

ANGULAR CORRELATIONS BETWEEN INTERNAL
CONVERSION ELECTRONS AND GAMMA RAYS
IN THE 620 keV CASCADE OF Ba¹³¹

A Thesis
submitted to the
Faculty of Graduate Studies
at the
University of Manitoba
in partial fulfillment of the
requirements for the degree of

MASTER OF SCIENCE

by

DONALD A. DOHAN

Winnipeg, Canada

July, 1966



TABLE OF CONTENTS

	<u>Page</u>
LIST OF FIGURES	1
LIST OF TABLES	2
ACKNOWLEDGEMENTS	3
ABSTRACT	4
INTRODUCTION	6
THEORY OF ANGULAR CORRELATIONS	9
APPARATUS	
Detectors	18
Electronics	19
Sources	21
Experimental Chamber	24
RESULTS ON Bi ²⁰⁷	26
RESULTS ON Ba ¹³¹	
Singles	32
Angular Correlation	37
APPENDIX A	48
APPENDIX B	53
REFERENCES	56

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Simple Angular Correlation Experiment	10
2	Simple Nuclear Cascade	10
3	Schematic Block Diagram	20
4	TAC Output	22
5	Apparatus	25
6	Decay Scheme of Bi ²⁰⁷	27
7	Bi ²⁰⁷ Gamma Spectrum	28
8	Bi ²⁰⁷ Conversion Electron Spectrum	29
9	Decay Scheme of Ba ¹³¹	33
10	Ba ¹³¹ Gamma Spectrum	34
11	Ba ¹³¹ Conversion Electron Spectrum	35
12	Conversion Electron Coincidence Spectrum of Ba ¹³¹	39
13	$W(\theta)/W(90)$ vs θ	40

LIST OF TABLES

Table		Page
1	K/L and K/(L + M) Conversion Ratios in Bi ²⁰⁷	30
2	K/(L + M) Conversion Ratios in Ba ¹³¹	36
3	Experimental A ₂₂ and A ₄₄ Coefficients	43
4	Theoretical A ₂₂ Coefficients	47

ACKNOWLEDGEMENTS

This thesis is based on work carried out at the University of Manitoba from May 1965 to June 1966.

The author wishes to express his sincere thanks to the director of the research - Dr. S. K. Sen - for his invaluable encouragement and extremely enthusiastic supervision throughout this work. His assistance in taking some of the data, without which the experiment would have been impossible, is gratefully acknowledged.

The work was supported by the National Research Council of Canada.

ABSTRACT

Systems for high resolution conversion electron and gamma ray detection are described. Results are given for Bi^{207} and Ba^{131} . Measurements on the K/L and K/(L + M) ratios for the 570 and 1064 keV transitions of Bi^{207} yielded the following values :

$$(\text{K/L})_{570} = 3.3 \pm 0.3 \quad (\text{K}/(\text{L} + \text{M}))_{570} = 2.5 \pm 0.3$$

$$(\text{K/L})_{1064} = 3.9 \pm 0.3 \quad (\text{K}/(\text{L} + \text{M}))_{1064} = 3.0 \pm 0.3$$

These measurements are in agreement with an E2 assignment for the 570 keV transition and an M4 assignment for the 1064 keV transition, which give theoretical K/L ratios of 3.4 and 3.7 respectively.

Measurements on the K/(L + M) ratios for the 1046, 922, 620, 496, 373, 216 and 124 keV transitions of Ba^{131} yielded the following values :

$$(\text{K}/(\text{L} + \text{M}))_{1046} = 5.8 \pm 0.7 \quad (\text{K}/(\text{L} + \text{M}))_{922} = 5.2 \pm 0.5$$

$$(\text{K}/(\text{L} + \text{M}))_{620} = 5.9 \pm 0.4 \quad (\text{K}/(\text{L} + \text{M}))_{496} = 5.9 \pm 0.3$$

$$(\text{K}/(\text{L} + \text{M}))_{373} = 4.1 \pm 0.3 \quad (\text{K}/(\text{L} + \text{M}))_{216} = 3.2 \pm 0.4$$

$$(\text{K}/(\text{L} + \text{M}))_{124} = 2.3 \pm 0.3$$

An internal conversion electron-gamma ray angular correlation spectrometer is described, in which a solid state detector is used for electron detection. Advantages for

performing electron-gamma angular correlation experiments are discussed. The system has been applied in measuring the 620-124-0 cascade angular correlation in Ba^{131} for both the electron-gamma and gamma-electron cases. The values obtained for the coefficients .

$$A_{22} = -0.01 \pm 0.01 ; A_{44} = -0.02 \pm 0.02 \text{ (gamma-electron)}$$

$$A_{22} = -0.10 \pm 0.03 ; A_{44} = -0.12 \pm 0.04 \text{ (electron-gamma)}$$

indicate a spin sequence of $\frac{5^+}{2} \rightarrow \frac{7^+}{2} \rightarrow \frac{5^+}{2}$ for the cascade.

INTRODUCTION

In the attempt to construct a logical and satisfying nuclear theory a fairly disperse group of nuclear phenomena has been studied in the last half century. Nuclear structure, and the quantum mechanical properties of the nuclear ground states and excited levels constitute the most important and by far the largest field of investigation. In recent years, many different methods of approach have been developed quite extensively to achieve this.

Electron-gamma angular correlation experiments, which are closely related to the conventional gamma-gamma directional correlations, have recently become subject to a great deal of interest as a technique of nuclear spectroscopy. The information provided by electron-gamma experiments, allows us to determine the multipolarities and mixing ratios of the nuclear transitions. From this we are able to decide about the spins of the excited nuclear energy levels. Contrary to gamma-gamma correlations, electron-gamma experiments distinguish between electric and magnetic characters. Hence we are able to obtain not only the spin but also the parity change caused by the nuclear transition. Another desirable feature of electron-gamma directional correlation measurements is that they are sometimes even more sensitive to minute admixtures of multipole radiations than are the gamma-gamma correlations. Electron-gamma correlations also yield information about the particle

parameters and conversion coefficients, which is important in internal conversion theory. The correlation, in some cases, may be perturbed by electric and magnetic fields acting on the electric and magnetic moments of the nuclei. These fields may, at least in principle, be calculated, or in the case of the magnetic field, externally applied, and hence it is possible to measure magnetic moments and electric quadrupole coupling energies of excited states from the electron-gamma angular correlation experiments.

The electron-gamma angular correlation measurement described in this thesis utilizes a solid state detector for electron detection and a NaI scintillation counter for gamma ray detection. The solid state detector has the following advantages over the usual method employing a beta-ray spectrometer :

1. A solid state detector enables the simultaneous measurement of gamma - K conversion electron and gamma - L conversion electron correlations.
2. The size and geometrical construction of solid state detectors allow convenient and well defined solid angle corrections.

Other experimental advantages of electron-gamma angular measurements are :

1. The detectors (solid state for electron and NaI crystals for gamma detection) act as particle identifiers. This reduces interference between counters, such as back scattering and Compton events, which may distort the angular correlation.
2. Solid state electron detectors have high detection efficiencies and are capable of much better energy resolution than obtained in gamma-gamma experiments using NaI crystals.

Recently, the theoretical values of the particle parameters for the L conversion electrons have been published. This enables one to obtain another piece of information concerning the nuclear cascade.

ANGULAR CORRELATIONS

Since the introduction, in 1940 (1) of the study of angular correlations of gamma rays in cascade, a great deal of progress has been made in this field especially with the advent of scintillation counters. Many theoretical and experimental papers have since been written on the subject, which is now quite well developed.

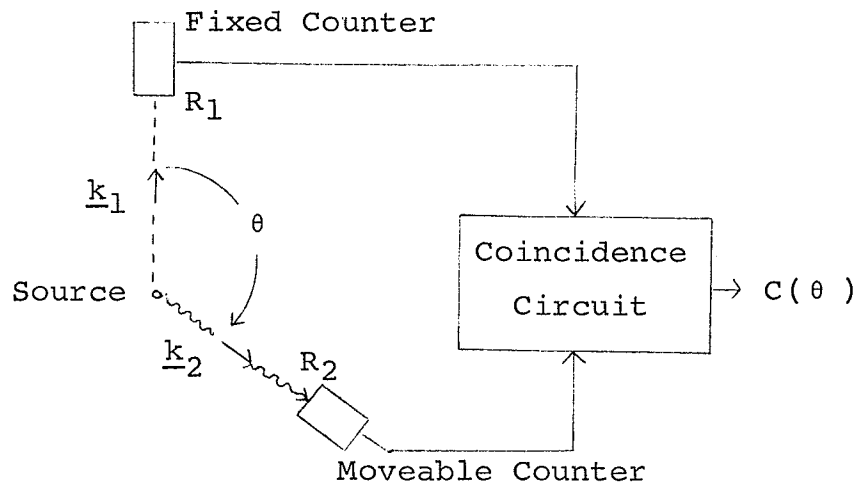
The reader is referred to the literature (2) for a detailed description of the theory. The results of the theory for gamma-gamma angular correlation will be summarized and extended to the electron-gamma case.

THE THEORY OF ANGULAR CORRELATIONS

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. Specifically, we pick out only those nuclei whose spins lie in a preferred direction. Figure 1 shows a typical angular correlation experiment.

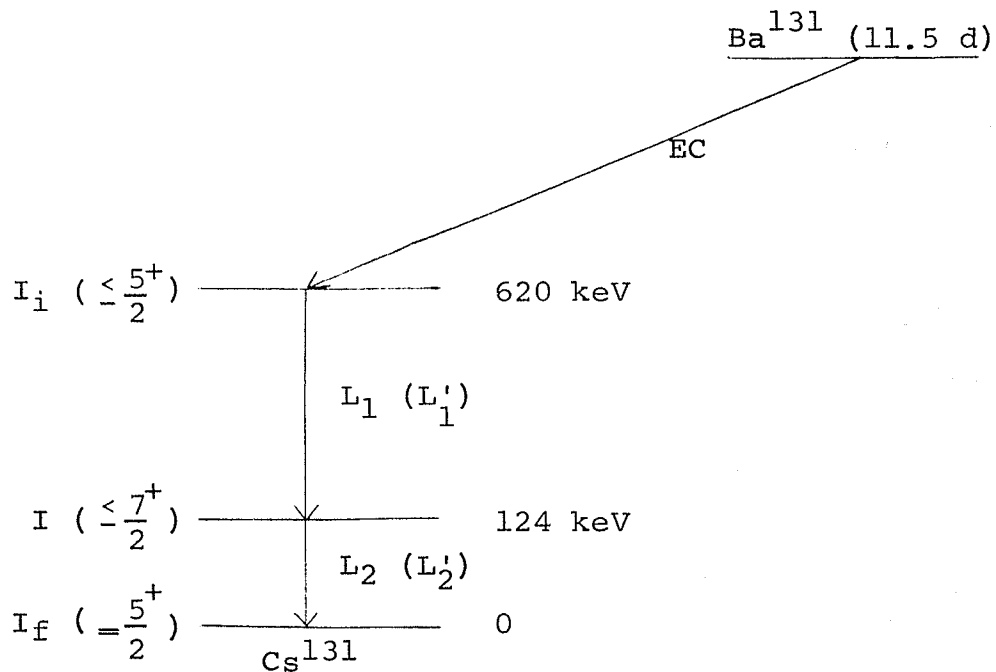
The observation of radiation R_1 in a fixed direction \underline{k}_1 selects an ensemble of nuclei that has a nonisotropic distribution of spin orientations. The succeeding radiation

Figure 1 : Angular Correlation Experiment



R_2 , in direction \underline{k}_2 , then shows a definite angular correlation with respect to \underline{k}_1 . Consider the figure below :

Figure 2 : Simple Cascade



A nucleus, (in our case, Ba^{131}), decays to an excited state of its daughter nucleus, (here the 620 keV level of Cs^{131}). The excited nucleus, in the initial state of spin I_i , decays to an intermediate state, with spin I , by emission of a gamma ray, or a conversion electron. This is followed by another transition, in cascade, to the final state, of spin I_f . The transitions are classified according to the amount of angular momentum, L , carried away by the emitted particle or quantum. The radiation is, then, called a 2^L -pole radiation. Again, for each value of L there are two different types of transitions - electric or magnetic depending on the change of the nuclear parity. The radiation is electric for even L with no change of parity, and for odd L with change in parity. A magnetic transition occurs for even L if there is a change in the nuclear parity or for odd L if the nuclear parity is unchanged. This is summarized as follows :

<u>Angular Momentum (L) Carried Away (Multiple Order of the Radiation</u>	<u>Parity Change</u>	<u>Type of Radiation</u>
0	No	E0 (electric monopole)
1	Yes	E1 (electric dipole)
1	No	M1 (magnetic dipole)
2	No	E2 (electric quadrupole)
2	Yes	M2 (magnetic quadrupole)
3	Yes	E3 (electric octopole)
3	No	M3 (magnetic octopole)

and so on.

The total angular momentum is conserved between the gamma ray and the emitting (or absorbing) system so that if I_i and I_f are the initial and final state spins of the nuclear levels which give rise to the gamma radiations, the angular momentum carried by the gamma ray is given by the rules of addition of angular momentum :

$$|I_i - I_f| \leq L \leq |I_i + I_f|$$

It follows from the above that the transition between two excited nuclear levels may be of a mixed character. That is, the orbital angular momentum carried away by the emitted particle may be either L or L' , (usually $L' = L + 1$).

The mixing ratio for the transition is defined by,

$$\delta^2 = \frac{\text{Number of } L' \text{ - pole gamma rays}}{\text{Number of } L \text{ - pole gamma rays}}$$

Referring to Figure 1, if we consider the direction of emission of the first gamma ray as fixed in space, then the probability of emission of the second gamma ray as a function of the angle θ between the gamma ray directions can be expanded in a series of Legendre Polynomials in $\cos \theta$:

$$W(\theta) = 1 + A_{22} P_2(\cos \theta) + A_{44} P_4(\cos \theta) + \dots + A_{mm} P_m(\cos \theta)$$

where the value of m which terminates the series is given by the rule :

$m \geq 2I, 2L_1, 2L_2$ where I is the spin of the intermediate state.

The coefficients $A_{\nu\nu}$ are the product of two factors. Each factor corresponds to only one transition of the cascade and is a function of the multipolarity of the transition and the spins of the states involved in the transition. Thus if the two transitions are pure, and are of multipole order L_1 and L_2 respectively, then

$$A_{\nu\nu} = F_{\nu} (L_1 L_1 I_i I) F_{\nu} (L_2 L_2 I_f I)$$

If, however, one of the transitions, say the first, is mixed, then the F coefficient for the transition, $F_{\nu} (L_1 L_1 I_i I)$ is replaced by a function containing the mixing ratio of the transitions given by,

$$\frac{F_{\nu} (L_1 L_1 I_i I) + 2 \delta_1^2 (\gamma_1) F_{\nu} (L_1 L_1' I_i I) + \delta_1^2 (\gamma_1) F_{\nu} (L_1' L_1' I_i I)}{1 + \delta_1^2 (\gamma_1)}$$

where $\delta_1^2 (\gamma_1)$, as previously described, is the mixing ratio of the first transition. If the second transition is also mixed, a similar expression involving the mixing ratio δ_2 of the second transition can be written.

The F coefficients have been calculated by Biedenharn and Rose (3) and have been tabulated by Ferentz and Rosensweig (4).

We have thus far discussed gamma-gamma angular correlations. In the following paragraphs we shall briefly discuss the theory underlying conversion electron - gamma angular correlation measurements. The relativistic theory of the directional correlations involving conversion electrons is due to Rose,

Biedenharn and Arfken (5). Since one of the gamma rays, of multipole order L , is now replaced by a conversion electron, (say a K conversion electron), the corresponding F coefficient for the electron becomes :

$$F_{\nu}(e) = \alpha_K b_{\nu}(LL) F_{\nu}(LLI_i I)$$

where α_K is the K conversion coefficient for the transition, and b_{ν} is the K electron particle parameter.

If the converted transition is mixed, the coefficient $F_{\nu}(e)$ is written as

$$\frac{b_{\nu}(LL) F_{\nu}(LLI_i I) + 2p(e) b_{\nu}(LL') F_{\nu}(LL'I_i I) + p^2(e) b_{\nu}(LL'') F_{\nu}(LL''I_i I)}{1 + p^2(e)}$$

Here, $p^2(e)$ is the mixing ratio defined by

$$p^2(e) = \frac{\text{Number of } L' \text{- pole electrons}}{\text{Number of } L \text{- pole electrons}}$$

and is numerically equal to

$$p(e) = \delta(\gamma) \sqrt{\frac{\alpha_K(L')}{\alpha_K(L)}}$$

where the α_K 's are the K conversion coefficients of the two multipolarities, L' and L .

Experimentally, we find the number of coincidences between the conversion electrons and the cascade gamma rays as a function of the angle between the directions of emission of the two particles. The curve,

$$W(\theta) = \sum_{\nu=1}^m A_{\nu\nu} R_{\nu}(\cos \theta)$$

is fitted to the data. The coefficients $A_{\nu\nu}$ thus obtained are then compared to theoretical coefficients which have been calculated for various spin assignments of the excited levels and using previously measured mixing ratios for the transitions. In this way, we obtain the spin assignments for the nuclear levels.

EXTRANUCLEAR PERTURBATIONS

It is well known (2) that extranuclear fields can cause strong perturbations of the angular correlation. The nuclear magnetic dipole moment couples to external magnetic fields and the nuclear quadrupole moment will cause a similar coupling to electric field gradients. In both cases, the coupling results in a precession of the nucleus around the external field gradient axis. If the coupling is sufficiently strong, the nuclei change their initial orientations resulting in an attenuation of the angular correlation. Quantum mechanically, if the quantization axis were chosen to coincide with the direction of the first radiation, then the extranuclear interactions cause transitions among the m-states. That is, projections of the nuclear angular momentum along the axis of quantization can undergo transitions.

It has also been proposed (2) that a time-dependent hyperfine structure interaction might appear due to a coupling between the nucleus and the atomic core, which as a result of the preceding K conversion will be in an excited and heavily ionized state. As a consequence, the nucleus is exposed to a strong magnetic field and the nuclear spin will precess about the atomic spin. This difficulty can be overcome by mounting the source on an electrically conducting backing, for which the recovery time of the atomic shell is too short to allow for any attenuation of the angular correlation pattern.

When these extranuclear effects are taken into account, the appropriate theoretical expression then becomes :

$$A_{\nu\nu}^{\text{exp}} = G_{\nu}(Q)G_{\nu}(\text{hfs})A_{\nu\nu}^{\text{unperturbed}}$$

where $G_{\nu}(Q)$ represents the attenuation due to the static quadrupole interaction and $G_{\nu}(\text{hfs})$ is the attenuation due to the hfs interaction mentioned above. For metallic source environments, $G_{\nu}(\text{hfs}) \approx 1$.

APPARATUSDETECTORS

The electron detector was a gold-silicon surface barrier p-n junction type obtained from ORTEC. It has a circular area of 50 mm² and the depletion depth at a bias of 475 volts was 1500 microns which is the range of a 980 keV electron in silicon. For singles counts, the pulses from solid state detector were led through the low noise Nuclear Enterprises' 5231 preamplifier and were further amplified by a Nuclear Enterprises' 5230 RC amplifier. In the coincidence work, the output pulse from the preamplifier was also fed simultaneously into an ORTEC 203 amplifier which produces double decay line clipped pulses necessary to drive a crossover pick-off unit. At room temperature the system is capable of an energy resolution of 10 keV. For the angular correlation experiment, the detector was cooled to liquid nitrogen temperature, giving a total energy resolution of about 5 keV for the system.

For the gamma spectra of Bi²⁰⁷ and Ba¹³¹, a Ge(Li) gamma ray detector was used. This was a 2 mm depletion depth by 2.78 cm² active area device obtained from R.C.A. These pulses were amplified by a Nuclear Enterprises' 5231-5230 amplifying system. The detector and amplifying system were capable of a total energy resolution (F W H M) of 4.2 keV at 660 keV.

In the angular correlation work, a $1\frac{1}{2}$ " x 1" NaI(Tl) crystal mounted in an integral line assembly with a Dumont 6292 photomultiplier tube was used for the gamma ray detection. Pulses from this unit were fed, via a cathode follower, to a Nuclear Enterprises' 5202 double delay line amplifier.

ELECTRONICS

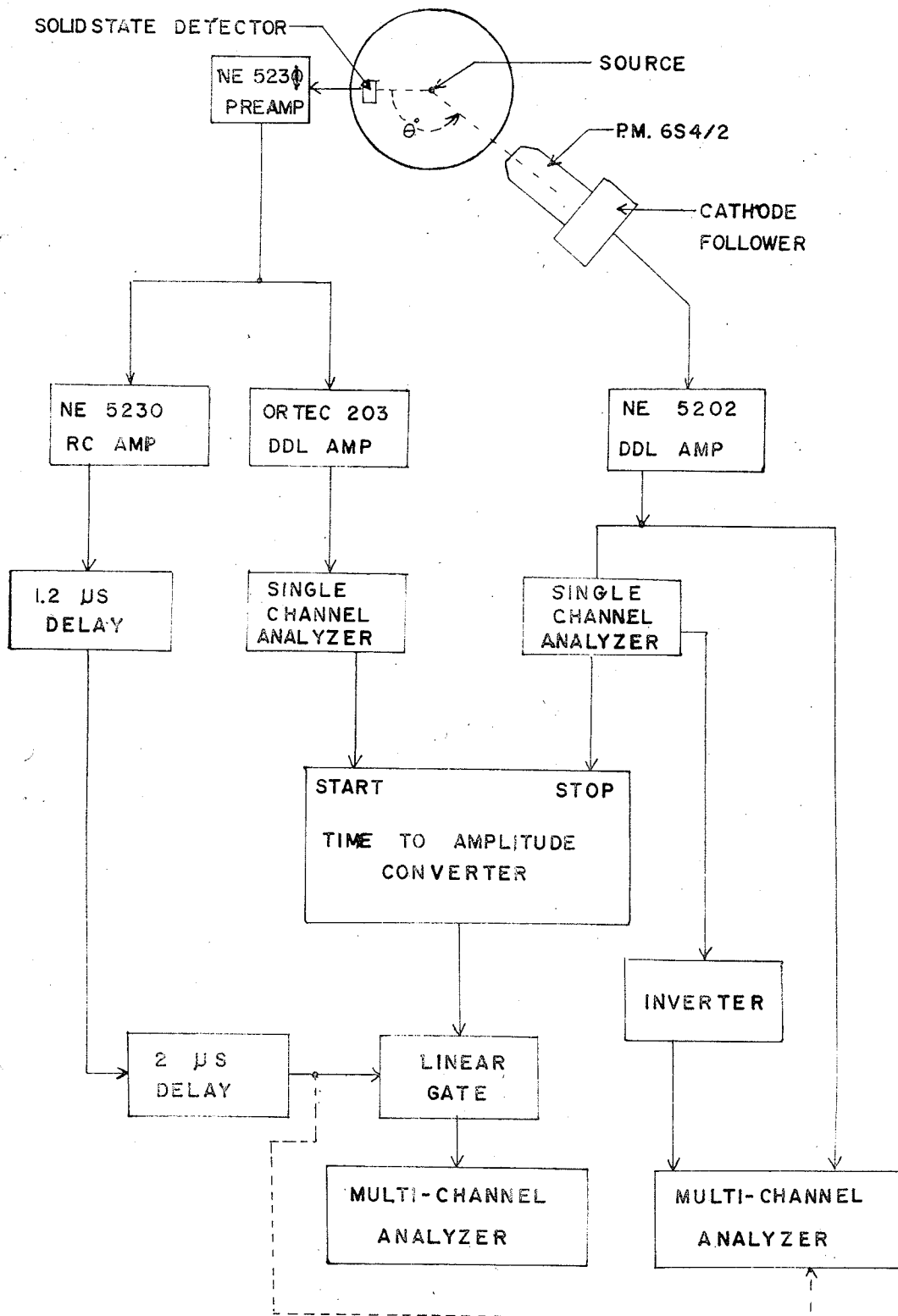
The block diagram for the electronics is given in Figure 3. The function of the apparatus was to select those pulses of the correct energy from the detector which were in coincidence with a detected gamma ray, also of the correct energy. These electron pulses were then analyzed in a Nuclear Data 1024 channel analyzer and from this data, the angular correlation function was obtained.

The pulses obtained from the electron and gamma ray detectors, (see the block diagram), after being amplified by the double delay line amplifiers, are fed into two single channel analyzers. The windows of the single channel analyzers were set to accept the appropriate electron and gamma ray energies of the particular cascade. The output pulses of the single channel analyzers, which were initiated at the cross-over point of the double delay line pulses, are fed into a time-to-amplitude converter (TAC) type fast coincidence system. The TAC gives an output pulse whose amplitude is proportional to the time difference between the two input pulses. These output pulses are fed through a single channel analyzer, which

FIGURE 3

Schematic Block Diagram

BLOCK DIAGRAM



accepts only those pulses which have the correct amplitude, corresponding to the coincidence events. A typical TAC spectrum, showing the single channel analyzer settings is illustrated in Figure 4. The dotted line represents the background due to chance events, which occur randomly in time. The output pulse from the TAC is used to gate the 1024 channel analyzer which analyzes the electron pulses obtained from the good resolution RC amplifier. The coincidence rate was thus taken at several angles to obtain the angular distribution curve. Simultaneously, the single channel analyzer output was used to gate another multichannel analyzer, which accumulated the gamma ray singles spectrum that was allowed through the window of the single channel analyzer. The singles counting rate of the moveable gamma counter was used to normalize the coincidence counting rate at each angle. This is to correct for the finite size of the source and for the possibility of the source not being in the exact center of the arc described by the gamma counter.

SOURCES

Our choice of source was limited by the following considerations :

1. We required a source which decayed by 100% electron capture so that there would be no background beta radiation. The presence of this radiation would make background subtraction extremely difficult.

FIGURE 4

TAC Output

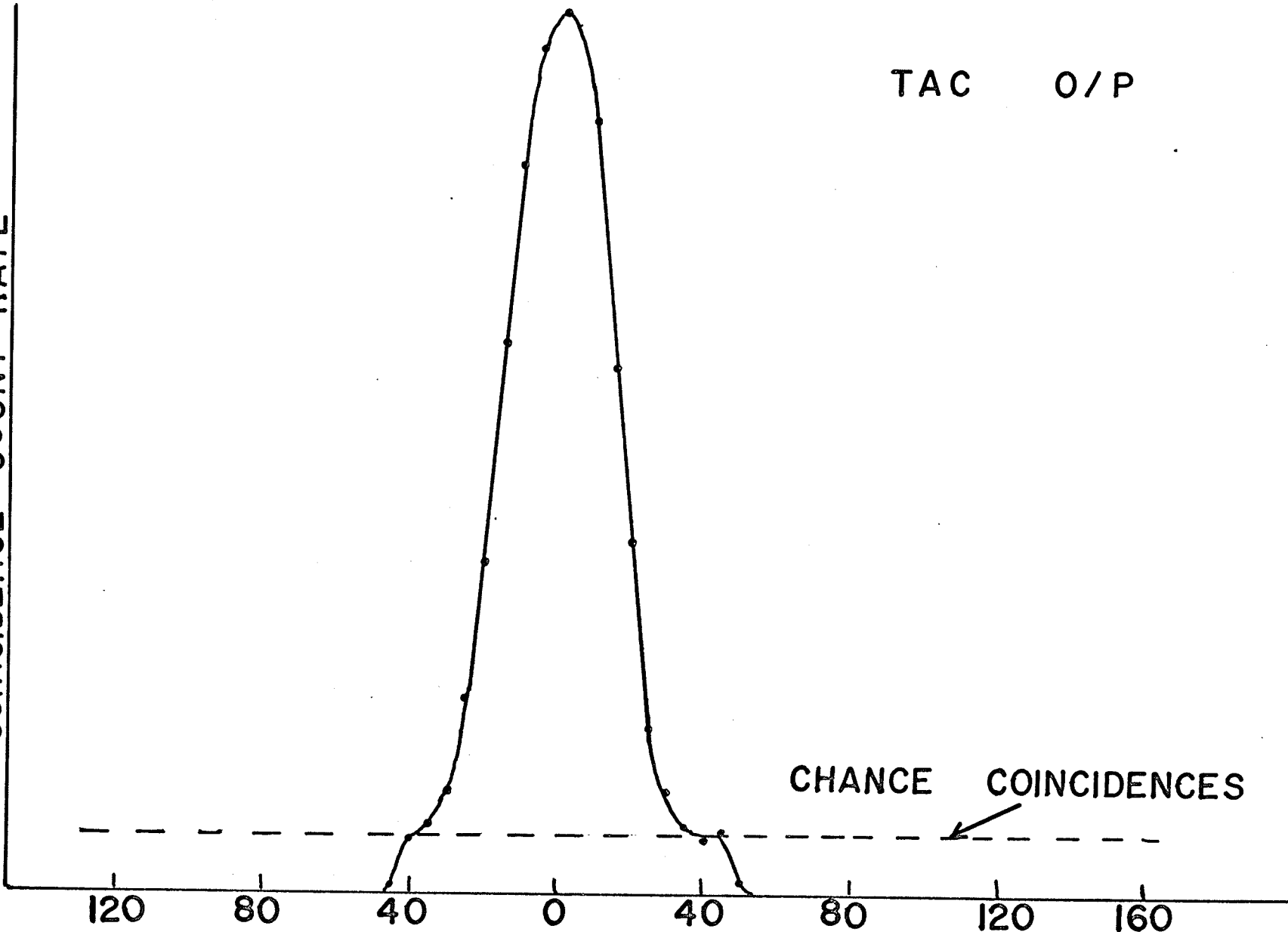
TAC O/P

COINCIDENCE COUNT RATE

CHANCE COINCIDENCES

120 80 40 0 40 80 120 160

DELAY (ns)



2. The conversion electrons should have energies within the energy range from 50 to 1000 keV. The lower limit is due to the rapid build up of pulses due to the backscattering of the electrons from the detector and thus distorting the conversion peak. The upper limit is set by the depletion depth of the detector. Also, in this region, the internal conversion coefficients become quite low thereby greatly reducing the coincidence counting rate.

3. The transition energies should differ sufficiently in order that the conversion lines could be separated in interfering cascades.

Two sources chosen were Bi^{207} (28 years) and Ba^{131} (11.5 days), which decay by electron capture to excited states of Pb^{207} and Cs^{131} respectively. Bi^{207} is a well known source and was used as a calibration in the singles work. In the past, several gamma-gamma angular correlations have been performed on Ba^{131} but the results do not agree well. To our knowledge there has been no measurement on the electron-gamma angular correlation of Ba^{131} . The present work was carried on with much improved resolution in the electron channel in order to remove some of the ambiguities of the earlier work and also to provide new information on this cascade.

For the Bi^{207} work, we used a 1 microcurie source which was deposited on 0.001" thick plastic to minimize scattering.

A preliminary experiment was carried out to measure the 496 gamma - 124 K electron angular correlation in Ba^{131} and in that experiment, the source consisted of a drop of radioactive source solution dried at the centre of a gold plated V.Y.N.S. film which was supported on an aluminum planchette ring.

However, in the final run, in which both the 496 gamma - 124 K electron and the 496 K electron - 124 gamma correlations were measured, the source was sublimed onto a 180 microgram/cm² Al foil.

EXPERIMENTAL CHAMBER

The experimental chamber is shown in Figure 5. The chamber is constructed with cylindrical symmetry about the axis of rotation of the moveable scintillation counter. This is to minimize possible anisotropies from being introduced due to scattering. The aluminum planchette source rings were held vertically in the geometrical centre of the vacuum chamber 1.7 cm from the solid state electron detector. Figure 5 also illustrates the mounting of the electron detector of the cold finger of the cryostat. This cold finger is readily interchangeable for various sizes of detectors. The chamber was evacuated by a Balzers rotary pump and a liquid nitrogen cold trap was used to prevent oil vapours from entering the chamber containing the detector and the source. A constant check was made on the temperature of the detector using a thermocouple and of the vacuum in the chamber during the runs.

FIGURE 5APPARATUS

a - Angular correlation table

b - Vacuum chamber, coldfinger and detector mount