

XIX

ANGULAR DISTRIBUTION OF NUCLEAR RADIATION

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(A) ANGULAR CORRELATIONS

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§ 1. Introduction

1.1. The probability of emission of a particle or quantum by a radioactive nucleus depends in general on the angle between the nuclear spin axis and the direction of emission. Under ordinary circumstances, the total radiation from a radioactive sample is isotropic because the nuclei are randomly oriented in space. An anisotropic radiation pattern can be observed only from an ensemble of nuclei that are not randomly oriented.

One method of arriving at such an ensemble consists in placing the radioactive sample at a very low temperature in a strong magnetic field or electric field gradient, thereby polarizing or aligning the nuclei, and then measuring the angular distribution of the emitted radiation with respect to the direction of the applied field. This topic will be dealt with in Part B of this chapter.

Another method, the subject of the present survey, consists in picking out only those nuclei whose spins happen to lie in a preferred direction. This case can be realized if the nuclei decay through successive emission of two radiations R_1 and R_2 . The observation of R_1 in a fixed direction k_1 selects an ensemble of nuclei that has a nonisotropic distribution of spin orientations. The succeeding radiation R_2 then shows a definite *angular correlation* with respect to k_1 .

We use in the following the term *angular correlation* as comprising *directional correlation* and *polarization correlation*. In directional correlation only the directions of the two radiations are observed; in polarization correlation one determines also the linear or circular polarization of one or both of the radiations.

1.2. In angular correlation work, a typical situation is as follows: a nucleus emits in rapid succession two γ -rays γ_1 and γ_2 . We ask for the relative probability $W(\theta) d\Omega$ that γ_2 is emitted into the solid angle $d\Omega$ at an angle θ with respect to k_1 . The theoretical expression for the correlation function $W(\theta)$ for γ -rays has been worked out for all cases of interest. Experimentally, one records the number of coincidences between γ_1 and γ_2 as a function of the angle ϑ subtended by the axes of the two counters. Because of the finite solid angles of the counters these numbers $C(\vartheta)$ are

averages of the true correlation $W(\theta)$ over angles θ distributed around ϑ (cf. Fig. 1). Hence the $C(\vartheta)$ must be properly corrected and normalized to yield $W_{\text{exp}}(\theta)$. The comparison of $W_{\text{exp}}(\theta)$ with theory finally provides the desired information about the properties of the nuclear levels and the radiations.

1.3. Figure 1 shows a particularly simple angular correlation experiment. Generalizations are obvious. Instead of γ -rays, α - or β -particles or conversion electrons can

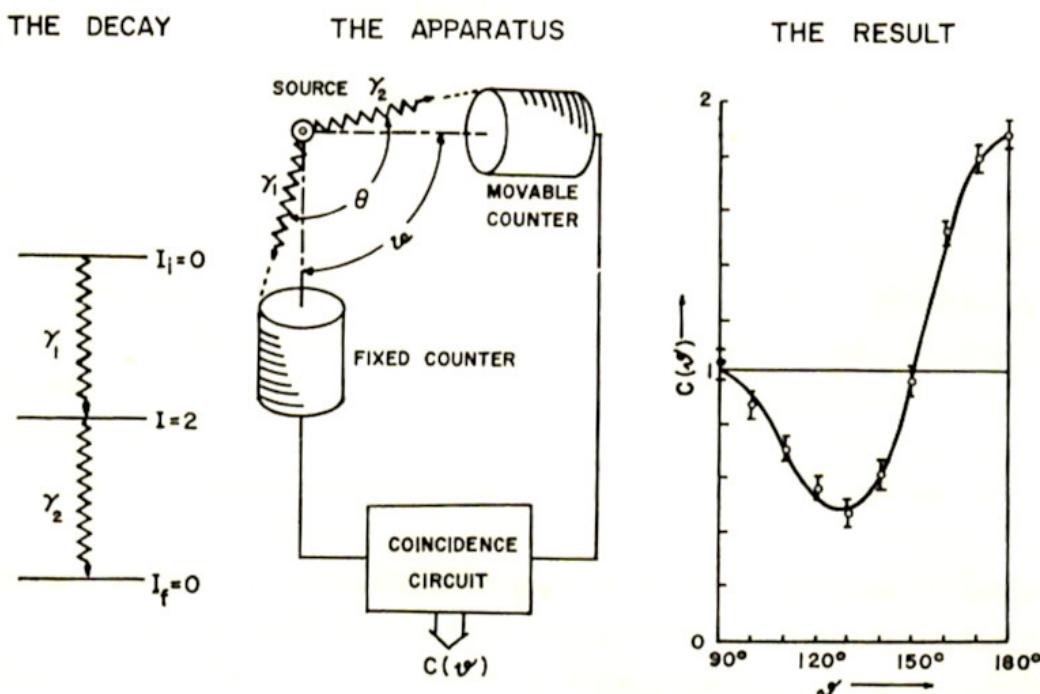


Fig. 1. Example of a directional correlation measurement

be used; instead of only observing the direction of the γ -rays, their linear polarization can also be measured. These correlations, discussed in the present chapter, do not exhaust all the possibilities. Angular correlations that involve either the polarization of β -particles or the circular polarization of γ -rays, yield information on the interaction in β -decay; they are discussed in Chapter XXIV. In nuclear reactions, one often observes the intensity or the polarization of particles or quanta at a certain angle with respect to the direction of the incoming beam. Such *angular distribution* experiments fall outside the scope of the present book. We will also omit angular correlations involving mesons and hyperons.

1.4. The information that can be obtained from angular correlation work depends on the type of radiation observed (α , β , γ , e^-), on the properties that are singled out by the experiment (direction, polarization, energy), and on the extranuclear fields acting on the nucleus.

In *Part I*, we assume that the decaying nuclei are free, i.e., that no extranuclear fields act on the nucleus and disturb its orientation in the intermediate state. To the extent that this can be realized in practice, angular correlation measurements provide

information about the properties of the nuclear levels involved and about the angular momenta carried away by the radiations. To be more precise, $\alpha-\gamma$ and $\gamma-\gamma$ directional correlation yield the spins of the nuclear levels, but not the parities. The relative parities can be determined, however, if one observes in addition to the direction also the polarization of the γ -rays, or if one measures the directional correlation between conversion electrons (e^-). The directional correlation of a $\beta-\gamma$ cascade depends not only on the nuclear spins and parities, but also on the matrix elements involved in the β -transition.

In *Part II*, we discuss the information that can be obtained from the influence of extranuclear fields on the nuclear angular correlation. Very often, one can determine the quadrupole coupling $Q\partial^2V/\partial z^2$ from the change of the correlation due to the extranuclear fields. (The calculation of the quadrupole moment Q from the value of $Q\partial^2V/\partial z^2$ is difficult because the electric field gradient $\partial^2V/\partial z^2$ in solids and liquids cannot be measured, but must be calculated.) In many cases, one can measure the g -factor of an excited nuclear state by observing the directional correlation as a function of an external magnetic field. From the g -factor one gets the magnetic moment if the spin of the nuclear state is known, e.g. fromn the unperturbed directional correlation.

In *Part III*, we briefly describe those aspects of the experimental technique that have not already been sketched in Parts I and II. In particular, we outline the steps leading from the raw data to the final result of an angular correlation experiment.

1.5. The pioneer work on the theory of directional correlation was done in 1940 by Hamilton¹. In the following years, some attempts were made to verify experimentally the predictions of Hamilton's theory, but they were unsuccessful. In 1946, Goertzel investigated theoretically the influence of extranuclear fields on the directional correlation². Such an interaction could have explained the negative experimental results in at least some cases. However, the first successful measurement, performed by Brady and Deutsch³ in 1947, showed that inadequate experimental techniques, and not extranuclear fields, were responsible for the earlier failures.

These first experiments of Brady and Deutsch were done with Geiger counters. A major improvement was the introduction of the scintillation counter into angular correlation work in 1948 (Brady and Deutsch^{4,5}, Metzger and Deutsch^{6,7}). It is difficult today to realize how tedious $\gamma-\gamma$ correlation experiments with Geiger counters were. A typical run that takes a day with scintillation counters would require more than a thousand years of continuous work with Geiger counters. Scintillation counters

¹ D. R. Hamilton, Phys. Rev. **58** (1940) 122.

² G. Goertzel, Phys. Rev. **70** (1946) 897.

³ E. L. Brady and M. Deutsch, Phys. Rev. **72** (1947) 870.

⁴ E. L. Brady and M. Deutsch, Phys. Rev. **74** (1948) 1541.

⁵ E. L. Brady and M. Deutsch, Phys. Rev. **78** (1950) 558.

⁶ M. Deutsch and F. Metzger, Phys. Rev. **74** (1948) 1542.

⁷ F. Metzger and M. Deutsch, Phys. Rev. **78** (1950) 551.

offered three major advantages over Geiger counters: high counting efficiency, speed, and energy sensitivity. The development of fast electronics and reliable multichannel analyzers at about the same time permitted full use of these three properties. Angular correlation measurements thus rapidly became a well-established tool in nuclear spectroscopy.

The first angular correlation experiments were performed to determine spins and parities of nuclear states and radiations. It soon turned out, however, that the correlation pattern is sensitive to extranuclear fields if the lifetime of the intermediate nuclear state is longer than, say, 10^{-11} sec⁸. This effect permits the determination of the g-factor⁹ and of the quadrupole moment¹⁰ of excited nuclear states. It furthermore constitutes a tool to study properties of solids and liquids¹¹.

Another field in which angular correlation experiments have yielded essential information is β -decay, particularly since the discovery of parity nonconservation. The correlation between β -particles and circularly polarized γ -rays was one of the early demonstrations of parity nonconservation^{12, 13}.

The theory of angular correlation developed as fast as the experimental techniques. The original calculations^{1, 2} were cumbersome and applied only to very special cases. Step by step, the theory was improved and generalized¹⁴⁻²⁵ and perturbations due to extranuclear fields were taken into account^{22, 26-30}.

1.6. The section on angular correlation in *Beta- and Gamma-Ray Spectroscopy* was prepared in 1954. The major advances since then have occurred in the field of β -decay;

- ⁸ H. Aeppli, A. S. Bishop, H. Frauenfelder, M. Walter and W. Zünti, Phys. Rev. **82** (1951) 550.
- ⁹ H. Aeppli, H. Albers-Schönberg, H. Frauenfelder and P. Scherrer, Helv. Phys. Acta **25** (1952) 339.
- ¹⁰ H. Albers-Schönberg, F. Hänni, E. Heer, T. B. Novey and P. Scherrer, Phys. Rev. **90** (1953) 322.
- ¹¹ E. Heer and T. B. Novey, Solid State Physics **9** (ed. F. Seitz and D. Turnbull; Academic Press, New York and London, 1959).
- ¹² H. Schopper, Phil. Mag. **2** (1957) 710.
- ¹³ F. Boehm and A. H. Wapstra, Phys. Rev. **106** (1957) 1364; **107** (1957) 1202.
- ¹⁴ J. W. Gardner, Proc. Phys. Soc. (London) **A62** (1949) 763.
- ¹⁵ D. S. Ling and D. L. Falkoff, Phys. Rev. **76** (1949) 1639.
- ¹⁶ M. Fierz, Helv. Phys. Acta **22** (1949) 489.
- ¹⁷ J. A. Spiers, Nat. Res. Council Canada, Publ. No. 1925 (1949), Phys. Rev. **80** (1950) 491.
- ¹⁸ D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79** (1950) 323.
- ¹⁹ G. Racah, Phys. Rev. **84** (1951) 910.
- ²⁰ U. Fano, Nat'l. Bureau of Standards Report 1214; Phys. Rev. **90** (1953) 577.
- ²¹ S. P. Lloyd, Phys. Rev. **85** (1952) 904.
- ²² K. Alder, Helv. Phys. Acta **25** (1952) 235.
- ²³ S. R. de Groot, Physica **18** (1952) 1201.
- ²⁴ F. Coester and J. M. Jauch, Helv. Phys. Acta **26** (1953) 3.
- ²⁵ L. C. Biedenharn and M. E. Rose, Rev. Mod. Phys. **25** (1953) 729.
- ²⁶ A. Abragam and R. V. Pound, Phys. Rev. **92** (1953) 943.
- ²⁷ K. Alder, H. Albers-Schönberg, E. Heer and T. B. Novey, Helv. Phys. Acta **26** (1953) 761.
- ²⁸ F. Coester, Phys. Rev. **93** (1954) 1304.
- ²⁹ H. Paul and W. Brunner, Ann. Physik **9** (1962) 316, 323; **10** (1963) 327.
- ³⁰ K. Alder, E. Matthias, W. Schneider and R. M. Steffen, Phys. Rev. **129** (1963) 1199.

very short-lived states ($\tau < 10^{-12}$ sec), this condition is always fulfilled. For states with longer lifetimes, perturbations are possible. In order to observe the unperturbed correlation (maximum correlation) one must find a nuclear environment where the extranuclear fields are very small (e.g. a regular lattice site in a cubic crystal) or where their time average vanishes (e.g. certain liquids). Except for occasional remarks, we postpone until Part II all questions concerning how one finds such an environment.

§ 2. Naive theory of γ - γ directional correlation in free nuclei

2.1. SINGLE γ -TRANSITION

As will be shown in section 3, the theory of angular correlation is highly developed and general expressions can be written down for even very complex situations. Quite

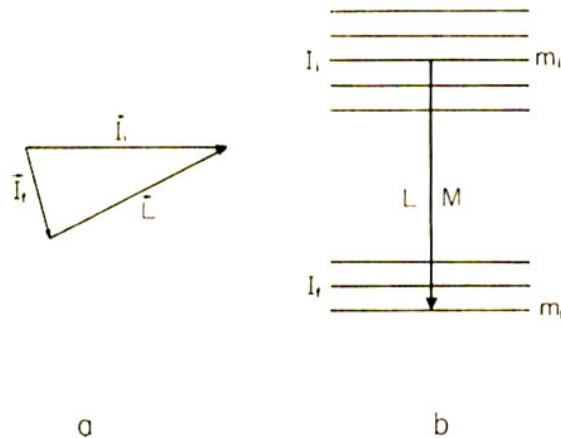


Fig. 2. Single γ -transition

often, however, particularly when one first studies a subject, the most general derivation of a result does not lead to a clear understanding of the basic physical processes. We therefore present in this section a simple treatment of γ - γ directional correlation that, while restricted in its usefulness, provides some insight into the correlation mechanism^{31, 32, 36}.

We investigate first a single γ -transition, of angular momentum L , between two nuclear levels i and f with spins I_i and I_f . For conservation of angular momentum, we have $I_i = I_f + L$. The projection of L on the arbitrary axis of quantization (z-axis) shall be denoted by L_z . The emitted γ -ray is then characterized by the angular momentum quantum number or multipolarity L , and the magnetic quantum number M , with $L^2 = L(L+1)\hbar^2$, $L_z = M\hbar$. The quantum numbers of the two nuclear states are I_i , m_i and I_f , m_f . Clearly, $m_i = m_f + M$ (Fig. 2).

Each component* $m_i \rightarrow m_f$ between specified magnetic sublevels (m -states) possesses a characteristic directional distribution $F_L^M(\theta)$ that is independent of I_i and I_f . Here θ denotes the angle between emitted γ -ray and z-axis. For γ -rays, the distribution

* We follow Condon and Shortley³⁸ in designating the radiation resulting from a transition between a particular pair of m -states as a *component*. The total radiation associated with all possible transitions between two levels is called a *line*.⁴⁰

function $F_L^M(\theta)$ can be found by calculating the energy flow (Poynting vector) as a function of θ for multipole radiation characterized by the quantum numbers L and M (for discussions of multipole radiation see Chapter XV and refs. 15, 33, 40–44). For dipole radiation ($L=1$), one obtains

$$\begin{aligned} F_1^0(\theta) &= 3 \sin^2 \theta = 2 - 2P_2(\cos \theta), \\ F_1^{\pm 1}(\theta) &= \frac{3}{2}(1 + \cos^2 \theta) = 2 + P_2(\cos \theta). \end{aligned} \quad (1)$$

The first of these functions, $F_1^0(\theta)$, is the familiar radiation pattern from a dipole lying in the z -axis.

In atomic spectroscopy, one observes the individual components (1) and their directional distributions in the Zeeman effect: the different m -states of the two atomic levels i and f split up in a magnetic field and the components associated with F_1^1 , F_1^0 , and F_1^{-1} thus differ in energy sufficiently to be observed individually. Usually, one selects only the two positions $\theta=0^\circ$ and $\theta=90^\circ$ and calls the corresponding observations longitudinal and transverse Zeeman effect.

The nuclear Zeeman effect was unobservable until a few years ago. The splitting of nuclear energy levels even in very strong magnetic fields is only of the order of 10^{-8} eV, many orders of magnitude too small to be resolved by conventional γ -ray spectrometers. Mössbauer's discovery of the recoilless nuclear resonance absorption in 1958 has changed this situation⁴⁵. The Zeeman effect of certain low-lying nuclear states can be observed and the angular distribution and the linear⁴⁶ and circular⁴⁷ polarization of the γ -ray components can be detected. The Mössbauer effect is discussed in Chapter XXIB; relevant references can be found there.

Unfortunately, the separation of the γ -ray components is possible only in a few cases; for most γ -rays, one observes the unresolved line. In order to calculate its directional distribution $F_L(\theta)$, we must know not only the functions $F_L^M(\theta)$, but also the relative population $P(m_i)$ for each sublevel m_i , and the relative transition probability $G(m_i m_f)$ for each component $m_i \rightarrow m_f$. The directional distribution $F_L(\theta)$ of the total line then is given by

$$F_L(\theta) \propto \sum_{m_i m_f} P(m_i) G(m_i m_f) F_L^M(\theta), \quad \text{where} \quad M = m_i - m_f. \quad (2)$$

⁴⁰ E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, 1951).

⁴¹ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (J. Wiley and Sons, New York, 1952).

⁴² J. D. Jackson, *Classical Electrodynamics* (J. Wiley and Sons, New York, 1962).

⁴³ D. M. Brink and G. R. Satchler, *Angular Momentum* (Oxford Univ. Press, London, 1962).

⁴⁴ M. E. Rose, *Polarization Phenomena in Beta and Gamma Emission*, in *Lectures in Theoretical Physics 2* (W. A. Benjamin, Inc., New York, 1962).

⁴⁵ R. L. Mössbauer, *Z. Physik* **151** (1958) 124.

⁴⁶ S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston and D. H. Vincent, *Phys. Rev. Letters* **4** (1960) 177.

⁴⁷ H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. F. Cochran and W. M. Visscher, *Phys. Rev.* **126** (1962) 1065.

The absolute transition probability for a component $m_i \rightarrow m_f$ can be written as a product of a ‘nuclear’ factor and a ‘geometrical’ factor. The nuclear factor depends on specific nuclear properties, but not on m_i and m_f , and enters into eq. (2) only as a constant factor. The geometrical factor depends explicitly on m_i and m_f and yields the desired relative transition probability $G(m_i m_f)$.

The factoring into a geometrical and a nuclear part can be seen most easily in the case of the matrix element for the radiation from an electric dipole lying in the z -axis:

$$\langle f|z|i\rangle = \int \psi_f^* z \psi_i d\tau. \quad (3)$$

Assuming wavefunctions of the form $\psi = R(r) Y_I^m(\vartheta, \phi)$, where $Y_I^m(\vartheta, \phi)$ denotes a normalized spherical harmonic, and setting $z = r \cos \vartheta$, $\langle f|z|i\rangle$ becomes the product of two integrals. One depends only on the radial factor of ψ and cannot be evaluated further without specific assumptions about $R(r)$, the other yields the geometrical factor and can be calculated explicitly. A more appropriate derivation can be given by using the properties of the Clebsch–Gordan coefficients.

We give here a short definition of Clebsch–Gordan (Wigner) coefficients. Details can be found in refs. 33, 40, 41, 43, 48–52. Let $|I_i, m_i\rangle$ be one of an orthonormal set $|I'_i, m'_i\rangle$ of eigenfunctions of the operators I_i^2 and $(I_i)_z$, with eigenvalues $I_i(I_i+1)\hbar^2$ and $m_i\hbar$, respectively. Let $|I_f, m_f\rangle$ and $|L, M\rangle$ be similar eigenfunctions for the vector operators I_f and L , and $I_i = I_f + L$, $m_i = m_f + M$. The Clebsch–Gordan coefficients are the coefficients in the expansion of $|I_i, m_i\rangle$ in terms of products $|I_f, m_f\rangle |L, M\rangle$

$$|I_i, m_i\rangle = \sum_{m_f, M} |I_f, m_f\rangle |L, M\rangle \langle I_f m_f LM | I_i m_i \rangle. \quad (4)$$

The Clebsch–Gordan coefficients $\langle I_f m_f LM | I_i m_i \rangle$ are the elements of a unitary matrix. The phases of the eigenfunctions are so chosen that the coefficients are real numbers and hence the matrix is orthogonal. The inversion of eq. (4) thus yields

$$|I_f, m_f\rangle |L, M\rangle = \sum_{I'_i, m'_i} \langle I_f m_f LM | I'_i m'_i \rangle |I'_i, m'_i\rangle. \quad (5)$$

To continue with the evaluation of eq. (3) we write $z = r \cos \vartheta = r (\frac{4}{3}\pi)^{\frac{1}{2}} Y_1^{0*}$, and assume as before $\psi = R(r) Y_I^m$. The functions Y_1^{0*} , $Y_{I_i}^{m_i}$, and $Y_{I_f}^{m_f}$ are all eigenfunctions of the operators I^2 and I_z . Moreover, $I_i = I_f + L$, where $L=1$. We can thus, after inserting the expressions for z and ψ into eq. (3), expand the product $Y_{I_f}^{m_f} Y_1^{0*}$ in terms of $Y_{I_i}^{m_i}$ by using eq. (5). Because of the orthogonality of the spherical harmonics, all integrals vanish except that with $I'_i = I_i$ and $m'_i = m_i$. The matrix element $\langle f|z|i\rangle$ therefore becomes proportional to the Clebsch–Gordan coefficient $\langle I_f m_f 10 | I_i m_i \rangle$, and the relative transition probability is proportional to its square.

⁴⁸ A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton Univ. Press, Princeton, 1957).

⁴⁹ E. Merzbacher, *Quantum Mechanics* (J. Wiley and Sons, New York, 1961).

⁵⁰ W. S. C. Williams, *Introduction to Elementary Particles* (Academic Press, New York, 1961).

⁵¹ A. Messiah, *Quantum Mechanics* (North-Holland Publ. Co., 1962).

⁵² B. Kursunoglu, *Modern Quantum Theory* (W. H. Freeman, San Francisco, 1962).

The outlined calculation of the relative transition probability was performed with single particle wave functions and for electric dipole radiation only. The result, however, is independent of the first assumption and can easily be generalized for all multipole orders. We can thus define the relative transition probability to be equal to the square of the Clebsch-Gordan coefficient for the vector addition $\mathbf{I}_i = \mathbf{I}_f + \mathbf{L}$, $m_i = m_f + M$:

$$G(m_i, m_f) = \langle I_f m_f LM | I_i m_i \rangle^2. \quad (6)$$

We will derive eq. (6) in section 3. (The most general derivation of eq. (6) is based on group theoretical methods; see refs. 53, 54.)

The relative populations $P(m_i)$ depend, in contrast to $F_L^M(\theta)$ and $G(m_i m_f)$, on the energies of the different m_i -states and on the way in which level i was created. If the nuclei are randomly oriented, i.e. if all m_i -states are equally populated for any choice of the z -axis, then $F_L(\theta)$ is independent of θ , i.e. the distribution is isotropic. This result can be confirmed by evaluating eq. (6) for equally populated m_i -states.

2.2. GAMMA-GAMMA CASCADE

In this subsection we assume that a nucleus decays through a cascade $I_i \rightarrow I \rightarrow I_f$ (Fig. 1) by successive emission of two γ -rays of multipolarity L_1 and L_2 , and we ask for the directional correlation function $W(\theta)$ between the two γ -rays.

In order to simplify the calculation we let the direction of the quantization axis coincide with the direction of emission of the first γ -ray. The directional correlation $W(\theta)$ between the two γ -rays then becomes identical with the directional distribution $F_L(\theta)$ of the second γ -ray with respect to the z -axis. It can thus be calculated from eq. (2) and (6) once each relative population $P(m)$ is known. This, however, is given by the sum of all transitions $m_i \rightarrow m$ leading into the state m . Assuming all m_i -states equally populated, we have, with $M_1 = m_i - m$:

$$P(m) \propto \sum_{m_i} G(m_i m) F_{L_1}^{M_1}(\theta=0). \quad (7)$$

The special choice of the z -axis introduces a second simplification. A photon propagating in a definite direction can carry in its direction of motion only the angular momentum $+h$ or $-h$ (refs. 41, 55-57). In our case, the first γ -ray travels along the z -axis and thus M_1 is restricted to the two values ± 1 . Only the functions $F_{L_1}^1(0)$

⁵³ C. Eckart, Rev. Mod. Phys. 2 (1930) 305.

⁵⁴ E. P. Wigner, Gruppentheorie (F. Vieweg and Sohn, Braunschweig, 1931); Group Theory and its Application to the Quantum Mechanics of Atomic Spectra (Academic Press, New York, 1959).

⁵⁵ W. Heitler, Quantum Theory of Radiation (Oxford Univ. Press, London, 1954) Appendix I.

⁵⁶ J. M. Jauch and F. Rohrlich, Theory of Photons and Electrons (Addison-Wesley, Reading, Massachusetts, 1955) section 2.8.

⁵⁷ J. Hamilton, The Theory of Elementary Particles (Oxford Univ. Press, London and New York, 1959). Section I, 14.

and $F_{L_1}^{-1}(0)$ then appear in eq. (7). For the special case of dipole radiation, this can be seen from eq. (1): F_1^0 vanishes for $\theta = 0$.

The desired directional correlation function $W(\theta)$, which for physical reasons must be independent of our special choice of the z -axis, finally follows from the combination of eq. (2), (6) and (7), with $M_2 = m - m_f$:

$$W(\theta) \propto \sum_{m_f m m_i} \langle I_m L_1 \pm 1 | I_i m_i \rangle^2 F_{L_1}^{\pm 1}(0) \langle I_f m_f L_2 M_2 | I_m \rangle^2 F_{L_2}^{M_2}(\theta). \quad (8)$$

We still have to justify one point in the derivation of eq. (8). The summation over m in eq. (8) has been done incoherently. In other words, the various possible ways $m_i \rightarrow m \rightarrow m_f$ in which a nucleus can decay from a given initial state m_i into a given final state m_f are assumed to be independent, and thus no interference terms appear. This simplification is due to the special choice of the z -axis, which allows only the values ± 1 for $M_1 = m_i - m$. Each of these two values describes one mode of circular polarization, as one can most easily verify by discussing the longitudinal Zeeman effect. In principle, we can now observe the circular polarization of each individual photon γ_1 , in addition to its direction. Such a polarization measurement would yield a definite value of M_1 , either $+1$ or -1 . Starting from a given state m_i , therefore, only one state m is reached (either $m_i - 1$ or $m_i + 1$) and no interference occurs^{58, 59}. The fact that in the actual experiment one does not measure the circular polarization, but averages over the two modes, does not change the conclusions. It is the possibility of performing the measurement, and the fact that the observation would yield a sharp value of M_1 , that destroys the interference. This possibility, however, is lost if one determines in addition to the direction also the linear polarization of the photons. In this case, interference terms appear.

In simple cases, $W(\theta)$ can be obtained from eq. (8) without excessive labor. As an example, we have evaluated three cascades, using the $F_L^M(\theta)$ from eq. (1) and taking the Clebsch-Gordan coefficients from ref. 40. The cascades and the resulting directional correlation functions are given in Fig. 3.

2.3. LIMITATIONS OF THE NAIVE THEORY

The derivation of eq. (8) in the last subsection gives a good insight into some aspects of the correlation process, but at the same time suffers from three kinds of shortcomings: (1) it is not rigorous; (2) the final eq. (8) contains tedious sums over unobserved magnetic quantum numbers; (3) it applies only to a very restricted case, namely, the directional correlation of two pure γ -rays emitted by free and unpolarized nuclei.

A more rigorous derivation of eq. (8) is not difficult. Hamilton, who first derived it¹, used second order damping theory⁵⁷. In the next section, an approach will be given that is based on first order perturbation theory. The removal of the other two types of deficiencies from the theory will also be discussed in the next section.

⁵⁸ B. A. Lippmann, Phys. Rev. **81** (1951) 162.

⁵⁹ O. Halpern and B. A. Lippmann, Phys. Rev. **87** (1952) 1128.

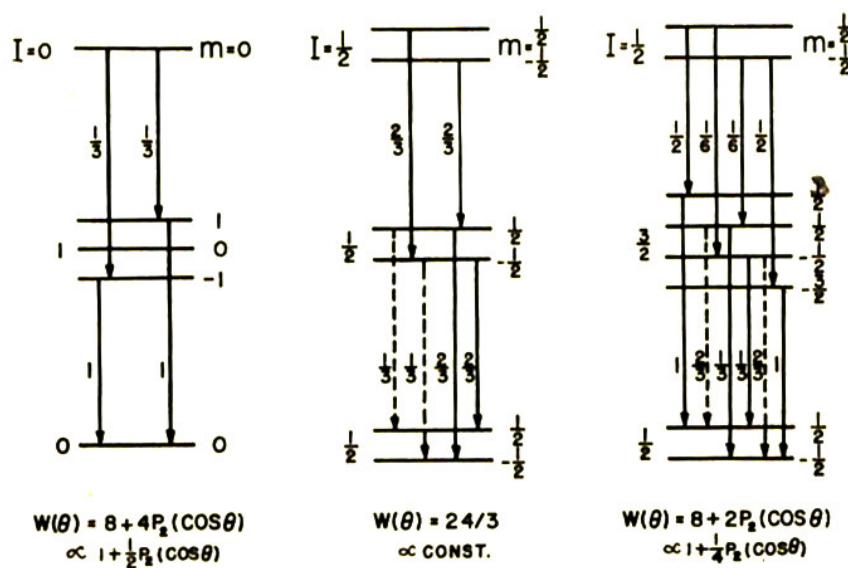


Fig. 3. Directional correlation of simple cascades. The first γ -ray of each cascade is observed in the quantization axis. Therefore, only transitions with $M=1$ occur (solid lines). The second γ -ray is detected in an arbitrary direction, thus no such restriction exists and transitions $M=1$ (solid lines) and $M=0$ (dotted lines) are allowed for dipole radiation. The relative intensities for the components are shown in the figure; their angular distributions are given by eq. (1)

§ 3. General theory of angular correlation

In the last section, we gave a simple theory of the directional correlation of successive γ -rays; here, we present a general theory of the angular correlation of successive radiations. An understanding of the naive approach of section 2 allows one to appreciate most of the physical ideas underlying the experiments that will be discussed in the later sections. However, for a complete understanding and a satisfactory description of the experiments a theory is required that overcomes the deficiencies mentioned in subsection 2.3. Such a theory indeed exists and it can be formulated in a number of equivalent ways^{19–25, 33–35a}. All of these treatments presuppose familiarity with the quantum theory of angular momentum. There exist a number of excellent introductions into this subject^{33, 43, 44, 48, 51} and any one of these contains enough information for an understanding of the present section. Some relations that will be needed in the development of the theory are collected in subsection 3.2.

In the following theory, some restrictions are kept: the nuclear states are assumed to have *well-defined symmetry properties*, i.e., to be characterized by single values of the total angular momentum and the parity. Furthermore, the radiations are assumed to be emitted in succession and not simultaneously.

3.1. THE DEVELOPMENT OF THE THEORY

The progress in the theory of angular correlation after Hamilton's work is largely due to the use of three tools: (i) group theory, (ii) Racah algebra, and (iii) density matrix formalism.

⁶⁰ V. F. Weisskopf, Ann. der Physik 9 (1931) 23.

The term U_k does not depend on the nature of the unobserved radiation, and no interference terms appear. The contributions to U_k from the different L_2 values add incoherently and are weighted by the square of the reduced nuclear matrix element $|\langle I_b \| L_2 \pi_2 \| I_a \rangle|^2$.

§ 4. Gamma-gamma angular correlation

In this section, we derive in 4.1 the radiation parameters $c_{k\tau}$ for γ -rays and present in 4.2 those theoretical results that are needed in the evaluation of the data from $\gamma-\gamma$ directional correlation measurements. In 4.3, we discuss some typical results. In 4.4, we treat the polarization correlation.

4.1. THE RADIATION PARAMETERS FOR γ -RAYS

The radiation parameters $c_{k\tau}(LL')$ are defined by eqs. (62) and (63). We outline here two different ways to calculate these parameters for γ -rays. We start from eq. (53) and note that only the factor $\langle k\sigma|LM\pi\rangle$ on the right hand side depends on the direction of emission k of the γ -ray. The directional distribution function $F_L^M(\theta)$, introduced in section 2, is proportional to the square of the transition probability $\langle Imk\sigma|H|I_im_i\rangle$ and hence can be written as

$$F_L^M(\theta) \equiv F_L^M(k) \propto S \langle k\sigma|LM\pi\rangle \langle k\sigma|LM\pi\rangle^*, \quad (82)$$

where S again denotes summation over unmeasured quantities. The radiation parameters thus are connected to the functions F_L^M by

$$c_{k0}(LL) \propto \sum_{\mu} (-1)^{L-\mu} (2k+1)^{\frac{1}{2}} \begin{pmatrix} L & L & k \\ \mu & -\mu & 0 \end{pmatrix} F_L^M(0). \quad (83)$$

The distribution functions $F_L^M(k)$ can be found by calculating the Poynting vector as a function of θ for multipole radiation characterized by the quantum numbers L and M . They are proportional to the functions Z_{LM} of Blatt and Weisskopf (p. 594 of ref. 41) and the function $|X_{LM}|^2$ of Jackson⁴². The derivation of F_L^M for mixed radiation can be found in ref. 15.

In the second approach to determine the radiation parameters, we calculate the eigenfunctions $\langle k\sigma|LM\pi\rangle$. According to eq. (53), these are defined as the coefficients in the transformation

$$\langle k\sigma| = \sum_{LM\pi} \langle k\sigma|LM\pi\rangle \langle LM\pi| \quad (84)$$

that leads from the plane wave state $\langle k\sigma|$ to the state $\langle LM\pi|$ characterized by sharp values of total and z -component of angular momentum and parity. To find these functions, one considers the vector potential of the electromagnetic field as the wavefunction for the photon. In order to write down this correspondence, we consider the expansion of a plane wave state into multipole fields. The plane wave corresponds to the situation in which the γ -rays are observed and the multipole expansion, characterized by eigenstates of parity and angular momentum, is appropriate to the theoretical description. We take circularly polarized plane waves as basic states; any other

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 $\mathbf{A}_\sigma(0)$, 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 $\mathbf{A}_\sigma(0)$ can be expanded into multipole fields:

$$\mathbf{A}_\sigma(0) = \sqrt{2\pi} \sum_{L=1}^{\infty} (2L+1)^{\frac{1}{2}} \{ \mathbf{A}_L^\sigma(m) + \sigma \mathbf{A}_L^\sigma(e) \}. \quad (85)$$

Here, $\mathbf{A}_L^\sigma(m)$ 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 $L(L+1)\hbar^2$, $\sigma\hbar$, and $(-1)^L$. Similarly, $\mathbf{A}_L^\sigma(e)$ 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

$$\begin{aligned} \langle 0\sigma | &= \mathbf{A}_\sigma(0), \\ \langle LM\pi | &_{\text{magnetic}} = \sqrt{4\pi} \mathbf{A}_L^M(m), \quad \pi = (-1)^L, \\ \langle LM\pi | &_{\text{electric}} = \sqrt{4\pi} \mathbf{A}_L^M(e), \quad \pi = (-1)^{L+1}. \end{aligned} \quad (86)$$

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

$$\begin{aligned} \langle 0\sigma | LM\pi \rangle &_{\text{magnetic}} = 2^{-\frac{1}{2}} (2L+1)^{\frac{1}{2}} \delta_{\sigma M}, \quad \pi = (-1)^L, \\ \langle 0\sigma | LM\pi \rangle &_{\text{electric}} = 2^{-\frac{1}{2}} (2L+1)^{\frac{1}{2}} \delta_{\sigma M} \sigma, \quad \pi = (-1)^{L+1}. \end{aligned} \quad (87)$$

The radiation parameters $c_{kr}(LL')$ 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)

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

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

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

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

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

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

$$c_{k0}(LL') = (-1)^{L-1} (2L+1)^{\frac{1}{2}} (2L'+1)^{\frac{1}{2}} (2k+1)^{\frac{1}{2}} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix}. \quad (89)$$

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 $\gamma-\gamma$ 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 $I_i \rightarrow I \rightarrow I_f$ 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

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

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

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

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)

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

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

where

$$F_k(LL I_i I) = A'_k(LL I_i I)/A'_{00}(LL I_i I). \quad (93)$$

An explicit expression for the normalized coefficients $F_k(LL I_i I)$ is obtained from eqs. (73) and (93) after inserting the radiation parameters (88) and using the following special value for the 6-j symbol⁴⁸:

$$\left\{ \begin{array}{c} j_1 j_2 j_3 \\ 0 j_3 j_2 \end{array} \right\} = (-1)^{j_1 + j_2 + j_3} [(2j_2 + 1)(2j_3 + 1)]^{-\frac{1}{2}}. \quad (94)$$

This procedure yields

$$F_k(LL I_i I) = (-1)^{I_1 + I - 1} (2L + 1)(2I + 1)^{\frac{1}{2}}(2k + 1)^{\frac{1}{2}} \begin{pmatrix} L & L & k \\ 1 & -1 & 0 \end{pmatrix} \left\{ \begin{array}{c} LL k \\ I I I_i \end{array} \right\}. \quad (95)$$

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 Rosenzweig⁹⁶ 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:

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

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

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.

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

$$\delta_1(\gamma) \equiv \langle I \| L'_1 \pi'_1 \| I_i \rangle / \langle I \| L_1 \pi_1 \| I_i \rangle \quad (96)$$

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 real⁸¹.

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

$$F_k(LL' I_i I) = (-1)^{I_1+I-1} [(2L+1)(2L'+1)(2I+1)(2k+1)]^{\frac{1}{2}} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} LL' k \\ I I I_i \end{Bmatrix}. \quad (96)$$

These coefficients satisfy

$$F_0(LL' I_i I) = \delta_{LL'}. \quad (97)$$

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

$$A_{kk} = A_k(L_1 L'_1 I_i I) A_k(L_2 L'_2 I_f I), \quad (98)$$

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

$$A_k(L_1 L'_1 I_i I) = \frac{F_k(L_1 L_1 I_i I) + 2\delta_1(\gamma) F_k(L_1 L'_1 I_i I) + \delta_1^2(\gamma) F_k(L'_1 L'_1 I_i I)}{1 + \delta_1^2(\gamma)} \quad (99)$$

and a similar expression for $A_k(L_2 L'_2 I_f I)$. (See also ref. 101.)

The coefficients $F_k(LL' I_i I)$ are tabulated in Appendix 8 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_i , I , I_f , L_1 , L'_1 , $\delta_1(\gamma)$, L_2 , L'_2 , and $\delta_2(\gamma)$. One

⁸⁹ 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 γ - γ 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 measurements¹⁰², they concluded that the γ - γ cascade probably occurs between spins 4, 2, and 0. An

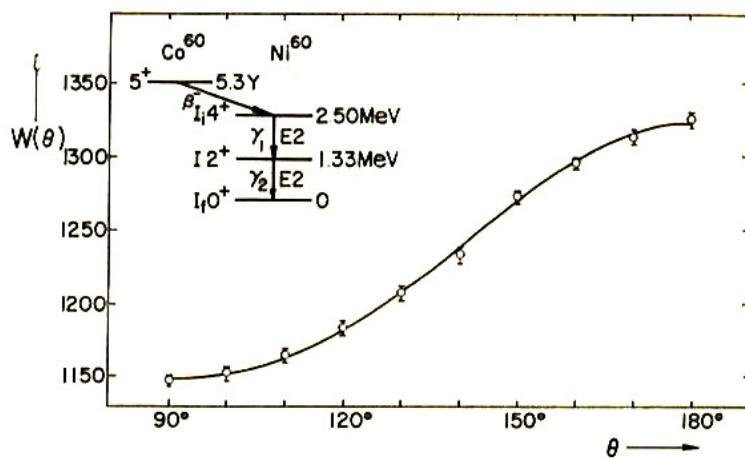


Fig. 5. Decay of Co⁶⁰ and directional correlation of the Ni⁶⁰ γ - γ cascade (Steffen³⁷). The solid line is the theoretical function $W(\theta)$ for the cascade 4(2)2(2)0

* We denote γ - γ cascades by the nuclide in which the cascade takes place, e.g. Ni⁶⁰ and not Co⁶⁰. β - γ and α - γ cascades are named after the parent nucleus.

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

accurate determination of the conversion coefficients¹⁰³ confirmed their conclusions. Since the first report by Brady and Deutsch, a large number of publications contain data on Ni⁶⁰, 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 Co⁶⁰ → Ni⁶⁰ and the directional correlation function measured by Steffen³⁷ 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 Ba¹³⁴ and Ce¹⁴⁰ 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 McGowan¹⁰⁶ 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} :

$$\begin{aligned} \text{0.624 MeV–0.513 MeV cascade: } & A_{22} = 0.346 \pm 0.008, A_{44} = 1.109 \pm 0.012 \\ \text{1.045 MeV–0.513 MeV cascade: } & A'_{22} = 0.092 \pm 0.003, A'_{44} = 0.035 \pm 0.009 \end{aligned}$$

The interpretation of these coefficients is best made by using the spin assignments that were deduced by Alburger¹⁰⁵ from measurements of the β -spectrum of Rh¹⁰⁶ and

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.

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

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 coincidences¹⁷⁷. 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 (Ni^{60} , Pd^{106}).

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 γ - γ directional correlation measurement. Other experiments involve obvious generalizations.

Before a run, and at intervals during runs, resolving time τ_0 and background counts $N_1^0(\vartheta)$, $C^0(\vartheta)$ are determined. N and C denote single counts and coincidences, respectively, per unit time. ϑ 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.:

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

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

The contribution $C^d(\theta)$, 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

$$N_i^t(\theta) = M p_i \Omega_i \varepsilon_i, \quad (410)$$

$$C^t(\theta) = M p_1 p_2 \Omega_1 \varepsilon_1 \Omega_2 \varepsilon_2 \varepsilon_c K(\theta). \quad (411)$$

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_c the efficiency of the coincidence circuit. $K(\theta)$ 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 $C^t(\theta)/N_1^t(\theta)$ is formed. One then finds for the desired uncorrected experimental correlation function from the eqs. (410) and (411):

$$K(\theta) = \frac{1}{p_2 \Omega_2 \varepsilon_2 \varepsilon_c} \frac{C^t(\theta)}{N_1^t(\theta)}. \quad (412)$$

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 $K(\theta)$ 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 $K(\theta)$ are now fitted by

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

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

^{451a} R. E. Bell, Nucl. 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}^{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}^{exp} for the finite solid angle of the counters. Using a method first developed by Frankel⁴⁵⁶, and neglecting the correction for the extension of the source, we write the corrected coefficients A_{kk} as⁴⁵⁷

$$A_{kk} = A_{kk}^{\text{exp}} / Q_{kk}. \quad (414)$$

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 Frankel⁴⁵⁸.

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

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

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

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

with

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

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.

⁴⁵⁵ 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 $\varepsilon_i(E, \alpha)$ 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

$$\begin{aligned} Q_2 &= \frac{1}{2} \cos \alpha_0 (1 + \cos \alpha_0), \\ Q_4 &= \frac{1}{8} \cos \alpha_0 (1 + \cos \alpha_0) (7 \cos^2 \alpha_0 - 3), \\ \alpha_0 &= \tan^{-1} (r/h). \end{aligned} \quad (418)$$

(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 calculated⁴⁵². For γ -rays, the efficiency $\varepsilon(E, \alpha)$ is well represented by

$$\varepsilon(E, \alpha) = 1 - \exp \{-\tau(E) X(\alpha)\}, \quad (419)$$

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.

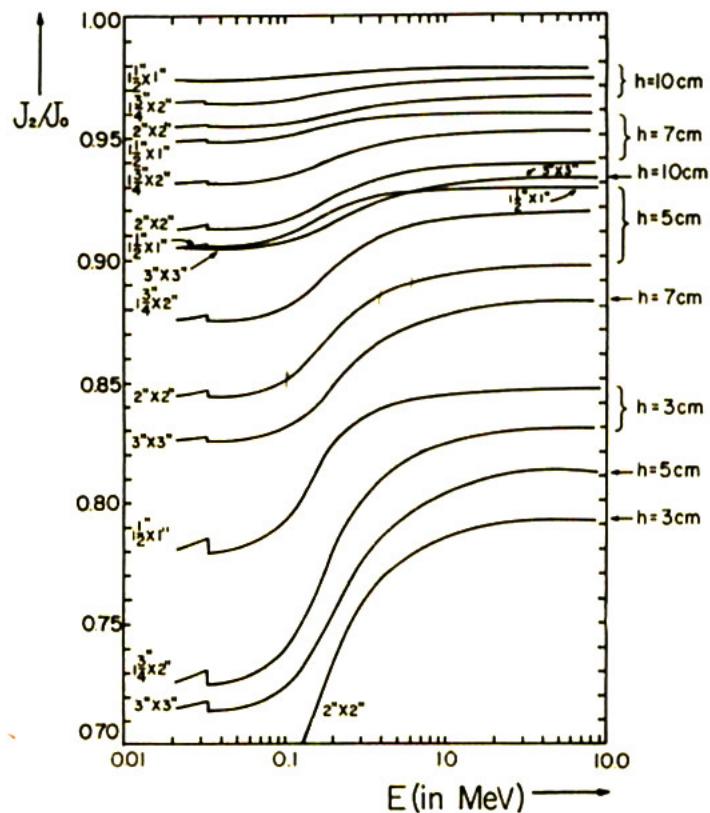


Fig. 72. Angular resolution correction factor $Q_2 = J_2/J_0$ for cylindrically symmetric NaI detectors of dimension $d \times r$ (d = thickness of NaI crystal, r = radius) at a distance h from the (point) source. (West⁴⁶⁰)

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

⁴⁶⁰ H. West, UCRL-Report 5451 (1959) unpublished, TID 4500 (1959).

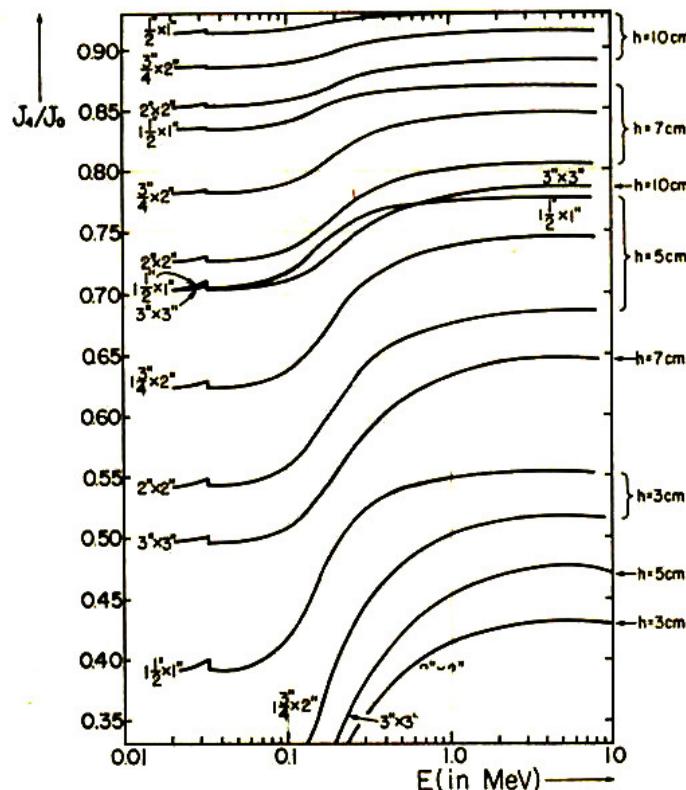


Fig. 73. Angular resolution correction factor $Q_4 = J_4/J_0$ for cylindrically symmetric NaI detectors of dimension $d \times r$ (d =thickness of NaI crystal, r =radius) at a distance h from the (point) source.
(West⁴⁶⁰)

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 radiation⁴⁶². The coefficients Q_k are found by a numerical integration⁴⁵⁷.

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

⁴⁶¹ 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.

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.

APPENDIX B

TABLES OF F -COEFFICIENTS

Numerical values of $F_k(LL'I'I)$ are tabulated for $k=1, 2, 3, 4$, for intermediate spins $I=1, \frac{3}{2}, 2, \frac{5}{2}, 3, \frac{7}{2}, 4$, and for $L \leq L' \leq 3$. The F -coefficients are symmetric in the multipole orders L and L' : $F_k(LL'I'I) = F_k(L'L'I'I)$. For $k=0$, $F_0(LL'I'I) = \delta_{L'L}$.

L	L'	P	I	$k=1$
0	1	$\frac{1}{2}$	$\frac{1}{2}$	1.732
1	1	$\frac{1}{2}$	$\frac{1}{2}$	-1.000
1	1	$\frac{3}{2}$	$\frac{1}{2}$	0.500
1	2	$\frac{3}{2}$	$\frac{1}{2}$	0.866
2	2	$\frac{3}{2}$	$\frac{1}{2}$	-0.500
2	2	$\frac{5}{2}$	$\frac{1}{2}$	0.333
2	3	$\frac{5}{2}$	$\frac{1}{2}$	0.943
3	3	$\frac{5}{2}$	$\frac{1}{2}$	-0.333
3	3	$\frac{7}{2}$	$\frac{1}{2}$	0.250

TABLE B1 ($I=\frac{1}{2}$)

TABLE B2 ($I=1$)

L	L'	P	I	$k=1$	$k=2$
1	1	0	1	-1.225	0.707
0	1	1	1	1.732	0
0	2	1	1	0	-2.236
1	1	1	1	-0.612	-0.354
1	2	1	1	0.612	-1.061
2	2	1	1	-0.612	-0.354
1	1	2	1	0.612	0.071
1	2	2	1	0.822	0.474
2	2	2	1	-0.204	0.354
2	3	2	1	0.730	-0.632
3	3	2	1	-0.408	-0.424
2	2	3	1	0.408	-0.101
2	3	3	1	0.873	0.378
3	3	3	1	-0.102	0.530
3	3	4	1	0.306	-0.177

TABLE B4 ($I=2$)

TABLE B3 ($I=\frac{3}{2}$)

L	L'	P	I	$k=1$	$k=2$	$k=3$
1	1	$\frac{1}{2}$	$\frac{3}{2}$	-1.118	0.500	0
1	2	$\frac{1}{2}$	$\frac{3}{2}$	0.387	-0.866	0.775
2	2	$\frac{1}{2}$	$\frac{3}{2}$	-0.671	-0.500	0.895
0	1	$\frac{3}{2}$	$\frac{1}{2}$	1.732	0	0
0	2	$\frac{3}{2}$	$\frac{1}{2}$	0	-2.236	0
1	1	$\frac{3}{2}$	$\frac{3}{2}$	-0.447	-0.400	0
1	2	$\frac{3}{2}$	$\frac{3}{2}$	0.693	-0.775	-0.346
2	2	$\frac{3}{2}$	$\frac{3}{2}$	-0.477	0	-0.894
2	3	$\frac{3}{2}$	$\frac{3}{2}$	0.566	-0.632	-0.283
3	3	$\frac{3}{2}$	$\frac{3}{2}$	-0.447	-0.600	0.447
1	1	$\frac{5}{2}$	$\frac{3}{2}$	0.671	0.100	0
1	2	$\frac{5}{2}$	$\frac{3}{2}$	0.794	0.592	0.076
2	2	$\frac{5}{2}$	$\frac{3}{2}$	-0.075	0.357	0.383
2	3	$\frac{5}{2}$	$\frac{3}{2}$	0.806	-0.338	0.227
3	3	$\frac{5}{2}$	$\frac{3}{2}$	-0.261	0.150	-0.671
2	2	$\frac{7}{2}$	$\frac{3}{2}$	0.447	-0.143	-0.064
2	3	$\frac{7}{2}$	$\frac{3}{2}$	0.828	+0.463	-0.069
3	3	$\frac{7}{2}$	$\frac{3}{2}$	0	+0.500	0.226
3	3	$\frac{7}{2}$	$\frac{3}{2}$	0.335	-0.250	-0.075

L	L'	P	I	$k=1$	$k=2$	$k=3$	$k=4$
2	2	0	2	-0.707	-0.598	1.414	-1.069
1	1	1	2	-1.061	0.418	0	0
1	2	1	2	0.473	-0.935	0.632	0
2	2	1	2	-0.589	-0.299	0	0.713
2	3	1	2	0.421	-0.535	-0.316	0.996
3	3	1	2	-0.472	-0.717	0.707	0.089
0	1	2	2	1.732	0	0	0
0	2	2	2	0	-2.236	0	0
1	1	2	2	-0.353	-0.418	0	0
1	2	2	2	0.725	-0.612	-0.414	0
2	2	2	2	-0.354	0.128	-0.808	-0.305
2	3	2	2	0.676	-0.571	-0.084	-0.798
3	3	2	2	-0.353	-0.179	0.354	-0.134
1	1	3	2	0.708	0.120	0	0
1	2	3	2	0.775	0.655	0.111	0
2	2	3	2	0	0.341	0.505	0.076
2	3	3	2	0.828	-0.175	0.242	0.326
3	3	3	2	-0.177	0.329	-0.471	0.089
2	2	4	2	0.471	-0.171	-0.101	-0.008
2	3	4	2	0.797	0.505	-0.100	-0.063
3	3	4	2	0.059	0.448	0.471	-0.030
3	3	5	2	0.353	-0.299	-0.118	0.004

TABLE B5 ($I=\frac{1}{2}$)

L	L'	P	I	$k=1$	$k=2$	$k=3$	$k=4$
2	2	$\frac{1}{2}$	$\frac{5}{2}$	-0.683	-0.535	1.095	-0.617
2	3	$\frac{1}{2}$	$\frac{5}{2}$	0.138	-0.378	-0.258	1.091
3	3	$\frac{1}{2}$	$\frac{5}{2}$	-0.488	-0.802	0.913	0.154
1	1	$\frac{3}{2}$	$\frac{5}{2}$	-1.025	0.374	0	0
1	2	$\frac{3}{2}$	$\frac{5}{2}$	0.520	-0.949	0.555	0
2	2	$\frac{3}{2}$	$\frac{5}{2}$	-0.537	-0.191	-0.313	0.705
2	3	$\frac{3}{2}$	$\frac{5}{2}$	0.528	-0.587	-0.262	0.326
3	3	$\frac{3}{2}$	$\frac{5}{2}$	-0.415	-0.441	0.091	-0.077
0	1	$\frac{5}{2}$	$\frac{5}{2}$	1.732	0	0	0
0	2	$\frac{5}{2}$	$\frac{5}{2}$	0	-2.236	0	0
1	1	$\frac{5}{2}$	$\frac{5}{2}$	-0.293	-0.428	0	0
1	2	$\frac{5}{2}$	$\frac{5}{2}$	0.741	-0.507	-0.445	0
2	2	$\frac{5}{2}$	$\frac{5}{2}$	-0.293	0.191	-0.704	-0.397
2	3	$\frac{5}{2}$	$\frac{5}{2}$	0.727	-0.498	+0.013	-0.798
3	3	$\frac{5}{2}$	$\frac{5}{2}$	-0.293	0.027	-0.517	-0.077
1	1	$\frac{7}{2}$	$\frac{5}{2}$	0.732	0.134	0	0
1	2	$\frac{7}{2}$	$\frac{5}{2}$	0.761	0.694	0.136	0
2	2	$\frac{7}{2}$	$\frac{5}{2}$	0.049	0.325	0.574	0.118
2	3	$\frac{7}{2}$	$\frac{5}{2}$	0.835	-0.071	0.236	0.447
3	3	$\frac{7}{2}$	$\frac{5}{2}$	-0.098	0.401	-0.304	0.103
2	2	$\frac{7}{2}$	$\frac{5}{2}$	0.488	-0.191	-0.130	-0.015
2	3	$\frac{7}{2}$	$\frac{5}{2}$	0.774	0.530	-0.121	-0.102
3	3	$\frac{7}{2}$	$\frac{5}{2}$	0.098	0.401	0.517	-0.044
3	3	$\frac{11}{2}$	$\frac{5}{2}$	0.073	-0.334	-0.152	0.007

TABLE B6 ($I=3$)

L	L'	P	I	$k=1$	$k=2$	$k=3$	$k=4$
3	3	0	3	-0.500	-0.866	1.080	0.213
2	2	1	3	-0.667	-0.495	0.926	-0.447
2	3	1	3	0.356	-0.463	-0.289	1.045
3	3	1	3	-0.458	-0.650	0.540	0.036
1	1	2	3	-1.000	0.346	0	0
1	2	2	3	0.548	-0.949	0.507	0
2	2	2	3	-0.500	-0.124	-0.463	0.670
2	3	2	3	0.585	-0.592	-0.211	0
3	3	2	3	-0.375	-0.274	-0.180	-0.107
0	1	3	3	1.732	0	0	0
0	2	3	3	0	-2.236	0	0
1	1	3	3	-0.250	-0.433	0	0
1	2	3	3	0.750	-0.433	-0.463	0
2	2	3	3	-0.250	0.227	-0.617	-0.447
2	3	3	3	0.756	-0.436	0.068	-0.739
3	3	3	3	-0.250	0.144	-0.540	-0.036
1	1	4	3	0.750	0.144	0	0
1	2	4	3	0.750	0.722	0.154	0
2	2	4	3	0.083	0.309	0.617	0.149
2	3	4	3	0.836	0	0.226	0.520
3	3	4	3	-0.083	0.433	-0.180	0.104
2	2	5	3	0.500	-0.206	-0.154	-0.020
2	3	5	3	0.756	0.546	-0.136	-0.134
3	3	5	3	0.125	0.361	0.540	-0.055
3	3	6	3	0.375	-0.361	-0.180	0.010

TABLE B7 ($I=\frac{7}{2}$)

L	L'	P	I	$k=1$	$k=2$	$k=3$	$k=4$
3	3	$\frac{1}{2}$	$\frac{7}{2}$	-0.490	-0.818	0.957	0.171
2	2	$\frac{3}{2}$	$\frac{7}{2}$	-0.654	-0.468	0.821	-0.358
2	3	$\frac{3}{2}$	$\frac{7}{2}$	0.404	-0.505	-0.295	0.967
3	3	$\frac{3}{2}$	$\frac{7}{2}$	-0.436	-0.546	0.319	-0.019
1	1	$\frac{5}{2}$	$\frac{7}{2}$	-0.982	0.327	0	0
1	2	$\frac{5}{2}$	$\frac{7}{2}$	0.567	-0.945	0.474	0
2	2	$\frac{7}{2}$	$\frac{7}{2}$	-0.473	-0.078	-0.547	0.637
2	3	$\frac{7}{2}$	$\frac{7}{2}$	0.622	-0.583	-0.171	-0.186
3	3	$\frac{7}{2}$	$\frac{7}{2}$	-0.346	-0.164	-0.319	-0.108
0	1	$\frac{7}{2}$	$\frac{7}{2}$	1.732	0	0	0
0	2	$\frac{7}{2}$	$\frac{7}{2}$	0	-2.236	0	0
1	1	$\frac{7}{2}$	$\frac{7}{2}$	-0.218	-0.436	0	0
1	2	$\frac{7}{2}$	$\frac{7}{2}$	0.873	-0.378	-0.474	0
2	2	$\frac{7}{2}$	$\frac{7}{2}$	-0.218	0.249	-0.547	-0.478
2	3	$\frac{7}{2}$	$\frac{7}{2}$	0.774	-0.387	0.103	-0.673
3	3	$\frac{7}{2}$	$\frac{7}{2}$	-0.218	0.218	-0.522	-0.007
1	1	$\frac{9}{2}$	$\frac{7}{2}$	0.609	0.153	0	0
1	2	$\frac{9}{2}$	$\frac{7}{2}$	0.742	0.742	0.169	0
2	2	$\frac{9}{2}$	$\frac{7}{2}$	0.087	0.296	0.647	0.174
2	3	$\frac{9}{2}$	$\frac{7}{2}$	0.835	0.052	0.215	0.567
3	3	$\frac{9}{2}$	$\frac{7}{2}$	-0.044	0.447	-0.087	0.102
2	2	$\frac{11}{2}$	$\frac{7}{2}$	0.508	-0.218	-0.174	-0.025
2	3	$\frac{11}{2}$	$\frac{7}{2}$	0.742	0.556	-0.148	-0.161
3	3	$\frac{11}{2}$	$\frac{7}{2}$	0.146	0.327	0.551	-0.063
3	3	$\frac{13}{2}$	$\frac{7}{2}$	0.382	-0.382	-0.203	0.012

TABLE B8 ($I=4$)

L	L'	P	I	$k=1$	$k=2$	$k=3$	$k=4$
3	3	1	4	-0.484	-0.783	0.874	0.145
2	2	2	4	-0.645	-0.448	0.749	-0.304
2	3	2	4	0.436	-0.530	-0.295	0.900
3	3	2	4	-0.420	-0.470	0.175	-0.048
1	1	3	4	-0.968	0.313	0	0
1	2	3	4	0.581	-0.940	0.449	0
2	2	3	4	-0.452	-0.045	-0.599	0.609
2	3	3	4	0.647	-0.571	-0.139	-0.304
3	3	3	4	-0.323	-0.085	-0.397	-0.101
0	1	4	4	1.732	0	0	0
0	2	4	4	0	-2.236	0	0
1	1	4	4	-0.194	-0.439	0	0
1	2	4	4	0.760	-0.335	-0.481	0
2	2	4	4	-0.194	0.265	-0.490	-0.498
2	3	4	4	0.786	-0.347	0.126	-0.614
3	3	4	4	-0.194	0.269	-0.493	0.013
1	1	5	4	0.774	0.160	0	0
1	2	5	4	0.735	0.757	0.181	0
2	2	5	4	0.129	0.285	0.667	0.194
2	3	5	4	0.832	0.092	0.205	0.601
3	3	5	4	-0.032	0.453	-0.016	0.098
2	2	6	4	0.632	-0.228	-0.191	-0.030
2	3	6	4	0.730	0.564	-0.157	-0.184
3	3	6	4	0.161	0.299	0.556	-0.069
3	3	7	4	0.387	-0.399	-0.222	0.014

TABLE 3
 Correction factors for $2'' \times 2''$ crystals.
 Calculated by E. Matthias using the present programme

h (cm)	E (MeV)	ϵ		Q_2		Q_4	
		Whole spectrum	Photopeak	Whole spectrum	Photopeak	Whole spectrum	Photopeak
10.0	1.5	0.4163	0.1108	0.9657	0.9692	0.8888	0.8999
	1.0	0.4781	0.1648	0.9653	0.9686	0.8876	0.8979
	0.7	0.5417	0.2449	0.9649	0.9678	0.8861	0.8955
	0.5	0.6116	0.3539	0.9642	0.9671	0.8842	0.8933
	0.3	0.7523	0.6167	0.9624	0.9643	0.8785	0.8845
	0.2	0.8640	0.8103	0.9597	0.9607	0.8700	0.8732
	0.15	-	-	0.9576	0.9576	0.8635	0.8646
	0.10	0.9694	0.9603	0.9557	0.9557	0.8573	0.8575
	0.05	0.9950	0.9933	0.9545	0.9545	0.8538	0.8538
	0.03	0.9907	0.9869	0.9547	0.9547	0.8544	0.8544
7.0	0.02	0.9971	0.9958	0.9544	0.9544	0.8535	0.8535
	1.5	0.3796	0.0993	0.9378	0.9440	0.8032	0.8222
	1.0	0.4375	0.1485	0.9369	0.9441	0.8006	0.8225
	0.7	0.4976	0.2202	0.9359	0.9422	0.7976	0.8167
	0.5	0.5648	0.3265	0.9346	0.9403	0.7935	0.8109
	0.3	0.7055	0.5736	0.9306	0.9345	0.7815	0.7932
	0.2	0.8288	0.7757	0.9246	0.9265	0.7636	0.7694
	0.10	0.9599	0.9507	0.9152	0.9153	0.7358	0.7363
	0.05	0.9934	0.9917	0.9124	0.9124	0.7278	0.7278
	0.03	0.9877	0.9839	0.9129	0.9129	0.7292	0.7293
5.0	0.02	0.9961	0.9948	0.9122	0.9122	0.7272	0.7272
	1.5	0.3472	0.0925	0.8947	0.9057	0.6801	0.7114
	1.0	0.4014	0.1421	0.8930	0.9046	0.6754	0.7086
	0.7	0.4584	0.2017	0.8911	0.9016	0.6699	0.7001
	0.5	0.5230	0.3007	0.8885	0.8981	0.6626	0.6901
	0.3	0.6633	0.5366	0.8808	0.8879	0.6411	0.6612
	0.2	0.7963	0.7439	0.8693	0.8727	0.6094	0.6188
	0.10	0.9510	0.9419	0.8503	0.8505	0.5587	0.5593
	0.05	0.9919	0.9902	0.8444	0.8444	0.5436	0.5436
	0.03	0.9850	0.9813	0.8455	0.8455	0.5462	0.5463
	0.02	0.9952	0.9939	0.8439	0.8439	0.5423	0.5424

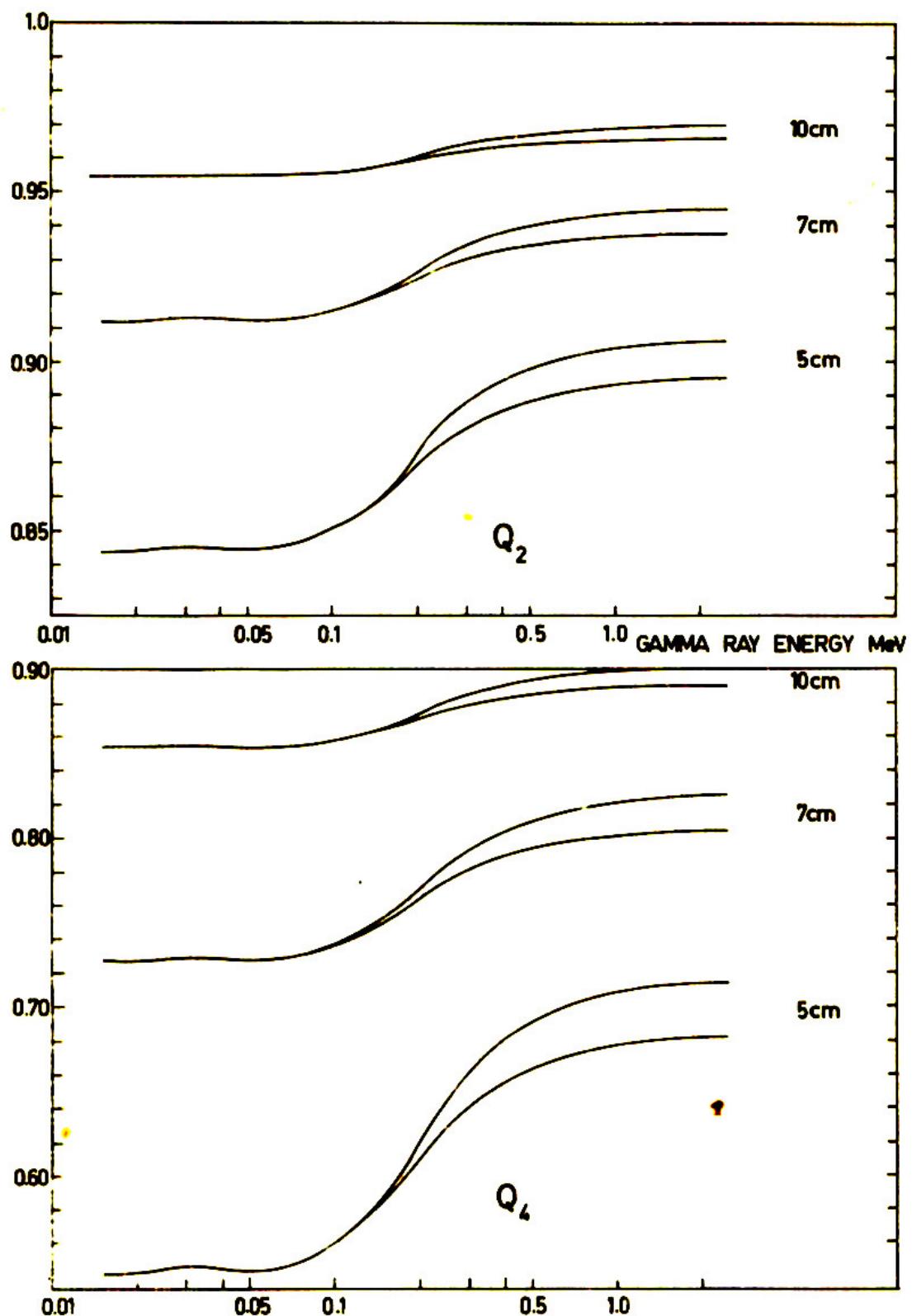


Fig. 4. Correction factors for 2" dia \times 2" long crystals. Calculated by E. Matthias using the present programme