AN INVESTIGATION OF THE ANGULAR CORRELATIONS OF THE NEUTRON INDUCED EUROPIUM ACTIVITIES

by

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INTRODUCTION

One of the important problems of modern nuclear physics is the determination of the properties of nuclear energy levels. Angular correlation is a recently developed method for investigating some of the properties of short lived, excited nuclear states. The information obtained by correlation measurements depends on the type of radiation observed, on the specific properties measured by the experiment, and on the external fields acting on the nucleus.

This paper deals explicitly with a type of angular correlation generally described as gamma-gamma directional correlation, i.e.; only the angle between the two emitted photons is observed. Under these circumstances the correlation yields the angular momenta of the nuclear levels, but not the parities.

The ultimate objective of this investigation was to determine the angular momenta of the nuclear energy levels of the decay products of Europium 152. To accomplish this objective it was necessary to construct and calibrate an apparatus which would be capable of determining the directional correlation identifiable gamma ray cascades in radio-isotopes.

A simple physical description of angular correlation can be given: In a gamma ray cascade, if we observe only one transition then it is obvious that the directional distribution of the radiation is isotropic since no direction in space is unique and the excited nuclei are randomly oriented. However, if the nucleus decays through the successive emission

of two gamma rays r_1 and r_2, the observation of only those r_1 rays emitted in a fixed direction selects a set of nuclei which has a non-isotropic distribution of orientations of the nuclear spin axis for a "short" time after the emission. The observation by a second counter of the succeeding gamma ray, r_2 in coincidence with r_1, then displays a directional correlation with respect to r_1.

The theory of directional correlation of successive gamma rays was first developed by Hamilton (8). Other workers, notably Falkoff and Uhlenbeck (5), have extended the theory and derived the correlation function in terms of Legendre Polynomials. Their distribution function is:

$$W(\theta) = 1 + A_2P_2(\theta) + A_4P_4(\theta)$$

where θ is the polar angle between the successive gamma rays and the coefficients A_2 and A_4 are determined explicitly by the spin quantum numbers of the respective nuclear levels. Tables of directional correlations have been prepared by Biedenharn and Rose (1). It is assumed that only pure dipole and quadrupole emission is involved, and that the nuclei are not perturbed by extra-nuclear fields. The latter assumption is justified when the mean lifetime of the intermediate state is much shorter than the interaction time between the nucleus and the extra-nuclear fields. Angular correlation in which the second transition is octopole has never been observed because of the longer lifetime of the intermediate state.

EXPERIMENTAL APPARATUS

Two scintillation spectrometers and a Rossi type coincidence counter were employed to measure the gamma radiation produced by the radio-isotopes investigated. Each spectrometer consisted of a scintillation counter (Radiation Instrument Development Laboratory, Model 43A), a high voltage power supply (Radiation Instrument Development Laboratory, Model 114), a pulse height analyzer (Radiation Instrument Development Laboratory, Model 115), and a decade scalar (Nuclear Instrument and Chemical Corporation, Model 162).

Due to temperature fluctuations in the electronic equipment the channel level voltage of the gamma ray peaks tended to change with time. To minimize this effect, air blowers were mounted under each chassis, the scintillation counters were wrapped with two inches of glass wool insulation and an outer covering of cloth, and the room temperature regulated by means of an air-conditioner. Even with these precautions it was necessary to reset the pulse height selectors after each thirty minutes of counting.

For a complete discussion of the operation of the electronic equipment the reader is referred to the thesis by Butler (3), and the thesis by Trimble (10).

The scintillation counters were mounted on a circular platform with a cource holder in the center. One counter was permanently located in a fixed position while the other counter was mounted on a carriage which was free to rotate

around the central axis of the platform. The source holder was designed to permit precise centering of the source with respect to the longitudinal and rotational axis of the counters. To obtain meaningful angular correlations it is necessary to center the source with sufficient precision that the radiation in the plane of the counters is isotropic. The condition is fulfilled when the variation of the singles counting rate of each counter at all angles of θ is less than 1 per cent. It was experimentally determined that the maximum deviation of the singles counting rate of the fixed counter was approximately 0.1 per cent and the deviation of the movable counter was approximately 0.3 per cent for all sources studied.

The radio-isotopes studied were in the form of a fine powder. A thin walled glass capillary tube was formed and the powder placed near one end to form a cylinder about 0.5 millimeter in diameter and one millimeter in length. The dimensions of the source were sufficiently small so that it was unnecessary to correct for finite source size (9). With this source arrangement the absorption of gamma radiation was radially uniform and scattering was held to a minimum.

EXPERIMENTAL PROCEDURE AND TREATMENT OF DATA

Before one is able to begin correlation studies one must have a complete and accurate decay scheme of the radio-isotope to be investigated. The decay schemes of the isotopes we investigated, Gadolinum 152, Samarium 152, and Cobalt 60 have

been published (4),(6). These decay schemes are in agreement in the gamma cascades studied here.

The investigator must then select two specific gamma transitions which are in cascade. In the case of Cobalt 60 there is no lattitude for choice since there are only two transitions both gamma rays are in cascade.

The channel levels of the pulse height analyzers are then set to accept only gamma rays of the selected energies from the scintillation counters. For example, in investigating Cobalt 60 one analyzer is set to accept only 1.33 Mev radiation and the other is set to accept only 1.17 Mev radiation.

Coincidence spectra were obtained for each source investigated by fixing one spectrometer on one gamma peak and scanning the spectrum with the other spectrometer and recording the coincidence counts at each energy setting. From the resulting curves one is able to establish which gamma transitions are in cascade. If the radio-isotope has a complex decay scheme with a large number of energy levels and gamma transitions, there will usually be several competing cascades producing coincidence peaks. Another process leading to coincidence peaks is the summing of non-coincident gamma rays with coincident gamma rays. Coincidence peaks can also be produced by photons downgraded in energy by Compton scattering between the scintillation crystals.

These three processes can mask or completely destroy the angular correlation. The first two effects are eliminated if the coincidence peaks are completely resolved by the spectrometers.

The coincidences due to Compton scattering can be eliminated by the proper use of shielding (2). In this investigation lead shields in the shape of truncated cones were placed over the scintillation crystals, effectively eliminating coincidences due to crystal scattering.

The experimental coincidence counting rate is the sum of the "true" coincidence rate due to the two gamma rays emitted in cascade and the "accidental" coincidence rate resulting from the finite resolving time of the coincidence circuit:

$$N_{exp} = N_t + N_{acc}$$

The accidental coincidence counting rate is given by:

$$N_{acc} = 2\tau N_1 N_2$$

where τ is the resolving time in seconds and N_1 and N_2 are the singles counting rates of the two counters[1]. Thus to obtain the true coincidence rate we subtract the accidental rate from the observed rate. The deviation of the true coincidence rate is determined by (9):

$$\sigma_t = \left[N_{exp} + N_{acc}\right]^{\frac{1}{2}}$$

Thus it can be seen that to minimize the relative error the ratio of accidental rate to true rate should be made as

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[1] A resolving time of 0.72×10^{-6} second was used throughout our investigation.

small as possible. Therefore, the best results are obtained by the use of large solid angles, short resolving times, and relative weak sources. Unfortunately other experimental factors place a limitation upon the preceding factors so that in actual practice the above ratio can seldom be made much smaller than 0.5.

The directional correlation measurements were obtained in the following manner: The spectrometers were set to count the individual transitions of the gamma cascade. The singles counts and the coincidence counts were recorded after a counting time of thirty minutes; the movable counter rotated 30°; the spectrometers re-zeroed on the gamma peaks; and a new counting cycle commenced. In this manner singles and coincidence counts were obtained at intervals of 30° over a range of 90° to 180°.

From this data the accidental coincidences were computed and the true coincidence count obtained at each angle of θ. Repeated measurements were made at each angle and the separate true coincidence counts at each angle summed. A correlation function of the form

$$W(\theta) = A_0 + A_2 P_2(\theta) + A_4 P_4(\theta)$$

was fitted to the total experimental values by a least squares method. This function was then normalized to a value of unity at $\theta = 90°$ by dividing $W(\theta)$ by A_0. The normalized experimental coefficients were then corrected for the crystal efficiency and finite solid angles of the counters by the method of Frauenfelder (7). The correction factors were obtained experi-

mentally by Trimble (9).

The corrected coefficients were then compared with the coefficients of the various theoretical correlation functions. If the experimental coefficients were found to match a particular pair of theoretical coefficients within the experimental error then the energy levels of the cascade investigated could be assigned specific spin quantum numbers.

EXPERIMENTAL RESULTS ON COBALT 60

The directional correlation of the gamma cascade in Cobalt 60 has been accurately determined by several workers. The properties of the nuclear levels are therefore well established. For this reason this radio-isotope was utilized in calibrating the correlation apparatus and as a check on the experimental procedure followed.

The decay scheme of Cobalt 60 is reproduced on Plate I. The parent nucleus is Cobalt 60 which decays by negatron emission to an excited state of Nickel 60. The nickel nucleus then releases its excitation energy by the emission of two successive gamma rays. Both gamma transitions are electric. quadrupole. The angular momenta of the three nuclear levels are: $J_1 = 0$, $J_2 = 2$, and $J_3 = 4$. The parities are all even.

The gamma spectra and coincidence spectra obtained are plotted in Plate I. The resolution of the spectrometer is indicated by the separation of the gamma peaks. Compton escape peaks are observed at 9 volts and 14 volts. The coincidence

curves were obtained by fixing one spectrometer on one of the gamma peaks and scanning the gamma spectra with the other spectrometer. The broad coincidence peaks at 9 volts and 14 volts are produced by Compton escape photons as one would expect.

The results of the directional correlation measurements made on Cobalt 60 are displayed graphically in Plate II. The solid curve is the least squared fit of the experimental points. In terms of Legendre polynomials the normalized correlation function is

$$W(\theta) = 1 + A_2P_2(\theta) + A_4P_4(\theta)$$

The experimentally determined coefficients are $A_2 = 0.098 \pm .005$ and $A_4 = 0.007 \pm .002$. The corresponding theoretical coefficients are $A^2 = 0.102$ and $A_4 = 0.0091$. Even though the deviations of the experimental points are significantly large the agreement between the experimental and theoretical coefficients is quite good. Greater accuracy could have been obtained by collecting further counts. However, the experimental results we obtained are sufficiently accurate to justify the conclusion that the experimental procedure followed was correct.

EXPERIMENTAL RESULTS ON EUROPIUM ACTIVITIES

For this research the source was a sample of natural Europium that had been pile irradiated and was therefore a mixture of Eu^{152} and Eu^{154}. These two nuclei are unstable with half lives of 5.3 years and 5.4 years respectively.

EXPLANATION OF PLATE I

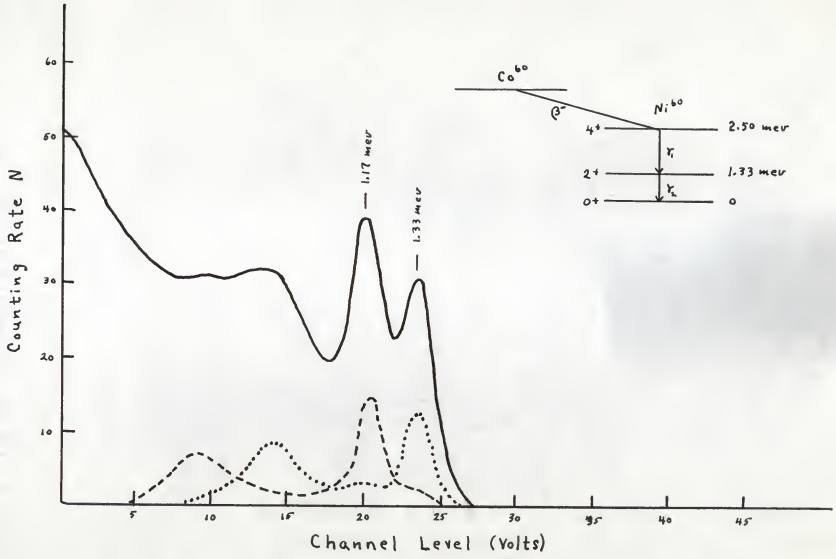
Gamma and coincidence spectra of Cobalt 60

Single spectrometer gamma spectrum _____

Coincidence spectrum with one spectrometer fixed on 1.33 Mev radiation _ _ _ _ _ _ _

Coincidence spectrum with one spectrometer fixed on 1.17 Mev radiation .....

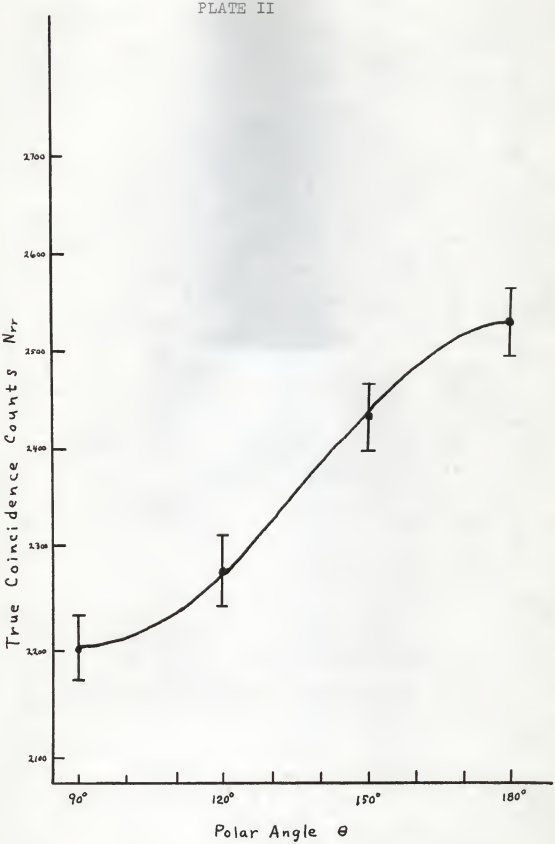
PLATE I


Counting Rate N
60
50
40
30
20
10
1.17 mev
1.33 mev
5
10
15
20
25
30
35
40
45
Channel Level (Volts)
Co 60
β-
Ni 60
4+
2.50 mev
γ1
2+
1.33 mev
γ2
0+
0

EXPLANATION OF PLATE II

Experimental directional correlation of Cobalt 60 gamma radiation.

PLATE II


True Coincidence Counts N_{rr}
2700
2600
2500
2400
2300
2200
2100
90°
120°
150°
180°
Polar Angle θ

Eu^{152} may decay by K-capture to Sm^{152} or by beta-emission to Gd^{152}. Eu^{154} decays only by beta-emission to Gd^{154}. The three daughter products are always left in an excited state and will decay by either internal conversion or by successive gamma emission. The decay schemes of the Europium activities (4) are reproduced in Plate V. Each of these isotopes has a complex decay scheme with several separate gamma cascades in each. As a result only the most intense gamma rays are resolved as individual peaks in the gamma spectrum. In the coincidence spectra only a very few coincidence peaks are completely resolved. This leads to expreme difficulties in the angular correlation measurements since it is a prime prerequisite to accurate correlations that competing cascades be eliminated by the spectrometers.

The decay scheme of Gd^{152} proposed by Cork (4) differs from the decay scheme proposed by Fowler (6) in the energy levels above the 1.127 mev level. In the decay scheme of Gd^{152} reproduced in Plate V, figure 2, the 203 kev and 520 kev gamma rays found by Fowler but not reported by Cork are drawn as dashed lines.

In this investigation the coincidence spectrum obtained by fixing one counter on 345 kev radiation (Plate III) showed coincidence peaks at 203 kev, 410 kev, 520 kev and 782 kev. Coincidence peaks were also found at 203 kev, 345 kev, and 520 kev when one counter was fixed at 782 kev and the gamma spectra scanned by the other counter. Thus, the coincidence

spectra obtained in this investigation support the decay scheme of Gd^{152} proposed by Fowler.

In addition, an investigation of the directional correlation of the 410-345 kev cascade in Gd^{152} indicated that the correlation was masked by a competing cascade. Since the coincidence peak at 345 kev was completely resolved, the competing cascade must be in Gd^{152}.

The coincidence peaks at 410 kev and 520 kev, which were obtained by fixing one counter on the 345 kev gamma ray peak, were not completely separated by the spectrometers. From this it may be inferred that the competing cascade masking the 410-345 kev correlation is a 520-345 kev cascade in Gd^{152}.

The angular correlations of several cascades in the Europium activities were investigated.

A study of the 245-122 kev cascade in Sm^{152} was made but a competing cascade, 248-123 kev in Gd^{154} masked the correlation.

The 1416-122 kev cascade in Sm^{152} was investigated. The gamma and coincidence peaks were easily resolved but a very large ratio of accidental coincidences to true coincidences would have necessitated an extremely long counting time for the correlation measurements. For this reason a correlation of this cascade was not attempted.

The most promising cascade was the 782-345 kev cascade in Gd^{152}. The coincidence peaks were fully resolved and the ratio of accidentals to trues was 0.5. For these reasons the major objective of this investigation was to obtain the

EXPLANATION OF PLATE III

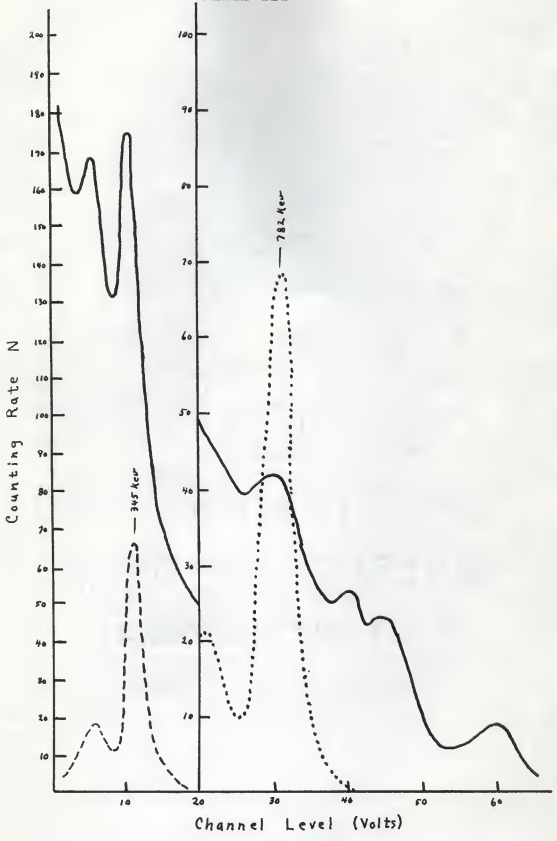
Gamma and coincidence spectra of Gadolinium 152

Single spectrometer gamma spectrum _____

Coincidence spectrum with one spectrometer fixed on 782 kev radiation _ _ _ _ _ _ _ _

Coincidence spectrum with one spectrometer fixed on 345 kev radiation . . . . . .

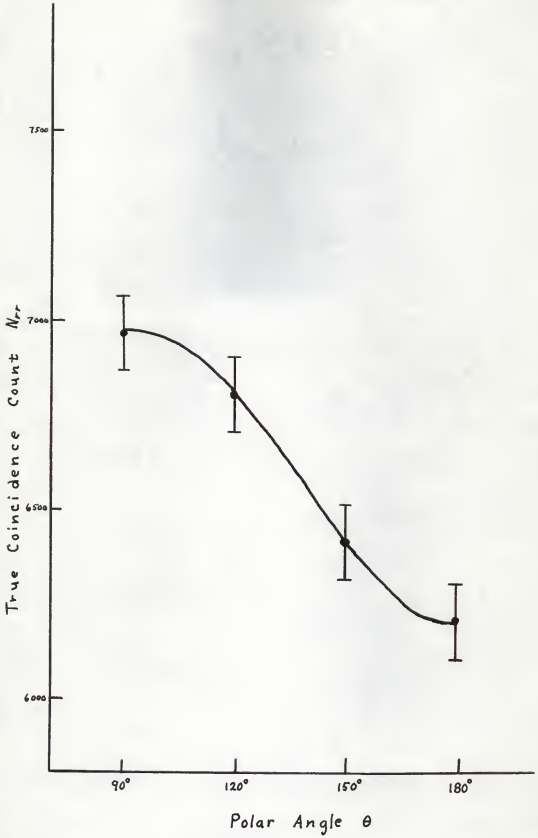
PLATE III



EXPLANATION OF PLATE IV

Experimental directional correlation of the 345-782 kev gamma cascade in Gadolinium 152.

PLATE IV


7500
7000
6500
6000
True Coincidence Count N_T
90°
120°
150°
180°
Polar Angle θ

EXPLANATION OF PLATE V

Figure 1. Decay scheme of Sm^{152}

Figure 2. Decay scheme of Gd^{152}

Figure 3. Decay scheme of Gd^{154}

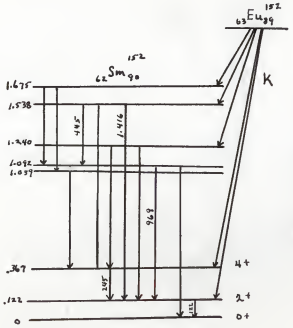


Figure 1

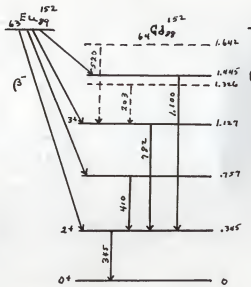


Figure 2

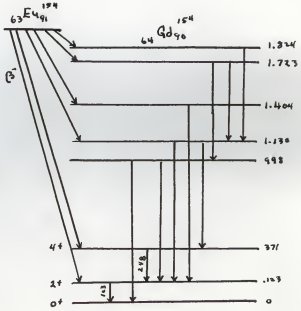


Figure 3

correlation of this cascade.

The gamma spectra and coincidence spectra for the 782-345 kev range is shown in Plate III. The gamma peaks at 345 and 782 kev are clearly resolved as are the coincidence peaks at these energies. It was therefore assumed that the correlation measurements on the 782-345 kev cascade are not mixed with competing cascades.

The experimental correlation function obtained on the 782-345 kev cascade in Gadolinium 152 is displayed in Plate IV. This curve is the least squared fit to the same equation in Legendre polynomials employed in the Cobalt 60 correlation. The experimental function was then normalized to a value of unity at $\Theta = 90^{\circ}$ and corrected for the crystal efficiencies and finite solid angles of the scintillation counters.

The corrected and normalized experimental correlation coefficients are $A_2 = -0.0815 \pm .003$ and $A_4 = -0.0058 \pm .005$.

The values of the signs and magnitudes of the experimental correlation coefficients exclude all but two theoretical correlation functions from consideration: The theoretical coefficients for a dipole-quadrupole cascade with nuclear spins of $J_1 = 0$, $J_2 = 2$, and $J_3 = 3$ are $A_2 = -0.0714$ and $A_4 = 0$. The theoretical coefficients for a quadrupole-quadrupole cascade with the same spins are $A_2 = -0.204$ and $A_4 = -0.0815$.

The measured coefficients exclude pure dipole radiation because A_4 is not equal to zero. However, the experimental

values are clearly in disagreement with the theoretical coefficients for pure quadrupole-quadrupole transitions. Furthermore, the anisotropy of the experimental correlation is 12.1 percent whereas the anisotropy of the pure quadrupole correlation is 33.3 percent. The anisotropy of the pure dipole-quadrupole correlation is 10.4 percent and agrees more closely with the measured value.

Since no other spin assignment fits the experimental data, one must assume that the 782 kev gamma ray between the nuclear level with spin 3 and the level with spin 2 must be a mixture of magnetic dipole and electric quadrupole radiation. The accuracy of the experimental coefficients precludes obtaining an accurate mixing ratio of the two types of transitions; however, a mixture of no more than 5 percent of electric quadrupole radiation is indicated.

The K/L ratio for the 345 kev gamma ray measured by Cork et al (4) was 4.6, which indicates that the transition is electric quadrupole. In addition the collective model of the nucleus predicts for even-even nuclei rotational levels with even spin and parity: $J_0 = 0+$ and $J_2 = 2+$. The prediction for states of higher energy have not been so well verified experimentally. These considerations are in agreement with the measured correlation.

There can be no parity change as a result of magnetic dipole or electric quadrupole emission, therefore the parity of the J_3 level must be even since the J_2 level has even parity.

In conclusion, an unambiguous nuclear spin assignment of $J_0 = 0+$, $J_2 = 2+$, and $J_3 = 3+$ can be made of the 345-782 kev cascade in Gadolinium 152.

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The author wishes to express his sincere appreciation and gratitude to Dr. Clarence M. Fowler for his excellent advice and suggestions throughout this investigation and to Dr. Robert Katz for his advice and helpful suggestions during the preparation of this thesis. Mr. G. Trimble and Mr. R. K. Fry are to be thanked for their cooperation in this investigation.

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In this investigation an apparatus was constructed and calibrated to obtain gamma-gamma directional correlation measurements of gamma ray cascades in radio-nuclei.

For calibration purposes a directional correlation of the gamma radiation from Cobalt 60 was obtained. The coefficients of the experimental correlation function, which was expressed in terms of Legendre polynomials, were in close agreement with the theoretical coefficients. It was therefore concluded that the apparatus and experimental procedure was correct.

Various gamma ray cascades in the Europium activities were investigated. The 345-782 kev cascade in Gadolinium 152 was selected for extensive directional correlation measurements. As a result of these measurements a correlation function was obtained and it was determined that the gamma transitions in this cascade are a mixture of magnetic dipole and electric quadrupole radiation. The nuclear angular momentum spins of the three levels of this cascade were assigned values of $J_0 = +0$, $J_2 = +2$ and $J_4 = +3$. The accuracy of the experimental values is sufficient to justify the conclusion that these spin assignments are correct and unambiguous.