yields very definite information concerning the parities of the nuclear levels involved. The ambiguities left in the examples discussed in this paper arise from a paucity of theoretical predictions for higher multipole transitions, to the possibility of interference for mixed radiation, and to the preliminary character of the angular correlation data. If these obstacles are eliminated the experiments with the polarimeter have only to decide the existence or non-existence of a polarization in the

position (θ) at which we expect the largest effect.[15] For this purpose the polarimeter described in this paper has sufficient sensitivity. For gamma-ray energies above a few hundred kev the correlation method compares favorably with other methods which lead to the assignment of spins to nuclear levels and of parity changes to the transitions between these levels.

[15] p is largest at $\theta=90°$ in most of the cases, but not in all of them.

Angular Correlation of Successive Gamma-Rays*

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The angular correlation of successive gamma-rays emitted by six even-even nuclei has been investigated and found to be anisotropic in every case, and of the magnitude expected theoretically. Effects of external magnetic fields and of chemical binding on the correlations are found to be smaller than the experimental uncertainty for some exploratory experiments. Interpretation of the results in terms of the nuclear states involved is in general possible by use of additional evidence such as relative transition probabilities.

1. INTRODUCTION

CONSIDERABLE progress has been made during the last few years in studies of nuclear radiation spectra from radioactive processes. The experiments have mainly yielded results concerning the energies of excited states. Only little unambiguous systematic evidence on the spins of these states has appeared. Coefficients of internal conversion have revealed the multipole character of nuclear gamma-rays in some cases. Although this method is in principle universally applicable, limitations on the precision of both theoretical and experimental results have so far restricted it to the more favorable cases. No values of nuclear angular momenta are obtained directly by this method, but probable values can frequently be assigned from the multipole orders.

If the life time of a gamma-ray transition is long enough to be measured a probable multipole order can be obtained. This is usually done by means of Bethe's life-time formulas as modified by Segrè. With a few notable exceptions only transitions of rather low energy or high multipole order have been studied so far by this method because of the difficulty in measuring extremely short life-times. There is considerable uncertainty about the exact validity of the life-time formula because it neglects necessarily all details of the nuclear configuration.

It would be most desirable to have a method of the general validity and applicability available to atomic spectroscopy in the Zeeman effect. Unfortunately, it seems at present entirely unfeasible to separate the nuclear magnetic substates by application of external fields sufficiently to observe the transitions between the individual m-values separately. It has been proposed to change the relative populations of the several magnetic substates of an excited state by applying a magnetic field at very low temperatures where the magnetic energy μH is comparable to the thermal agitation energy kT.[1] Information about the multipole nature of the radiation and about the angular momenta of the initial and final states could then be obtained from the space distribution and the polarization of the emitted radiation. This method has not yet been tried. It presents formidable experimental difficulties and is probably limited to long-lived excited states.

Although it is difficult to change the relative populations of the several substates it is frequently possible to make measurements on a sample of nuclei for which these relative populations are different from their equilibrium values. This is in general the case when two successive rays are emitted by one nucleus. If we observe the first ray to be emitted in the z-direction the probability of the intermediate state being in any one of the possible m_z states is no longer the same as in the absence of this information. Thus, by observing the space distribution and polarization of those gamma-rays which

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[1] J. A. Spiers, Nature 161, 807 (1948).

coincide with the emission of another gamma-ray in the z-direction the results are similar to those obtained if the populations of the states had been changed by an external field. In fact, if nuclear spectroscopy had to deal practically only with dipole radiation as does optical spectroscopy, unique spin assignments could be made in virtually every case from the angular correlation of successive gamma-rays. Because of the occurrence of higher multipoles the situation is somewhat more complicated. Hamilton[2] and Goertzel[3] have calculated explicitly the angular correlation of successive gamma-rays for all possible combinations of pure dipole and quadrupole radiations. A general discussion of the correlations of successive radiations has been given by Yang.[4]

The probability, per unit solid angle, that two successive gamma-rays are emitted at an angle θ is proportional to

$$W(\theta) = 1 + \sum_1^l a_i \cos^{2i}\theta$$

where $2l$ is the order of the lowest multipole in the cascade. Thus, if both gamma-rays are quadrupoles $W(\theta) = 1 + a_1 \cos^2\theta + a_2 \cos^4\theta$. If one is a dipole $W(\theta) = 1 + a_1 \cos^2\theta$, etc. A further restriction on the number of terms in $W(\theta)$ is $a_i = 0$ for $i > J_2$. J_2 is the spin of the intermediate state in the cascade. Thus if J_2 is zero or $\frac{1}{2}$, the angular correlation will always be isotropic; if J_2 is 1 or $\frac{3}{2}$, the correlation will at most contain terms in $\cos^2\theta$. The coefficients a_1 and a_2 have been given by Hamilton[2] for all possible combinations of angular momenta. In Table I we have listed the values of these coefficients from Hamilton's paper for the values of J which are of interest in connection with our experiments. Coefficients for octupole radiation should be very useful, but have not yet been published. If the transition involves mixed multipoles, e.g., electric quadrupole and magnetic dipole components, the situation becomes very complicated and the coefficients depend not only on the relative intensities of the two components but also on their relative phases.[5]

Following the original suggestion of Dunworth[6] that an angular correlation between gamma-rays might exist a number of experiments were performed to find the effect.[7-9] In all cases but one it was reported that any anisotropy in the correlation, if it existed, was smaller than the estimated experimental uncertainty. This was true for Na^{24}, Br^{82}, Y^{88} and Co^{60}. In one case, Cl^{38}, Kikuchi et al.[7] reported a smaller probability for emission at 180° than at 90°. They gave the result

$$(W(\pi) - W(\pi/2))/W(\pi/2) = \sum a_i = -0.15 \pm 0.06.$$

[2] D. R. Hamilton, Phys. Rev. 58, 122 (1940).
[3] G. Goertzel, Phys. Rev. 70, 897 (1946).
[4] C. N. Yang, Phys. Rev. 74, 764 (1948).
[5] D. S. Ling and D. L. Falkoff, Phys. Rev. 74, 1224 (1948).
[6] J. V. Dunworth, Rev. Sci. Inst. 11, 167 (1940).
[7] Kikuchi, Watase, and Itoh, Zeits. f. Physik 119, 185 (1942).
[8] R. Beringer, Phys. Rev. 63, 23 (1943).
[9] W. M. Good, Phys. Rev. 70, 978 (1948).

TABLE I. Coefficients for the angular correlation with the spin of the ground state $J_1 = 0$.

J_2	J_3	Multipoles	a_1	a_2
1	0	Dipole–Dipole	1	0
1	1	Dipole–Dipole	−1/3	0
1	2	Dipole–Dipole	−1/3	0
1	1	Quadrupole–Dipole	−1/3	0
1	2	Quadrupole–Dipole	3/7	0
1	3	Quadrupole–Dipole	−3/29	0
2	3	Dipole–Quadrupole	−3/29	0
2	2	Dipole–Quadrupole	3/7	0
2	1	Dipole–Quadrupole	−1/3	0
2	0	Quadrupole–Quadrupole	−3	4
2	1	Quadrupole–Quadrupole	5	−16/3
2	2	Quadrupole–Quadrupole	−15/13	16/13
2	3	Quadrupole–Quadrupole	0	−1/3
2	4	Quadrupole–Quadrupole	1/8	1/24

However, Beringer[8] reported isotropic correlation for this case, claiming slightly greater precision.

It seemed extremely difficult to reconcile these results with the theoretical predictions. In all cases studied the ground state of the nucleus in question has almost certainly zero spin, containing even numbers of neutrons and protons so that the intermediate state must have at least $J = 1$. Except in the case of Br^{82} the gamma-rays are all emitted in simple two-quantum cascades. Theory would predict isotropic correlation in such cases only for accidental combinations of multipoles. The entire theory is, of course, based on the assumption that the orientation of the nucleus is not appreciably disturbed during the lifetime of the intermediate state. In a free atom a disturbance which may interfere with the correlation is provided by the magnetic field of the orbital electrons which has random direction with respect to the chosen axis of quantization. The rate at which this field will cause transitions between the nuclear m states is of the order of the frequency of the Larmor precession which is known from the observed hyperfine structure splittings to be at most about 10^9 sec.$^{-1}$. According to present views the life times of the nuclear states should in general be of the order of 10^{-12} sec. so that this disturbance should be negligible. Furthermore it is well known that in non-paramagnetic solids the atomic moments are largely quenched so that the rate of transition should be of the order of the reciprocal relaxation times observed in nuclear resonance experiments, i.e., 10^{-5} sec. or even much longer. One must, of course, consider the fact that the atom in question may be in an excited state due to recoil and the recent change in atomic number but it is still hard to visualize a process which will reduce the relaxation time much below the precession time in a free atom. If we had to accept the above negative results of search for an angular correlation we should be forced to assume either that the gamma-ray widths are much smaller than is generally assumed or that some unknown mechanism creates extremely strong magnetic fields or electric field gradients at the recoiling nuclei or, even less probable, that some
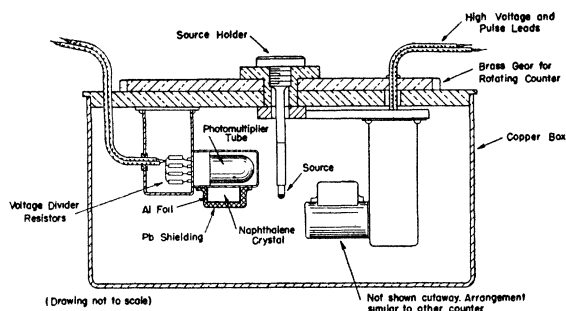
FIG. 1. Apparatus for observing angular correlations.

unknown circumstance favors particular nuclear configurations which will give rise to isotropic emission.

In view of this conclusion it seemed desirable to reexamine the problem with improved experimental techniques. Our results, confirming the basic predictions of the theory have already been briefly reported.[10, 11]

2. EXPERIMENTAL PROCEDURE

The principal limitation in previous attempts to observe angular correlation between successive gamma-rays arose from the low intrinsic efficiency of the Geiger-Müller counters used. This efficiency for gamma-rays is about one percent. The experiment consists in observing the coincidence rate between two counters when the angle θ between the lines joining the counters with the source is varied. In order to maintain a useful angular resolution the solid angle subtended by each counter must be limited to about 0.25 steradian. The net efficiency of each counter, including solid angle is therefore about 2×10^{-4}. Since the shortest useful resolving time of a coincidence circuit used with Geiger counters is about 3×10^{-7} sec., the source strength must be kept to about 10^6 disintegrations per second in order to keep the accidental coincidence rate manageable.[6] Thus the genuine coincidence rate was only about 0.1 count per second. While we were able to prove the existence of a correlation of the expected order of magnitude in preliminary experiments[10] further, improved measurements were all made with the use of scintillation counters. Such counters, in the arrangement used by us have an intrinsic efficiency five to ten times as great as does a Geiger counter. Furthermore, they permit the use of shorter coincidence resolving times, 1×10^{-7} sec. in our arrangement. Thus true coincidence rates of the order of 5 counts per second are obtained while keeping the accidental rate substantially smaller than the true rate. In the experiments reported here pure naphthalene blocks and selected 931A photo-multipliers were used. It is expected that the recent great strides in scintillation counter techniques will permit further refinement of the experiments. The multipliers were operated at a total voltage of about 600

volts. Los Alamos model 501 amplifiers and pre-amplifiers were used with a total gain of about 3×10^4. The coincidence circuit consisted of blocking oscillator pulse shapers and a 6AS6 mixer tube. The experimental arrangement is shown in Fig. 1. The dimensions of the naphthalene blocks were approximately $1\times1\times2$ cm. One counter is mounted in a fixed position on the brass cover while the other counter is mounted on a heavy brass gear which can be rotated from the outside. The angular position is read on a revolution counter. The entire assembly was placed in a thermally insulated box filled with dry ice to reduce the dark current of the multiplier tubes.

The angular resolution of the arrangement was determined by observing the coincidence rate due to annihilation quanta from a Na^{22} source as a function of angle. Since these quanta are known to be practically exactly collinear[12, 13] the curve shown in Fig. 2 represents directly the angular resolution of the apparatus. The small residual coincidence rate at large angles is due to the nuclear gamma-ray accompanying the decay of Na^{22}. Correction for the finite angular resolution was made in all cases but only in one case did the correction exceed the experimental uncertainty of the data. In this one case (Rh^{106}) the validity of the correction was verified by repeating the experiment with better angular resolution. Sources were mounted in small cups of brass, aluminum or plastic, covered with wax and inserted in the source holder shown in Fig. 1. It is important that the size and self-absorption of the source be very small. Otherwise the counts recorded by the two counters will, on the average, come from different parts of the source when the counters are 180° apart, but from the same part of the source when the counters are close together. Thus the coincidence rate at large angles would appear smaller than at small angles, even though the individual counting rates would be unaffected.

The other main danger of systematic error arises from the possibility that a single primary quantum may
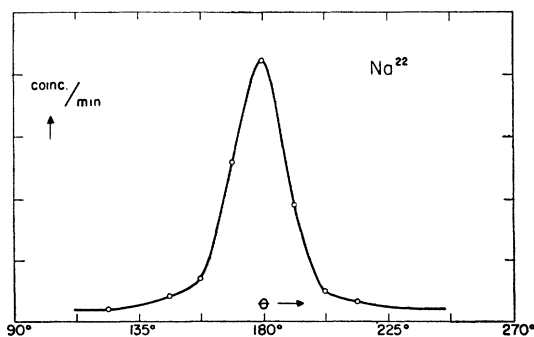

FIG. 2. Angular correlation of coincidences due to annihilation quanta from Na^{22}.

[10] E. L. Brady and M. Deutsch, Phys. Rev. 72, 870 (1947).
[11] E. L. Brady and M. Deutsch, Phys. Rev. 74, 1541 (1948).
[12] R. Beringer and C. G. Montgomery, Phys. Rev. 61, 222 (1942).
[13] deBenedetti, Cowan, and Konneker, Phys. Rev. 76, 440 (1949).

cause a coincidence by Compton scattering from one counter to the other. This effect will generally be greatest when the two counters are close together. It was minimized by covering the sides of the naphthalene blocks with tapered lead shields about $\frac{1}{8}$ in. to $\frac{1}{4}$ in. thick. In angular positions near 180° the backscattered quanta can enter through the front opening of this lead shield. The energy of the scattered quanta at this angle will always be less than 0.25 Mev and definite improvement was obtained by covering the front of the crystals with $\frac{1}{32}$ in. to $\frac{1}{16}$ in. lead foil. The primary radiation is not appreciably attenuated by this but the soft scattered quanta which have to traverse *both* shields are greatly reduced. The small residual effect of scattering of primary photons, remaining after these precautions, can be determined and corrected for by observing the coincidence rate due to a source emitting only single gamma-rays. Fe[59] and Mn[54] were used for this purpose and the very small correction found was applied to all data. This correction should be practically exact for gamma-rays near 1 Mev, i.e., for Co[60] and Sc[46] but may be slightly in error for some of the other substances. This error is almost certainly less than the indicated experimental uncertainty due to other causes. It should be emphasized that unless proper precautions are taken scattering effects may completely mask the true angular correlation. The magnitude of the effect depends on the energy of the primary radiation and the type of detector used.

A slight asymmetry in the source position will affect the observed correlation. However, the effect on the coincidence rate will, in first order, be the same as on the individual counting rates. The observed number of coincidences was therefore always divided by the product of the individual counts. This procedure also corrects in first order for changes in counter efficiency and effects of scattering by the apparatus and surrounding material. The latter phenomenon may reduce the effective angular resolution at small angles compared with that
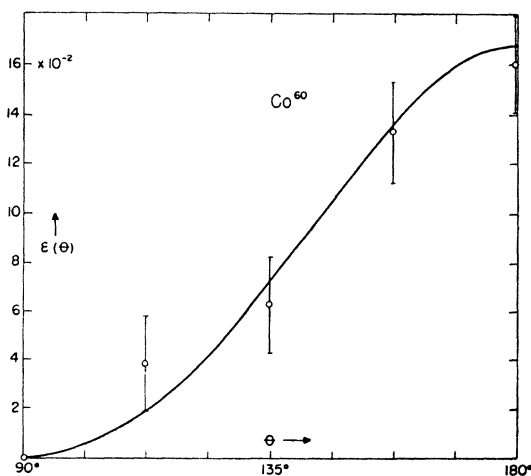

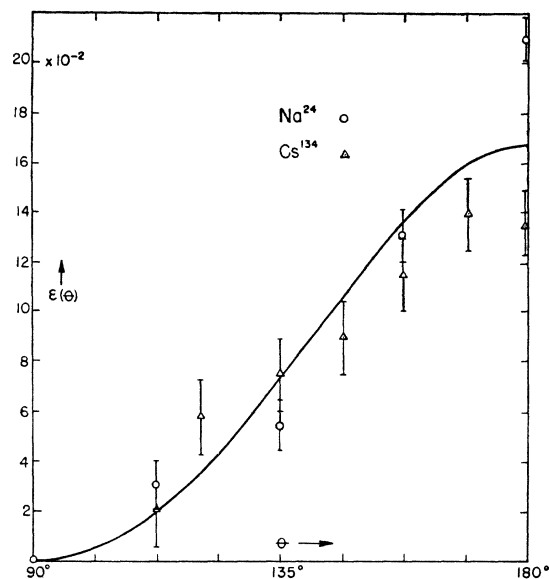FIG. 4. Correlation of gamma-rays from Cs[134] and Na[24].

shown in Fig. 2. It is very unlikely that the disturbance due to this can be comparable to our estimated experimental uncertainty.

3. RESULTS

Figures 3 to 6 show the results obtained with a number of substances. The ordinate represents

$$\epsilon(\theta) = [n(\theta) - n(\tfrac{1}{2}\pi)]/n(\tfrac{1}{2}\pi) = W(\theta) - 1.$$

The solid lines in Fig. 3 and 4, and the upper line in Fig. 5 represent the equation

$$\epsilon(\theta) = 0.125 \cos^2\theta + 0.042 \cos^4\theta$$

characteristic of two successive quadrupole transitions with angular momenta $J_1 = 0$, $J_2 = 2$, $J_3 = 4$ for the three states in order of increasing excitation energy. The line in Fig. 6 has the equation $\epsilon(\theta) = -1.5 \cos^2\theta + 2 \cos^4\theta$. Its significance and that of the lines in Fig. 4 will be discussed below.

Co[60]

The decay of this nuclide was shown by Deutsch, Elliott, and Roberts[14] to lead to an excited state of Ni[60] which decays by the successive emission of two quanta whose energies have been measured by Lind, Brown, and duMond[15] to be 1.172 ± 0.001 Mev and 1.332 ± 0.001 Mev, respectively. Ni[60] being an even-even nucleus its ground state probably has spin zero.

Deutsch *et al.*[14] showed that the beta-decay of the 10.7′ isomer of Co[60] probably leads to the intermediate excited state of Ni[60] and W. C. Peacock (private communication) confirmed this, showing that the energy of

FIG. 3. Correlation of Co[60] gamma-rays.

[14] Deutsch, Elliott, and Roberts, Phys. Rev. **68**, 193 (1945).
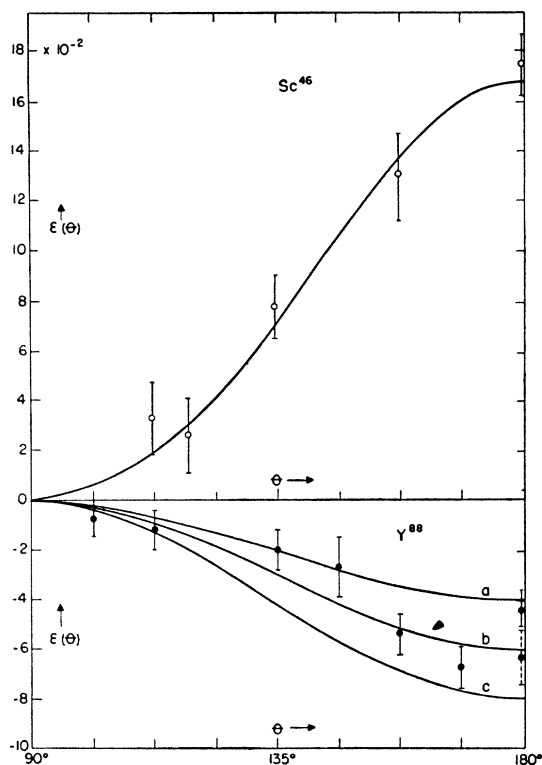[15] Lind, Brown, and duMond, Phys. Rev. **76**, 591 (1949).

FIG. 5. Correlation of gamma-rays from Sc[46] and Y[88]. The point marked by the broken line has been corrected for an estimated effect of positrons.

this level corresponds to the 1.332-Mev gamma-ray. The electromagnetic transition from the 10.7' state to the 5 yr. state of Co[60] probably involves no parity change and an angular momentum change of 3 or 4.

Our experimental points in Fig. 3 follow the solid line within the experimental error and it appears very probable that the angular momenta of the excited states are 2 and 4, respectively. Metzger and Deutsch[16] have observed a polarization-direction correlation for Co[60] which indicates that if the above spin assignment is correct the parities of all three states of Ni[60] are the same, i.e., both transitions electric quadrupole. Siegbahn and Deutsch[17] have measured the internal conversion of the two gamma-rays and their results also indicate that the multipole nature of the two transitions is the same. The observations of Fluharty[18] indicating that the direct transition from the 2.5-Mev level to the ground state of Ni[60] occurs in less than 2.5×10^{-7} of the disintegrations favor the assignment of different parities to the 2.5-Mev state and the ground state. However, this conclusion is based on the very rough lifetime formula of Bethe and Segrè. The matrix element for the "cross-over" transition may easily be a hundred times smaller than calculated from this formula. A spin and parity

assignment consistent with all our present information is shown in Fig. 7.

Cs[134]

A sample of this nuclide was obtained from the Isotopes Division. Its decay scheme is somewhat more complex[19] than those of the other activities reported here but the main mode of decay is very similar to that of Co[60], the energies of the two gamma-rays being 0.79 Mev and 0.60 Mev, respectively. The results shown in Fig. 6 are not too dissimilar to those for Co[60] and Sc[46]. Because of the complexity of the decay scheme it is difficult to draw definite conclusions. One might assume that the main cascade involves the same spins as in the case of Co[60]. In this case the polarization measurements of Metzger[16] indicate that the parities are also the same. Such a spin and parity assignment would imply that the 1.35 Mev gamma-ray reported by Siegbahn and Deutsch[20] should accompany only the 0.09-Mev beta-ray spectrum reported by Elliott and Bell.[19] This is consistent with the fact that Fluharty[18] found indication of a transition from the 1.96 Mev level directly to the ground state with an abundance just under one per million disintegrations. While these interpretations are still somewhat speculative, it can be stated with certainty that the main gamma-ray cascade cannot involve spins giving rise to very great anisotropy, e.g., 0, 2, 0. Our failure to observe the correlation in the preliminary experiments with Geiger counters[10] is inexplicable.

Na[24]

The decay of this nuclide is well known to involve the successive emission of two gamma-rays of 1.38 Mev and 2.76 Mev. It was prepared in the cyclotron by deuteron bombardment of sodium. As seen in Fig. 4 the angular correlation is again rather similar to that obtained with Co[60] and Sc[46], indicating that the spins of the states involved may also be 0, 2, 4. No indirect support for this assignment is available, though, as in
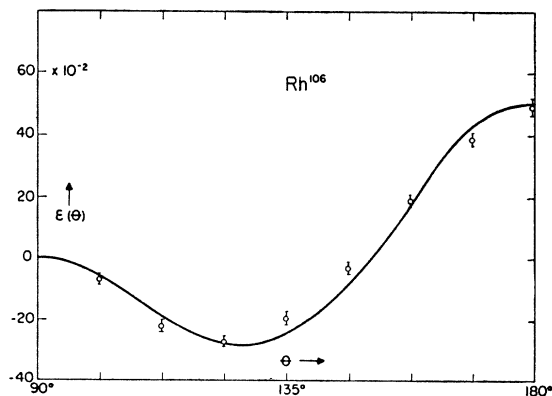

FIG. 6. Correlation of Rh[106] gamma-rays.

[16] F. Metzger and M. Deutsch, Phys. Rev. 78, 551 (1950).
[17] M. Deutsch and K. Siegbahn, Phys. Rev. 77, 680 (1950).
[18] R. G. Fluharty and M. Deutsch, Phys. Rev. 76, 182 (1949).
[19] L. G. Elliott and R. G. Bell, Phys. Rev. 72, 979 (1947).
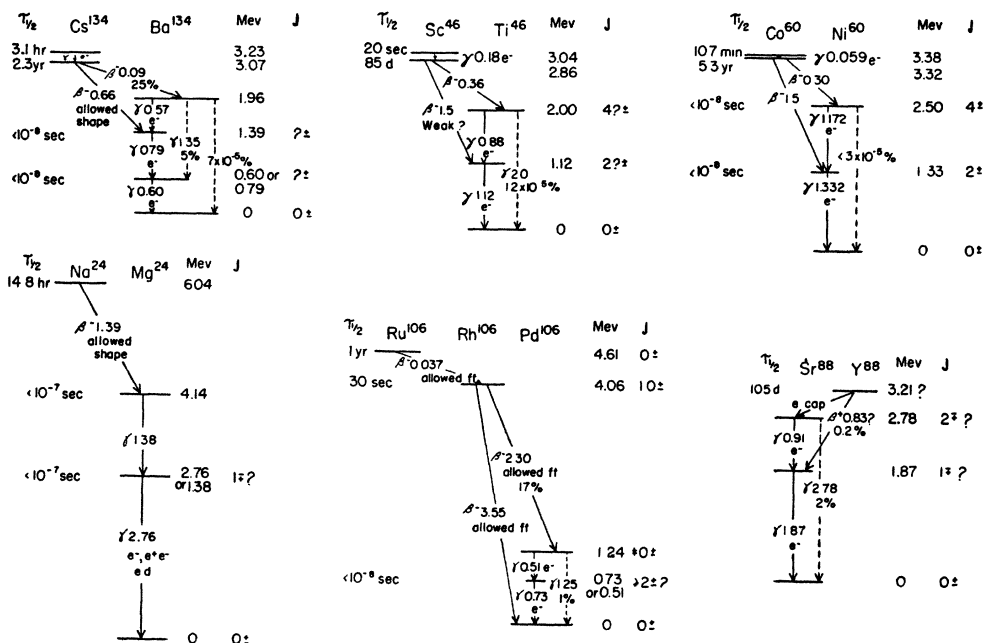[20] K. Siegbahn and M. Deutsch, Phys. Rev. 71, 483 (1947).

FIG. 7. Summary of information concerning the decay schemes discussed. States of opposite parity are indicated by $\pm$ and $\mp$, respectively.

the case of Co^{60} and Sc^{46}. The point at 180° in Fig. 4 is definitely higher than would be expected from this assignment. This may be caused by internal pair conversion of the 2.76-Mev gamma-ray. The annihilation radiation due to the resulting positrons may cause the observed effect. By changing the thickness and material of the source cup it was shown that the increased counting rate at 180° was not due to pairs created by the gamma-rays in the cup. This point should be investigated further since this type of measurement may be used as a sensitive determination of internal pair creation.

Sc^{46}

This nuclide was obtained from the Isotopes Division of the Atomic Energy Commission. Its decay scheme has been shown[21-23] to be very similar to that of Co^{60}, the gamma-ray energies being 1.2 Mev and 0.88 Mev. The results, shown in Fig. 5 are again consistent with spin assignments 0, 2, 4 to the three states of Ti^{46}. Metzger[16] demonstrated from the polarization correlation that the parities of all three states must be the same as in the case of Co^{60}. Fluharty[18] found evidence of a "cross-over" gamma-ray, corresponding to a direct transition from the 2.0 Mev level to the ground state in about 1.2×10^{-7} of the transitions. This is consistent with the proposed spin and parity assignments. Figure 7 summarizes our present knowledge of this nuclide.

Y^{88}

This nuclide was obtained by deuteron bombardment of strontium metaborate in the M.I.T. cyclotron. The

target was allowed to cool for three weeks. After removing strontium by precipitation in conc. HNO_3 and copper by precipitation with H_2S, yttrium was precipitated with HF, dissolved in $HClO_4$ and finally reprecipitated with NH_4OH.

The orbital electron capture decay of Y^{88} has been shown[24] to lead to the successive emission of two gamma-rays of 0.91 Mev and 1.86 Mev energy.[25] Gamertsfelder[26] has also shown that in about 2 percent of the disintegrations a single "cross-over" gamma-ray of 2.8 Mev is emitted instead of the two quantum cascade. This relatively high abundance of the "cross-over" transition makes it obvious that the spins must be different from those in Co^{60} and Sc^{46}, where there is only very little direct one-quantum de-excitation. Indeed the points shown in Fig. 5 indicate a very different angular correlation. They cannot be fitted by any of the curves calculated by Hamilton for pure dipoles and quadrupoles (see Table I). It is, of course, possible that higher multipoles for which calculations are not available, are involved in Y^{88} or that the ground state of Sr^{88} does not have $J=0$. But the results can also be fitted by assuming that the first gamma-ray of the cascade is a mixture of electric quadrupole and magnetic dipole radiation. If the ground state has spin zero, the second gamma-ray must, of course be a pure multipole and agreement is obtained by assuming it to be a dipole. The angular momenta involved must then be 0, 1, 2. Ling and Falkoff[5] have calculated the coefficients of the correlation to be expected in such a case. We are indebted to them for having made their more

[21] I. Feister and L. F. Curtiss, J. Research Nat. Bur. Stand. 38, 411 (1947).
[22] A. E. Miller and M. Deutsch, Phys. Rev. 72, 527 (1947).
[23] C. Peacock and R. G. Wilkinson, Phys. Rev. 72, 251 (1947).

[24] Downing, Deutsch, and Roberts, Phys. Rev. 60, 470 (1947).
[25] W. C. Peacock and J. W. Jones CNL-14 (1948), also declassified manuscript.
[26] G. R. Gamertsfelder, Phys. Rev. 66, 288 (1944).

complete calculations available to us in advance of publication. We made the somewhat arbitrary assumption that the electric and magnetic components are in phase. Although the coefficient a_1 for pure electric quadrupole or pure dipole radiation with the above spin assignments is positive, it is negative for the mixture unless the relative intensity of the quadrupole component is either less than 2 percent or more than 85 percent. The lower solid curves in Fig. 5 represent the correlation $W(\theta) = 1 - a \cos^2\theta$, with $a = 0.04$, 0.06, 0.08. Our results may be fitted satisfactorily by assuming that the relative quadrupole intensity is either between 1 and 2 percent or between 80 and 85 percent of the magnetic dipole intensity. The resulting spin and parity assignments are shown in Fig. 7. They are in good agreement with the observed "cross-over" intensity and in fair agreement with the observed internal conversion coefficients.[25] The latter agreement had previously been reported to be excellent but the improved calculations of Rose, Goertzel et al. have made it somewhat less disintegrations a single "cross-over" gamma-ray of 2.8 favorable. The observed conversion coefficients definitely indicate that no multipoles higher than quadrupole are involved. The possibility that the positron spectrum observed by Peacock and Jones[25] may really be at least partly due to internal pair conversion cannot be entirely ruled out.

Rh[106]

Rh[106] is the 30″ daughter of the 1 yr. Ru[106] fission product. It was used in equilibrium with its parent which emits only very soft beta-radiation and no gamma-rays.[27] The ruthenium was received from Oak Ridge in a solution containing long lived Tc, Ru, Rh, and Te, all carrier free. Ruthenium was separated from the mixture by distillation of ruthenium tetroxide from perchloric acid solution, and subsequent reduction with ethyl alcohol. The sample was old enough for the 42 day Ru[103] to be negligible. Rh[106] has been reported by Peacock[28] to decay to the ground state of Pd[106] with the emission of a 3.55-Mev beta-ray in 82 percent of the disintegrations. Eighteen percent of the beta-decays lead to a state of Pd[106] with 1.25 Mev excitation. This state decays mostly with the emission of two successive gamma-rays of 0.73 Mev and 0.52 Mev. Peacock presents evidence for a 1.25 Mev "cross-over" gamma-ray appearing in about 1 to 2 percent of the total number of disintegrations. Both beta-transitions appear to be allowed and the existence of this gamma-ray is a strong argument for Gamow-Teller selection rules since it indicates that the 1.25-Mev state cannot have $J = 0$ if the ground state of even-even Pd[106] is assumed to have zero spin. Unfortunately the abundance of the hard gamma-ray is so low that it is difficult to be certain of its assignment in the decay scheme.

Figure 6 shows that the angular correlation of the gamma-rays from Rh[106] is very different and much more anisotropic than for any other substance investigated. The fact that the curve of $\epsilon(\theta)$ vs. θ crosses the axis shows that at least two coefficients in the expansion in powers of $\cos^2\theta$ must be different from zero so that both gamma-rays must be at least quadrupole radiation. Thus the spin of the first excited state of Pd[106] must be at least 2. The data cannot be fitted with any of the coefficients calculated by Hamilton (see Table I). This would seem to establish definitely that the spin of the second excited state cannot be zero since the correlations for the assignments 0, 1, 0 and 0, 2, 0 are unique because no cross terms between electric and magnetic components can occur in these cases.

Ling and Falkoff[29] report that no combination of dipole and quadrupole radiations can explain the observed correlation for Rh[106] even if one includes interference terms. One must then conclude that either higher multipoles occur in the decay of Rh[106] or that our results do not represent the true correlation but are distorted by instrumental effects or by disturbing fields at the nucleus. Indeed the solid line in Fig. 6 represents the correlation expected for spins 0, 2, 0 but with the *coefficients reduced to half* of their calculated values. The rather good agreement with the experimental points is probably fortuitous but sufficiently suggestive to have caused us to investigate carefully the possibility of instrumental effects. The observed correlation could have been reduced by scattering of the gamma-rays in the source cup which has to be fairly heavy to stop all of the energetic beta-rays from the source. However, no measurable change occurred when the necessary absorber was placed at the counters instead of the source. Another possibility is the presence in the decay of some undiscovered gamma-ray cascade with nearly isotropic correlation, superimposed on the main process. However the observed correlation was not measurably affected when the amplifier gain was changed to favor higher energy gamma-rays. This indicates that the disturbing gamma-rays must have very nearly the same energy as those giving rise to the correlation, an unlikely coincidence which we may disregard. The possibility of internal magnetic fields disturbing the nucleus during the life time of the intermediate state was investigated by the method discussed in the next section. The result was negative. The solution of the riddle of Rh[106] will probably be provided by measurements of internal conversion coefficients. If octupole radiation is indeed involved it must be the second gamma-ray and the spin of the first excited state must be 3, provided the ground state has spin 0, since the fact that both beta-transitions are allowed fixes the spin of the second excited state as 0, 1 or 2. Higher multipoles are definitely ruled out by life-time considerations.

[27] L. E. Glendenin, NNES-PPR Vol. 9B (1946).
[28] W. C. Peacock, Phys. Rev. 72, 1049 (1947).

[29] D. S. Ling and D. L. Falkoff, Phys. Rev. 76, 431 (1949).

4. EFFECT OF A MAGNETIC FIELD

It was pointed out in the introduction that the theory of angular correlation as used here can be expected to hold only if there is no appreciable reorientation of the nuclear moment during the lifetime of the intermediate state. In particular such reorientation might occur unless the width of the intermediate state is considerably greater than the hyperfine structure splitting of the state involved. While reasons were adduced in Section 1 which make it very unlikely that the known fields existing in solids could destroy the correlation in most cases, it does not follow necessarily that the observed correlation represents the one which would be obtained with an entirely undisturbed nucleus. Particularly with quadrupole and higher multipole radiation of relatively low energy, as in Rh^{106} and possibly Cs^{134}, the lifetime of the intermediate state may accidentally be long enough for a residual hyperfine structure splitting of 0.01 cm^{-1} to cause an appreciable distortion of the correlation. Deutsch and Stevenson[30] have recently demonstrated an abnormally long lifetime for a transition whose internal conversion[31] shows it to be almost certainly magnetic dipole or electric quadrupole radiation. Goertzel[3] has calculated the effect of a disturbing field on the coefficients of the angular correlation and has also pointed out that an external magnetic field of the order of 10^4 gauss may in many cases be sufficient to remove the perturbation to a considerable extent provided one of the gamma-rays is observed in the direction of the field. Since it is only necessary to make the coupling between the electronic shell and the external field stronger than the hyperfine structure coupling to make the direction of the average magnetic field at the nucleus coincide with that of the applied field, such moderate fields suffice to fix a "good" axis of quantization. Furthermore it is evident that one need in general only observe the coincidence rate for one position of the counters with and without the magnetic field to establish the effect of the field on the angular correlation since the total intensity of radiation must remain unchanged and for every increase in one position there must be a corresponding decrease in another.
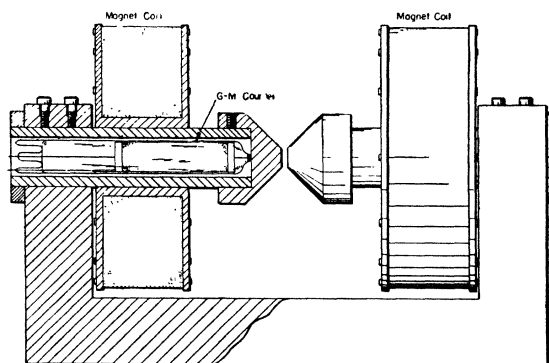
Since it did not seem practicable to provide simul-

FIG. 8. Apparatus for studying the effect of a magnetic field on the coincidence rate.

taneously cooling and adequate magnetic shielding for the multiplier tubes while maintaining a favorable geometry, Geiger counters were used in these experiments. The experimental arrangement is shown in Fig. 8. The counter tubes were placed inside the hollow pole pieces of the magnet, at opposite sides of the source and the coincidence rate was measured with and without a magnetic field of about 12,000 gauss. The results are summarized in Table II. The statistical accuracy of the results is not as good as might be desired since only Geiger counters were used. It seems probable, though, that there is no drastic change in the angular correlation due to reorientation of the intermediate state. Recent progress in scintillation counter technique should make it possible to investigate the effect further.

It should be noted that while a relatively weak external field will remove the distortion of the angular correlation by perturbing atomic fields, a field of this magnitude will not affect an undistorted correlation unless the life-time of the intermediate state is at least of the order of 10^{-8} sec. Indeed, if it should be found that the magnetic fields in solids or solutions are sufficiently weak even at the position of a recoiling nucleus to permit observation of the angular correlation when the lifetime of the intermediate state exceeds this time, then it should become possible to measure the magnetic moment of the nucleus in the intermediate state by a careful observation of the effect of an external field. The experiment would essentially compare the Larmor precession period of the nucleus with the lifetime of the state. If delayed coincidences are observed it will be found that the effect of the field is greatest on the correlation of coincidences with the longest delay. Experiments of this type are in preparation in this laboratory.

5. EFFECT OF CHEMICAL BINDING

If the angular correlation is greatly disturbed by some unsuspected mechanism involving collisions of the recoiling nucleus one might expect that the observed correlation would depend strongly on the medium in

TABLE II. Effect of magnetic field on coincidence rate at 180°.

Sample	Conc. per minute Magnet off	Magnet on	Percent difference
Co^{60} (solution)	5.4±0.13	5.2±0.2	−4±4
Co^{60} (gas)	8.9±0.2	8.6±0.3	−3±5
Y^{88} (solid)	13.6±0.2	14.1±0.3	4±3
Cs^{134} (solid)	5.2±0.2	5.2±0.2	0±6
Cs^{134} (solution)	5.2±0.2	5.2±0.3	0±6
Sc^{46} (solid)	8.4±0.2	8.3±0.2	−1±3
Rh^{106} (solid)	10.0±0.4	9.6±0.4	4±6

[30] M. Deutsch and D. T. Stevenson, Phys. Rev. 76, 184 (1949).
[31] Boehm, Hubei, Marmier, Preiswerk, and Steffen, Helv. Phys. Acta 22, 69 (1949).

which the atom finds itself. It seemed, therefore, worth while to examine at least one of the decay processes when the radioactive material was in gaseous, solution and solid form. Co^{60} was the material chosen since it seemed easiest to prepare as a gas in the form of $CoNO(CO)_3$ according to the procedure of Blanchard and Gilmont.[32] The solid was in the form of Co_2O_3 and the solution of $CoCl_2$. Because of the need to cool the multipliers the samples had to be compared using Geiger counters. In order to speed the collection of data four counters were arranged in the manner of a cross around the source. All six possible twofold coincidence combinations were counted simultaneously, two corresponding to $\theta = \pi$ and four to $\theta = \pi/2$. The samples were changed repeatedly to average possible drift in the apparatus and were chosen to give very nearly the same counting rates to avoid effects due to possible differences in recovery time of the several counters. Within the accuracy of the experiment, about 2 percent, no differences among the samples were found. As in the case of the effect of magnetic fields such experiments should be carried out in cases where the intermediate state has a measurable lifetime.

6. CONCLUSIONS

In these experiments we have investigated gamma-ray cascades in even-even nuclei only. If the ground states of all of these have zero angular momentum an anisotropic correlation should be observed in every case except for accidental cancellation of terms by interference. Such a correlation was observed in all cases, proving the applicability of the theory to experiments of this type in contradiction to previous results by other observers. The choice of even-even nuclei also reduces the number of possible combinations of spins which must be considered. Table I lists the coefficients for all the possible combinations up to quadrupole

transitions. It appears from the table that for example, the Co^{60}, Sc^{46}, Cs^{134}, Na^{24} correlations can only be fitted by the 0, 2, 4 spin sequence. All other combinations differ at least by a factor two from the data. This result alone does not establish the spins because only the simplest combinations are considered in Table I. On the other hand Y^{88} and Rh^{106} *must* involve either octupole radiations or interference terms. It should be noted that for zero spin ground states the second of the two gamma-rays must always be a pure multipole. This again reduces the number of possibilities that need be considered. Despite this simplification, unique spin assignments cannot be made from the angular correlation alone, but certain conclusions can be drawn. For example, the first-excited state of Pd^{106} must have a spin of at least 2. The second-excited state of Ni^{60}, Ti^{46}, and Mg^{24} cannot possibly have spin zero, etc. It is also interesting that at least half, and probably all but one, of the gamma-rays involved are quadrupole or higher multipole transitions. The evidence very strongly supports the view that the spins of the odd-odd nuclei Co^{60}, Sc^{46}, and Na^{24} are high. The spin of Rh^{106} must, of course be 0 or 1 because of the allowed beta-transition from the ground state of Ru^{106} and to the ground state of Pd^{106}. The most urgently needed data are precise measurements of the internal conversion coefficients. It seems certain that the combination of such data with the angular correlation and polarization experiments will make all spin and parity assignments unique.

It is a pleasure to acknowledge the help of Professor J. W. Irvine with the chemical phases of this work. All of the theoretical physicists quoted have contributed further through private discussions.

Note added in proof.—Since this paper was written a number of related results and suggestions have been published: Waggoner, Moon, and Roberts (Bull. Am. Phys. Soc. 25, No. 3, 45 (1950)) measured the internal conversion coefficients for Co^{60} and find both gamma-rays to be electric quadrupole radiations. Rae (Phil. Mag. 40, 1155 (1949)) measured the internal pair conversion of the 2.76-Mev gamma-ray of Na^{24} and showed it to be an electric dipole transition. Calculations of Spiers (Phys. Rev. 78, 75 (1950)) indicate that our results for Rh^{106} could not be explained by octupole terms.

[32] A. A. Blanchard and P. Gilmont, J. Am. Chem. Soc. 62, 1192 (1940).