polarization state can be constructed by superposition from these basic states. (For a detailed description of this multipole expansion see for instance Chapter XV, or section 27 of ref. 33.)

The vector potential of a circularly polarized plane wave propagating parallel to the z-axis shall be denoted by $A_{\sigma}(0)$, where $\sigma=+1$ indicates right circular polarization, $\sigma=-1$ left circular polarization. A right (left) circularly polarized plane wave corresponds to photons whose projection σ of the intrinsic spin on the propagation direction (the z-axis here) is $\sigma=+1(-1)$. The absence of the state $\sigma=0$ is classically due to the transverse nature of the electromagnetic radiation. Quantum-mechanically, it comes from the fact that the photon has rest mass zero 41,55-57,94,95.

The vector potential $A_{\sigma}(0)$ can be expanded into multipole fields:

$$A_{\sigma}(0) = \sqrt{2\pi} \sum_{L=1}^{\infty} (2L+1)^{\frac{1}{2}} \left\{ A_{L}^{\sigma}(m) + \sigma A_{L}^{\sigma}(e) \right\}.$$
 (85)

Here, $A_L^{\sigma}(m)$ is the vector potential of a magnetic multipole of order L; it is an eigenfunction of the operator of the square of the total angular momentum, z-component of angular momentum, and parity, with eigenvalues $L(L+1)h^2$, σh , and $(-1)^L$. Similarly, $A_L^{\sigma}(e)$ is the vector potential of an electric 2^L -pole field; its parity is $(-1)^{L+1}$.

We now establish the connection between the vector potentials and the photon wavefunctions by identifying

$$\langle 0\sigma | \rangle = A_{\sigma}(0),$$

$$\langle LM\pi | \rangle_{\text{magnetic}} = \sqrt{4\pi} A_L^M(m), \quad \pi = (-1)^L,$$

$$\langle LM\pi | \rangle_{\text{electric}} = \sqrt{4\pi} A_L^M(e), \quad \pi = (-1)^{L+1}.$$
(86)

The constant $\sqrt{4\pi}$ determines the normalization⁽⁷⁹⁾ of $\langle 0\sigma | LM\pi \rangle$. Inserting the expressions (86) into (85) and comparing the result with the expansion (84) for k=0 gives

$$\langle 0\sigma|LM\pi\rangle_{\text{magnetic}} = 2^{-\frac{1}{2}}(2L+1)^{\frac{1}{2}}\delta_{\sigma M}, \qquad \pi = (-1)^{L},$$

$$\langle 0\sigma|LM\pi\rangle_{\text{electric}} = 2^{-\frac{1}{2}}(2L+1)^{\frac{1}{2}}\delta_{\sigma M}\sigma, \qquad \pi = (-1)^{L+1}.$$
(87)

The radiation parameters $c_{k\tau}(LL')$ follow after inserting the coefficients (87) into (63). The δ functions restrict the sum over M and M' in eq. (63) to two terms, M = M' = 1 and M = M' = -1. In particular we find for directional correlation ($\tau = 0$) and for pure electric or pure magnetic radiation, using eq. (18)

$$c_{k0}(LL) = (-1)^{L-1} \frac{1}{2} (2L+1)(2k+1)^{\frac{1}{2}} {L \choose 1-1} {L \choose 0} [1+(-1)^{2L+k}].$$

Since 2L is even, the radiation parameter vanishes unless k is even also. For even k, we thus find

$$c_{k0}(LL) = (-1)^{L-1} (2L+1)(2k+1)^{\frac{1}{2}} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix}.$$
 (88)

For mixed multipole transitions, the parity selection rules expressed in eq. (87) demand that L+L' be odd if we have a mixture of an electric and a magnetic multipole, and even if the mixture consists of like multipoles. In both cases, the radiation parameter vanishes if k is odd. This argument verifies the statement made in subsection 3.7 that only even k appear. The radiation parameters for mixed multipoles then are

$$c_{k0}(LL') = (-1)^{L-1} (2L+1)^{\frac{1}{2}} (2L'+1)^{\frac{1}{2}} (2k+1)^{\frac{1}{2}} {L \choose 1-1} {L \choose 0}.$$
 (89)

The radiation parameters (88) and (89) apply to directional correlations, i.e., to the situation where only the directions of the radiations are determined. It is straightforward to write down the radiation parameters for circular polarization characterized by a unique value of the quantum number σ , and for linear polarization.

The radiation parameters for circularly polarized and for unpolarized γ -rays are independent of the parity quantum numbers π . A γ - γ directional or circular polarization measurement hence does not give any information about the parity of nuclear levels. In order to obtain the relative parities, the linear polarization must be determined.

The fact that the directional correlation function $W(\theta)$ does not depend on the parities of the nuclear states can be understood classically. Electric and magnetic radiations of the same multipole order L are related by the transformation $E \rightarrow H$, $H \rightarrow -E$. This transformation leaves the Poynting vector and therefore the angular distribution of the radiation unaltered. It is thus impossible to distinguish between electric and magnetic radiation by means of a directional correlation measurement.

4.2. GAMMA-GAMMA DIRECTIONAL CORRELATION. THEORETICAL RESULTS

4.2.1. We consider first a $\gamma - \gamma$ cascade $I_1 \rightarrow I \rightarrow I_f$ in which both γ -rays, of multipole order L_1 and L_2 , respectively, are pure. According to eq. (71), the most convenient form of describing the directional correlation $W(\theta)$ between γ_1 and γ_2 is Legacite polynemials

$$W(\theta) = 1 + A_{22} P_2(\cos \theta) + \dots + A_{k_{\max}k_{\max}} P_{k_{\max}}(\cos \theta).$$
 (90)

The highest term in this expansion is determined by the selection rules (70):

$$k_{\text{max}} = \text{Min}(2l, 2L_1, 2L_2).$$
 (91)

The explicit calculation of the coefficients A_{kk} is facilitated by the fact that they can be broken up into two similar factors, each factor depending on only one transition of the cascade. This factorization is expressed by eq. (72) and the unnormalized factors are given by the eq. (73). The normalized form (90) is easily obtained from eqs. (72)

⁹⁴ M. Fierz, Helv. Phys. Acta 13 (1940) 95.

⁹⁵ E. Wigner, Rev. Mod. Phys. 29 (1957) 255.

and (73) by dividing each coefficient A'_{kk} by A'_{00} . Introducing coefficients F (not to be confused with the F^M_L of section 2), we write

$$A_{kk}(\text{pure}) = F_k(L_1 L_1 I_1 I) F_k(L_2 L_2 I_1 I),$$
 (92)

where

$$F_k(LL\,I_1I) = A'_k(LL\,I_1I)/A'_0(LL\,I_1I). \tag{93}$$

An explicit expression for the normalized coefficients $F_k(LLI_iI)$ is obtained from eqs. (73) and (93) after inserting the radiation parameters (88) and using the following special value for the 6-*i* symbol ⁴⁸:

$$\begin{cases}
j_1 j_2 j_3 \\
0 j_3 j_2
\end{cases} = (-1)^{j_1 + j_2 + j_3} [(2j_2 + 1)(2j_3 + 1)]^{-\frac{1}{2}}.$$
(94)

This procedure yields

$$F_k(LL I_1 I) = (-1)^{I_1 + I - 1} (2L + 1)(2I + 1)^{\frac{1}{2}} (2k + 1)^{\frac{1}{2}} \begin{pmatrix} L & k \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} L L k \\ I & I_1 \end{Bmatrix}.$$
(95)

Numerical values and formulas for the 3-j and 6-j coefficients, or the equivalent Clebsch-Gordan and Racah coefficients, are available (see Table 1) and hence the coefficients F_k can be calculated for any desired case. Numerical tables of the F-coefficients have been computed by Biedenharn and Rose²⁵, and by Ferentz and Rosenzweig⁸⁶ for even values of k. Some of these tables are reprinted in ref. 36, 97.

F-coefficients for odd values of k are used in calculations involving parity effects (see Chapters XXIVD-XXIVG). Such coefficients have been calculated by Alder et al. 98; they are also reprinted in ref. 97. Tables of frequently used F-coefficients are given in Appendix B of this chapter and Appendix 7.

As an example of the use of the F-coefficients, we calculate the coefficients A_2 and A_4 for the cascade 4(2)2(2)0:

$$F_0 = 1$$
, $F_0 = 1$, $A_{00} = 1$, $F_2(2242) = -0.1707$, $F_2(2202) = -0.5976$, $A_{22} = 0.1020$, $F_4(2242) = -0.0085$, $F_4(2202) = -1.069$, $A_{44} = 0.0091$.

4.2.2. The directional correlation for double cascades in which one of the two γ -rays is mixed, i.e. must be described by more than one value L of the angular momentum, was first treated by Ling and Falkoff¹⁵. Their calculations were restricted to dipole and quadrupole radiation. The further development of the theory yielded formulas that are easier to evaluate.

We consider a $\gamma-\gamma$ cascade $I_i \rightarrow I \rightarrow I_f$ in which two multipole components L_n and L'_n

contribute to each of the two γ -transitions. The restriction to two multipole components is no limitation in practice, since the contributions of components of higher order than the two lowest ones permissible by angular momentum and parity selection rules are negligible. Only two types of mixed γ -transitions have so far been observed experimentally, the rather frequent M1+E2 and the rare E1+M2 transitions.

We define the (amplitude) mixing ratio of the transition 1 as the ratio of the reduced matrix elements $\delta_1(\gamma) \equiv \langle I \| L_1' \pi_1' \| I_1 \rangle / \langle I \| L_1 \pi_1 \| I_1 \rangle$ (96)

and similarly for the second transition. The ratio of the total (i.e. angle-integrated) intensity of the L'-pole to that of the L-pole is then equal to δ^2 . (This can be seen from eq. (56) by using the orthogonality relations of the Clebsch-Gordan coefficients.) The reduced matrix elements for γ -emission can always be chosen to be real, as indicated in eq. (58); thus the mixing ratio δ is real.⁸¹.

For a given intensity ratio δ^2 , the mixing ratio δ can have either a positive or negative sign, depending on the relative phase of the reduced matrix elements. However, the sign depends on the *definition* of the reduced matrix elements. This fact must be kept in mind when one compares the sign of δ as determined from a correlation experiment with that calculated on the basis of a certain nuclear model. (See the discussion by Biedenharn and Rose, ref. 25, section II-C, D and F; and refs. 99, 100.)

To write the directional correlation for mixed transitions in a convenient form, we generalize the *F*-coefficients (95) to read

$$F_k(LL'I_iI) = (-1)^{I_i+I-1} \left[(2L+1)(2L+1)(2I+1)(2I+1)(2k+1) \right]^{\frac{1}{2}} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} LL'k \\ I & I_i \end{Bmatrix}. (96)$$

These coefficients satisfy

$$F_0(LL'I_iI) = \delta_{IJ'}. (97)$$

The normalized coefficients A_{kk} in the expansion then can be written as

$$A_{kk} = A_k (L_1 L_1' I_1 I) A_k (L_2 L_2' I_1 I), \tag{98}$$

and one obtains easily from the eqs. (73), (88), (96) and (97)

$$A_{k}(L_{1}L'_{1}I_{1}I) = \frac{F_{k}(L_{1}L_{1}I_{1}I) + 2\delta_{1}(\gamma)F_{k}(L_{1}L'_{1}I_{1}I) + \delta_{1}^{2}(\gamma)F_{k}(L'_{1}L'_{1}I_{1}I)}{1 + \delta_{1}^{2}(\gamma)}$$
(99)

and a similar expression for $A_k(L_2L'_2I_lI)$. (See also ref. 101.)

The coefficients $F_k(LL'I_1I)$ are tabulated in Appendix 7 and in refs. 96, 97; related coefficients G_k are given in refs. 25, 36.

4.2.3. The coefficients A_{kk} (98) of the directional correlation function (90) depend in general on nine quantum numbers, namely I_1 , I_2 , I_3 , I_4 , I_5 , I_6 , I_8 ,

⁹⁶ H. Ferentz and N. Rosenzweig, Argonne National Laboratory Report 5324 (1954).

⁹⁷ A. H. Wapstra, G. J. Nijgh and R. van Lieshout, Nuclear Spectroscopy Tables (North-Holland Publ. Co., Amsterdam, 1959).

⁹⁸ K. Alder, B. Stech and A. Winther, Phys. Rev. 107 (1957) 728.

⁹⁰ G. R. Satchler, Proc. Phys. Soc. A67 (1954) 1024.

¹⁰⁰ R. Huby, Proc. Phys. Soc. A67 (1954) 1103.

¹⁰¹ M. E. Rose, Phys. Rev. 93 (1954) 477.

can safely assume that L'=L+1 and hence is left with seven quantities to be determined from experiment. The experimental investigation of a $\gamma-\gamma$ directional correlation, however, will at best yield the two constants A_{22} and A_{44} . (Higher terms have not yet been observed: the energies that normally are available in radioactive decays are such that multipoles with L>2 possess a half life $>10^{-6}$ sec. It is experimentally difficult or even impossible to measure a directional correlation function of a cascade involving an intermediate state with so long a half-life.) Thus the directional correlation measurement alone never allows a determination of all nuclear parameters involved. Only in conjunction with other methods is it possible to establish a complete decay scheme.

4.3. GAMMA-GAMMA DIRECTIONAL CORRELATION. EXPERIMENTAL TECHNIQUES AND RESULTS

In the present subsection, we discuss some typical experiments, indicate the various ways in which the experimental technique has progressed since the early experiments, and outline methods used to evaluate directional correlation measurements.

4.3.1. As our first example, we choose an even-even nucleus*, Ni⁶⁰ (Fig. 5). Even-even nuclei are especially well suited for angular correlation measurements. They presumably have spin 0 in their ground state and the second transition $I(\gamma_2)$ 0 must therefore be pure. Only the first γ -ray can be mixed and this reduces considerably the number of possible interpretations of a measured correlation.

The directional correlation of Ni⁶⁰ was first investigated successfully by Brady and Deutsch^{3,4,5}. On the basis of their correlation data and of earlier measurements ¹⁰², they concluded that the $\gamma-\gamma$ cascade probably occurs between spins 4, 2, and 0. An

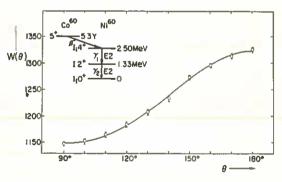


Fig. 5. Decay of Co⁶⁰ and directional correlation of the Ni⁶⁰ $\gamma - \gamma$ cascade (Steffen ³⁷). The solid line is the theoretical function $W(\theta)$ for the cascade 4(2)2(2)0

accurate determination of the conversion coefficients 103 confirmed their conclusions. Since the first report by Brady and Deutsch, a large number of publications contain data on Ni 60 , and this cascade has become a 'standard cascade' for directional correlation work. We have already calculated the coefficients A_2 and A_4 for the cascade 4(2)2(2)0 in 4.2.1. Using those values, we get for the correlation function $W(\theta) = 1 + 0.1020P_2(\cos\theta) + 0.0091P_4(\cos\theta)$. The main features of the decay $Co^{60} \rightarrow Ni^{60}$ and the directional correlation function measured by Steffen 37 are shown in Fig. 5. The theoretical curve and the experimental values agree very well.

4.3.2. For some years after the pioneer work of Brady and Deutsch, only nuclei emitting two γ -rays in cascade could be investigated with a fair degree of accuracy. The extension of directional correlation measurements to cascades of more than two γ -rays failed because one could not unambiguously single out the desired events. Today, most of the directional correlation work is performed by using scintillation spectrometers as counters and it is thus possible to attack even very complicated decays.

The investigation of the triple cascades in Ba^{134} and Ce^{140} by Robinson and Madansky¹⁰⁴ constitutes one of the first successful applications of scintillation spectroscopy to directional correlation. In addition to selecting the γ -ray energies, they used a third counter with a very large solid angle to detect β -particles in (triple) coincidence with the γ -rays. This method – accepting only γ -ray coincidences that are in coincidence with a β -branch of a given energy – may be very useful for correlation work.

The work of Klema and McGowan ¹⁰⁶ on Pd¹⁰⁶ (cf. Fig. 6) well exemplifies the highly refined technique of directional correlation measurements. The directional correlation of Pd¹⁰⁶ was a long-standing puzzle. Brady and Deutsch⁵ first measured the correlation of the 0.624 MeV-0.513 MeV cascade and found that the form of $W(\theta)$ agreed with that expected for a 0-2-0 cascade, but that the coefficients A_{22} and A_{44} were only about half as large as theory predicted. Many explanations were advanced, but none proved to be completely satisfactory. Klema and McGowan finally found the solution. The γ -spectrum of Pd¹⁰⁶ as seen by their detectors is given in Fig. 7. By measuring $W(\theta)$ for each of the energy ranges indicated by A, B, ... in Fig. 7 and by suitably combining the resulting data, Klema and McGowan obtained the following coefficients A_{22} and A_{44} :

0.624 MeV-0.513 MeV cascade: $A_{22} = 0.346 \pm 0.008$, $A_{44} = 1.109 \pm 0.012$. 1.045 MeV-0.513 MeV cascade: $A'_{22} = 0.092 \pm 0.003$, $A'_{44} = 0.035 \pm 0.009$.

The interpretation of these coefficients is best made by using the spin assignments that were deduced by Alburger 105 from measurements of the β -spectrum of Rh¹⁰⁶ and

^{*} We denote $\gamma - \gamma$ cascades by the nuclide in which the cascade takes place, e.g. Ni⁶⁰ and not Co⁶⁰. $\beta - \gamma$ and $\alpha - \gamma$ cascades are named after the parent nucleus.

¹⁰² M. Deutsch, L. G. Elliott and A. Roberts, Phys. Rev. 68 (1945) 193.

¹⁰³ M. A. Waggoner, M. L. Moon and A. Roberts, Phys. Rev. 80 (1950) 420.

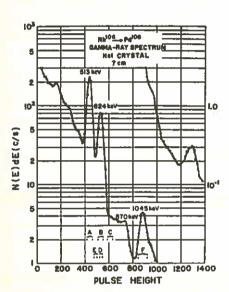
¹⁰⁴ B. L. Robinson and L. Madansky, Phys. Rev. 84 (1951) 604, 1067.

¹⁰⁵ D. E. Alburger, Phys. Rev. 88 (1952) 339.

¹⁰⁶ E. D. Klema and F. K. McGowan, Phys. Rev. 92 (1953) 1469.

from conversion data. These spin assignments are shown in Fig. 6. The 0.624 MeV-0.513 MeV cascade occurs between spins $I_1 = 0$, I = 2, $I_r = 0$. If this spin assignment is correct, then both γ -rays must be pure quadrupoles, and the theoretical coefficients are $A_{22} = 0.3571$, $A_{44} = 1.143$. The agreement between these coefficients and the ones determined by Klema and McGowan is good, and it confirms the assumed spin assign-

Fig. 6 (right). The decay Rh¹⁰⁶→Pd¹⁰⁶ (Alburger¹⁰⁵)



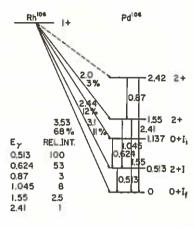


Fig. 7. Gamma-spectrum of Rh¹⁰⁶ Pd¹⁰⁶, measured by Klema and McGowan¹⁰⁶. The limits indicated by A, B, C, etc., show the positions of the windows (i.e., the energy ranges) of the two scintillation spectrometers used as detectors. One counter was fixed at position A, the other counter was set at the various other positions and the directional correlation function was determined for each setting

ment. Moreover, the old puzzle is solved by this measurement: in the earlier work the true correlation was masked by competing cascades.

The measured coefficients for the 1.045–0.513 MeV cascade exclude pure dipole radiation, because $A'_{44} \neq 0$. However, the theoretical coefficients for a cascade 2(2)2(2)0, $A'_{22} = -0.0766$ and $A'_{44} = 0.3265$, are in clear disagreement with the experiment alvalues given above. As long as one assumes that both γ -rays are pure multipoles, no spin assignment fits the experimental data. One is thus forced to assume that the 1.045 MeV γ -ray between the two states with spin 2 must be a mixture of M1 and E2.

4.3.3. In the past few years, a number of cascades have been investigated in which at least one of the γ -rays was found to be mixed. As an example, we discuss the main γ - γ cascade in Cd¹¹¹.

The energy levels of Cd¹¹¹ have been very carefully investigated and an unambigu-

ous spin assignment (Fig. 8) can be constructed without the help of directional correlation data. The main $\gamma-\gamma$ cascade observed in the decay of \ln^{111} occurs between states $I_1 = \frac{7}{2}$, $I = \frac{5}{2}$ and $I_f = \frac{1}{2}$. The measured conversion coefficients of γ_1 and γ_2 indicate that γ_1 is an M1, γ_2 an E2 radiation and that both γ -rays involve less than 15% admixture of higher multipoles. From the tables of F-coefficients, we find the coefficient A_{22} for the cascade $\frac{7}{2}(1) \frac{5}{2}(2) \frac{1}{2}$: $A_{22} = -0.0714$; the anisotropy

$$A \equiv \frac{W(180^{\circ}) - W(90^{\circ})}{W(90^{\circ})},\tag{100}$$

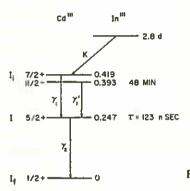


Fig. 8. The decay In 111 → Cd 111

then becomes A = -0.1034. The first experimental determination of the anisotropy ¹⁰⁷ yielded $A_{\text{exp}} = -0.07 \pm 0.04$ and thus seemed to confirm this decay scheme.

However, the investigation 8 of the influence of extranuclear fields on the directional correlation of Cd^{111} revealed the surprising result that the magnitude of the anisotropy could be made considerably larger than the theoretical value 0.103 by embedding the radioactive atoms in a suitable environment. In some sources, anisotropies as large as $A = -0.18 \pm 0.01$ were found. This value was in marked disagreement with the theoretical predictions and necessitated a reinvestigation of the Cd^{111} decay 108 .

We repeat here briefly the discussion 108 of the Cd¹¹¹ decay, but use the value 37 $A=-0.245\pm0.015$ for the 'unperturbed correlation'. The spin and parity assignments are unambiguous and one must thus assume that at least one of the γ -rays is mixed. An admixture of M3 to E2 in γ_2 is very unlikely (see Chapters XXIII and XV). Much more probably is the mixing of E2 and M1 in γ_1 . We thus assume that γ_1 consists of a mixture of E2 and M1, with an intensity ratio $\delta^2 = \text{Int}(E2)/\text{Int}(M1)$. Using eqs. (96)-(99) and the tables of F, we find for the coefficients A_{22} and A_{44} as function of the mixing ratio δ :

$$A_{22} = -(0.0714 + 0.7424 \delta + 0.1734 \delta^2)/(1 + \delta^2), \qquad A_{44} = -0.0726 \delta^2/(1 + \delta^2).$$

¹⁰⁷ F. Boehm and M. Walter, Helv. Phys. Acta 22 (1949) 378.

¹⁰⁸ H. Aeppli, H. Frauenfelder and M. Walter, Helv. Phys. Acta 24 (1951) 335.

It is interesting to note that the conserved vector current theory predicts the ratio of (a)/(ip) 300, 301;

$$\frac{\langle \alpha \rangle}{\langle i \hat{r} \rangle} = W_0 R - \Delta M R + \frac{7}{6} \alpha Z,$$

where ΔM is the neutron-proton mass difference in units mc^2 . The predicted value of this ratio for the Eu¹⁵² β ⁻-transition is + 0.55, which agrees with the experimental value $0.57^{+1.1}_{-0.26}$

7.5.5. The modified Bii-approximation

It is sometimes convenient to analyze β - γ angular correlation experiments on the basis of a somewhat artificial approximation that was first suggested by Matumoto et al. 302. This 'modified B_{ij} -approximation' presupposes that the $\langle iB_{ij} \rangle$ component contributes strongly to a nonunique first-forbidden β -transition, but that the vector and scalar type nuclear matrix elements contribute only through their presence in the linear combinations V'_0 and Y'_1 . In terms of the parameters defined in eqs. (172), this corresponds to:

$$z_2 \neq 0$$
, $Y_1' \neq 0$, $V_0' \neq 0$, $x_1 = u_1 = w_0 = 0$. (194)

This approximation is useful for an estimate of the relative contribution of the $\langle iB_{ij} \rangle$ component to a nonunique β -transition, because it leads to rather simple expressions for the factors S and $A_k(\beta)$:

$$S_B = V_0^{\prime 2} + Y_1^{\prime 2} + \frac{1}{12} z_2^2 (q^2 + \lambda_1 p^2), \tag{195}$$

$$A_{2}(\beta)_{B} = S_{B}^{-1} \left\{ -F_{2}(02I_{1}I)\sqrt{\frac{2}{75}}\lambda_{2}V_{0}'z_{2} + F_{2}(12I_{1}I)\sqrt{\frac{2}{15}}\lambda_{2}Y_{1}'z_{2} + F_{2}(22I_{1}I)\frac{7}{60}z_{2}^{2}\lambda_{1}W\right\} p^{2}W^{-1},$$
(196)

$$A_{1}(\beta)_{B} = \pm S_{B}^{-1} \left\{ \frac{2}{3} F_{1}(01 I_{1} I) V_{0}' Y_{1}' + \frac{2}{3} F_{1}(11 I_{1} I) Y_{1}'^{2} + \sqrt{\frac{10}{27}} F_{1}(12 I_{1} I) \lambda_{4} Y_{1}' z_{2} W + F_{1}(22 I_{1} I) \left(\frac{1}{12} q^{2} + \frac{1}{20} \lambda_{1} p^{2}\right) z_{2}^{2} \right\} p W^{-1},$$
 (197)

$$A_3(\beta)_B = \pm S_B^{-1} \frac{1}{20} F_3(22 \, l_1 l) \, z_2^2 \lambda_1 \, p^3 W^{-1} \,. \tag{198}$$

This procedure allows one to estimate the relative contribution of the tensor type component $\langle iB_{II} \rangle$ to a nonunique first-forbidden β -transition. This method has been applied to a number of β -transitions in connection with predictions of nuclear models to obtain information on the structure of nuclear states 303, 304.

PART II. THE INFLUENCE OF EXTRANUCLEAR FIELDS

§ 8. Survey

8.1. The theory of extranuclear perturbations on angular correlations has been developed during the past 18 years to a high degree of completeness. The possibility of extranuclear perturbations was already mentioned by Hamilton in his pioneer paper¹. The first detailed study is due to Goertzel² who investigated the hyperfinestructure interaction and externally applied magnetic fields. The theory was extended by Alder 22 who presented expressions that could easily be applied to actual experimental situations. A thorough treatment of the magnetic dipole and the electric quadrupole interaction, including time dependent effects has been presented by Abragam and Pound 26. The theory of an axially symmetric quadrupole interaction in a single-crystal source has been developed by Alder et al.27. Extensive calculations on the influence of the combined magnetic dipole and electric quadrupole interaction have been done by Alder et al.30. Nonaxial electric field gradients have been considered by Matthias et al. 305. The consistent use of density matrices and tensor formalism in extranuclear perturbation calculations has been introduced by Coester 28. An elementary discussion of the theory on the basis of the vector model can be found in a review by Steffen 37, which also gives a survey of the experimental results. A complete discussion of extranuclear perturbations can be found in ref. 37a. These papers constitute an adequate basis for the understanding and the quantitative evaluation of most of the experimentally observed facts. A survey of the experimental determination of g-factors of excited states has been prepared by Bodenstedt 38. A paper by Alder and Steffen 306 reviews the theory and experiments on electromagnetic moments of excited nuclear states. Applications to problems of solid and liquid state physics are discussed by Heer and Novey 11.

8.2. Many of the considerations discussed here are not restricted to cascade decays, but apply also to radiations from oriented nuclei 307 and to nuclear reactions. Among nuclear reactions, Coulomb excitation is used for investigations along the lines discussed here. Excitation experiments using external magnetic fields provide information about the magnetic dipole moments of excited states of nuclei 308, or about the undisturbed directional distribution of the de-excitation γ -rays. The use of single-crystal films may provide information about electric quadrupole moments. Of particular interest is the 'reorientation effect' in Coulomb excitation 309. This effect is caused by the interaction of the electric quadrupole moment in the excited state with the field gradient produced by the hyperbolic orbit of the exciting ion. The fact that the field

³⁰⁰ J. I. Fujita, Phys. Rev. 126 (1962) 202.

³⁰¹ J. Eichler, Z. Physik, 171 (1963) 463.

³⁰² Z. Matumoto, M. Morita and M. Yamada, Bull. Kobayashi Inst. Phys. Research 5 (1955) 210 (in Japanese).

³⁰³ Z. Matumoto, M. Yamada, I. T. Wang and M. Morita, Phys. Rev. 129 (1963) 1308.

³⁰⁴ H. J. Fishbeck and R. W. Newsome, Phys. Rev. 129 (1963) 2231.

²⁰⁵ E. Matthias, W. Schneider and R. M. Steffen, Physics Letters 4 (1963) 41.

³⁰⁶ K. Alder and R. M. Steffen, Ann. Rev. of Nucl. Science 14 (1964), to be published.

³⁰⁷ N. R. Steenberg, Phys. Rev. 95 (1954) 982.

²⁰⁸ R. F. Christy, Phys. Rev. 94 (1954) 1077.

²⁰⁹ G. Breit, R. L. Gluckstern and E. J. Russell, Phys. Rev. 103 (1956) 727.

gradient can be exactly calculated in this case may lead to the accurate determination of electric quadrupole moments in excited nuclear states.

- 8.3. The angular correlation of a cascade $I_i \rightarrow I \rightarrow I_f$ will, in general, be altered as soon as the nuclei in their intermediate level I are subject to torques, due to the interaction of either the magnetic dipole moment μ with an extranuclear magnetic field B, or of the electric quadrupole moment Q with electric field gradients $\partial^2 V/\partial z^2$. In the semiclassical picture, these interactions produce a precession of the nuclei around the symmetry axis. The changing nuclear orientation results in an altered angular correlation. In quantum-mechanical language, if the quantization axis was chosen to coincide with the direction of the first radiation, the interactions cause transitions among the m-states. The second radiation is emitted from a level with an altered population distribution and this change is responsible for the attenuation of the correlation. In the language of density matrices, the transitions among the m-states are described by a time dependence in the density matrix of the intermediate state.
- 8.4. The semiclassical picture of a precessing nucleus adequately describes the situation if the extranuclear fields are static. However, there also exist time-dependent fields that produce a change in the correlation function. In this case, realized for example in certain viscous liquids, the fluctuating electric field gradients at the site of the nucleus induce transitions among the m-states.

One major difference between static and time-dependent interactions can be described as follows. Consider first a static interaction in an axially symmetric single crystal. The populations of the m-states quantized along the symmetry axis remain constant, hence the correlation is undisturbed if the symmetry axis points in the direction of one counter (i.e. coincides with the direction of emission of one radiation). If it points in any other direction, then the populations of the m-states quantized along the direction of emission change in time and this change causes an attenuation of the angular correlation. In a polycrystalline source, this description applies for each microcrystal and ultimately for each nucleus. The angular correlation in a poly-crystalline source is never wiped out completely because a certain fraction of nuclei experience the static field in such a direction that their correlation is unperturbed or only slightly attenuated. Static interactions in polycrystalline sources thus never reduce the correlation below the 'hard core' value. Time-dependent interactions, however, can wipe out the correlation completely, because the direction of the field at each nucleus changes continuously in a random manner. No quantization axis exists for which the populations of the m-states remain constant. Eventually, all m-states are equally populated for any choice of the quantization axis and the directional correlation becomes isotropic.

8.5. When will a particular cascade be influenced by an extranuclear field? It is clear that the perturbation depends primarily on the magnitude of the interaction and on the length of time it can act, i.e., on the mean life τ of the intermediate level I.

For static perturbations, the magnitude can be described by a precession frequency ω .

For magnetic interactions, ω is equal to the Larmor frequency ω_B , i.e., proportional to μ and H. In the quadrupole case, ω is proportional to Q and $\partial^2 V/\partial z^2$. Time-dependent perturbations can be characterized by an inverse time constant λ : the m-states approach a uniform population exponentially and λ is the corresponding relaxation constant.

The angular correlation of a cascade is perturbed if at least one of the following crude criteria holds: $\omega \tau \approx 1$, $\lambda \tau \approx 1$. A more refined criterion depends on the sensitivity of the experimental arrangement to alterations in the correlation function. A detailed discussion 37 leads to the conclusion that with the present experimental techniques the limits are given by $\omega \tau > 0.01$, $\lambda \tau > 0.01$.

For a large number of solids and liquids, precession frequencies have been measured by microwave and nuclear resonance techniques (see ref. 310, Appendix, Table A 6). For quadrupole interaction, values for $\omega/2\pi$ as large as 3000 MHz have been found. According to our criterion for perturbation ($\omega\tau > 0.01$), cascades with a lifetime $\tau > 10^{-12}$ sec can thus be measurably perturbed in extreme cases. The magnetic interaction can be extremely strong in free atoms 311. In solids, however, the orbital contribution to the magnetic moment of the electron shell is quenched (see, e.g., ref. 312) and the magnetic interaction can usually be neglected in comparison with the quadrupole interaction.

8.6. Here, we compare briefly some of the possibilities and difficulties of the angular correlation method with those of the microwave and nuclear resonance techniques.

The magnetic and electric interaction between the nucleus and its environment are well known from studies of the hyperfine structure of spectral lines, both by optical means311 and with microwaves310, and from nuclear resonance absorption work 312-315. These conventional methods have, beside the much higher accuracy and much broader range of applicability, two main advantages over angular correlation: (a) By choosing a nucleus with spin $\frac{1}{2}$, one can eliminate the quadrupole interactions and study the magnetic effects. This separation of the interactions is impossible in angular correlation work, for in order to observe an anisotropic correlation, the spin of the intermediate nuclear level must be at least one, and hence the nucleus can possess a quadrupole moment. (b) In conventional methods, the environment of the nucleus under investigation is in its ground state. In angular correlation work, the nuclear decay leading to the cascade (or the first radiation of the cascade itself) very often excites the atomic shell. The nuclear environment is thus in an unknown state. This

W. Gordy, W. V. Smith and R. F. Trambarulo, Microwave Spectroscopy (J. Wiley and Sons, New

³¹¹ H. Kopfermann, Nuclear Moments, (Academic Press, Inc., New York, 1958).

³¹² C. Kittel, Solid State Physics (J. Wiley and Sons, New York, 1953).

³¹³ A. Abragam, The Principles of Nuclear Magnetism (Oxford Univ. Press, London, 1961).

³¹⁴ N. Bloembergen, Nuclear Magnetic Relaxation; Reprint volume (W. A. Benjamin, New York,

³¹⁵ C. P. Slichter, Principles of Magnetic Resonance, with Examples from Solid State Physics (Harper and Row, New York, 1963).

situation is especially pronounced in a cascade preceded by a K-capture or an α-decay.

Fortunately, angular correlation offers some possibilities that compensate for these disadvantages. We have already mentioned the possibility of finding the g-factor and the quadrupole moment of short-lived excited nuclear levels. The internal fields in solids and liquids can be investigated even if they are so large that they wash out nuclear resonance lines. It is also possible to work with metal crystals without difficulties arising from the skin effect. Moreover, the changes in the atomic shell due to K-capture and α-decay can be studied.

- 8.7. It is clear that the *measured* correlation function for a given decay depends on the physical and chemical state of the radioactive source and on the time delay between the two channels of the coincidence circuit. Two different approaches are thus possible.
- (a) One can use a given source state and change the delay between the two channels. By measuring $W(\theta)$ as a function of the delay, one gets information about the time dependence of the perturbation. (The finite resolving time of the coincidence circuit causes a smearing-out of the time dependence, so one must choose a resolving time as short as possible.) This approach ('delayed coincidence') was first proposed by Lundby 316 and the corresponding theory was constructed by Lloyd 317. Fraser and Milton performed the first completely successful experiment 315.
- (b) The second, more widely-used method consists in keeping the time delay and the resolving time constant and varying the physical state of the source. This approach was first tried by Brady and Deutsch⁵ in the case of the Ni⁶⁰ γ - γ cascade. However, the lifetime τ_I of Ni⁶⁰ is so short that no influence is to be expected and none was found.

A positive effect was first found with the $\gamma-\gamma$ cascade of Cd¹¹¹ by the Zürich group^{8,318}. By embedding the radioactive nuclei in different media, anisotropies ranging from zero to -0.18 were found. Since this first unambiguous evidence for the influence of the nuclear environment on angular correlation, many experiments have been performed with the $\gamma-\gamma$ cascade of Cd¹¹¹ and the investigations have been extended to other nuclides also. Most of the experiments are in satisfactory agreement with theory and the main aspects of the interaction of the nucleus with its environment are well understood. However, there still remain some unsolved problems, mostly connected with the processes following a K-capture. Also, since no reliable way of determining the magnitude of the electric field gradient has been found, the magnitudes of the quadrupole moments as measured by angular correlation are uncertain.

§ 9. Theory of extranuclear perturbations

9.1. GENERAL DESCRIPTION

9.1.1. We start with the expression (46) for the unperturbed angular correlation

which we write in the following form:

$$W(k_{1}, k_{2}) = \sum_{\substack{m_{1}m_{n}m_{b} \\ m_{n}^{*}m_{b}^{*}m_{f}^{*}}} \langle m_{f}|H_{2}|m_{b}\rangle \langle m_{a}|H_{1}|m_{i}\rangle \delta_{m_{n}m_{b}} \times \times \langle m_{f}|H_{2}|m_{a}^{\prime}\rangle^{*} \langle m_{b}^{\prime}|H_{1}|m_{i}\rangle^{*} \delta_{m_{n}^{\prime}m_{b}^{\prime}},$$
(199)

where H_1 and H_2 represent the interaction between nucleus and radiation field only. In the absence of an extranuclear perturbation, the final states $\langle m_a |$ and $\langle m'_a |$, after emission of the first radiation, are identical with the initial states $|m_b\rangle$ and $|m'_b\rangle$ of the second radiation.

We assume now the presence of an interaction of the nucleus in its intermediate state I with some extranuclear fields. This interaction, which we describe by the Hamiltonian K, is assumed to act from the time the first radiation is emitted (t=0) until the time t at which the second radiation is emitted. During this time interval the states $|m_a\rangle$ change to different states $|m_b\rangle$ under the influence of the extranuclear perturbation. This change can be represented by an unitary operator $\Lambda(t)$ that describes the evolution of the state vectors $|m_b\rangle$, and the perturbed angular correlation can be expressed as:

$$W(k_1, k_2, t) = \sum_{\substack{m_1 m_1 \\ m_2 m_1 = t}} \langle m_f | H_2 \Lambda(t) | m_a \rangle \langle m_a | H_1 | m_1 \rangle \langle m_f | H_2 \Lambda(t) | m_a' \rangle^* \langle m_a' | H_1 | m_1 \rangle^*.$$
(200)

The states $|m\rangle$ form a complete set and the state vector $\Lambda(t)|m_a\rangle$ can be expressed as:

$$A(t)|m_{a}\rangle = \sum_{m_{b}} |m_{b}\rangle \langle m_{b}|A(t)|m_{a}\rangle, \qquad (201)$$

and similar for $\Lambda(t)|m'_a\rangle$. The expansion coefficients are the matrix elements of the time-evolution operator $\Lambda(t)$ in the representation m.

The time-evolution operator satisfies the Schrödinger equation:

$$\frac{\partial}{\partial t} \Lambda(t) = -\frac{\mathrm{i}}{\hbar} K \Lambda(t). \tag{202}$$

If K does not depend on the time t (static interaction) the solution of this equation is simply:

$$\Lambda(t) = \exp(-i Kt/h). \tag{203}$$

For a time-dependent interaction, the solution of (202) can be written symbolically

$$\Lambda(t) = \exp\left(-\frac{\mathrm{i}}{h} \int_{0}^{t} K(t') \,\mathrm{d}t'\right),\tag{204}$$

where the integral must be evaluated according to Feynman's rules for ordered operators 319.

319 R. P. Feynman, Phys. Rev. 84 (1951) 108.

³¹⁶ A. Lundby, Phys. Rev. 76 (1949) 993.

³¹⁷ S. P. Lloyd, Phys. Rev. 82 (1951) 277.

³¹⁸ H. Frauenfelder, Phys. Rev. 82 (1951) 549.

12.3.3. The situation is much more complicated if the decaying atom recoils in a solid or liquid. In addition to the I-J coupling and the shake-off effect discussed above, other processes occur that change the perturbation. The recoiling atom can pass through regions of very large field gradients; inelastic collisions may excite or ionize the electron shell; the time-dependent processes in the electron shell that occur after the α -decay may be drastically altered as discussed in subsection 12.1.1; the symmetry of the surroundings of the nucleus, after it has come to rest, may be different from those of the original site. In addition, the new sites will, in general, not be the same for all nuclei and a frequency distribution of the interacting fields is to be expected. The experiments indeed show that most of these effects are present. More work is required, however, to study all of these processes in detail.

PART III. EXPERIMENTAL METHODS

In various sections of Part I, particularly in subsection 4.3.6, we have briefly discussed some of the experimental arrangements used in angular correlation work. These arrangements are basically the same as those used in conventional coincidence work, with the exception that the angle ϑ , subtended at the source by the axes of the two counters, can be varied. We refer to the pertinent chapters for experimental details (counters, electronic equipment, source preparation, determination of the resolving time, stability checks, energy selection) and discuss here only those problems that are specific to angular correlation work.

§ 13. The determination of the true correlation

In several earlier sections, we mentioned source conditions under which one expects the angular correlation to be unperturbed. We summarize here these conditions.

- (i) Cubic metal crystals. The radioactive atoms must sit in regular lattice sites and the environment of each radioactive atom must possess cubic symmetry. Moreover, imperfections in the crystal should be avoided as much as possible. The recoil of the decaying atom can displace it from a regular lattice site ($\alpha-\gamma$ correlations thus are attenuated even in cubic crystals).
- (ii) Noncubic single crystals. Such crystals yield the unperturbed correlation if the radioactive atoms sit in regular lattice positions and the symmetry axis of the crystal points toward the fixed counter (Figs. 63, 64).
- (iii) Liquids. Liquids generally yield the unperturbed correlation if the correlation time is of the order of 10⁻¹¹ sec or smaller. Molten metals are often very suitable for determining the maximum correlation. The observation of the anisotropy as a function of the viscosity of a liquid and a fit of the resulting data with the theoretical curve may yield a good value for the unperturbed correlation (cf. Fig. 51).
- (iv) Delayed coincidences. The determination of the anisotropy as a function of the delay time allows an extrapolation to the unperturbed value (cf. Fig. 52).
- (v) Magnetic decoupling. Goertzel² suggested that a strong magnetic field, applied to the source in the direction of the fixed counter, should 'decouple' the nucleus from

the atomic field and yield the unperturbed correlation. However, this method works only in some special cases (Figs. 50, 71).

None of the methods (i)-(v) is guaranteed to yield the unperturbed correlation. It is thus advisable to use more than one method to obtain a reliable value for the true angular correlation function, if the lifetime of the intermediate state is longer than about 10^{-11} sec. Caution is especially necessary when one works with cascades following or involving K-capture, α -decay, and conversion electrons.

§ 14. The treatment of the experimental data

Three aspects are involved in the evaluation of an angular correlation experiment: Tests must establish that the equipment works properly; a procedure for getting reliable data has to be worked out; the raw data must be corrected for all deviations of the actual set-up from the ideal one.

14.1. EXPERIMENTAL TESTS

Tests form an essential part in any experiment. During the setting-up of the equipment, ideas for tests occur naturally. Here, we outline a number of tests that are useful. The list is far from complete and it is intended only as an example.

In a directional correlation experiment, one can perform the following tests:

- (i) For each channel, one determines stability, energy resolution, energy calibration, linearity of response, efficiency, and total delay with pulse generators and with radio isotopes of well-known radiation characteristics.
- (ii) The performance of the channels and the coincidence system as a function of source strength is observed. (Difficulties due to losses in the electronic system arise particularly when decays with small branching ratios are investigated.)
- (iii) The resolving time τ_0 of the coincidence circuit is investigated as a function of the source strength and the energy of the incident radiation (cf. subsection 9.1.3). τ_0 can be found either by measuring the coincidences from a 'prompt' cascade (i.e., a cascade with a lifetime of the intermediate state short compared to the resolving time) with a delay $\tau > \tau_0$ in one channel, or by using two independent sources and separating the counters.
- (iv) Scattering from one counter into the other, or from material close to source or counters, is checked.

Effects caused by scattering can mask the true correlation. If the radiation under investigation is scattered in the source, the correlation function is falsified in a manner treated in 14.3.3. Moreover, scattering of any radiation in or near the counters can give rise to spurious coincidences ¹⁷⁷. These in general are not isotropic and therefore simulate a correlation. One way of avoiding such spurious coincidences consists in shielding the counters frontally and laterally with lead. The necessary wall thicknesses for a given γ -ray energy can be found in ref. 32. If the lead shields are formed as cones that accept only γ -rays from a point source in the center, a slight maladjustment of the source changes the solid angle markedly. Thus the lead shields should be so constructed that the size of the source and slight decentering do not affect the solid

angle. A more refined protection against scattered γ -rays and spurious coincidences consists in using scintillation or solid state spectrometers as detectors and accepting only the photopeaks of the desired γ -rays (cf. Fig. 7).

The effectiveness of the protection against spurious coincidences can be checked by using a strong source and preventing its γ -rays from entering one counter directly by placing heavy shielding between the source and this counter. All measured coincidences are then spurious.

(v) The over-all performance of the directional correlation system is checked by measuring $W(\theta)$ for well-known cascades (Ni⁶⁰, Pd¹⁰⁶).

Most of these tests apply also to *polarization-direction correlations*. In addition, however, the asymmetry ratio R, defined in eq. (107) must be determined by one or more of the following experiments:

- (vi) R can be found by measuring the linear polarization of γ -rays that were scattered by an angle of about 90° 7, 150, 151.
- (vii) The polarization-direction correlation of a well-known cascade (Ni^{60} , Pd^{106}) fixes R.
- (viii) The linear polarization of γ -rays following Coulomb excitation in even-even nuclei is well known and can hence also be used to find R^{155} .
- (ix) The linear polarization of annihilation quanta yields a value for R at an energy of 0.51 MeV. This experiment, however, requires two polarimeters.

14.2. PROCEDURE

In order to outline the procedure, we discuss a $\gamma-\gamma$ directional correlation measurement. Other experiments involve obvious generalizations.

Before a run, and at intervals during runs, resolving time τ_0 and background counts $N_1^0(9)$, $C^0(9)$ are determined. N and C denote single counts and coincidences, respectively, per unit time. 9 denotes the angle subtended at the source by the counters (Fig. 1).

A source of proper strength is prepared. The maximum source strength is determined by the ratio of accidental to true coincidences. For accurate measurements, this ratio should be smaller than about 0.2; under unfavorable conditions, one may be forced to make it as high as unity.

The source is then 'centered' to about 1% or less; i.e. the counting rate $N_m(\vartheta)$ of the movable counter as a function of the angle ϑ should be constant to about 1% or less.

After the source is centered, the coincidences and the single counts are recorded for various angles. The angle should be changed frequently, e.g., every 10 or 20 min. For short-lived radioisotopes, more frequent changes are advisable. The choice of angles depends on the form of the correlation function. In general, it is best to use a few angles only (e.g. 90°, 120°, ... 270°) and to obtain at each angle a large number of coincidences. For extended experiments, automatic equipment for changing the angles and recording the data is very convenient.

From the measured data, one obtains the 'true' values $C^{t}(\vartheta)$ and $N_{i}^{t}(\vartheta)$ by subtracting background, accidental coincidences, and contributions due to disturbing

radiations; e.g.:

$$C^{t}(\vartheta) = C^{\text{meas}}(\vartheta) - C^{0}(\vartheta) - C^{\text{acc}}(\vartheta) - C^{d}(\vartheta),$$
 (408)

$$C^{\text{acc}}(\vartheta) = 2\tau_0 N_1^{\text{meas}}(\vartheta) N_2^{\text{meas}}(\vartheta). \tag{409}$$

The contribution $C^{d}(\vartheta)$, due to other coincident γ -rays present in the source, must be found in a separate measurement, with the energy selection adjusted in such a way that the various contributions can be singled out. In calculating the accidental coincidences (409), higher order corrections may have to be included $^{451-451b}$.

The true number of single counts and coincidences can now be written as

$$N_i^t(\vartheta) = M \, p_i \, \Omega_i \, \varepsilon_i \,, \tag{410}$$

$$C^{i}(9) = M p_1 p_2 \Omega_1 \varepsilon_1 \Omega_2 \varepsilon_2 \varepsilon_c K(9). \tag{411}$$

M is the number of nuclear disintegrations per unit time, p_i the probability per disintegration that the radiation selected in counter i is emitted, Ω_i the solid angle in units of 4π , ε_i the efficiency of channel i, and ε_e the efficiency of the coincidence circuit. K(9) denotes the directional correlation function, as measured with the equipment under discussion.

Comparison of eq. (411) with (409) shows that the accidental coincidences increase with the square of the source strength, the true coincidences only linearly. As mentioned above, an optimum source strength exists which can be calculated from eqs. (409)–(411). Decreasing the resolving time τ_0 permits the use of correspondingly stronger sources. The eqs. (409)–411) also show that the ratio of true to accidental coincidences is unaffected by the solid angle. Very large solid angles hence are often advantageous. Optimum solid angles are discussed by Devons and Goldfarb³⁵.

For further evaluation, the coincidence rate $C^{\dagger}(\vartheta)/N_1^{\dagger}(\vartheta)$ is formed. One then finds for the desired uncorrected experimental correlation function from the eqs. (410) and (411):

$$K(\vartheta) = \frac{1}{p_2 \Omega_2 \, \varepsilon_2 \, \varepsilon_c} \frac{C^{\iota}(\vartheta)}{N_1^{\iota}(\vartheta)}. \tag{412}$$

If one uses movable counters it is seen from this equation that one best divides through by the counting rate in the movable counter, i.e. that one identifies counter 1 with the movable one. The ratio $K(\vartheta)$ then becomes independent of the (slightly variable) solid angle of the movable counter. Furthermore, it is independent of the decay of the source, of the branching probability p_1 , and the efficiency of the movable counter. Small errors in the centering of the source are corrected in first order.

The numbers $K(\vartheta)$ are now fitted by

$$K(\theta) = K_0 \left\{ 1 + A_{22}^{\exp} P_2(\cos \theta) + A_{44}^{\exp} P_4(\cos \theta) + \dots \right\},\tag{413}$$

⁴⁵¹ H. Paul, Nucl. Instr. and Meth. 9 (1960) 131.

⁴⁵¹a R. E. Bell, Nuci. Instr. and Meth. 12 (1961) 199.

⁴⁵¹b E. B. Shera, K. J. Casper and B. L. Robinson, Nucl. Instr. and Meth. 24 (1963) 482.

using the method of least squares. All correlation functions measured to date can be fitted by using only the three terms written in eq. (413). Details concerning the determination of the coefficients $A_{kk}^{\rm exp}$ and their errors are given in refs. 452-454; examples are given in refs. 455, 456. [Sometimes, the constant K_0 in eq. (413) has been chosen equal to $K(90^{\circ})$. This choice lends infinite weight to the measurement at 90° and is clearly incorrect.]

14.3. CORRECTIONS

The coincidence rate $K(\theta)$ corresponds to the correlation function $W(\theta)$ only under the assumption of centered point sources and point detectors. In order to compare the experimental results with theoretical calculations, the coefficients A_{kk}^{\exp} must be corrected for the deviations from an ideal arrangement.

14.3.1. The first step consists in correcting the coefficients $A_{kk}^{\rm exp}$ for the finite solid angle of the counters. Using a method first developed by Frankel ¹⁷⁶, and neglecting the correction for the extension of the source, we write the corrected coefficients A_{kk} as ⁴⁶⁷

$$A_{kk} = A_{kk}^{\exp}/Q_{kk}. \tag{414}$$

In order to determine the solid angle correction factors Q_{kk} , we assume here that the efficiency ε of the detectors is a function only of the angle α between the counter axis and the direction of propagation of the incident particle, and of the energy E of the radiation. We call such a detector cylindrically symmetric or circular (most scintillation counters fall in this category). The correction formulas for arbitrary detectors have been worked out by Feingold and Frankel 458.

For a circular detector the correction factor Q_{kk} can be expressed as the product of two factors

$$Q_{kk} = Q_k(1) Q_k(2), (415)$$

where $Q_k(1)$ and $Q_k(2)$ are the correction factors for the individual detectors 1 and 2. These correction factors, normalized to $Q_0 = 1$, are given by

$$Q_k(i) = J_k(i)/J_0(i),$$
 (416)

with

$$J_k(i) = \int_0^{\frac{1}{2}\pi} \varepsilon_i(E, \alpha) P_k(\cos \alpha) \sin \alpha \, d\alpha. \tag{417}$$

The correction factors $Q_k(i)$ can be determined in various ways:

452 M. E. Rose, Phys. Rev. 91 (1953) 610.

453 E. Breitenberger, Proc. Phys. Soc. (London) A69 (1956) 489.

⁴⁵⁴ P. C. Price, Phil. Mag. (7) 45 (1954) 237; Proc. Cambridge Phil. Soc. 50 (1954) 491.

456 E. D. Klema and F. K. McGowan, Phys. Rev. 91 (1953) 616.

456 E. Breitenberger, Proc. Phys. Soc. (London) A69 (1956) 453.

457 J. S. Lawson and H. Frauenfelder, Phys. Rev. 91 (1953) 649.

458 A. M. Feingold and S. Frankel, Phys. Rev. 97 (1955) 1025.

(i) As an approximation one can assume the efficiency $\varepsilon_l(E,\alpha)$ to be independent of the angle α . This is, in general, a good approximation for thick β -scintillation detectors and for solid state detectors. For a circular detector of radius r at a distance h from the source, the correction factors then are given by

$$Q_{2} = \frac{1}{2}\cos\alpha_{0}(1 + \cos\alpha_{0}),$$

$$Q_{4} = \frac{1}{8}\cos\alpha_{0}(1 + \cos\alpha_{0})(7\cos^{2}\alpha_{0} - 3),$$

$$\alpha_{0} = \tan^{-1}(r/h).$$
(418)

(ii) The angular efficiencies $\varepsilon(E,\alpha)$ of the two detectors are determined experimentally by using a well-collimated γ -ray beam. A numerical integration then yields Q_k^{457} .

(iii) The correction factors can be calculated 452. For γ -rays, the efficiency $\varepsilon(E,\alpha)$ is well represented by $\varepsilon(E,\alpha) = 1 - \exp\{-\tau(E)X(\alpha)\}, \tag{419}$

where $\tau(E)$ is the total absorption coefficient in the crystal for γ -rays of energy E and $X(\alpha)$ is the distance traversed in the crystal by the radiation at an angle α with respect to the axis of the cylindrical detector. Using the eqs. (416), (417) and (419), the correction factors Q_k have been evaluated for various sizes of sodium iodide crystals at various distances h from the source $^{452,\,459,460}$. Figures 72 and 73 display the results of such calculations for some typical cases.

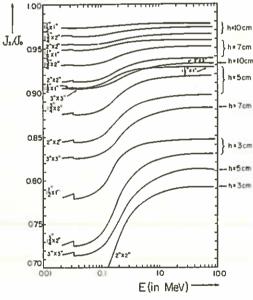


Fig. 72. Angular resolution correction factor $Q_2 = J_2/J_0$ for cylindrically symmetric NaI detectors of dimension $d \times r$ (d=thickness of NaI crystal, r=radius) at a distance h from the (point) source. (West ⁴⁶⁰)

⁴⁵⁹ A. L. Stanford and W. K. Rivers, Rev. Sci. Instr. 30 (1959) 719.

⁴⁰⁰ H. West, UCRL-Report 5451 (1959) unpublished, TID 4500 (1959).

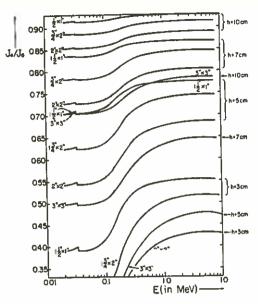


Fig. 73. Angular resolution correction factor $Q_4 = J_4/J_0$ for cylindrically symmetric NaI detectors of dimension $d \times r$ (d=thickness of NaI crystal, r=radius) at a distance h from the (point) source. (West 460)

The method (iii) works best if the scintillation crystals are perfect and not shielded. It has been shown experimentally 106,461 that both methods (ii) and (iii) then yield identical results. If the crystals are shielded, however, the solid angle is enlarged because some of the γ -rays scattered in the shielding are also counted. Method (ii) then must be applied.

Both methods (ii) and (iii) are applicable regardless of the energy of the γ -rays, provided the coefficients Q_k are properly calculated.

(iv) If the energies of the γ -rays are not very different from that of annihilation radiation, one can measure the angular resolution of the counters with annihilation radiation 462 . The coefficients Q_k are found by a numerical integration 457 .

14.3.2. In most correlation experiments, the source can be so chosen that no correction for source size is necessary. In some cases (e.g. very small specific activities) one may be forced to use large sources and the following consideration then applies 35, 457, 458, 462, 463.

The only source size correction that has been treated rigorously is that for an 'axial' source, i.e., a line source located at the origin and oriented perpendicular to the plane of the two counters. Thus in experimental work, one best prepares the source as a

long thin cylinder and corrects only for the axial extension. All necessary formulas are given in refs. 458, 463.

14.3.3. After having corrected the coefficients A_{kk} for finite solid angle and finite source extension, one is left with the problem of correcting them for scattering in the source. Unfortunately, this correction is more complex and more difficult to perform than the others.

Suppose the activity to be dispersed uniformly in a cylindrical source. Two effects then mask the correlation function: (i) Some of the γ -rays are deflected before being counted. This effect, caused by Compton scattering, tends to 'smear out' the correlation, i.e., it always decreases the magnitude of the coefficients A_{kk} . (ii) The γ -ray pairs that give coincidences in the 90° position are on the average less absorbed than those that give coincidences in the 180° position. Thus one misses more coincidences at $\theta = 180^{\circ}$ than at $\theta = 90^{\circ}$. A correlation function with negative anisotropy is enhanced, one with positive anisotropy is attenuated. The absorption occurs mainly through the photoeffect. However, the Compton effect also contributes to the 'absorption', because the γ -rays can lose so much energy that they no longer are counted. The absorption is very pronounced for low γ -ray energies and sources with high atomic number Z.

If the radioactive source itself is very thin, but surrounded by scattering material, only effect (i) is present and the correlation is attenuated.

The corrections due to scattering in the source are treated in ref. 464.

Experimentally, the correction for scattering in the source can be performed by measuring the anisotropy for various source diameters and plotting it against the probability that a γ -ray pair escapes from the source without undergoing either photoeffect or Compton effect.

⁴⁸¹ D. W. Glasgow, L. W. Coleman and L. Schecter, Rev. Sci. Inst. 32 (1961) 683.

⁴⁶² E. L. Church and J. J. Kraushaar, Phys. Rev. 88 (1952) 419.

⁴⁶³ D. H. White, UCRL-Report 6994 (1962), unpublished.

⁴⁶⁴ E. Breitenberger, Proc. Phys. Soc. (London) A67 (1954) 1108.

APP. 9

TABLE 1 Correction factors for $1\frac{1}{2}$ " \times 1" crystals

h (cm)	E (MeV)	ε		Q ₂		Q_4	
		Whole spectrum	Photopeak	Whole spectrum	Photopeak	Whole spectrum	Photopeak
7.0	1.5 1.0 0.5 0.2 0.1 * 1.5 1.0	0.2886 0.3418 0.4726 0.8298 0.9685 0.2706 0.3208 0.4450	0.0545 0.0886 0.2253 0.7605 0.9588 0.0516 0.0816 0.2132	0.9781 0.9781 0.9778 0.9766 0.9745 0.9591 0.9589 0.9583	0.9800 0.9798 0.9794 0.9771 0.9745 0.9620 0.9626 0.9612	0.9284 0.9281 0.9274 0.9233 0.9166 0.8680 0.8675 0.8657	0.9343 0.9335 0.9324 0.9250 0.9167 0.8773 0.8792 0.8747
5.0	0.2 0.1 * 1.5 1.0 0.5 0.2 0.1	0.7957 0.9575 0.2525 0.2997 0.4171 0.7606 0.9458	0.7281 0.9482 0.0479 0.0758 0.1989 0.6957 0.9362	0.9553 0.9501 0.9283 0.9279 0.9267 0.9200 0.9087	0.9563 0.9502 0.9338 0.9343 0.9318 0.9221 0.9089	0.8561 0.8400 0.7747 0.7735 0.7700 0.7503 0.7173	0.8595 0.8403 0.7913 0.7934 0.7854 0.7564

* Note added by the editor:

For energies below 0.1 MeV, the calculations by Rose [Phys. Rev. 91 (1953) 610] are valid to a very good approximation. We obtain for

h=10 cm	E = 0.082	$Q_2 = 0.9741$	$Q_4 = 0.9155$
4000 -	0.048	=0.9737	= 0.9143
$h=7\mathrm{cm}$	-0.082	=0.9493	=0.8375
	= 0.048	=0.9483	=0.8346

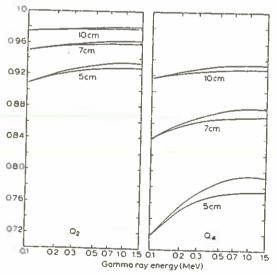


Fig. 2. Correction factors for 1½" dia ×1" long crystals

This is a problem which is not amenable to analytic solution, but an alternative method which can be employed is the use of Monte Carlo calculations. In these the interactions of the primary photons and all the secondary radiations produced within the crystal are simulated by an electronic computer. The outcome of each event is decided by sampling an internally generated set of random numbers in such a way as to produce the required probability distribution function. Several such programmes have been described in the literature, e.g., those of Berger and Dogget⁹, Beach and Davisson¹⁰, Miller and Snow¹¹, Zerby and Moran¹² and Weitkamp¹³, and used to calculate response spectra and photofractions. The problem of angular correlation correction factors has received much less attention^{14,15} although their calculation usually requires little or no more calculation.

One of the above programmes (that of Zerby and Moran) has been translated into FORTRAN by the author, and adapted to calculate the angular correlation correction factors Q_2 and Q_4 . (For γ -ray measurements, ignoring centre of mass motion, only the even order Legendre polynomials are required.) The main characteristics of this programme have already been described ^{12,15}. The basic principle is, of course, that each contribution to the photopeak is multiplied by a weight proportional to P_I (cos β) before being added in, where β is the angle of entry of the original photon.

For energies above 1.5 MeV where bremsstrahlung losses become increasingly important, the photopeak has been defined to include all contributions greater than $E_{\rm cut} = \frac{1}{2}(E_{\gamma} + E_{\rm c})$, where E_{γ} is the γ -ray energy and $E_{\rm c}$ is the energy given to a Compton electron by a photon scattered through 180°. As the energy increases $E_{\rm cut} \rightarrow E_{\gamma} - 0.128$ MeV. For energies less than 1.5 MeV the bremsstrahlung losses are small, and have been neglected. In this case the processes which are simulated are total absorption (photoelectric absorption), Compton scattering, and pair production. The necessity for introducing $E_{\rm cut}$ is a consequence of the small bremsstrahlung losses which build up near (but not in) the photopeak. When the instrumental resolution is folded into the spectrum these events cannot be distinguished from the total absorption peak itself, and must therefore be included with it.

§ 3. The calculated corrections

The results of these calculations are set out in Tables 1-5 and Figs. 2-7. Also given, for completeness, are the intrinsic efficiency and the photopeak efficiency, which are the

⁹ M. J. Berger and J. Dogget, Rev. Sci. Instr. 27 (1956) 269; J. Research Natl. Bur. Standards 56 (1956) 355.

¹⁰ C. M. Davisson and L. A. Beach, Naval Research Lab. Report NRL-5408 (1960).

W. F. Miller and W. J. Snow, Rev. Sci. Instr. 31 (1960) 905; Argonne Report ANL-6318 (1961).
 C. D. Zerby and H. S. Moran, Nucl. Instr. and Meth. 14 (1961) 115; Oak Ridge Report ORNL-3169 (1962).

¹³ C. Weitkamp, Nucl. Instr. and Meth. 23 (1963) 13.

¹⁴ C. R. Gosset and C. M. Davisson, Bull. Am. Phys. Soc. Ser. II, 7 (1962) 9; Navai Research Lab. Quarterly Report NP-11098 (1961) p. 38.

¹⁵ M. J. L. Yates, Nucl. Instr. and Meth. 23 (1963) 152.

¹⁶ P. J. Twin and J. C. Willmott, Nucl. Instr. and Meth. 22 (1963) 109.

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TABLE 2

Correction factors for 1½"×2" crystals.

Calculated by E. Matthias using the present programme

h (cm)	E (MeV)	€		Q_2		Q_4	
		Whole spectrum	Photopeak	Whole spectrum	Photopeak	Whole spectrum	Photopeal
10.0	1.5	0.4126	0.0916	0.9804	0.9822	0.9355	0.9416
	1.0	0.4741	0.1454	0.9801	0.9822	0.9348	0.9417
	0.7	0.5374	0.2113	0.9799	0.9818	0.9340	0.9401
	0.5	0.6071	0.3263	0.9795	0.9811	0,9328	0.9381
	0.3	0.7480	0.5874	0.9784	0.9796	0.9294	0.9330
	0.2	0.8607	0.7967	0.9769	0.9775	0.9243	0.9262
	0.10	0.9684	0.9588	0.9745	0.9745	0.9166	0.9167
	0.05	0.9948	0.9931	0.9738	0.9738	0.9144	0.9144
	0.03	0.9904	0.9866	0.9739	0.9739	0.9148	0.9148
	0.02	0.9970	0.9957	0.9737	0.9737	0.9142	0.9143
7.0	1.5	0.3726	0.0816	0.9638	0.9674	0.8829	0.8942
	1.0	0.4296	0.1302	0.9633	0.9669	0.8813	0.8928
	0.7	0.4891	0.1962	0.9627	0.9663	0.8794	0.8908
	0.5	0.5557	0,2962	0.9619	0.9651	0.8768	0.8871
	0.3	0.6963	0.5447	0.9595	0.9617	0.8693	0.8765
	0.2	0.8213	0.7586	0.9559	0.9570	0.8580	0.8617
	0.1	0.9575	0.9482	0,9501	0.9502	0.8400	0.8403
	0.05	0.9929	0.9912	0.9483	0.9483	0.8347	0.8347
	0.03	0.9869	0.9831	0.9486	0.9487	0.8356	0.8357
	0.02	0.9958	0.9945	0.9482	0.9482	0.8342	0.8342
5.0	1.5	0.3348	0.0760	0,9371	0.9439	0.8015	0.8222
	1.0	0.3875	0,1161	0.9360	0.9430	0.7984	0.8195
	0.7	0.4432	0,1763	0.9348	0.9417	0.7946	0.8157
	0.5	0.5066	0.2711	0.9332	0.9392	0.7897	0.8082
	0.3	0.6459	0.5035	0.9284	0.9327	0.7753	0.7882
	0.2	0.7813	0,7198	0.9211	0.9233	0.7536	0.7602
	0.1	0,9457	0.9361	0.9087	0.9089	0.7173	0.7178
	0.05	0.9909	0.9891	0.9047	0.9047	0.7058	0.7058
	0.03	0.9832	0.9794	0.9054	0.9054	0.7078	0.7079
	0.02	0.9946	0.9933	0.9044	0.9044	0.7048	0.7048

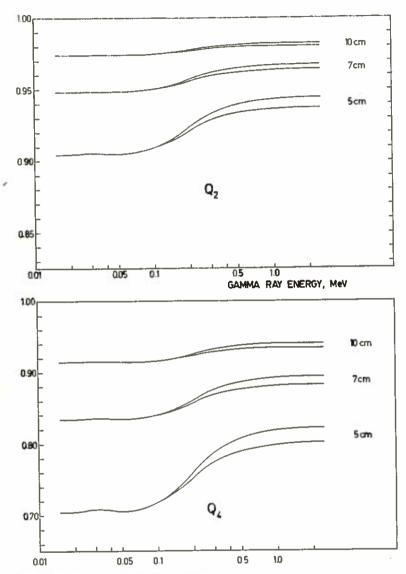


Fig. 3. Correction factors for 1\frac{1}{2}" dia \times 2" long crystals. Calculated by E. Matthias using the present programme