Angular Correlation of Nuclear Radiations[1]

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THE PROBLEMS OF NUCLEAR PHYSICS have been a major concern of physicists for at least two decades. In the concerted effort which has been brought to bear on these problems, the primary question has been concerned with nuclear structure. That is, how are we to understand the existence of the known stable nuclei with specified neutron and proton numbers, the occurrence of unstable nuclei with their various modes of spontaneous decay, and the rates of such decay processes? These questions, as well as many others concerned with the binding energies, or, more generally, with the energy levels of stable as well as unstable nuclei, could be understood (in principle) if we were in possession of all the facts concerning nuclear forces and had the means of applying this knowledge in a detailed solution of the problems of nuclear dynamics. During the past twenty years considerable progress has been made toward the solution of this difficult problem; but, in many important areas, our understanding is not only qualitative but, frankly speaking, rather crude. In part, the difficulty is that while the general principles of the dynamics are well known, sufficiently complete information concerning nuclear forces is not yet available.

In view of the formidable character of the problem with which the nuclear physicist is faced, it is understandable that considerable effort has been expended in recent years along the lines of a phenomenological approach. Short-circuiting the nuclear force problem, at least temporarily, one asks if a more or less simple model can be invented which will account for the experimental facts, or at least some features of them. This type of *ad hoc* attack has the virtue that (*a*) progress can be made, (*b*) a "set of rules" can be established whereby experimental results can be interpreted and our knowledge advanced, and (*c*) an insight into the nature of the operative forces can be gained.

Once a model has been postulated, its validity (or degree thereof) can be demonstrated only by comparison with experiment and this in itself is no trivial program. The example, *par excellence,* of such models is the so-called shell model (*1, 2*) which makes a specific assumption with regard to the dependence of the forces on the angular momenta of interacting particles (coupling scheme). From this deductions as to the angular momenta of ground and excited states, the occurrence of isomers (long-lived energy states), the systematics of beta-decay and many other aspects of nuclear structure can be made. However, to take the example of angular momentum values of nuclear energy levels, the deduction of these from experiment is by no means direct, and a considerable body of auxiliary theory must be developed for this purpose. We return to this question below.

The general objective to be considered now is the determination of the properties of nuclear levels and the investigation of dynamic characteristics of the radiations emitted when an unstable nucleus is formed. To take a simple example, Co^{59} irradiated in a pile provides radioactive Co^{60} by capturing a neutron and emitting γ-rays ($n-\gamma$ reaction). From the ground state, as well as from the first excited state electron emission (β^-) takes place, forming Ni^{60} in second and first excited states respectively. Stable Ni^{60}, in the ground state ensues by γ-emission. In the case of the 0.32-Mev β^--emission, there is a cascade of two γ-rays of energies 1.17 and 1.33 Mev in temporal order. Figure 1 shows the decay scheme for this chain of events.

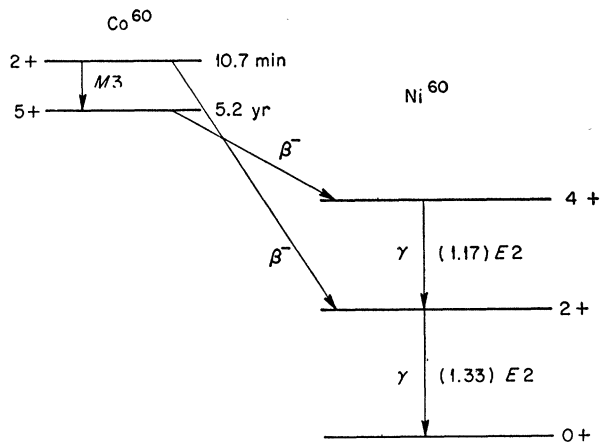

FIG. 1. Decay scheme of Co^{60}. The γ-ray energies in Mev are shown in parentheses (*3*).

The genealogy of the Co^{60} decay is established by β–γ coincidence studies (*3*). The angular momentum and parity[2] changes involved in the β-transitions are established from a study of the shapes of the β-spectra and the measured β-lifetimes. The angular momentum and parity change between the two lowest Co^{60} states

[1] A much more detailed and comprehensive survey of this subject appears in the article by L. C. Biedenharn and M. E. Rose, *Rev. Mod. Phys.*, **25**, 729 (1953). In that paper, which will be referred to as BR, numerical results as well as a complete quantitative account of the theory will be found. A much more complete list of references is also to be found in that survey article.

[2] Parity is a certain symmetry property of the wave function describing the nuclear state considered. The concept of parity has no classical analogy. The parity of a state is "even" if an inversion of all spatial coordinates does *not* change the sign of the associated wave function. When the inversion of the spatial coordinates in the wave function *does* change the sign, then the parity of the state is said to be "odd."

is determined by measuring the internal conversion coefficient (ratio of conversion electrons to γ-rays) emitted in this transition. The designation $M3$ in Fig. 1 means a magnetic octupole γ-ray. The angular momentum and parity of the Ni^{60} ground state is almost certainly 0^+, since without exception this is true of all nuclei with even numbers of protons (28 here) and neutrons (32 here). The remaining information (angular momentum and parity of the two excited states of Ni^{60}) can also be obtained by internal conversion and relative gamma intensity measurements. Internal conversion effects, however, are quite weak for these gamma rays, and lifetimes for γ-transitions are here too short to be measured. Consequently the independent determination of the angular momenta and the parities of the Ni^{60} states by angular correlation measurements is of major interest. This particularly straightforward case illustrates the fact that the unraveling of decay schemes involves the combined use of several kinds of experimental techniques and the pooling of information derived from all of them.[3] In very many cases the method of angular correlation measurements is the only way to determine angular momenta of nuclear states.

Description of the Angular Correlation Process

Whenever a nucleus in an unstable state emits two (or more) radiations in succession there is a possibility that the directions of emission are not randomly distributed with respect to each other but are *correlated* (*4*). Some angles θ between the two propagation vectors are more or less favored over others. This result will also apply, in general, if the radiations are not consecutive. Only in special cases will this correlation fail to appear, and all relative directions are equally probable. One then speaks of *isotropy*. When the correlation is isotropic one can often deduce valuable information from this fact as well. Two facts must be emphasized: (*a*) The radiations need not be two γ-rays, as in Ni^{60}. Either one or both transitions may involve emission of α-particles, β^+- or β^--particles, protons, neutrons, deuterons, conversion electrons, or even x-rays following conversion or K-capture. We refer in the sequel to a set of nuclear levels with specified properties (energy, angular momentum j, parity ±), but these levels need not belong to the same nuclear species. (*b*) In the above we refer to a correlation measurement in which the directions of emission of the two radiations are observed. This is the easiest measurement to make and, in a large number of cases, it suffices to provide all the information needed. However, in one important case, that is, γ-ray emission, the parity change is not fixed by the angular correlation observations. This information is provided by a so-called polarization-direction correlation measurement. This is discussed more completely later.

[3] Ni^{60} is also formed by the decay of Cu^{60} which emits positrons (β^+). In other cases there is appreciable competition between β^+-emission and the capture of orbital electrons (mainly from the K-shell). The measurement of the branching ratio, K-captures to β^+-emissions, is also useful in some cases.

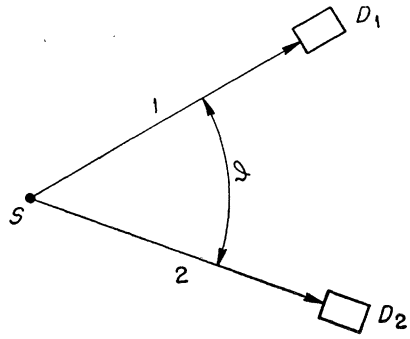

Fig. 2. Schematic angular correlation arrangement.

The experiment envisaged (*5*) is the measurement of the coincidence rate of two detectors, see Fig. 2, as a function of the angle θ. In most cases one can easily arrange matters so that one particular detector records only one of the two radiations, the other detector recording the other radiation (*6, 7*). When this is not convenient it is only necessary to consider the weighted average of the expected coincidence rate for radiation 1 to detector D_1 and radiation 2 to detector D_2 with the rate for the radiations interchanged. The weight factors are overall detector efficiencies which are easily measured.

It is important to understand why there should be an anisotropic correlation at all. For this purpose only very simple facts need be adduced. First, consider a single transition taking place from a level of angular momentum j to another of arbitrary angular momentum. All angular momenta and components are measured in units of $\hbar$ = Planck's action constant divided by 2π; i.e., $\hbar = 1.05 \times 10^{-27}$ erg sec. The first fact is that, as a consequence of space quantization of all angular momentum vectors, these vectors can assume only a finite number of orientations. For the initial level there are $2j+1$ orientations possible corresponding to the fact that the component of angular momentum in any[4] *one* direction can assume the values $m = -j, -j+1, \ldots j-1, j$. In the absence of magnetic fields (internal or external) and homogeneous electric fields, these $2j+1$ states are degenerate, that is, they have the same energy. Consequently all these states with different m are equally populated. The total intensity radiated in any direction is the sum due to transitions from the $2j+1$ substates and these enter with equal weights. As a consequence, one finds the expected result that the radiation is isotropic. On the other hand, if an external magnetic field were present the $2j+1$ substates would be no longer degenerate but would show a very small Zeeman splitting. The ensuing unequal populations distributed according to the Boltzmann law, would have the effect of introducing anisotropy (with respect to the magnetic field direction) because the unequal weights with which each substate enters would prevent the exact cancellation of anisotropic

[4] The direction is arbitrary because there is no physical distinction between various directions in the situation considered. Thus, in the absence of a magnetic field, which would make one direction preferred, space is isotropic.

contributions arising from each of these substates.

How then does anisotropy arise in a correlation of radiations in coincidence, even though there are no fields producing level splitting? The answer is that by selecting a specific direction for one of the radiations (say, the first) we select a particular subset of all possible transitions originating from the intermediate level (see Fig. 3). While all $2j+1$ substates are equally populated the transitions from the separate m values do not contribute equally. Selecting a direction for the first transition makes space anisotropic. In detail, suppose we consider two γ-rays emitted successively and further, suppose that $j_1 = 0$, $j = 1$, $j_2 = 0$ in Fig. 3. The angular dependence of the intensity of γ-radiation from j_1 to j is:

$$I_{+1} = \tfrac{1}{2}C(1+\cos^2\theta), \quad I_0 = C\sin^2\theta \tag{1}$$

where the subscripts are the values of $\Delta m = m_1 - m$ and here $m_1 = 0$. C is a constant and θ is the angle between the propagation vector of the first γ-ray and the axis of quantization-axis with respect to which m is measured. This axis can be taken arbitrarily. If it is taken along the direction of the first γ-ray, $\theta = 0$ and only $I_{\pm 1} \neq 0$. The state $m = 0$ is not populated at all so far as the experiment is concerned. Consequently, in the second transition only $\Delta m = \pm 1$ is possible and the correlation function giving the coincidence rate is proportional to $1 + \cos^2\theta$ – anisotropic! This result can be expressed by the observation that a γ-ray can have components of angular momentum along its direction of propagation equal to ± 1 only. Similarly, for an α-particle only a zero component of angular momentum along the direction of motion is possible. In the above example the correlation for an $\alpha - \gamma$ or $\gamma - \alpha$ transition is proportional to $\sin^2\theta$. In all cases only the angular shape is important and proportionality constants are irrelevant.

The application of the data can now be seen. If one considers the scheme of Fig. 3 and the case of γ-rays emitted in both transitions as an example, the first transition will be a 2^{L_1} pole γ-ray, where L_1 is an integer between the limits $|j_1 - j|$ and $|j_1 + j|$ and the second will be a 2^{L_2} pole γ-ray, $|j_2 + j| \geq L_2 \geq |j_2 - j|$. Because the radiation wavelength is always considerably larger than the size of the nucleus, usually the smallest L value ($L > 0$) consistent with parity conservation will be important. This is the case of pure multipoles. It is possible that either or both transitions contain an admixture of the next higher pole (2^{L_1+1} and 2^{L_2+1}), where L_1 and L_2 are the minimum values. This will not occur unless both j's involved in the transition differ from zero. In any case, the theoretical evaluation of the correlation function will give a coincidence rate $W(\theta)$ of the form (*8–10*)

$$W(\theta) = \sum_{\nu=0}^{\nu_{max}} A_\nu P_\nu(\cos\theta) \tag{2}$$

where P_ν is a Legendre polynomial and the A_ν depend on j_1, j, j_2 and on L_1, L_2. In the mixed case A_ν will also contain the relative intensity of the 2^{L_1+1} to 2^{L_1} poles if, say, only the first γ-ray is mixed. In Eq. (2) ν is always an even integer and ν_{max} is the smallest of $2L_1$, $2L_2$ and $2j$. Since $P_0 = 1$, it follows that for $j = 0$ or $j = \frac{1}{2}$ only isotropy results. This is obvious, at least in the $j = 0$ case. In the case $j = \frac{1}{2}$ the isotropy is a consequence of the fact that there are only two substates, and the increase of population of one is equal to the depletion in the population of the other. The anisotropy results from an effective alignment of the nuclear spins in the *intermediate* level. That is, considering the substates of this level in the way they actually contribute, then the pertinent populations are not uniform. Since L_1 and $L_2 \geq 1$ for γ-rays, the observation of isotropy tells us at once that $j = 0$ (for even mass nuclei) and $j = \frac{1}{2}$ (for odd mass nuclei), provided there are no disturbing effects which wash out the angular correlation (see below). Otherwise when anisotropy occurs, and this is the more frequently occurring case, comparison of observed results and theory enables one to assign j-values to the nuclear levels and L-values to the radiation. Numerical values of A_ν for pure and mixed multipoles are given in BR. Comparison of these results with the observations (*5*) leads to the assignment shown in Fig. 1. Both Ni^{60} γ-rays are electric quadrupole radiations ($E2$).

In an $\alpha - \gamma$ or $\gamma - \alpha$ correlation, to take another example, the A_ν are different. In fact, isotropy here can occur for $L_\alpha = 0$ ($j_1 = j$ or $j = j_2$) which means that the α-particle has zero orbital angular momentum. The difference between the A_ν coefficients for the $\alpha - \gamma$ and $\gamma - \gamma$ cascades is

$$A_\nu(\alpha - \gamma) = b_\nu(\alpha) A_\nu(\gamma - \gamma) \tag{3}$$

where $b_\nu(\alpha)$ is a so-called particle parameter which depends on properties of the α-particle only and not on the nuclear properties like j_1, j, j_2. There is an indirect dependence on the parities of the nuclear states in that between two states with the same parity, parity change "no," only α-particles with even L can be emitted and for parity change "yes," only α-particles with odd L are possible. In general, for a transition $j_1 \rightarrow j$, the possible L values are $|j_1 - j| \leq L \leq |j_1 + j|$ and for α-particles only every other value is permitted because of the parity conservation. In many cases the barrier due to centrifugal forces depresses the contri-

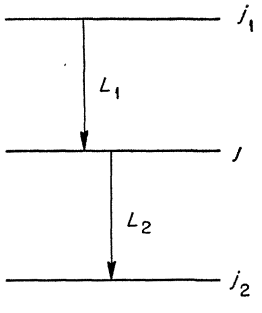

FIG. 3. Level diagram for angular correlation of successive radiations. The three levels are $2j_1+1$, $2j+1$, $2j_2+1$-fold degenerate, reading from top to bottom. The symbols L_1 and L_2 refer to the angular momentum of the emitted radiations.

butions of all but lowest possible L value. In this case we have a situation analogous to the pure multipole γ-ray case. For α-particles with a pure L the parameter $b_\nu(\alpha)$ depends only on ν and L and not on the α-particle energy or other nuclear properties.

The results expressed by Eq. (3) is a special case of a general rule (*10–12*). A standard correlation, such as the $\gamma-\gamma$, can be adopted. Then all correlation functions are of the form Eq. (2) and if the radiations are generally labeled x and y

$$A_\nu(x-y) = b_\nu(x)\, b_\nu(y)\; A_\nu(\gamma-\gamma) \qquad (4)$$

where the particle parameters $b\nu(x)$ depend only on the properties of the radiation x and A_ν contains all the dependence on nuclear j-values. The introduction of these particle parameters greatly simplifies the theory and the presentation of the results, see BR.[5] It also enables one to view the angular correlation process from a simple point of view in that part of the theory which is concerned with the properties of angular momentum and this is the same no matter what kind of radiations are emitted as long as they carry given amounts of such angular momentum. This part is expressed essentially by the coefficients A_ν. The remainder of the considerations pertinent here is the physics of the radiations. For example, this involves the potential field in which a K-electron moves in the internal conversion process (*11*). All such factors, depending on a physical model, are contained in the b_ν. In particular, for the $\beta-\gamma$ or $\beta-\alpha$ (in fact $\beta-x$) correlations, the b_ν depend not only on the energy of the β-particle and the Coulomb field in which it moves but also on the form of the β-decay theory and on quantities (nuclear matrix elements) whose values are dependent on the details of nuclear forces. When the form of the β-decay theory is firmly established by purely β-spectroscopic experiments, the correlation involving β-transitions would be useful for the purpose of fixing the values of these nuclear matrix elements and hence obtaining a clue as to the fine points of nuclear structure.

Other applications of angular correlation observations are considered later. At this juncture it suffices to say that, except in the case of γ-ray emission, a parity change determination is also provided by the angular correlation measurements and the analysis thereof. As mentioned, the directional correlation with α-particles fixes L, the α-particle orbital angular momentum, and the parity change is $\Delta\pi = (-)^L$ where $\Delta\pi$ is the product of parities of the initial and final states of the transition ($+1$ for even or symmetrical states, -1 for odd or antisymmetrical states). This statement is also true for any particle which can be treated nonrelativistically, in which case intrinsic spin effects do not enter. Thus, it applies for neutrons, protons, deuterons, etc. But for conversion electrons and β-particles, which must be treated relativistically, the parity change cannot be expressed in such a simple way. Nevertheless, the correlation processes involving such particles do depend on the parity changes involved in the corresponding transitions. This is clear when it is noted that for conversion electrons, the total intensity, or alternatively the internal conversion coefficient, is strongly parity-change dependent (*11, 13*). The nucleus, regarded as an electric dipole (of dynamic character) has a different probability for ejecting orbital electrons than a nucleus which behaves like an oscillating current loop (magnetic dipole). For β^--transitions the shape of the energy spectrum of the β^+ or β^- is parity dependent and the β-decay lifetime is strongly so. In fact, for conversion electrons and β-particles parity determinations are usually more convenient by total intensity measurements.

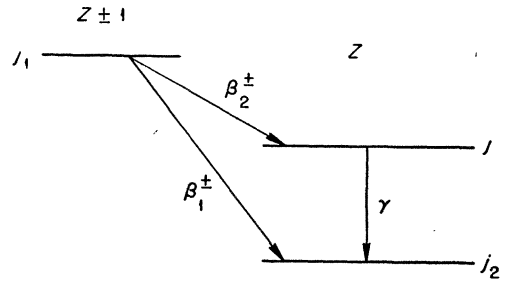

FIG. 4. Decay scheme showing a β-branching. The allowed shape occurs for β_2 if $|j_1 - j| = 0$ or 1 and the parities of these levels are the same.

This brings the discussion to the question of the relative advantage of correlation measurements over other procedures for obtaining the same information. With regard to directional $\gamma-\gamma$ correlations the information provided (L and j values) is also obtainable from separate internal conversion measurements for each transition. Aside from the fact that an angular correlation provides information about both transitions (three levels) in one experiment, there is also the important factor that both for low atomic numbers (Z less than about 20) and high transition energy (E greater than about 2 Mev) internal conversion becomes a rather weak effect. On the other hand, $\gamma-\gamma$ correlations are not affected by either of these two considerations. The coefficients A_ν $(\gamma-\gamma)$ are neither Z nor E dependent. On the other hand, we shall later discuss an important effect which causes some trouble in the interpretation of all correlation measurements but does not affect total intensity measurements. However, as will be evident, these effects are of considerable interest in that they may provide a means of studying some very interesting extranuclear phenomena which would be difficult to investigate in any other way.

Another advantage of correlation measurements can be illustrated by the simple decay scheme of Fig. 4. The excited state of the nucleus of charge $Z \pm 1$ branches, giving two β groups, β_1 and β_2. Unless the lower energy group β_2 is very much more intense than β_1 the shape of the β_2-spectrum would be difficult to measure in the presence of β_1. However, a $\beta_2-\gamma$ cor-

[5] The only important cases where the codification expressed by Eq. (4) is not applicable is in the case of neutron or proton emission (or absorption). In this case one pair of j-values differs by $n + \frac{1}{2}$ where n is an integer while in the γ-ray case the difference is always an integer.

relation is unaffected by the presence of β_1 and this provides the same (and, in fact, additional) information. If β_2 has an "allowed" shape, which occurs, for example, if in the β-transition $\Delta j = 0, \pm 1$ and $\Delta\pi = 1$, the angular correlation will be isotropic. In measuring the β-spectrum there are always instrumental deviations at low energies from the theoretically permitted shapes of the β-spectrum due to backing and source thickness distortion. This need not be a source of difficulty in the correlation measurement because it is permissible, and moreover advisable, to use pulse height discrimination in such a way that only the higher energy β-particles are accepted. If one accepts the low energy β-particles the correlation tends to be more isotropic. This is because the low energy β-particles carry essentially zero orbital angular momentum and the direction of motion of such particles is not defined.

Additional Applications of Correlation Processes

Polarization-Direction Correlations. In the foregoing mention was made of the fact that the directional $\gamma - \gamma$ cascade provides no parity-change information. The reason for this is to be found in classical terms. An electric multipole wave is one with no radial component of magnetic field strength. It is a TM wave in another terminology. A magnetic multipole wave has a zero radial component of electric field strength. It is a TE wave. In fact these two waves are dual to each other; namely, aside from constant multiplicative factors of no importance here, the electric and magnetic fields, **E** and **H** respectively, of one are obtained from the other by the duality transformation

$$\mathbf{E} \rightarrow \mathbf{H}, \qquad \mathbf{H} \rightarrow -\mathbf{E}$$

Now the Poynting flux ($\sim \mathbf{E} \times \mathbf{H}$) is unchanged by this. Hence electric and magnetic multipoles of the same angular momentum have the same Poynting flux, and hence the same intensity, at all angles.

The above statement applies only to unpolarized waves. Hence, if the polarization of one of the γ-rays is measured, as well as its direction, one can expect to find the parity change involved in the transition in which this γ-ray was emitted (*14*).

The arrangement, originally used by Metzger and Deutsch (*15*) is schematically shown in Fig. 5. Here the polarization sensitive detector is a Compton scatterer C and an ordinary (scintillation crystal) detector D_1 for the Compton scattered radiation. The other detector D_2 is as before. The angle θ is as defined above and ϕ is the angle between the electric field vector of the γ-ray going to the polarization-sensitive detector and the normal (n) to the plane containing the two γ-ray directions from the source S. One measures the number of triple coincidences in which one γ goes to D_2 and the other to C and D_1 as a function of ϕ. The theory gives a correlation function

$$W_1(\theta\phi) = \sum_\nu A_\nu P_\nu(\theta, \varphi) \qquad (5)$$

where A_ν is as before and

$$P_\nu(\theta, \phi) = P_\nu(\cos\theta) + \alpha_\nu(-)^{\sigma_1}\cos 2\phi P_\nu^2(\cos\theta) \qquad (6)$$

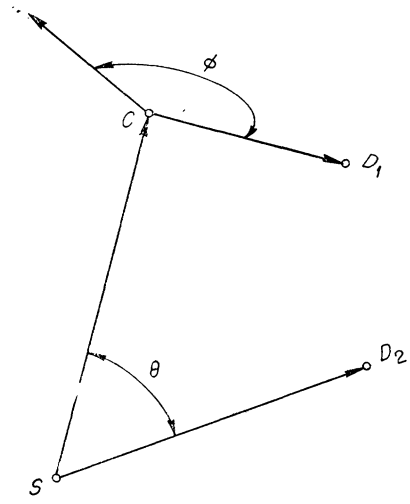

FIG. 5. Schematic diagram for polarization-direction correlation.

Here $\sigma = 0$ for electric and 1 for magnetic radiation and the parity dependence enters through the factor $(-)^{\sigma_1}$. The subscript 1 implies that it is the electric or magnetic character of the first photon which is pertinent. In Eq. (6)

$$P_\nu^2(\cos\theta) = \sin^2\theta \frac{d^2 P_\nu}{d\cos\theta^2} \qquad (7)$$

is the usual associated Legendre polynomial and α_ν depends on L_1, see BR. Pure multipoles are assumed for the sake of discussion.

One can expect that the second photon will also go to the polarization-sensitive detector ($C - D_1$), the first to the polarization-insensitive one (D_2). The correlation function for this is obtained by replacing σ_1 by σ_2 and L_1 by L_2 in α_ν. Let this correlation function be denoted by $W_2(\theta, \phi)$. If the overall efficiencies for the W_1 and W_2 arrangements are η_{12} and η_{21} then the measured quantity is

$$W = \eta_{12} W_1 + \eta_{21} W_2 \qquad (8)$$

These results provide the basis for the interpretation of all experiments with pure multipoles. If there are mixtures present the correlation function is somewhat more complicated but has the essential properties of that given above, see BR. In general it suffices to make measurements at $\phi = 0$ and $\phi = \pi/2$. If the coincidence rates in these two positions are denoted by $W_\parallel$ and $W_\perp$, it is possible to deduce information concerning relative parities from the *sign* of $(W_\parallel/W_\perp) - 1$. In particular, for transitions with $L_1 = 1$, $L_2 = 2$, or $L_1 = 2$, $L_2 = 1$ with $\sigma_1 = \sigma_2$ and for $L_1 = L_2$ but $\sigma_1 \neq \sigma_2$ it is possible to determine both parity changes simply by measurements of the above-mentioned sign and that of $[W(\pi)/W(\pi/2)] - 1$, which is referred to as the anisotropy in the directional correlation. This is also possible in the case $L_1 = 1$, $L_2 = 2$ or $L_1 = 2$, $L_2 = 1$ but with $\sigma_1 \neq \sigma_2$ and $L_1 = L_2$ with $\sigma_1 = \sigma_2$. These are all very frequently occurring cases.

Correlation of Nonconsecutive Radiations. Many

decay schemes are quite complex, and it is not unusual that great difficulty is encountered in fitting the observed γ-rays into a proper order. If there are three (or more) γ-rays in cascade, the order of these can be determined by measuring their correlations in pairs. Thus, if the γ-rays are denoted by their energies E_1, E_2, E_3 one measures the three correlations $W(E_1\ E_2)$, $W(E_2\ E_3)$, $W(E_3\ E_1)$. In this connection it is to be noted that in any correlation, if the time order of events is reversed, the correlation function is unchanged.

If, for the sake of argument, we assume that the order of emission is E_1, E_2, E_3 or E_3, E_2, E_1, then the first two correlation functions are described by the theory outlined in the preceding section while the third one is given by

$$W(E_3E_1) = \sum_{\nu=0}^{\nu_{max}} B_\nu P_\nu(\cos\theta) \qquad (9)$$

The coefficients B_ν, different from A_ν, depend on three L values and four j-values. They are given in BR. There is an extra limitation on ν_{max}; that is, ν_{max} is the smaller of the four integers $2L_1$, $2L_3$, $2j$, $2j'$, where L_1 and L_3 refer to the observed (first and third) γ-rays, j and j' are the momenta of the two intermediate states, see Fig. 6.

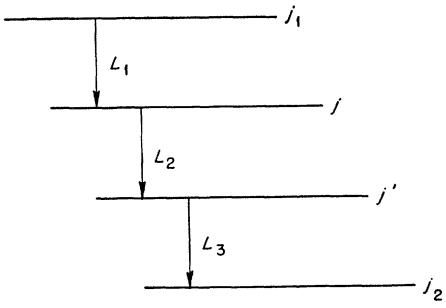

FIG. 6. Level diagram for correlation process involving three γ-rays. The correlations between the three pairs of γ-rays is to be measured.

All three correlations give information concerning at least some of the three L's and four j's and the information from each experiment overlaps. In general, only when one assigns the order of the γ-rays correctly will this overlapping information be free of contradiction.

Another possible application of the measurement to which Eq. (9) would apply, arises when the intermediate (unobserved) γ-ray is of such low energy and low intensity that its observation would be very difficult.

Measurement of Magnetic Moments of Excited States. It has been emphasized that the correlation depends in a vital way on the fact that the populations of the substates of the intermediate level were nonuniform. Clearly any type of extranuclear field which can cause transitions between these substates will alter the correlation function (*16*). If the strength and direction of this external field is given, the observed alteration of the correlation provides information concerning those parameters of the nucleus which measure the amount of interaction or coupling energy to the field. Now, it was seen that for an anisotropic correlation $j \geqslant 1$, and this means that in the intermediate state the nucleus will, in general, have both a magnetic dipole and electric quadrupole moment. Higher moments (magnetic octupole, etc.) could exist if $j \geqslant 3/2$, but are far too small to give rise to observable effects. For historical reasons we confine our attention to coupling to the magnetic dipole moment, and this implies a comparison of the correlation with and without an external magnetic field.

It is important in such an experiment that only the external magnetic field acts. Otherwise one has a hopeless tangle of effects of a quadrupole moment coupled to an inhomogeneous electric field and/or the coupling to an essentially unknown magnetic field of variable direction arising from the magnetic moment of electrons in the immediate vicinity of the nucleus. These effects can be made negligible in a manner to be described in the following section.

Whatever the nature of the coupling, two things are clear. First, the coupling will split the levels and there will be a characteristic frequency whose magnitude is a measure of this splitting. In the case of the magnetic field this is the well-known Larmor frequency

$$\omega = \frac{\mu H}{j\hbar} \qquad (10)$$

where μ is the nuclear magnetic moment of the state j. In the magnetic field **H**, the nuclear angular momentum j precesses with this frequency. If the mean life of the intermediate state is τ, the number of radians through which the precession progresses is $\omega\tau$. Unless this is comparable to unity the effect will be too small to detect. On the other hand $\omega\tau \gg 1$ is undesirable since this will give isotropy. Since $\mu \sim 10^{-23}$ cgs, this implies $\tau \sim 10^{-8}$ sec for $\mathbf{H} \sim 5000$ gauss. This is just in the range of many dipole and quadrupole γ-rays. For high-energy dipole and quadrupole radiations for which τ is less than about 10^{-12} sec the requisite field strength is too large. Moreover, a measurement of τ in this range is not feasible at present. For slower transitions (τ greater than about 10^{-4} sec) other difficulties arise.

The second fact, which emerges clearly, is that any coupling to the nuclear spin will attenuate, that is, "wash out" the correlation so that the anisotropy will tend to disappear. This comes about because precessions, or more exactly transitions between different substates will proceed most rapidly from rich populated to poorly populated substates. Hence, in certain cases, if allowed to proceed long enough the system saturates when the populations are equal. For this reason τ must not be too large.

Because there are now three directions specified, the magnetic field direction and that of the two γ-rays, the correlation function takes on a somewhat more complex form than Eq. (1). However, a practical arrangement is one in which the magnetic field is perpendicular to the plane of the two γ-rays (12). Then

$$W(\theta) = \sum_{\nu=0}^{\nu_{max}} \frac{S_\nu}{1+(\nu\omega\tau)^2} \cos\nu\theta \qquad (11)$$

where S_ν are the coefficients without magnetic field and are simply related to the A_ν of Eq. (1). Equation (11) shows that isotropy results for large $\omega\tau$.

It is clear that a comparison of anisotropies gives $\omega\tau$ and a life-time measurement gives τ. Since j is known from the usual directional correlation in zero field the magnetic moment μ in the intermediate state can be measured. Since only ω^2 enters it appears that only the magnitude of the magnetic moment can be measured. However, Eq. (11) refers to the case that the overall efficiencies of the detectors is the same when the photon directions are interchanged. It is clear that interchange of photon directions is equivalent to reversing the direction of the magnetic field. Therefore, with unequal efficiencies (using energy discrimination) one can also measure the sign of μ.

While other means for obtaining this information have been developed recently, using polarized nuclei at low temperature, the procedure described here has some obvious advantages. It has been carried out successfully in one case thus far (*17, 18*).

Very recently attenuation effects of the quadrupolar coupling in single crystals have been discovered (*19*). It must be recognized, however, that here one starts with an additional unknown factor. The inhomogeneous electric field is not a "synthetic" one and its properties are not specified a priori.

Spin Coupling in Angular Correlation

The use of a magnetic field as just described constitutes what might be called a beneficial effect of spin-coupling. As already intimated it may be that other couplings due to fields whose properties are not so well known are also present. Moreover, these fields cannot be turned off in any obvious way. They arise from magnetic interactions, which are also manifested by hyperfine structure, and from quadrupole interactions.

Let us consider a nuclear cascade initiated by K-capture. Before the K-capture takes place one may assume, for the sake of simplicity, that the electronic environment possesses no magnetic moment. In this state no magnetic interaction occurs. However, the K-capture results in a K-vacancy. In a time of order 10^{-14} sec this will be filled by x-ray emission or Auger transitions, thus removing the hole to outer orbits, and to some extent, producing new holes. The K-capture is followed by a cascade of two γ-rays or other radiations.

We further assume that the ion is embedded in an environment in which, by some process involving electron transitions, the ground state with zero coupling can be reestablished. There will be a characteristic relaxation time τ_{el} for this process. Then one of several possibilities may apply:

1) The lifetime τ_1 in the initial (j_1) nuclear state is long compared to τ_{el}. Then if the first radiation does not produce any appreciable disturbance in the electronic environment, the intermediate state is established without spin-coupling and the theory described above for an isolated nucleus applies. If by recoil or sudden charge change, the first radiation excites or ejects an electron the intermediate state is subject to spin coupling.

2) If the spin-coupling attenuation is operative in the intermediate state, the effect will be large or small according as to whether τ is about equal to or greater than about τ_{sp} or $\tau \ll \tau_{sp}$. Here τ is the intermediate state mean life and τ_{sp} is the precession period ($\sim 1/\omega$).

3) If $\tau_{el} \ll \tau$ the ion will "heal" rapidly, but for no appreciable attenuation it is necessary that $\tau_{sp} \gg \tau_{el}$ as well.

It is clear that what is going on is a series of complex electronic transitions which are concomitant with and strongly conditioned by the nuclear transitions. Everything depends on a comparison of τ_{el}, τ, and τ_{sp} in the intermediate state. The first is a function of only the electronic environment, the second is essentially a purely nuclear property, and the third is a property of the coupling between nucleus and environment. This strong interplay of nuclear and nonnuclear phenomena is due to two facts. First, we are dealing with an angular distribution. In contrast, if we require a knowledge of the total number of transitions regardless of which substate of the intermediate level was involved, the precession of the angular momentum j is irrelevant. Hence total intensity measurements are free of these effects. Second, the nuclear lifetimes are often of the same order of magnitude as the time constants for the coupling and extranuclear processes.

From the viewpoint of the nuclear physicist there are two ways to treat the situation in which spin-coupling modifies the correlation. First, one may attempt to include it in the calculations. Secondly, one may attempt to remove spin-coupling or minimize its effects. These effects are often too small to be important, and even where they are observable they may not becloud the issue entirely because the experimental results are to be compared with a finite number (often a small number) of distinct possible theoretical curves. However, in a second class of correlations the degree of spin-coupling is too large to ignore.

The first possible course, wherein spin-coupling effects are calculated, can be carried out only if one assumes that the electrons remain in a fixed state. Then, as is expected, the correlation function assumes the form

$$W = \sum_{\nu=0}^{\nu_{max}} Q_\nu A_\nu P \ (\cos\theta) \qquad (12)$$

where Q_ν are the attenuation coefficients. Of course, $Q_0 = 1$ and $0 < Q \leqslant 1$. They depend only on parameters describing the physical situation in the intermediate state. Thus, for magnetic interactions Q_ν depends on τ, j, μ, ν, and on H_{eff} where the latter is the effective magnetic field produced by the environment. This latter, unfortunately, is not known but an empirical

set of attenuation coefficients may be determined from comparison of (12) with the data and the ν-dependence of these compared with what is expected.

It is of interest to note that the same attenuation factors Q_ν apply whatever the nature of the radiations and whatever the nature of the observations made on the radiations. Thus, in the polarization-direction correlations the A_ν are replaced by $Q_\nu A_\nu$ as above. It can be shown quite easily that, to a large extent, conclusions concerning parity changes as deduced from polarization correlation measurements are independent of the precise form of the coefficients A_ν. Thus, if the experimental coefficients are used here spin-coupling is automatically taken into account in a correct manner. This fact that the attenuation is independent of the nature of the radiations or observations made is understandable since, for given spin-coupling, it does not matter how the intermediate state is reached.

As to the second possible course, two procedures to remove spin-coupling effects have been developed. The first involves the use of thin evaporated or electrodeposited metallic films. This method has been more or less outmoded by the second, which involves the use of dilute aqueous solutions. However, since some interesting solid state phenomena are involved in the thin film method, a few words regarding it may be in order. The method was developed and used mainly by the Swiss group of experimenters (*20*). By using an In^{111} source (decaying to Cd^{111}) the In^{111} ions were embedded in a thin metallic film by evaporation of both the In and the metal from two sources. Oddly enough it was found that most metallic environments did not give the maximum anisotropy, corresponding to no spin coupling, but only Ag worked well. The results depended on the film thickness (of order 10^3–10^4 A), the nature of the backing, temperature, and rate of evaporation or electrodeposition. For the most part the pertinent properties of such films have not been studied. The angular correlation is very sensitive to the various properties listed above, and the results could presumably shed some light on the structure of such thin layers.

With regard to the dilute solution sources (*21–23*) it is clear that one once again is confronted with a physical problem of which little is known. The evidence that dry sources of a large variety of compounds give a diminished anisotropy and that dilute solutions of these same sources uniformly exhibit a maximum correlation is now quite convincing (*21, 22*).

In a liquid one would expect a fairly homogeneous environment. Due to the averaging effect of the Brownian motion there should be only small magnetic fields and inhomogeneous electric fields, if any at all. That residual small quadrupolar couplings may exist seems to be rather plausible, and the possibility of studying these, using angular correlations measurements as a tool, has recently opened up.

It should be noticed that the use of dilute solutions would make correlation measurements with charged particles (conversion electrons and β-particles) impossible. The amount of liquid is necessarily such that a large amount of scattering will occur. On the other hand, by using a dry thin source for which the scattering is such that corrections can easily be made, one can eliminate attenuation effects by comparing two cascades $\gamma-\gamma$ and γ-conversion electron. The intermediate state is the same in these two cases so that the Q cancel out in the ratios of experimental coefficients. However, this procedure cannot apply to β-particles. It is also clear that correlation measurements in which the β-particle is emitted as a second radiation are not feasible. This follows since β-emitting states are long-lived and all types of weak spin-coupling would then play a role.

In addition to scattering corrections for electrons or positrons, corrections for the finite solid angle subtended by the detectors are also important. Both of these introduce attenuation (or "smearing") factors so that experimentally Q_ν may be the product of as many as three factors. Of course, for scattering the attenuation factors depend on the nature of the particle since they involve the scattering properties. The finite solid angle corrections are geometrical and in addition depend on detector efficiency as a function of position in the detector.

In summary, it may be said that the study of angular correlation processes has already been demonstrated to be one of the more valuable tools in the attack on the problems of nuclear structure. This is based in no small measure on the circumstance that the theory rests on as firm a foundation as any modern development in physics. For the rest, the utilization of this tool has been made possible by the very gratifying development in detecting techniques which has taken place in recent years.

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