GAMMA - GAMMA COINCIDENCE AND DIRECTIONAL CORRELATION STUDIES

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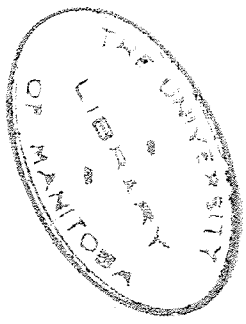

by

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ABSTRACT

A coincidence spectrometer suitable for directional correlation measurements has been used to investigate the decay schemes of several radioisotopes. High energy selectivity was secured through the use of two differential discriminators and conventional scintillation counters. The resolving time of the coincidence stage was about 1.1×10^{-7} seconds.

Reports of additional positron and gamma ray components accompanying the decay of Zn^{65} have been discounted by coincidence experiments carried out with the above spectrometer. Instrumental effects have been shown to give rise to asymmetrical time resolution curves.

The directional correlation between the two strong lines in the gamma ray spectrum of Se^{75} has been studied. The observed correlation is consistent with the spin assignment 5/2 (99.12% M1; .88% E2) 7/2 (E2) 3/2 for the levels concerned, if it is assumed that an interfering cascade is weak. Possible re-interpretations of the present data will have to await more accurate intensity measurements for the transitions involved.

The gamma ray spectrum of Sb^{124} was investigated, using a single channel spectrometer. Two new gamma rays with energies of 965 and 1365 kev were found during the course of the investigation. It was confirmed that the 1690 and 2090 kev gamma radiations were in coincidence with the 600 kev transition. The directional correlation of the 1690 - 600 kev cascade was studied and found to be consistent with a spin sequence 3 (D) 2 (E2) 0 for the levels concerned. The E1 assignment for the first transition, which has appeared in the literature, was discussed.

The directional correlation of the combined 67 - 1122 kev and 67 - 1222 kev cascades in W^{182} has been investigated. The results were discussed in the light of two independent spin assignments which have appeared in the literature. The results obtained favor one of these assignments slightly, but they are not conclusive.

Spins have been assigned to five states of the Pt^{192} level scheme on the basis of directional correlation studies undertaken with the 468 - 316 kev and 308 - 296 - 316 kev cascades. The net spin assignment 4 - 4 - 2 - 2 - 0 seems to be consistent with all the known details of the decay scheme of Ir^{192}. The expected correlation function for the triple gamma ray cascade was calculated and found to agree with experiment if the $2 \rightarrow 2$ (296 kev) transition is a 2.5% M1 - 97.5% E2 multipole mixture.

CHAPTER I.

THEORETICAL CONSIDERATIONS

a) Introduction

One of the principal aims of modern experimental nuclear physics is to supply a foundation of measurement upon which a theory of nuclear forces may be erected. To this end, a variety of techniques have been developed to determine the properties of nuclear energy levels. One of the most recent of these is the study of the angular correlation between particles or gamma rays emitted in cascade by an excited nucleus. Theoretical considerations show that when two radiations are emitted in this way, the directions of emission are not oriented randomly but are spatially correlated. Furthermore, the character of this correlation is determined solely by the spins or the spin-parity assignments of the nuclear states involved in the emission process. It is apparent, then, that by a careful study of the spatial correlation between nuclear radiations, one can learn something about the spins and parities of nuclear energy levels. Of course, this is not the only technique by which they may be determined. The shape of a β-ray spectrum, or the conversion coefficient of a gamma ray may yield the same information. It has been found, however, that angular correlation studies lead to less ambiguous results than either of the above techniques. This is particularly evident for gamma ray transitions involving mixed multipolarities. In this case the observed conversion coefficient will have a value lying somewhere between the theoretical values corresponding to pure multipole transitions. The relative intensities of the two types of radiation present in the transition cannot be unambiguously determined from these data. However, the angular correlation between this mixed radiation and some other radiation associated with it, might well be

expected to yield the mixing ratios with a fair degree of accuracy. This will be demonstrated in later sections of this thesis.

As mentioned above, the angular correlation between nuclear radiations is determined by the spins and parities of the nuclear states concerned. It is important to distinguish between two kinds of angular correlation experiment, however. Suppose, for example, we consider a simple two-component gamma ray cascade. If the spatial correlation between the two radiations is measured, a "directional" correlation experiment has been performed. On the other hand, if the polarization of one of the radiations is measured also, then a "polarization-direction" experiment has been performed. The former is dependent only on the spins of the nuclear levels, while the latter determines the relative parities of nuclear states. Hence it has become customary to refer to an experiment as a directional correlation investigation if the detectors are polarization insensitive, and as a polarization-direction experiment if the detectors are polarization sensitive. The term 'angular correlation' embraces both cases. This terminology will be used throughout the remainder of this treatise.

The purpose of the present work is to present gamma-gamma directional correlation and coincidence data for several selected radioisotopes. Since the experiments have been restricted to the study of radiative transitions, theoretical discussions will not be presented in their most general form, but rather in that form which pertains to radiative processes only. Before proceeding with this discussion, however, a brief outline of the history of the phenomenon will be given. This will be followed by the theoretical aspects of the subject mentioned above,

and an outline of the coincidence technique, errors, angular corrections, etc. A discussion of the apparatus, and the experimental results will be reserved for Chapters II and III respectively. A brief summary in Chapter IV will complete the work.

b) Historical Note

The theory of the angular correlation of nuclear radiations is one of the better developed physical theories. However, until recently, the experimental techniques had lagged behind their theoretical counterpart to a considerable extent. This was due to 1) a lack of sensitive detection equipment and 2) an unawareness of the care with which correlation measurements should be made. The first defect has been overcome by the wide use of scintillation spectrometers; the second deficiency still persists to some extent, although in recent months it has become evident from the literature on the subject that more attention is now being paid to the collection and correction of experimental data. However, much of the early work must now be regarded as ambiguous due to faulty technique. Some examples of this will be discussed in the later sections of this thesis. They will indicate clearly how much progress has been made in the last year or two.

Prior to 1940 there existed neither experimental evidence of, nor theoretical reason to expect, a spatial correlation between nuclear radiations emitted in cascade. In the subsequent thirteen years, however, the theoretical aspects of the subject have been developed to a high degree of perfection. The first substantial contribution in this regard came in 1940 when Hamilton (1) presented a theoretical paper on the

gamma-gamma directional correlation. In the same year, Dunworth (2) mentioned the possibility of directional experiments in his classic paper on coincidence techniques. After a lapse of some six years, a generalization of Hamilton's results was given by Goertzel (3) who, in discussing the effect of a magnetic field on a directional correlation, laid the foundation for the group-theoretic methods currently in vogue. The next important steps on the experimental scene were taken in 1942-43 by Kikuchi et al., and Beringer (4). Of the isotopes studied by these authors, only one - Cl^{38} - gave a result which indicated the existence of a directional correlation. For the others, any existing correlation was smaller in magnitude than the experimental errors. In 1947-48, however, Brady and Deutsch (5), using Geiger counters, finally confirmed the theoretical predictions for Co^{60}, Sc^{46}, Y^{86}, Cs^{134}, Na^{24} and Rh^{106}. In 1950, these authors (6) repeated their experiments using the newly-developed scintillation counters. Once again, the results were in good agreement with theory and the way was at last opened to the securing of reliable data. By this time, largely through the efforts of Racah, Lloyd, Falkoff and others, the theoretical aspects of the subject were finally consolidated using the techniques of group theory. In 1953, Biedenharn and Rose (7) published a long review article on the mathematical formulation, which was intended to be a source book for the experimentalist. On the experimental side, however, the picture was less promising. In the same year, the interpretation of the gamma-gamma directional correlation in Ni^{60} was again in doubt, and had to be settled once for all by an excellent experiment performed by Klema and McGowan (8) at Oak Ridge. Since the Ni^{60} cascade was the best known and most extensively studied gamma-ray cascade as far as correlation measurements were concerned, it

was apparent, prior to Klema's paper, that the experimentalists had a long way to go to equal the accomplishments of the theorists. At the present time the experimental picture is considerably brighter, however. The quality of the work done in the last year indicates that substantial progress has been made toward attaining the precision necessary for the elucidation of complex decay schemes.

Although brief, the above outline of the historical development of the subject should give the reader some perspective on the subject as a whole; that, at least, was its intention. Several of the remarks made regarding the inaccuracy of early experiments will come up for further elaboration in later sections. We shall now consider the theory proper.

c) The Method of D. R. Hamilton (1)

Before proceeding with the formulation due to Hamilton, it will be profitable to consider in qualitative terms why there should be a directional correlation between nuclear radiations. In addition, a tabulation of the characteristics of nuclear radiative transitions will be given.

Consider a large number of nuclei, all of which are in the same state. This state is characterized by a total angular momentum j, whose component along a reference axis is m. All the nuclei are eventually to undergo radiative transitions to a final state with arbitrary spin. Due to the $(2j + 1)$-fold degeneracy of the initial state, there will be a set of substates characterized by values of m. Since each substate is associated with the same energy, and if there are no external magnetic or electric fields present, then the substates will be equally populated. If transitions occur from the state j, the radiation from the ensemble will be isotropic,

since, although the radiation from a single transition is emitted anisotropically, the equal population of the substates leads to a net intensity independent of angle. If there is a magnetic field present, however, the degeneracy is removed (Zeeman effect) and the m substates in the ensemble become populated according to the Boltzmann distribution law. When transitions occur, the net effect is an anisotropy with respect to the magnetic field axis, since the substates are now unequally populated.

In the case of a coincidence measurement involving two correlated gamma rays, the selection of a definite direction in space by means of one detector means that only a particular set of transitions from the substates can be studied, viz. those whose emission directions pass through the detector. The situation is similar to that with the magnetic field mentioned above. In the coincidence experiment, the selection of a particular direction introduces an anisotropy into space, which prevents the substates from contributing equally to the radiation field detected experimentally, even though the substates are equally populated. Thus the Zeeman effect is strongly analogous to the directional correlation phenomenon.

The presentation of the above analogy is intended to give the reader an intuitive feeling for the phenomenon. This is most desirable. A purely mathematical argument is often less satisfying to the experimentalist, who invariably thinks in terms of a model. We pass now to a brief review of the selection rules for radiative processes in general.

When a nuclear state undergoes a radiative transition, the total

angular momentum (referred to as 'spin' hereinafter; denoted by $\underline{j}$) and the parity (π) of the resultant state are in general different from those of the former. Mathematical analysis of the expression for the transition probability indicates that both magnetic and electric multipole radiations may occur; the order of the multipole being determined by the spin change, and the character of the radiation by the parity change. The multipole order and parity of the radiations are not completely independent, as will be seen later.

If $\underline{j}_i$ and $\underline{j}_f$ are the spin vectors of the initial and final states respectively, and if the gamma ray carries away $\underline{L}$ units of angular momentum, the following selection rules can be shown to hold (9):

$$|j_i - j_f| \leq L \leq j_i + j_f$$

$$\pi_i = \pi_f \quad \text{for even parity radiation}$$

$$\pi_i = -\pi_f \quad \text{for odd parity radiation.}$$

where $|\underline{j}_i| = \hbar\sqrt{j_i(j_i + 1)}$ etc. and the transition $j_i = 0 \rightarrow j_f = 0$ is absolutely forbidden. The lowest order of multipole radiation consistent with these selection rules has been assumed for the transition. The following table summarizes the results in a convenient form.

Lowest Order of Multipole Radiation in a Transition j_i, π_i to j_f, π_f.

a) $j_i \neq j_f$

	Electric (E) radiation	Magnetic (M) radiation
Parity favored, $\pi_i \pi_f = (-1)^{j_i - j_f}$	$L = \lvert j_i - j_f \rvert$	$L = \lvert j_i - j_f \rvert + 1$ except $j_i = j_f = 0$
Parity unfavored, $\pi_i \pi_f = (-1)^{j_i - j_f + 1}$	$L = \lvert j_i - j_f \rvert + 1$ except j_i or $j_f = 0$	$L = \lvert j_i - j_f \rvert$

b) $j_i = j_f \neq 0$

$\pi_i = \pi_f$	$L = 2$ except $j_i = j_f = 1/2$	$L = 1$
$\pi_i = -\pi_f$	$L = 1$	$L = 2$ except $j_i = j_f = 1/2$

Since frequent use will be made of this table in discussing experimental results, it was felt necessary to reproduce it here. A full discussion of radiative processes cannot be given, however. For further details, the reader is referred to reference (9). We now proceed with the discussion of the main topic of this section.

Hamilton's approach to the correlation problem involves the standard techniques of time-dependent perturbation theory. The clarity of his calculations permits the experimentalist to follow the arguments without the necessity of first reading extensively of the literature on group theory. For this reason alone, it will be profitable to present Hamilton's arguments here. Following that, a summary of the results obtainable using group-theoretic methods will be given.

Throughout this thesis, the following notation will be used:

j_1, j, and j_2 are the angular momentum quantum numbers corresponding to nuclear states A, B and C respectively; m_1, m and m_2 are the quantum numbers for the z-components of $\underline{j}_1$, $\underline{j}$ and $\underline{j}_2$. The states A, B and C will describe not only the nucleus but the radiation field as well. When the nucleus is coupled to the field, a radiative transition is assumed to occur yielding a photon of a given type. The further coupling of the resultant nuclear state to the radiation field then results in another transition and the appearance of a photon of a second given type. In the final state C, the nucleus and the radiation field are assumed to be decoupled.

In carrying out his calculations, Hamilton assumes that
a) the intermediate state is unperturbed by external influences,
b) the substates of A and C are uniformly populated,
c) the emitted gamma radiations have pure multipolarities, and that
d) the detection equipment is to be polarization insensitive.
With the exception of the 'postulate of purity', these conditions are usually fulfilled in a normal case. However, for a triple gamma ray cascade, (b) does not hold since the sublevels of the second state are populated according to the transition probabilities for the different components of the first gamma ray.

Hamilton starts his solution by writing down the differential equations for the probability amplitudes of the three states A, B and C. He assumes a series solution for each amplitude and by direct substitution finds those series parameters which are consistent with the initial equations. The matrix elements for the entire process are then written down formally, and expressed in terms of the above parameters. The probability of the de-excitation of the nucleus through radiative transitions yielding two photons is written as

$$W(\underline{k}'_o\ \underline{e}'\ ,\ \underline{k}''_o\ \underline{e}'') = \sum_{m_2\ \underline{e}'\underline{e}''} \left\langle |C_{m_2}|^2 \right\rangle_{av} \qquad (1)$$

where C_{m_2} is the probability amplitude corresponding to state C, $\underline{k}'_o$ $\underline{k}''_o$ are propagation vectors for the first and second photons respectively, and $\underline{e}'$, $\underline{e}''$ are their corresponding polarization vectors. The summation extends over all the final magnetic substates and polarization directions. By using the definition of C_{m_2} as given by Hamilton, equation (1) can be rewritten in the form:

$$W = \sum_{m_1 m_2 \underline{e}'\underline{e}''} \left| \sum_m (A_{m_1} | H(\underline{k}'_o\ \underline{e}') | B_m)^* (B_m | H(\underline{k}''_o\ \underline{e}'') | C_{m_2})^* \right|^2 \qquad (2)$$

where H is the interaction term in the Hamiltonian operator, and the brackets (| |) are matrix elements describing transitions between adjacent states. It is apparent from equation (2) that the summation over m produces a number of interference terms, i.e. terms which contain a product of two matrix elements, each of which refers to a different transition. These interference terms can be removed by taking the direction of emission of the first gamma ray along the z-axis. Hamilton proves this as a theorem, the statement of which is: If the z-axis of quantization is along the emission direction of the first gamma ray, the probability of the double transition is given by the product of the probabilities of the separate single transitions. The reader is referred to Hamilton's paper (1) for the mathematical proof. Lippman (10) has given the following qualitative proof: The interference terms arising in a transition involving an intermediate state are due to a lack of information concerning that state. If, however, the first gamma ray moves along the z-axis, then its z-component of angular momentum can be measured without disturbing the system. Since the initial state is known in detail, it is possible to determine the pertinent characteristics of the intermediate state, and hence, for this case, no interference can occur.

Making use of this theorem, W can be written as:

$$W = \sum_{\substack{m_1 m\, m_2 \\ \underline{e}'\underline{e}''}} \left|(A_{m_1} \mid H(0) \mid B_m)\right|^2 \left|(B_m \mid H(\theta) \mid C_{m_2})\right|^2 \qquad (3)$$

where $\underline{k}_o'$ has been taken along the z-axis and θ is the angle between this axis and $\underline{k}_o''$. Thus any angular dependence in W comes from the matrix element for the second transition. It is convenient to define

$$P_{m_1 m}(0) = \sum_{\underline{e}'} \left|(A_{m_1} \mid H(0) \mid B_m)\right|^2$$

and

$$P_{m\, m_2}(\theta) = \sum_{\underline{e}''} \left|(B_m \mid H(\theta) \mid C_{m_2})\right|^2$$

then W becomes

$$W = \sum_{m_1 m\, m_2} P_{m_1 m}(0)\; P_{m\, m_2}(\theta) \qquad (4)$$

The P's are the probabilities for the transitions concerned. It can be shown (1) that

$$P_{m\, m_2}(\theta) = C(j\, L_2 j_2;\; m\; m_2 - m)\; F^M_{L_2} \quad , \; etc.$$

where $\underline{L}_1$, $\underline{L}_2$ are the angular momenta for the first and second photons respectively, $M = m_2 - m$, and the C's are the vector addition coefficients. $F^M_{L_2}$ is a function of (θ) only. The notation for the C's given above is related to that of Condon and Shortley (11) by: $C(a\ b\ c;\ d\ e) \equiv (a\ b\ d\ e \mid a\ b\ c\ d+e)$. The F-function in $P_{m_1 m}(0)$ is unity because we have chosen the z-axis as the direction of emission of the first photon.

Hamilton was able to carry out the summations indicated in equation (4), and tabulate the following results:

second transition dipole: $$W(\theta) = 1 + \frac{R'}{Q'} \cos^2 \theta \qquad (5)$$

second transition quadrupole: $$W(\theta) = 1 + \frac{R}{Q} \cos^2 \theta + \frac{S}{Q} \cos^4 \theta \qquad (6)$$

(Since only the dependence of W on θ is important for a directional correlation measurement, several common factors have been dropped from (5) and (6) in order to reduce them to simple forms.) In subsequent