrepancy between the decay scheme consistent with the data of Tomlinson and Ridgway[37] and Hubert[38] and the suggested scheme of the earlier workers, the gamma-ray spectrum was analyzed[39] in both singles and coincidences.

The singles spectrum clearly showed γ-rays at 0.56, 0.65, 1.21 Mev and a weaker high energy line of about 2.1 Mev. The coincidence spectrum showed strong lines at 0.56 and 0.65 Mev, and a weak and perhaps composite peak at 1.2 Mev. The ratio of the peak height of the singles to the coincidences for the 0.65-Mev line indicated that this gamma-ray was in direct cascade with the 0.56-Mev line. The ratio of singles to coincidences for the 1.2-Mev line was much greater than that to be expected were it in direct cascade with the prominent gamma-rays. The above results are in agreement with the data of Hubert except that a 1.41-Mev gamma-ray was not resolved.

The decay scheme that summarizes the data of Hubert, Tomlinson, and Ridgway, and the present work, is shown in Fig. 4.

In order to determine the spins of the excited states of Se^{76} the directional correlation of the successive gamma-rays has been measured. Sources were prepared by irradiating 10 mg of metallic As powder with slow neutrons for 6 minutes in the Brookhaven reactor. The results of the correlation measurements, taken with no bias on the counters, is shown in Fig. 5. The values of a_2 and a_4, corrected for the finite detector size, are -0.611 ± 0.030 and 0.700 ± 0.034, respectively. There is no assignment of spins, considering pure dipole, quadrupole, and octupole radiation that will present such a distribution, assuming a spin of 0 for the ground state of Se^{76}. Of the possibilities involving mixtures of dipole and quadrupole radiation in one of the transitions only 2–2–0 (D, Q–Q) with $\alpha/\beta=0.5$ to 1.5 has coefficients at all compatible with the measured values.

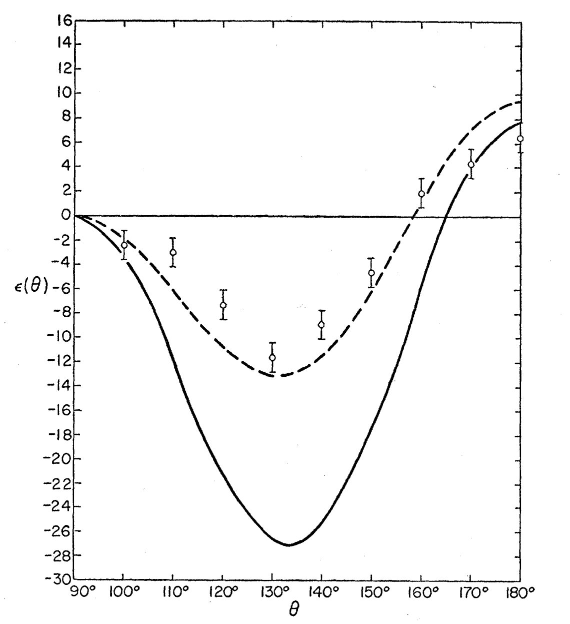

FIG. 5. The dashed curve is the least squares fit, corrected for the finite geometry, to the indicated experimental points obtained with As^{76}. The solid curve is the theoretical distribution for a 2–2–0 (Q–Q) transition.

[35] *Nuclear Data*, National Bureau of Standards, Circular No. 499 (1950).
[36] E. P. Tomlinson and S. L. Ridgway, Phys. Rev. **88**, 170 (1952), and private communication.
[37] J. K. Bair and F. Maienschein, Atomic Energy Commission Report NEPA 1620, unpublished.
[38] P. Hubert, J. phys. et radium **12**, 823 (1951).
[39] This work was done in cooperation with A. W. Sunyar of this laboratory, to whom our thanks are due.

On the basis of the intensity measurements of Hubert and considering the crystal efficiencies, 67 percent of the coincidences counted would be due to the 0.56–0.65 Mev cascade. Preliminary work using differential discrimination on one of the counters showed that the exclusion of a large portion of the higher energy cascades did not significantly change the anisotropy at 180°. This would not be inconsistent with the fact that the spin of the highest level might logically be taken as 3, from intensity considerations. The largest contribution to the coincidence background would then be from a cascade (2.1–0.56 Mev) which would offer little anisotropy.

It would appear that by taking into account the contribution from the other coincidence γ-rays, agreement could be obtained between the 0.56–0.65 correlation and that to be expected for a 2–2–0 spin assignment with $\alpha/\beta=0.5$ to 1.5 (20–66 percent D, 80–34 percent Q). In fact, simply the knowledge that a_2 is negative and a_4 is positive uniquely requires a 2–2–0 spin assignment for the 1.21-, 0.56-, and 0.0-Mev levels, if the possibility of a 0–2–0 assignment is disregarded because of the observed cross over. The phase difference is not determinable from our data.

Based on the directional correlation measurements, spins have been assigned to the excited levels of Se^{76} as shown in Fig. 4. The assignment of a spin of 2 to the first excited state is in agreement with the measurement of Tomlinson and Ridgway of the conversion coefficient of the 0.56-Mev gamma-ray, and the beta-gamma directional correlation measurements of Ridgway and Pipkin.[40] The assignment of an even parity to the second excited level is based on the fact that the mixture is probably between $M1$ and $E2$ radiations, both of which require no change of parity.

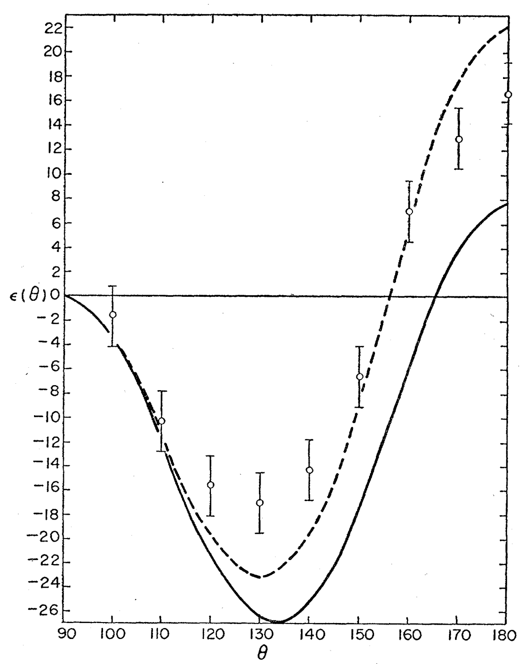

FIG. 6. The dashed curve is the least squares fit, corrected for the finite geometry, to the experimental points obtained with Ir^{194}. The solid curve is the theoretical distribution for a 2–2–0 (Q–Q) transition.

Ir^{194}

The decay of 19-hour Ir^{194} is known[35] to involve cascade gamma-rays of energies 0.33 and 1.48 Mev. However, due to uncertainties in some of the available information on this nuclide, a study of the gamma-radiation was undertaken as a preliminary to the directional correlation measurements.

Sources were prepared by neutron bombardment of a few mg of pure metallic iridium powder in the Brookhaven reactor. The decay of one source, irradiated for 5 minutes, was followed for a period of days, after letting the 1.5-minute activity of Ir^{192} decay. Two activities of 18.5 hours and 70 days were found. The 70-day activity of Ir^{192} accounted for about 21 percent of the gamma-rays at the beginning of the measurements.

Inasmuch as the data of Wilson[41] and Wattenberg[42,34] were in disagreement on the presence of Be photoneutrons from Ir^{194}, an experiment to detect such photoneutrons was carried out. In agreement with Wilson, photoneutrons were detected from Be, which decayed with a 19-hour half-life. The intensity of the responsible gamma-rays was estimated to be about 1 percent of the intensity of the two prominent lines.

The gamma-rays were also studied with a scintillation spectrometer. The two prominent lines of 0.33 and 1.48 Mev were observed and found to have about equal intensity. Weak, higher energy gamma-rays of 1.8 and 2.1 Mev were also in evidence. It would seem reasonable to assume that the 1.8-Mev gamma-ray is the cross-over transition. The 2.1-Mev gamma-ray could be the direct transition to the ground state that appears in the decay of Au^{194}.[43]

Inasmuch as the 70-day activity of Ir^{192} was also present in the sources used, precautions were necessary in order to insure that the measured directional correlation was only that due to the 1.48- and 0.33-Mev gamma-rays of Ir^{194}. The most intense gamma-rays of Ir^{192} are known to have energies of 0.3 and 0.47 Mev. One of the discriminator levels was therefore raised until annihilation radiation coincidences were no longer counted. The coincidences counted with the Ir source under these conditions decayed with a 19-hour half-life.

The measured directional correlation is shown in Fig. 6. The values of a_2 and a_4, obtained by a least squares procedure and corrected for the finite geometry, were -1.118 ± 0.061 and 1.340 ± 0.070, respectively. The situation is similar to As^{76}, and again only a 2–2–0 (D, Q–Q) assignment is compatible with the data, with

[40] S. L. Ridgway and F. M. Pipkin, Phys. Rev. **87**, 202 (1952).
[41] R. Wilson, Phys. Rev. **79**, 1004 (1950).
[42] A. Wattenberg, Phys. Rev. **71**, 497 (1947).
[43] Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).

$\alpha/\beta=0.13$ (98 percent Q, 2 percent D) and a phase difference of 180°. The agreement with this assignment is just outside the indicated errors.

It is interesting to note that a similar directional correlation has been observed[44] for the gamma-rays from the excited states of the neighboring even-even nucleus Pt^{196}. Further, the cross-over transition here is known also to be very weak (<1 percent). This, of course, as in Pt^{194}, is hard to reconcile with the 2–2–0 (Q–Q) assignment.

Measurements of the directional correlation of the gamma-rays of Pt^{194} have recently been reported by Whittle and Jastram.[45] While one of the alternative assignments 2–2–0 indicated by their data agrees with the assignment purported here, the measured directional correlation is apparently different.

There is no direct evidence on the energy of the first excited state. However, it seems most reasonable on the basis of systematics[11] of even-even nuclei of adjacent atomic number to take its energy as 0.328 Mev. Such an energy is also preferred over 1.48 on the basis of the low intensity of the cross-over transition. The K/L ratio of the 0.328-Mev gamma-ray has been determined[46] to be 2. This information, taken together with life-time considerations, would favor an $E2$ assignment for the transition. Such an assignment would be consistent with the directional correlation data and would add another member to the family of first excited states of even-even nuclei that have spin 2, even parity.

The decay of Au^{194} has been studied rather extensively by Steffen *et al.*[43] With the possible exception of the measured internal electron conversion coefficient for the 0.328-Mev gamma-ray, the information thus obtained on the excited states of Pt^{194} is in good agreement with that from Ir^{194}.

III. DIRECTION-POLARIZATION CORRELATIONS

The two counters used in the directional correlation work were placed so that they each subtended an angle of 90° at the source with a polarimeter. The value of θ was thus 90° and remained so. For such an arrangement, considering dipole and quadrupole radiation, the expressions of Hamilton[4] for J_θ/J_φ are reduced to $(1+a_2+a_4)/(1-a_2-a_4)$ or its reciprocal for yes-yes or no-no parity changes, respectively. For mixed parity changes, assuming equal coincidence counting efficiencies, J_θ/J_φ should be unity.

The polarimeter, based on Compton scattering, was similar to that employed by Metzger and Deutsch.[5] Its essential features are shown in Fig. 7. Another brass bearing to the rear of the chassis permitted rotation of the entire assembly through 360°. The lead shield was approximately symmetrical about the axis of rotation. The angular resolution in θ was about equal to that of the individual counters, i.e., 12.0°.

[44] B. M. Steffen and D. M. Roberts, Phys. Rev. **83**, 222 (1951).
[45] C. E. Whittle and P. S. Jastram, Phys. Rev. **87**, 203 (1952).
[46] J. W. Mihelich (unpublished data).

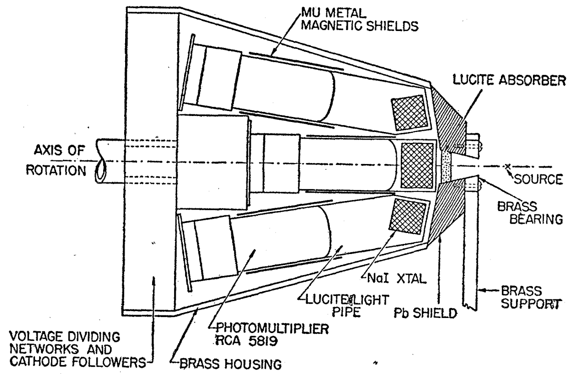

FIG. 7. The experimental arrangement for the measurement of the polarization of gamma-rays. A line joining the two single counters (not shown) would pass through the source perpendicular to the page.

The two individual counters were coupled together, as were the two side counters of the polarimeter. After amplification, the pulses from the three sets of counters went to integral pulse-height discriminators and blocking oscillators identical to the ones used in the directional correlation work. Three coincidence circuits monitored all double coincidence events so that the chance coincidences could be computed. The resolving time of the three circuits was 1.2×10^{-7} second. A slow resolving time coincidence circuit then registered double coincidences common to the center-side counter circuit and the center-individual counter circuit.

Data were taken by measuring the triple coincidence rate for half-hour periods for the four values of φ: 0°, 90°, 180°, and 270°. One determination of $N_{\parallel}/N_{\perp}$ consisted then of $(N_{0°}+N_{180°})/(N_{90°}+N_{270°})$, corrected for random coincidences.

The value of the angle δ (notation of Metzger and Deutsch[5]) was made 80°. Instead of attempting to evaluate the asymmetry ratio in terms of the spreads in δ and φ and correcting for scattering from the side to the center counter, an effective asymmetry ratio as a function of energy was determined in the following way. The collimated beam of gamma-rays from Co^{60} was scattered through 90° by a Cu plate. The value of $N_{\parallel}/N_{\perp}$ was measured for three thicknesses of the scatterer. The extrapolated value for zero thickness for $N_{\parallel}/N_{\perp}$ was thus determined to be 1.48 ± 0.05. From the Klein-Nishina formula, the value of J_θ/J_φ for the scattered beam is 2.19. The asymmetry ratio at 0.36 Mev is then 3.14 ± 0.1.

A more realistic determination of the effective asymmetry ratio was made using Co^{60} and Rh^{106} in a direction-polarization experiment. Inasmuch as the character of the prominent gamma-rays is known, a measurement of both the direction and polarization correlations should afford a determination of the effective asymmetry ratio at the energies involved. For Rh^{106}, the value of a_2+a_4 measured using the apparatus previously described was $+0.57\pm0.03$ for the 12.0° geometry. This is uncorrected

TABLE II. Values of $N_{\parallel}/N_{\perp}$ to be expected for the most probable spin and gamma-ray character assignments for the excited states of Pb^{208}.

Assignment	$N_{\parallel}/N_{\perp}$	Assignment	$N_{\parallel}/N_{\perp}$
3–1–0 ($E2$–$M1$)	1.02	4–2–0 ($E2$–$E2$)	0.97
3–2–0 ($E2$–$E2$)	1.06	4–2–0 ($E2$–$M2$)	1.00
3–1–0 ($E2$–$E1$)	1.00	4–2–0 ($M2$–$E2$)	1.00
3–2–0 ($M1$–$E2$)	1.02		

for detector size as the direction-polarization apparatus has approximately the same angular resolution. $N_{\parallel}/N_{\perp}$ was measured and a value of 0.62±0.03 obtained. The asymmetry ratio at the average energy of 0.57 Mev is then 2.39±0.20.

Likewise the experimental value of a_2+a_4 for Co^{60} was 0.1517±0.0126, and $N_{\parallel}/N_{\perp}$ was measured to be 0.959±0.02. The asymmetry ratio at the average energy of 1.22 Mev is then 1.31±0.03.

ThC″

Recently, Petch and Johns[47] made a careful determination of the directional correlation of ThC″. Although other gamma-rays are present, the 0.58–2.62 Mev gamma-ray cascade will be observed about 65 percent of the time. Their measured correlation agreed well with that expected from a 4–2–0 (Q–Q) transition. The conversion coefficients have been measured by Martin and Richardson.[48] Their work indicated that the 0.58-Mev gamma-ray is $E2$. The measured coefficient for the 2.62-Mev gamma-ray (1.8×10^{-3}) falls between the values to be expected for $E2(1.0\times10^{-3})$ and $M1(2.1\times10^{-3})$ radiation. The authors preferred the closer $M1$ assignment.

Because of this discrepancy in the assignment of spins to the 3.20- and 2.62-Mev states of Pb^{208}, a direction-polarization correlation was undertaken. Such a measurement will act as a check on the spins as determined by the directional correlation measurements and will serve to indicate the relative parities of the three levels involved.

Deposits of 10.6-hour ThB on Al foil were used as sources of about 0.1-mC strength. There was no appreciable bias on any of the counters; the value of $N_{\parallel}/N_{\perp}$ obtained was 0.958±0.03.

In view of the results of the measurements of the asymmetry ratio as a function of energy previously described, the asymmetry ratio for the gamma-ray cascade in Pb^{208} has been estimated as 1.2. Using this, the value of $N_{\parallel}/N_{\perp}$ to be expected for the most probable gamma-ray and spin assignments have been calculated and are shown in Table II.

The 4–2–0 ($E2$–$E2$) assignment is in more satisfactory agreement with our experimental value than the 3–1–0 ($E2$–$M1$) assignment given by Martin and Richardson. With this result, then, the parities of the first and second excited states are the same as the ground state, i.e., even, and the spins are in agreement with the result of the directional correlation measurements. The spin and parity of the first excited state lends support to the generalization[8–11] discussed previously.

Recently, the internal pair production coefficient of the 2.62-Mev gamma-ray has been measured by Slätis and Siegbahn.[49] The value obtained, agrees with that to be expected for an $E2$ transition.

IV. SUMMARY AND DISCUSSION

The occurrence of mixtures of radiation and the presence of extra-nuclear fields in directional correlation measurements has made necessary more accurate evaluations of the data and more care in correcting for instrumental effects. It is evident, also, that the further application of the techniques of directional correlations to the determination of spins and gamma-ray characteristics in the more complex decay schemes will be dependent on the energy selectivity of the detectors.

In the work reported here it has been demonstrated that the major contribution to the discrepancy between the earlier measured directional correlation of Rh^{106} and that expected for a 0–2–0 transition probably lay in the geometrical correction. The discrepancy that still exists after accounting for the effects of the detector size and the other gamma-rays is still a matter for speculation. The directional correlation of the 1.70–0.65 Mev gamma-ray cascade of Sb^{124} has been measured and indicates spins of 3, 2, and 0 for the three levels involved with a small admixture of quadrupole with the dipole radiation in the first transition.

The directional correlations of As^{76} and Ir^{194} are similar, and spin assignments of 2–2–0 (D, Q–Q) have been made. It is rather surprising to find such large ratios of quadrupole to dipole radiation in the mixed transitions and such weak intensities for the cross-over gamma-ray. This may support the contention[50] that the ground state of an even-even nucleus has a relatively purer wave function than the excited states.

The direction-polarization measurements with ThC″ substantiated the assignment of spins 4, 2, and 0, as determined by the directional correlation measurements, and determined the character of both gamma-rays to be $E2$.

It is interesting to note that the majority of the directional correlation measurements made recently require mixtures of dipole and quadrupole radiation in one of the transitions in order to interpret the data. Considering even-even nuclei and assuming that the first excited state has spin 2, even parity, mixtures can occur with spins of 1, 2, or 3 for the second excited state. Table III shows the results of directional corre-

[47] H. E. Petch and M. W. Johns, Phys. Rev. **80**, 478 (1950).
[48] D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) **A63**, 223 (1950).
[49] H. Slätis and K. Siegbahn, *M. Siegbahn Commemorative Volume* (Uppsala, Sweden, 1951), p. 153.
[50] A. de-Shalit and M. Goldhaber (unpublished).

TABLE III. The occurrence of mixtures of radiation from Lipher excited states of even-even nuclei.*

Parent nuclide	Assigned spins	Percent dipole radiation in the first transition	Phase difference	Energy of first transition in Mev	Cross-over transition energy (Mev)	percentage branching	Reference
Pr^{144}	1–2–0	94–100	0°	1.48	2.185	73	a
In^{114}	2–2–0	95.6	0°	0.548	1.26	3–6	b, c, d, e
Sb^{122}	2–2–0	20		0.68	1.24		f
As^{76}	2–2–0	20–66	180°	0.65	1.21	72	present paper
Au^{196}	2–2–0	{5	180°	0.33	0.69	<1	g, m
		{6	180°				e
Ir^{194}	2–2–0	2	180°	1.48	1.81	1	present paper
Rh^{106}	2–2–0	10–20	180°	1.045	1.55	24	h
Sb^{124}	3–2–0	99.9	0°	1.70	2.3	0.02	present paper
Cl^{38}	3–2–0	0		1.60	3.8	<0.02	i
Y^{88}*	3–2–0	99.9	0°	0.908	2.76	~1	j, k, l

* A spin and parity of 2+ has been assumed for the first excited state of Sr^{88}, however, as discussed before, 1+ cannot be excluded. The recent internal conversion measurements of Metzger and Amacher indicate that if there is a mixture of radiation in the first transition it must be of $E1$ and $M2$ radiation

[a] D. E. Alburger and J. J. Kraushaar, Phys. Rev. **87**, 448 (1952).
[b] Johns, Cox, and McMullen, Phys. Rev. **86**, 632 (1952).
[c] R. M. Steffen and W. Zoebel, Phys. Rev. **88**, 170 (1952).
[d] E. D. Klema and F. K. McGowan, Phys. Rev. **87**, 524 (1952).
[e] H. Heer and collaborators, reported at the Amsterdam Conference on beta- and gamma-rays.
[f] M. J. Glaubman and F. R. Metzger, Phys. Rev. **87**, 203 (1952).
[g] R. M. Steffen and D. M. Roberts, Phys. Rev. **83**, 222 (1951).
[h] Arfken, Klema, and McGowan, Phys. Rev. **86**, 413 (1952).
[i] R. M. Steffen, Phys. Rev. **80**, 115 (1950).
[j] E. L. Brady and M. Deutsch, Phys. Rev. **78**, 558 (1950).
[k] D. S. Ling and D. L. Falkoff, Phys. Rev. **76**, 1639 (1949).
[l] F. R. Metzger and H. C. Amacher, Phys. Rev. **88**, 147 (1952).
[m] R. M. Steffen, Bull. Am. Phys. Soc. **27**, No. 5, 18 (1952) and Phys. Rev. (to be published).

Note added in proof:—A private communication from Dr. H. Frauenfelder has informed us of the results of D. Schiff and F. Metzger for the directional correlation of the γ-rays of Hg^{198}. These are in the order given in the table: 2–2–0, 30–50 percent dipole, 0°, 0.68 Mev, cross-over 1.091 Mev, 20 percent. We wish to thank Dr. Frauenfelder for this information and for other valuable remarks.

lation measurements that have been interpreted as having either of these three spins for that excited state. The striking feature of the compilation is the sporadic behavior of the relative amounts of dipole radiation that are involved in 2–2 transitions.

Thanks are due E. L. Church for valuable contributions to the procedure for analyzing the data, to H. Finston who performed some chemical changes on the Rh^{106} sources, and to P. Prentkey who designed the amplifiers used.