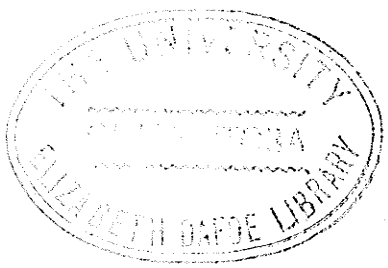
GAMMA - GAMMA COINCIDENCE
and ANGULAR CORRELATION INVESTIGATIONS

A Thesis
Submitted to the
Faculty of Graduate Studies
University of Manitoba
in partial fulfillment of the
requirements for the degree of
Master of Science (Physics)

by

ANDREW JOHN DILAY

July, 1965



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ABSTRACT

A gamma-gamma coincidence and 11-position angular correlation spectrometer capable of automatic operation is described and tested. In the decay of Se^{75}, gamma spectra in coincidence with the 122 kev and the weak 199 kev gamma rays have been investigated by a fast-slow coincidence technique (2τ = 70 nanoseconds). A gamma-gamma angular correlation run is performed on the 199-66 kev cascade and the value of $\frac{1}{2}$ for the spin of the 199 kev level in As^{75} is substantiated. Finally, some features of the high-energy (> 402 kev) region are described.

C O N T E N T S

		Page
	List of Plates and Figures	iii
	Preface	iv
	Abstract	v
Chapter 1.	INTRODUCTION	1
Chapter 2.	THE SCINTILLATION SPECTROMETER	6
Chapter 3.	THEORETICAL CONSIDERATIONS	11
3.1	Emission of Gamma Rays	11
	Multipoles	11
	Selection Rules	12
3.2	Gamma-Gamma Angular Correlations	13
	Gamma-Gamma Directional Correlations	13
	Geometrical Corrections	16
Chapter 4.	THE APPARATUS	19
4.1	General Description	19
4.2	Individual Descriptions	22
	The Detectors	22
	The Linear Amplifiers	25
	The Fast Speed Discriminator and Single Channel Analyzer	25
	The Fast-Slow Coincidence Base Unit	26
	The Biased Amplifier	27
	The Pulse Generator	27
	The Angular Correlation Spectrometer	28

Chapter 5.	PERFORMANCE TESTS	31
5.1	The Linear Amplifiers	31
5.2	The Single Channel Analyzer	31
5.3	The Fast Speed Discriminator and Coincidence Base Unit	34
5.4	Angular Correlation Spectrometer	38
	Co^{60} Asymmetry	38
	Na^{22} Isotropy	42
Chapter 6.	SELENIUM 75 INVESTIGATIONS	46
6.1	General	46
6.2	Gating on 122 kev Peak	51
6.3	Gating on 199 kev Peak	53
6.4	High Energy Region	61
6.5	Conclusion	65
	References	68

LIST OF PLATES AND FIGURES

		Page
PLATE 1.	Angular Correlation Spectrometer and Control Unit, NaI(Tl) detectors and Accompanying Electronics	21
2.	Fast-Slow Coincidence Spectrometer Control Panel	24
FIGURE 1.	Schematic Diagram of Gamma-Gamma Coincidence and Angular Correlation Spectrometer	20
2.	Linearity of Linear Amplifier #1	32
3.	Compton Portion of Co^{60} Spectrum with Corresponding Window of F.S.D. #1	33
4.	Delay Curve of F.S.D. #1, F.S.D. #2 Fixed	37
5.	Co^{60} Asymmetry	41
6.	Level Scheme of As^{75} Due to Decay of Se^{75}	47
7.	Se^{75} Singles Spectrum to 402 kev	48
8.	Low Energy Calibration Curve for NaI(Tl) Crystal	50
9.	Se^{75} Singles Spectrum (Gating Side) and Gate on 199 kev	55
10.	Se^{75} Coincidence Spectrum (Gating on 199 kev)	56
11.	Se^{75} Singles Spectrum (> 402 kev)	64

PREFACE

The work described herein was done at the University of Manitoba during 1964 and 1965.

The author wishes to express his heartfelt thanks and appreciation to Dr. S. I. H. Naqvi for his valuable advice, ideas, and his constant encouragement. The author is also indebted to Mr. W. R. Wall for his enlightening explanations on various topics in electronics, to Mr. R. S. Foulds for his aid in removing the "bugs" from the electronics, and to Drs. R. D. Connor, B. G. Hogg, and S. K. Sen for the loan of various scintillation detectors. Finally, the author would like to thank his fiancee, Miss Aloma Kadychuk, for her painstaking work and infinite patience in typing this thesis.

A research grant from the National Research Council of Canada supported the work and is gratefully acknowledged.

Chapter 1.

INTRODUCTION

A fundamental purpose of nuclear physics studies is the obtaining of information which will make it possible to understand the elemental basis of nuclear structure. Such an understanding is absolutely necessary if man desires to utilize fully the potentialities of the atomic nucleus, either as an energy source or as a means for providing knowledge that will contribute to the enrichment of human life.

A theoretical treatment of nuclear structure is considerably more complex than a corresponding treatment of atomic structure, primarily because the basic law of nuclear force still remains, to a great part, a mystery. With the advancement of the quantum theory came an essentially complete solution to the atomic problem since the only difficulties met lay in the application of quantum mechanics to systems governed entirely by the well known laws of electrodynamics. However, we have only the very barest phenomenological picture of the forces acting between nucleons. In fact, there is even doubt as to whether

we can justifiably regard the nucleons as elementary particles. Consequently, although there has been gathered an abundance of data and facts about nuclei, we have as yet been unsuccessful in our attempts to unite the facts into a consistent theory of the nucleus. We can merely describe various approaches to the problem, indicate the extent to which they have been successful, and also point out where they have broken down.

Historically, Becquerel's discovery of natural radioactivity in 1896, an indirect consequence of Roentgen's discovery of x-rays several months earlier, marked the birth of nuclear physics. Subsequent investigations divided the radiation into three distinct types now known as alpha, beta and gamma rays. In 1911 Ernest Rutherford laid the foundation of the modern theory of atomic structure with his classical paper in which he postulated that the atom consists not of a uniform sphere of positive charge (J. J. Thomson's plum-pudding model), but that all positive charge is concentrated in a relatively small region at the centre of the atom. This region was later called the nucleus of the atom. In 1919 Lord Rutherford broke up certain of these atomic nuclei and in 1934 Joliet and Curie showed that some of these artificial

nuclei were themselves radioactive, thus initiating an entirely new branch of research. Further studies have strongly pointed to the existence of energy levels within the nucleus itself, just as spectroscopic investigations showed the existence of atomic energy levels. This thesis will be concerned with a branch of nuclear spectroscopy, which is generally concerned with systematic arrangement and identification of the aforementioned nuclear energy levels.

In particular, we consider gamma ray spectroscopy. Gamma rays are electromagnetic radiation emitted from certain atomic nuclei due to some type of rearrangement within the nuclei leading to a lower energy content. Under the suppositions that there are a number of discrete energy levels within the nucleus and that ordinarily a nucleus is in the ground state, we consider the circumstances which must prevail in order for a nucleus to exist in an excited state. Experimentally, we find that alpha emitters exhibiting discrete alpha spectra are also gamma emitters, and similarly beta emitters are often gamma emitters. Subsequent energy measurements of the various radiations, together with the theory of beta decay, indicated that gamma emission is a result of a product nucleus being left in an excited state, i.e. a nucleus in its ground state cannot emit any gamma

radiation, so gamma decay occurs only as a consequence of those instances of alpha or beta emission, or electron capture, where the product nucleus is left in an excited state. A nucleus can also be excited in a scattering experiment, in which case it will emit gamma rays if the degree of excitation is insufficient for it to emit a particle. This implies that the last stages of most nuclear reactions involve gamma emissions.

Gamma ray spectroscopy provides a major source of knowledge of low-lying nuclear energy levels. This follows from the fact that gamma rays carry away angular momentum and explain changes in angular momentum, parity, and energy between the excited levels of a nucleus. Various specialized experimental techniques involve variations of what is referred to as the coincidence method, first introduced by Bothe and Geiger in 1925. One observes two or more ionizing events as detected by two or more respective detectors, which events have taken place within a predetermined time interval. In particular, we shall apply the coincidence method to gamma-gamma angular correlation studies wherein we shall record the coincidence counting rate of two successive gamma rays as a function of the angle between the propagation vectors of the gamma rays. Such a measurement enables one to calculate,

under favorable circumstances, the angular momenta of the relevant energy levels.

A pair of NaI(Tl) scintillation spectrometers served as the detectors in the coincidence and angular correlation work. The scintillation counter is discussed in the following chapter.

Chapter 2.

THE SCINTILLATION SPECTROMETER

Gamma ray spectroscopy is vitally dependent on accurate determinations of the gamma energies. However, one cannot, in general, use the same principles in gamma energy determinations as are used in energy determinations of charged particles. In the first place, gamma photons are uncharged, thus ruling out magnetic spectrometer methods. Secondly, the absorption of gamma rays by matter takes place not in a continuous fashion as does absorption of charged particles, but rather in a "yes - no" process in which aborption and scattering occur in single events. This implies that in observing a well collimated beam of gamma rays incident on some absorber, the collimated gamma rays which remain unattenuated have experienced virtually no absorption nor scattering, and conversely, any gamma rays which have experienced either absorption or scattering will be removed from the beam. i.e. There is either complete attenuation or none at all. Obviously this rules out energy determinations based on range measurements.

Techniques in gamma energy determinations must be based on interactions between gamma rays and matter and it

is found that the most significant of the possible interactions are the photo-electric effect, the Compton effect, and pair production. The definitions and discussions of these interactions can be found in many books on nuclear physics and in many theses (1), (2), (3), (4).

Since the end of World War II there have been many notable advancements in one of the earliest techniques of particle detection. This technique depends on the fact that certain materials scintillate on exposure to nuclear radiations. In particular, if a gamma ray is incident upon certain crystals, a rather complex process takes place, involving the initial formation of an excited electronic state of a small region of the crystal. This excess energy is promptly emitted as a very small flash of light, a scintillation.

Before the 1930's nuclear physicists had no efficient vacuum-tube circuits to simplify the counting of electrical pulses, and the scintillation method, involving wearisome visual counting and recording, was the sole method of alpha particle study. With the advent of vacuum-tube circuits and the Geiger-Müller tube, the scintillation method fell into disuse, but two developments in the mid-1940's hailed it back into popularity where it has remained ever since. First came the discovery of "phosphors" which not only scintillated on exposure to radiation, but which

were transparent to the light given off. This meant that one could use large volumes of these "phosphors" and thus enjoy an absorption efficiency far higher than ever before possible. A second obstacle to be overcome was the fact that a single scintillation is too feeble to be measured directly. The development of sensitive, efficient, high gain photomultiplier tubes (P. M. tubes) solved this problem.

Curran and Baker (5) in 1944 used a ZnS screen and an RCA 1P21 P. M. tube, connected to an oscilloscope, and detected 2 Mev alpha particles. Marshall and Coltman (6), (7), (8), in 1947-48 used a P. M. scintillation detector with a well-designed optical system to detect and count alpha particles, protons, fast electrons, gamma rays, x-rays, and neutrons. The main incentive to the scintillation method was provided in 1947 by H. Kallmann(9), who used a large clear crystal of naphthalene as a scintillator, and detected scintillations by beta and gamma rays with a P. M. tube. In 1948 Bell (10) found that crystalline anthracene is even better than naphthalene, giving pulses five times as great. It was then discovered (11) that there exists a class of scintillators, inorganic substances, which do not scintillate in the pure state but which scintillate strongly when "activated" with a certain substance. In particular Hofstadter (11) used thallium-activated sodium iodide crystals which produced pulses even

greater than anthracene, and the presence of the heavy element iodine made NaI(Tl) crystals especially useful for gamma ray detection. This follows because the high density NaI has a relatively high stopping power for gamma rays and has high photoelectric cross section and thus exhibits a high absorption efficiency. A 1950 report by McIntyre and Hofstadter (12) succeeded in establishing the NaI(Tl) scintillation spectrometer as an indispensible tool in gamma ray spectrometry.

Briefly describing the operation of a typical scintillation spectrometer, we have first the production of a scintillation described earlier (p. 7), followed by a photoelectric emission from the cathode of the P. M. as the feeble flash of light impinges on it, and finally the multiplication of the photoelectrons by secondary emission in successive stages until a measurable current is produced.

On examination of the output pulse from NaI(Tl), it is found that its size is very nearly in direct proportion to the energy of the incident gamma ray. It is interesting to look back and see how this question of linearity was finally settled. In 1950 Pringle and Standil (13) published a paper in which they reported, almost as an afterthought, a small degree of non-linearity in the scintillation response as a function of incident gamma energy, for $E_\gamma < 150$ kev. The nature

of the deviation was such that if E_γ is the abscissa, then the slope of the curve increased slightly and gradually from $E_\gamma \sim$ 150 kev to $E_\gamma \sim 0$. There arose an immediate storm of refutations from different research groups (14), (15), (16), (17), (18), who all claimed to have observed absolute linearity at least down to 1 kev, thus suggesting that the reported non-linearity (13) was due not to the crystal but rather to the accompanying electronics.

Since that time however, the result of Pringle and Standil has been confirmed by many researchers (19), (20), (21), and in fact the author has observed the effect, about which more will be said later.

Chapter 3.

THEORETICAL CONSIDERATIONS

3.1 Emission of Gamma Rays

Multipoles:

Details of this can be found in Segre (22). The electric and magnetic field vectors $\underline{E}$ and $\underline{H}$ can be shown (23) to be expressible as an expansion in terms of multipole components for outgoing waves. Performing this expansion yields two distinctly different types of terms, called electric and magnetic multipole components. Classically this corresponds to separating the radiation from some system of charges and currents into distinct types, depending on their angular distributions. Quantum mechanically, it corresponds to classifying the emitted gamma rays according to the amount of angular momentum $L\hbar$ they carry off. Several different angular momenta $L\hbar$ may be possible for a given gamma energy. L can take on only positive integral values, and L=0 is forbidden due to the transverse nature of electromagnetic waves. The component multipoles are specified by their order, 2^L, so that L=1 corresponds to a 2^1-pole, i.e. a dipole. Further, if the parity happens to be odd, then the radiation is referred to as E1 radiation, electric dipole radiation. If

the parity is even, then the radiation is denoted by M1, magnetic dipole radiation. In general, it is found that the parity change allowed equals the parity of the magnetic field, so E(L) transitions correspond with parity changes $(-1)^L$ and M(L) transitions correspond with parity changes $(-1)^{L+1}$. This rule regarding the parity changes is a consequence of the fact that the parity of the wave function of the entire system, which includes radiation as well as nucleus, must be conserved.

Selection Rules:

If $\underline{J}_i$ and $\underline{J}_f$ are total nuclear angular momentum vectors of initial and final states respectively, and if $\underline{L}$ is the angular momentum carried off by the gamma quantum, then conservation of angular momentum demands that

$$|J_i - J_f| \leq L \leq J_i + J_f \ .$$

The rules regarding the parities of the states can be written as

$$\pi_i \cdot \pi_f = (-1)^L \quad \text{for an E(L) pole,}$$

$$\text{and } \pi_i \cdot \pi_f = (-1)^{L+1} \quad \text{for an M(L) pole,}$$

where $\pi = +1$ corresponds to a state of even parity and $\pi = -1$ corresponds to a state of odd parity. Hence $\pi_i \cdot \pi_f = -1$ implies a parity change in the transition.

In considering transition probabilities, if it is assumed that the nuclear radius R is much smaller than the de Broglie wavelength $\lambda\!\!\!^{-}$ of the emitted gamma photon (i.e. $\frac{R}{\lambda\!\!\!^{-}} \ll 1$),

then it turns out that only the multipole components of lowest order will contribute appreciably. The assumption is good for most cases considered, so it is usually sufficient to consider only the lowest multipole order of each kind of radiation.

More explicitly, if $\pi_i \cdot \pi_f = (-1)^{J_i - J_f}$, then only $E(|J_i - J_f|)$ and $M(|J_i - J_f| + 1)$ radiation need be considered, whereas if $\pi_i \cdot \pi_f = (-1)^{J_i - J_f + 1}$, then only $M(|J_i - J_f|)$ and $E(|J_i - J_f| + 1)$ radiation are likely to be important. Furthermore, in the former case, where the electric component is of lower order than the magnetic component, the electric radiation generally eclipses the magnetic contribution. Such a transition is labelled a "parity-favored transition", while in a "parity-unfavored transition" the magnetic component is of lower order than the electric and the magnetic contribution is usually at least as important as is the electric.

3.2 Gamma-Gamma Angular Correlation

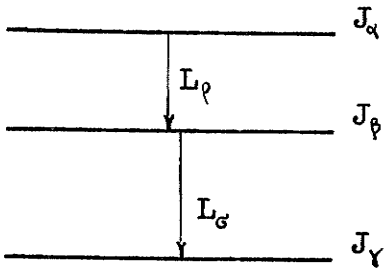
Gamma-Gamma Directional Correlations:

A considerably more general and exhaustive treatment of angular correlation of nuclear radiations is presented by Biedenharn and Rose (24), and in Siegbahn (25) and Segre (22). Only the basic principles involved and the major results required to interpret theoretically the particular angular cor-

relation work carried out by the author will be presented here.

The term "angular correlation" refers in general to both directional correlation and polarization correlation. The former allows determination of only (some of) the angular momenta of the pertinent nuclear energy levels and that of the emitted radiations, while the latter also enables one to determine the relative parities of the levels involved. This thesis will be concerned strictly with directional correlation in ~~shall further restrict the investigations to direction-direction~~ double cascades only.

Consider a simple double cascade, as illustrated below, where the initial state has angular momentum J_α, the intermediate state J_β, and the final state J_γ. Also, let the first quantum carry off angular momentum L_ρ and the second quantum L_σ. Then, from the selection rules, it can be said



$$|J_\alpha - J_\beta| \leq L_\rho \leq J_\alpha + J_\beta$$

and $$|J_\beta - J_\gamma| \leq L_\sigma \leq J_\beta + J_\gamma .$$

Of special interest here is the relative probability $W(\theta)d\Omega$ that quantum σ enters a solid angle $d\Omega$ at an angle θ with respect to quantum ρ. Experimentally, it is the detectors which each subtend a solid angle $d\Omega$ at the centrally located source, and the detectors are at an angle θ one

from another. $W(\theta)$ is the correlation function. Ideally $W(\theta)$ represents the coincidence rate between quanta ρ and σ for the angle θ. However, the source is not a point and the solid angles are not infinitesimal, and thus the experimental coincidence rates are merely averages of $W(\theta)$ over angles θ varying about the angle between the axes of the two detectors. The necessary corrections will be mentioned later.

Following Segre (22), after considerable manipulation and simplification the result obtained for the theoretical correlation function is

$$W(\theta) = \sum_{n=0}^{n^{max}} A_{2n} P_{2n}(\cos\theta) \quad \text{with } A_o=1$$

$$= \text{const.} \sum_{n=0}^{n^{max}} B_{2n} \cos^{2n}\theta \quad \text{with } B_o=1$$

where P_{2n} is the Legendre polynomial

$$P_{2n}(\cos\theta) = \left(\frac{4}{4n+1}\right)^{\frac{1}{2}} Y_{2n,0}(\theta)$$

and n^{max} is the smallest of the numbers J_β, L_σ, L_ρ (if J_β is half integer, replace it by $J_\beta - \frac{1}{2}$). The coefficients are given by

$$A_{2n} = F_{2n}(L_\rho J_\alpha J_\beta) F_{2n}(L_\sigma J_\gamma J_\beta)$$

where F_{2n} are expressible by Clebsch-Gordan coefficients and Racah coefficients and are tabulated in Segre (22), pp. 380-383.

Geometrical Corrections:

The corrections employed to allow for the finite length of the source and for the finite solid angles subtended by the detectors are essentially those developed by Feingold and Frankel (26) in the section headed "Axial Source Corrections for Circular Detectors". For corrections due to the finite radius of the source the reader is referred to Manning and Bartholomew (27). They claim that to a first approximation the finite radius introduces a small negative $P_2(\cos\theta)$ term into the observed correlation function. Specifically, for a source one inch in diameter and four inches from the detector, the coefficient A_2 is replaced by $(A_2-0.02)$ for a distribution normalized to unity at 90^o. For a source one half inch in diameter and four inches from the detector, A_2 is replaced by $(A_2-0.005)$. In general it is found that the correction depends on the square of the radius of the source. The author used sources only 0.08 inches in radius, (i.e. about 0.16 inches in diameter) and a source-to-crystal distance of about four inches. The error involved was thus extremely small and in fact quite negligible as compared with the solid angle and finite length corrections.

The following is a brief outline of the finite source and solid angle corrections based on reference (26).

Later, in sec. 5.4, the corrections will be applied to a particular experiment.

Let ϵ_o be the length of the line source and let r_o be the distance from the centre of this axially placed source to the centre of the circular crystal face. Also, suppose the crystal has diameter "a".

The article (26) gives the true point-point correlation as

$$W(\theta') = \sum_l \frac{2l+1}{4\pi} a_l P_l(\cos\theta') \quad \ldots\ldots\ldots\ldots (1)$$

which, aside from a factor $\frac{4\pi}{a_o}$, is identical with the expression for the correlation given earlier in this section, where $A_2 = 5\,\frac{a_2}{a_o}$ and $A_4 = \frac{9a_4}{a_o}$. The experimental correlation is then given as

$$W(\theta) = \sum_l \frac{2l+1}{4\pi} h_l P_l(\cos\theta) \quad \ldots\ldots\ldots\ldots (2)$$

where $h_l = a_l b_l c_l = a_l b_l^2$, since $b_l = c_l$ for two identical detectors, the Legendre coefficients describing the efficiency functions of each detector. In terms of A_{2n}'s, equations (1) and (2) become respectively

$$W(\theta') = 1 + A_2 P_2(\cos\theta') + A_4 P_4(\cos\theta') \quad \ldots\ldots (3)$$

and

$$W(\theta) = 1 + \left(\frac{b_2}{b_o}\right)^2 A_2 P_2(\cos\theta) + \left(\frac{b_4}{b_o}\right)^2 A_4 P_4(\cos\theta) \quad \ldots\ldots (4).$$

To obtain the geometrical correction factors the article (26) gives

$$b_1 = \pi\alpha^2 \left\{ 1 + F_1\left(\alpha^2 + \frac{2}{3}\gamma^2\right) + F_2\left(\alpha^4 + \frac{3}{5}\gamma^4 + 2\alpha^2\gamma^2\right) \right\},$$

where $\alpha = \frac{a}{2r_o}$, $\gamma = \frac{\epsilon_o}{2r_o}$, and the coefficients F_1, F_2, as listed in table V of reference (26), are:

	F_1	F_2
b_o	-3/4	5/8
b_1	-1	1
b_2	-3/2	15/8
b_3	-9/4	7/2
b_4	-13/4	25/4
b_5	-9/2	85/8
b_6	-6	69/4

Chapter 4.

THE APPARATUS

4.1 General Description:

A schematic diagram of our spectrometer for gamma-gamma coincidence and angular correlation work is shown in fig. 1. A gamma ray entering one of the crystals results in a pulse emitted from the P.M. tube, which pulse then passes through a preamplifier (not shown). The preamplifier emits negative pulses, and since the pulse will eventually enter a gated biased amplifier (if certain conditions are satisfied) which accepts only positive pulses, it must be inverted. This is accomplished by the linear amplifier (L.A.) which also shapes it into a double delay line pulse (for coincidence purposes) and amplifies it according to the desired gain (see D.D.A. in fig. 1). The L.A. has two outputs, prompt and delayed, the latter being delayed by two microseconds. The prompt pulse must then satisfy predetermined upper and lower limits imposed by a fast speed discriminator and single channel analyzer (F.S.D. and S.C.A.) which each are patched into a fast/slow coincidence base unit. If a prompt pulse from the other F.S.D. and S.C.A. should happen to be in coincidence with the former prompt pulse, as well as satisfying