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GAMMA-GAMMA AND CONVERSION ELECTRON-GAMMA DIRECTIONAL CORRELATIONS IN Cs\textsuperscript{133}

A THESIS

Presented to

The Faculty of the Graduate Division

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Frank Titus Avignone, III

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CHAPTER I

INTRODUCTION

Nuclear spectroscopy is concerned with the study of the various ways in which nuclei absorb and emit energy. The nucleus can reside in discrete nuclear states which can be described by appropriate parameters. The following are a few of the parameters which are of particular importance in the current work: (a) excitation energy, (b) total angular momentum of the nucleus, (c) the parity of the nuclear wave function, and (d) the electric and magnetic moments of the nucleus.

One of the objects of experimental nuclear spectroscopy is to obtain as much information about these parameters as possible in order to permit comparison of experimentally obtained values of the parameters with those calculated theoretically using one of the various models of the nucleus. The angular correlation technique is a powerful tool in nuclear spectroscopy and is used in this investigation.

The Angular Correlation

The probability of emission of a particle or quantum by an excited nucleus depends in general on the angle between the nuclear spin axis and the propagation direction of the emitted radiation. In an ordinary radioactive source the nuclear spin directions are randomly oriented and the radiations emerge isotropically from the source. In order to measure the angular dependence of the distribution of particles or quanta in nuclear emissions, the nuclear spin axes must be aligned.
One method of alignment of the nuclei consists of placing the radioactive source in a strong magnetic field or an inhomogeneous electric field at very low temperatures. The directional distribution of the radiations can then be measured with respect to the direction of the applied field and the pertinent nuclear information obtained.

An alternative method of obtaining much the same nuclear information is the directional correlation measurement. The directional correlation \( W(\theta) \) can be defined as the relative probability that the two radiations from a nuclear cascade decay will have the angle \( \theta \) between their propagation vectors. The directional correlation, if not perturbed by the environment, is completely specified by the angular momentum carried off by each radiation and the interaction giving rise to the transitions. Nuclear levels are in general degenerate with respect to the magnetic substates in the case that there are no extranuclear fields. A single transition then is composed of component transitions between the several substates of the initial level and those of the final one. All components of the transition will occur for which the laws of conservation of energy and angular momentum are obeyed. In nuclear spectroscopy the usual experiment cannot resolve the components of a transition hence a weighted sum over the components of a line must be made in the theoretical development of the correlation function. An outline of selected results of the theory is presented in Appendix B.

The measurement of directional correlations is done by means of two radiation detectors the signals from which enter a coincidence circuit. If the signals generated in the counters reach the coincidence circuit within a predetermined time interval of each other, the radiations are
assumed to have come from the same nuclear cascade and the event is recorded as a coincidence event. Energy restrictions are placed on the coincidence events by using pulses which are approximately proportional to the energy of the ionizing radiations. In the usual instrumentation one counter is fixed in space and the other counter is rotated about an axis through the radioactive source. The number of coincidence events recorded per unit time when the angle between the two propagation vectors is \( \theta \) is directly proportional to the value of the directional correlation, \( W(\theta) \), after certain instrumental corrections are made to the raw data. The interpretation of angular correlation experiments relies on the intimate relationship between the angular momenta and parities of the nuclear states and the emitted radiations and on the symmetry properties under spatial rotation and inversion of the nuclear wavefunctions and the radiated fields. For transitions between states of definite spin and parity, the conservation of angular momentum will restrict the range of angular momentum values of the emitted radiations. Angular correlation techniques are of interest in other than cases of nuclear decay cascades. The techniques of angular correlations are also used to investigate non-resonant or potential scattering, absorption and emission phenomena, Coulomb excitation, and various types of inelastic scattering. Further discussion of angular correlations in this thesis will be restricted, however, to the directional correlations of nuclear decay cascades.

Directional correlation experiments permit a determination of the angular momenta of the nuclear levels involved and the angular momenta carried by the radiations, which in the case of gamma emission is the multipole order of the emitted gamma ray.
The measurement of gamma-gamma directional correlations in which at least one of the transitions is not a pure multipole is a sensitive method of determining the multipole mixing ratio, which is the ratio of the intensities of the electric to magnetic multipoles. It is a more sensitive method of measuring this quantity than the measurement of the internal conversion coefficients for the mixed transition. In the case of conversion electron-gamma correlations, nuclear structure information can be obtained (1) as a consequence of the finite size of the nucleus and the fact that atomic electrons can penetrate the nucleus.

The above mentioned measurements depend to some extent on having the decay processes take place in a field-free environment. It may be the case, however, that interaction of the nuclear moments (e.g. magnetic dipole or electric quadrupole) with extranuclear magnetic or electric fields takes place during the time that the intermediate state exists. The intermediate state refers to the nuclear state after the first decay but prior to the second decay in a two-step decay cascade. Cascades that comprise more than two decays are not considered here. These interactions can cause transitions between the magnetic substates of the intermediate nuclear state which change the relative population densities of the substates (2). Classically this effect may be thought of as a precession of the nucleus during the time of existence of the intermediate state. It is necessary to determine the extent of extranuclear effects and to correct for them, when indicated, before the results of directional correlation measurements can be properly interpreted. The perturbation of directional correlations by external fields has become a tool for measurement of magnetic moments (3) and also for estimating the electric quadrupole
moments (4) of excited nuclear states. Perturbations of both types can also exist due to the fields of the electronic structure of the atom at the nucleus. These atomic structure interactions may become very important in cases of decay by internal conversion since strong electric field gradients can occur at the site of the nucleus due to the vacancy in the electronic shell which exists immediately after the decay. The importance of the extra-nuclear effects depends in general on three quantities: (a) the lifetime of the intermediate state, (b) the magnitude of the nuclear moments which are involved in the interaction, and (c) the strength of the perturbing fields or field gradients at the site of the nucleus. Perturbations of directional correlations by extranuclear interactions can then be an additional tool for probing the nucleus and measuring nuclear moments of fairly short-lived nuclear states. If the moments are known from other experiments, the radioactive nucleus can be used as a tool in solid state physics to probe crystalline fields (4).

**Survey of Historical Development of Angular Correlation Theory**

The initial application of angular correlations to successive nuclear radiations arose from coincidence studies of beta-gamma cascade decays of radioactive nuclei. The possibility of using angular correlations for gaining an understanding of the radiation processes was first suggested by Dunworth (5) in 1940. The first study of angular correlation theory was made by Hamilton (6) in the same year. Hamilton applied perturbation theory to the study of gamma-gamma cascade decays for a few special pure multipole emissions. The theoretical development was greatly facilitated and rendered in a more useful form by the development of the
appropriate quantum mechanical algebra by Racah (7) (8) in 1942, which he later applied directly to the theory of angular correlations of nuclear radiations (9). Goertzel (10), in 1946, extended the theory to include cases where the nuclear moments interacted with extranuclear fields during the existence of the intermediate state. In 1949 Ling and Falkoff (11) extended the theory to include gamma-gamma cascades in which one or both of the transitions result in the emission of mixed multipole gamma rays.

The earliest theoretical treatments were restricted to gamma-gamma correlations. The extension to include beta-gamma directional correlations was made by Falkoff and Uhlenbeck (12) in 1950 and a further extension to include internally converted transitions in electron-gamma and electron-electron directional correlations were made by Rose, Biedenharn and Arfken (13) in 1951. A recent extension of the theory was prompted by Church and Weneser (14) when they showed that the internal conversion coefficients may depend on the details of nuclear structure. The directional correlation theory including these considerations was first worked out by Church and Weneser (14) and later by Green and Rose (15).

Survey of Development of Experimental Techniques

The first attempts to measure the effect predicted by Hamilton's theory were unsuccessful. It was suspected at the time that the attenuation of the correlations due to extranuclear fields predicted by Goertzel (10) was strong enough to mask the correlation. The first successful directional correlation measurement was made by Brady and Deutsch (16) in 1947 with Geiger counters as the particle detectors. The introduction of scintillating crystals and photomultiplier tubes by Brady and Deutsch in 1947 (17) made a radical change in the counting time for a typical
experiment. The usual one day of counting with scintillation counters would require approximately one thousand years of counting with Geiger counters. Continuous improvements have been made in the properties of the scintillation crystals, the photomultiplier tubes, and the associated electronic circuitry. However, the essential technique in current usage derives from that employed by Brady and Deutsch (17).

Previous Investigations of Cs\textsuperscript{133} Ba\textsuperscript{133} decays to excited states of Cs\textsuperscript{133} by orbital electron capture. The excited states then decay to the ground state via a system of gamma cascades (see Fig. 1). There have been ten reported gamma rays, some of which have been confirmed several times (18) (19) (20) (21) (22) (23) (24) (25). A recent report (26) gives the conversion electron spectrum detected with semiconductor counters. Earlier work on comparison of experimental conversion coefficients with the theoretically calculated coefficients for the 81 keV and 355 keV transitions was done by Gupta et al. (20).

Decay schemes have been proposed by Hayward et al. (18), Craseman et al. (19), Gupta et al. (20), Koicki et al. (21), Stewart et al. (22), Ramaswamy et al. (23), Bodenstedt et al. (27) Fagg (28), Subba Rao et al. (25), and Yin et al. (29).

The spin and parity assignments of the nuclear levels made on the basis of previous research can be summarized in the following table:

<table>
<thead>
<tr>
<th>Reference</th>
<th>436 keV</th>
<th>382 keV</th>
<th>160 keV</th>
<th>81 keV</th>
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<tr>
<td>(20)</td>
<td>$\frac{1}{2}^+$ or $\frac{3}{2}^+$</td>
<td>$\frac{3}{2}^+$ or $\frac{5}{2}^+$</td>
<td>-</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(21)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(22)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(23)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(27)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(25)</td>
<td>$\frac{3}{2}^+$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>(29)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
<tr>
<td>(30)</td>
<td>$\frac{1}{2}^+$</td>
<td>$\frac{3}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
<td>$\frac{5}{2}^+$</td>
</tr>
</tbody>
</table>
Figure 1: Decay Scheme of Ba$^{133}$
Spin and parity assignments were determined by studies of gamma and conversion electron spectra by use of beta ray and scintillation spectrometers (20) (21) (22) (23) and directional correlations (25) (27) (29) (30). The spin of the ground state of Cs$^{133}$ has been measured by an atomic beam experiment (31) as $\frac{7}{2}$.

The parities of all the states in Cs$^{133}$ have been reported as even so that the multipolarities of the transitions between the states will be of even parity. Of particular interest are the multipolarities of transitions involving the nuclear states for which disagreement exists in the reported value of the spin. There are two such cases: the 436 kev level and the 160 kev level. In the case of the 160 kev level, the spin assignment of $\frac{3}{2}$ (21) (22) was made using coincidence spectrometry of conversion electrons and gamma rays. A third measurement (23) yielded a value $\frac{5}{2}$ by similar methods which agreed with directional correlation measurements (27) (29) (30) of the 160 kev gamma and the 81 kev gamma. The careful analysis of Yin and Wiedenbeck (29) provides reasonably strong evidence that the spin of the 160 kev state is $\frac{5}{2}$. The spin of the 436 kev level has been reported recently as $\frac{3}{2}$ (25) rather than $\frac{1}{2}$ which had been previously reported by several authors (27) (23) (22) (21). The multipolarity of the 355 kev transition, which previously was reported as E2, has now been reported as a predominantly M1 transition with an admixture of E2 (25).

Bodenstedt et al. (27) observed no attenuation of the 355 kev gamma-81 kev gamma directional correlation in a delayed coincidence experiment with the Ba$^{133}$ source in aqueous solution. Although several values of the 81 kev transition multipole mixing ratio have been
reported (27) (25) (26), the various articles agree that the transition is predominantly M1.

The half life of the 81 kev level of Cs\textsuperscript{133} has been measured as $6 \times 10^{-9}$ sec. (32) and $6.31 \times 10^{-9}$ sec. (27) by delayed coincidence techniques. Upper limits were placed on the half lives of the 160 kev and the 436 kev levels as $5 \times 10^{-10}$ sec. and $1.5 \times 10^{-9}$ sec., respectively (27).

The magnetic moment of the 81 kev state has been measured as $3.10 \pm 0.03$ nuclear magnetons (27). This value was determined by two separate measurements: (a) by observation of the rotation and attenuation of the 355 kev gamma-81 kev gamma directional correlation in a constant external magnetic field and (b) by measuring the same correlation as a function of the magnetic field. A recent report (33) gives a value of $3.25 \pm 0.15$ nuclear magnetons for the 81 kev level, which is the result of an azimuthal shift type directional correlation measurement between the 355 kev and 81 kev gamma rays. The magnetic moment of the ground state has been measured as 2.577 nuclear magnetons and the nuclear quadrupole moment has been measured as $-0.003 \ (e \times 10^{-24} \ cm^2)$ (34).

Nuclear structure effects on the internal conversion coefficient of the 81 kev transition have recently been reported (25), (26).

**Purpose of This Research**

The investigation of the decay scheme of Cs\textsuperscript{133} is of particular interest because certain properties of the nucleus including the spins and parities of several excited states are in good agreement with the shell model. Recently, a disagreement on the spin assignment to the
436 kev level has developed. The 355 kev gamma-81 kev gamma directional correlation experiments that have been performed have generally indicated that the spin sequence is $1/2 (E2) \ 5/2 (M1 + E2) \ 7/2$. This result conflicts with the spin sequence suggested by Subba Rao (25), based on the results of the directional correlation between the 355 kev gamma and the 81 kev conversion electron, who concludes that the spin sequence is $3/2 (M1 + E2) \ 5/2 (M1 + E2) \ 7/2$.

It is desirable from the point of view of nuclear model theory to resolve disagreements of this nature by performing an independent experiment. In this instance the directional correlation between the conversion electron from the 355 kev transition and the 81 kev gamma ray can serve as the independent experiment for verification of the spin sequence. It is also of interest to compare such a directional correlation with the 355 kev gamma-81 kev gamma directional correlation in order to verify, for this particular case, the validity of the particle parameters appropriate to interpretation of correlations involving conversion electrons.

In order to accomplish the objectives three directional correlation experiments must be performed: (a) between the 81 kev gamma and the conversion electron from the 355 kev transition, (b) between the 81 kev gamma and the 355 kev gamma, and (c) the gamma-gamma correlation again, as a function of delay between the 355 and 81 kev gammas, for delays comparable to the lifetime of the 81 kev level.

Experiments (a) and (b) permit an independent verification of the spin of the 436 kev level. These experiments also permit an independent determination of the multipole mixing ratio of the 81 kev transition and, if the results of the two experiments are mutually consistent,
the validity of the particle parameters used in the present instance will be supported. It may be noted that the multipole mixing ratio is sensitive to details of the nuclear model and unambiguous experimental determination of the mixing ratio is valuable for comparison with theoretical calculations which, in the Cs$^{133}$ nucleus, may be premised on the shell model. Experiment (c) is to be performed for the purpose of determining the extent, if any, of attenuation of the directional correlation by extranuclear fields. This information is needed in order to preclude an erroneous interpretation of the other experiments.

In directional correlation measurements involving internal conversion electrons, difficulties may arise due to scattering of the electrons in the source material and in the air surrounding the source (4). Scattering effects are more pronounced for low electron energies. These considerations suggest that a directional correlation experiment with conversion electrons from the 355 kev transition might be more reliable than an experiment with conversion electrons from the 81 kev transition, as used by Subba Rao (25). Furthermore, previous work strongly suggests that nuclear penetration effects are significant in the conversion of the 81 kev transition, hence, it is desirable to avoid the use of measurements on these conversion electrons when making spin and multipolarity assignments.
CHAPTER II

EXPERIMENTAL PROCEDURE

Introduction

The apparatus consisted of two counters and associated electronic circuitry and data recording equipment. The purpose of the mechanical part of the apparatus was to permit motion of one counter relative to the fixed radioactive source and the other counter which was also fixed. The coincidence counting rate between the two counters could then be studied as a function of the angle between the trajectories of the two radiations. During part of the experiment, the fixed counter detected internal conversion electrons. For those measurements a vacuum system was provided which contained the source and the fixed counter.

The principal purposes of the electronic part of the apparatus were: (a) to select both radiations according to their energy and (b) to select coincidence events between the two counters, i.e. events which occurred at the same time, within the resolving time of the circuitry, in the two detectors. The circuits also controlled the mechanical system and the data recording devices so that data could be recorded automatically over extended periods of time.

Electronic System

The instrumentation employed in this experiment was essentially a standard fast-slow coincidence, two counter, scintillation spectrometer capable of detecting coincidences between radiations of preselected energies. In the event that the directional correlation was measured as a function of
delay time, the fast coincidence circuit also acted as a pulse overlap time to pulse height converter. Integrated coincidence pulses from the converter were grouped according to their delay time by use of an Atomic Instrument Company model 520 twenty-channel pulse height analyzer. A block diagram of the instrumentation is shown in Fig. 2.

All gamma rays were detected in 1 $\frac{1}{2}$ inch diameter by 1 inch thick NaI(Tl) scintillation crystals supplied by the Harshaw Chemical Company. During the internal conversion electron-gamma directional correlation, the 319 kev electron was detected in a 1 mm. thick by $\frac{1}{2}$ inch diameter anthracene crystal bonded to a 3 mm. thick lucite light pipe. The anthracene was also supplied by the Harshaw Chemical Company. The scintillators were optically joined to the faces of the photomultiplier tubes with Dow Corning No. 200 silicon fluid. R. C. A. 6810-A, fourteen stage, curved cathode, photomultiplier tubes were used in both counters for all experiments discussed. The tubes were operated with -2350 volts applied to the cathodes and with the anodes near ground potential. Voltages were supplied to the various dynodes, etc. by an adjustable bleeder network. This circuit allowed the optimum gain or energy resolution conditions to be achieved for each individual tube. The circuitry and procedure for optimizing tube operating conditions is discussed by Belletini et al. (35).

Pulses used for fast coincidence timing were fed directly from the anodes of the multiplier tubes to transistorized limiter circuits designed by Dr. E. T. Patronis (see Fig. 3). The two limiter output pulses passed through RG-114/U cables into the two ends of a continuously variable delay line of matching impedance (36). The pulses were fed from the variable delay to the coincidence integrator via an RG 62/U cable. The
Figure 2: Block Diagram of Instrumentation

[Diagram showing a block diagram with various components such as Det. 1, Det. 2, DDL AMP, fast coinc. integrator, linear AMP, Cath. follower, Disc, 20 channel pulse sorter, Gate, Triple coinc, delay, scalers, and a printer.]
Figure 3: Limiter and Coincidence Circuitry
pulses were clipped at the entrance to the coincidence integrator, which was of the type introduced by Bell and Petch (37), by a shorted cable. The down and back time for the clipper was 15 ns. The pulses which passed the reverse biased diode then passed through a cathode follower stage to a model N-301 Hamner linear amplifier. During delayed directional correlation measurements, this amplifier output was sent to the 20 channel pulse height analyzer, which was gated by the slow coincidence pulse. The fast circuit amplifier output also went to a pulse height analyzer whose output, in turn, was fed to the slow coincidence circuit.

The energy selection was accomplished by the use of pulses from the eleventh dynode of each photomultiplier. The dynode pulses went directly to cathode follower stages then to double delay line amplifiers of the type designed by Patronis (38). The amplifier outputs were fed to Hamner model N601 single-channel pulse height analyzers. The two analyzer output pulses went to the slow coincidence circuit input. The slow coincidence circuit registered a triple coincidence in the event that it received (a) a pulse from the fast amplifier discriminator output and (b) pulses from each of the two single channel analyzers. All three pulses had to arrive within a time interval of about one microsecond of one another in order to give a triple coincidence. The slow coincidence circuit was a transistorized circuit designed and built by Dr. E. T. Patronis.

**Mechanical System**

A gamma-ray counter was mounted on a rotating table which was mounted on a hollow ring gear salvaged from an aircraft fire control director. The rotation was motorized and automatically controlled to permit counting in sequence at predetermined positions for preset time intervals.
The other counter, which detected gamma rays or conversion electrons, was in a fixed position. Both counters were aimed at the axis of symmetry of the ring gear on which the radioactive source was located during directional correlation measurements. The system was originally designed and built by Dr. E. T. Patronis and Mr. N. S. Kendrick and was altered to fit the needs of the present experiment. The counting geometry is shown in Fig. 4.

**Source Preparation**

The $^{133}$Ba was produced at the Oak Ridge National Laboratory by irradiation of $^{132}$Ba with thermal neutrons. The source sample was prepared by evaporating several drops of BaCl$_2$ in HCl solution on a 0.25 mil aluminized Mylar film. The source was sealed with a thin coating of spray lacquer and covered with a second Mylar film which was vented with a pumping port. This was done as a precaution against leakage of the source from its mount. It was found that uncovered sources did indeed leak even when sprayed with lacquer. The source strength was estimated as being approximately five microcuries by counting 319 kev conversion electrons in a known geometry in the vacuum chamber. A similar source was prepared for use in the chance coincidence rate measurement.

**Instrumental Asymmetry**

The photomultiplier tube associated with the movable gamma detector can demonstrate a position-dependent gain and cause subsequent instrumental asymmetries due to effects of the earth's magnetic field. Such effects were found to exist and were corrected by shielding the moving gamma counter with Netic S3-6 magnetic shielding alloy wrapped on the outside of Co-Netic AA foil. The foils were purchased from the Magnetic Shield Division,
Figure 4: Directional Correlation Experiment Geometry
Perfection Mica Company. The fixed counter photomultiplier was shielded in a comparable manner to prevent magnetic interaction between the counters.

The instrument was tested for possible inherent asymmetries by measuring the directional correlation between the allowed beta ray and subsequent gamma ray in the decay of Co\textsuperscript{60}. This correlation is theoretically predicted to be isotropic (39) and has been measured as isotropic (40). The measurement of the Co\textsuperscript{60} correlation on this instrument was isotropic to within the statistical error based on 20,000 counts at the 180 degree position. In all subsequent data analysis and in the assignment of error limits for the various directional correlation measurements, it has been assumed that no inherent instrumental asymmetry exists.

It may also be noted that in each directional correlation measurement the source was centered by adjusting its position until the singles counting rates in the movable counter at the 90°, 180°, and 270° positions were equal.

\textbf{Conversion Electron-Gamma Directional Correlation}

To prevent electron scattering the source was mounted inside a thin-walled aluminum vacuum chamber. The chamber was 7.6 cm. in diameter by 15 cm. high and was mounted with its axis vertical and coincident with the axis of the ring gear and the axis of rotation of the movable counter. A horizontal aluminum tube was welded into a hole in the side of the vertical chamber. The horizontal tube housed the electron counter so that the anthracene crystal used for detection of electrons was inside the evacuated region. The source holder was suspended from the top of the vertical chamber and was provided with a source centering device. The vacuum chamber was kept at a pressure of about 20 microns of mercury.
The source was oriented in the chamber so that the electrons had to pass through the 0.25 mil Mylar film onto which the radioactive material had been evaporated. The electron energy resolution measured under these conditions was not noticeably different from that measured with an uncovered source.

**The Electron Spectrum Measurement**

The electron energy spectrum in Cs\(^{133}\) was measured by setting the differential analyzer energy window to 0.25 volt and recording the count rate at 0.5 volt intervals over the range from 5 volts to 50 volts. This was done for various distributions of the potentials applied to the photomultiplier electrodes and the optimum distribution was that for which the largest energy separation occurred between the 267 kev and 319 kev lines in the conversion electron spectrum. The instrument still appeared to be approximately linear in energy to pulse height characteristics. The spectrum was interpreted with the aid of the spectrum published by Nieschmidt et al. (26). The observed spectrum is shown in Fig. 5 along with an interpretation of its main features: The notation 355 K denotes the electrons due to the internal conversion of the 355 kev transition in the K shell which yields electrons of 319 kev; the peak denoted as 302 K and 274 L is a composite peak due to conversion of the 302 kev transition in the K shell and the conversion of the 274 kev transition in the L shell, both of which yield electrons of about 267 kev. The part of the electron spectrum accepted was that indicated by the crosshatched region of Fig. 5. The spectrum was run daily at different times of day to make certain that the energy selection system was functioning properly.

**81 kev Gamma Ray**

The 81 kev gamma line in Cs\(^{133}\) was identified by comparison with
Figure 5: Internal Conversion Spectrum in Cs$^{133}$
the 84 kev gamma line in Tm\textsuperscript{170}. The entire 81 kev gamma line was accepted for purposes of gamma-ray energy selection.

**Time Alignment and Time Resolution Measurement**

The time resolution and optimum time alignment of the instrumentation were measured using a Bi\textsuperscript{207} source. This nuclide decays mainly by orbital electron capture followed promptly by a 570 kev gamma, or by electron capture to a higher metastable state which decays by a cascade of a 1060 kev gamma followed promptly by the 570 kev gamma. Both transitions are partly internally converted. The electron counter differential analyzer was set to accept pulses corresponding to the conversion line from the 570 kev transition (480 kev) while the gamma counter analyzer was set to accept the pulses corresponding to the Pb K series x-ray line (about 74 kev). An 8.3 nanosecond delay cable was installed between the gamma counter limiter and the coincidence circuit. Triple coincidence rates were recorded for each of a series of RG114 cables installed between the electron counter limiter and the coincidence circuit. The electron counter cables ranged in delay time from 8.3 nanoseconds to 30 nanoseconds in increments of 3 nanoseconds. The procedure was repeated with the roles of the counters interchanged. The continuously variable delay was then installed and the alignment repeated. The variable delay did not appear to introduce a noticeable change in the results. The cables were installed for which the triple coincidence rate was a maximum. The variable delay was then set at various positions at 1 ns intervals while the triple coincidence rate was recorded for each position. The time alignment for which the coincidence rate was maximized was assumed to be the optimum alignment.
The time resolution, $2\tau$, was deduced from a plot of the coincidence rate vs. time delay introduced. Fig. 6 is a typical curve of this type. The time resolution is defined, for the purposes of this discussion, as the width at half-maximum of a plot of coincidence rate vs. time delay introduced into one counting channel. The time resolution was set at $17 \pm 0.5$ nanoseconds by adjustment of the coincidence circuit diode bias and the fast coincidence amplifier discriminator level.

**Directional Correlation Measurement**

The directional correlation measurement consisted of counting triple coincidences at counting positions of 90°, 180°, and 270°. The counts were taken for 15 minute intervals for each position of the rotating counter. The following data were automatically recorded: gamma counts for a selected energy, conversion electron counts for a selected energy, fast coincidence pulses of pulse height above the discrimination level, and the triple coincidence events. The data were recorded on tape by a four channel printer. The data were normalized by multiplying the number of triple coincidences by the quantity

$$\Gamma(\theta) = N_\gamma(180°) N_e(180°)/N_\gamma(\theta) N_e(\theta).$$

Here $N_\gamma(\theta)$ and $N_e(\theta)$ are the gamma and electron counting rates, respectively, for an angle $\theta$ between the two detectors. A sample of the data is shown in Table 1.

**Chance Coincidence Correction**

The chance coincidence or accidental coincidence rate was measured daily by the method of separate sources. The gamma counter was moved to a position approximately 1 meter from the chamber. A second source of Ba$^{133}$
Figure 6: Prompt Curve for Conversion Electron - X-Ray Coincidences in Ba-207

Delay of X-Ray in ns.

Triple Coincidence Rate in Arbitrary Units

17 ± 0.5 ns = 2T
Table 1. Sample of Data for 319 kev Conversion Electron - 81 kev Gamma kev Gamma Directional Correlation.

<table>
<thead>
<tr>
<th>Angle in Degrees</th>
<th>Triple Coincidences</th>
<th>Gamma Counter Singles $\div 4 \times 10^3$ $N_2(\theta)$</th>
<th>Electron Counter Singles $\div 1 \times 10^3$ $N_1(\theta)$</th>
<th>Fast Coincidences $\div 10^3$ $N_E$</th>
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was placed at a distance such that the gamma counting rate was the same as the rate during the directional correlation experiment. The counters were then counting their normal rates but the events should be uncorrelated in time. Any coincidences were assumed to be uncorrelated in time. Any coincidences were assumed to be accidental coincidences. The average accidental rate was found to be 13% of the total coincidence rate.

The difference between the total triple coincidence rate and the accidental coincidence rate was assumed to be the real coincidence rate.

**Result for 319 kev Conversion Electron - 81 kev Gamma Directional Correlation**

Analysis of the data is summarized in Appendix C. The experimentally determined asymmetry parameter is defined as

\[ A = \frac{2N(180^\circ)}{N(90^\circ) + N(270^\circ)} \]

where \( N(\theta) \) is the observed triple coincidence rate at counting position \( \theta \) after correction by the normalizing factor \( \Gamma(\theta) \) and after correction for accidental coincidences. The average 319 kev conversion electron-81 kev gamma asymmetry parameter was found to be \( A = 1.09 \pm 0.01 \) where the error limits represent, essentially, the probable statistical error based on 14,210 real coincidence counts at the 180° position.

The directional correlation, \( W(\theta) \) is most conveniently expressed as an expansion in even order Legendre polynomials:

\[ W(\theta) = 1 + A_2P_2(\cos \theta) + A_4P_4(\cos \theta) + \ldots \]

In the present decay, terms of higher order than \( P_4 \) are excluded by virtue of the fact that the multipolarities of the transitions are presumed to be
no larger than two.

From a measurement of only the asymmetry parameter it is not possible to evaluate both $A_2$ and $A_4$. However, on the basis of gamma-gamma measurements for this same cascade (25) (27), it may be assumed that the $A_4$ coefficient is small. The $A_2$ coefficient may then be evaluated from $A_2 = 2(A-1)/(A+2)$, where one assumes $A_4 = 0$ for $i > 2$. In this manner one finds $A_2$ (uncorrected) = 0.061 ± 0.006.

The result must, finally, be corrected for the finite solid angles subtended by the two detectors, as outlined in Appendix A. The final experimental result is $A_2 = 0.074 ± 0.008$.

**Gamma-Gamma Directional Correlation**

Measurement of the directional correlation between the 355 kev gamma-ray and the 81 kev gamma ray in conjunction with measurement of the K conversion electron-gamma correlation of the same transitions yields a value for the particle parameter, $b_2$, of the 355 kev transition (41). For the gamma-gamma experiment the vacuum chamber was removed. The stationary counter was fitted with a NaI(Tl) crystal.

**Gamma-Ray Spectrum Measurement**

The moving counter was used to detect the 355 kev gamma. A 1 mm. thick lead shield was inserted in front of this counter in order to decrease the intensity of low-energy gamma rays and the x-rays. The reduction in rate of unwanted pulses enhances operation of the limiter by increasing the probability that it will be in a quiescent state when activated by a pulse. The fixed counter was used to detect the 81 kev gamma. A 0.1 mm. thick lead shield was inserted in front of this counter in order to decrease the 31 kev x-ray intensity. The energy region of
the gamma spectrum selected for the 355 kev gamma detector is indicated by the crosshatched area shown in Fig. 7. The low energy gamma ray detection system was set to accept the entire 79-81 kev gamma peak (see Fig. 8).

Time Alignment and Time Resolution Measurement

The procedure used in time alignment and the measurement of time resolution were essentially the same as that followed for the electron-gamma experiment. In this case, however, Eu$^{154}$ was used as a test source and the coincidences detected were between the 1280 kev gamma ray and the 123 kev gamma ray. The results of this experiment are shown in Fig. 9. The resolving time was set, in the manner discussed previously for the electron-gamma experiment, to be 16.5 ± 0.5 ns. The asymmetry in the curve is due to the finite half life of 1.2 ns. for the 123 kev first excited state in Eu$^{154}$.

Gamma-Gamma Directional Correlation Measurement

Triple coincidences were counted for 10 minute intervals at positions of 90°, 180°, and 270°. A daily measurement of the accidental coincidence rate was made and daily time alignment was performed to check the stability of the instrumentation. The accidental rate was measured by use of two sources and was found to be approximately 8% of the triple coincidence rate. A typical sample of the data is shown in Table 2.

Result of 355 kev Gamma - 81 kev Gamma Directional Correlation Measurement

Analysis of the data was similar to that employed for the conversion electron-gamma correlation and is outlined in Appendix C. The asymmetry parameter was found to be $A = 1.058 ± 0.008$. Assuming $A_4 = 0$,
Figure 8: Gamma Ray Single Spectrum of Cs133 with 0.1 mm of Lead Shielding
Setting of Differential Pulse Height Analyzer in Volts

Figure 7: Gamma Ray Singles Spectrum of Cs$^{133}$ with 1.0 mm. Lead Shield
Delay Time of 123 keV Gamma Ray in ns.

Figure 9: Gamma-Gamma Coincidence Curve in Eu^{154}
Table 2. Sample of Data for the 355 kev Gamma – 81 kev Gamma Directional Correlation

<table>
<thead>
<tr>
<th>Angle in Degrees $\theta$</th>
<th>Triple Coincidences</th>
<th>Moving Counter Gamma Singles $\div 16 \times 10^3 N_2(\theta)$</th>
<th>Fixed Counter Gamma Singles $\div 16 \times 10^3 N_1(\theta)$</th>
<th>Fast Coincidences $\div 10^3 N_f$</th>
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A_2\ (\text{uncorrected}) = 0.038 \pm 0.005 \text{ and the final result corrected for finite detector size, as discussed in Appendix A, is } A_2 = 0.040 \pm 0.006. \text{ The probable statistical error is based on 20,194 real coincidence counts at 180°.}

Delayed Gamma-Gamma Directional Correlation

The perturbing effect of extranuclear fields on the gamma-gamma directional correlation was probed by sorting out the correlation data into groups of events according to the delay time of the 81 kev gamma ray with respect to the 355 kev gamma ray. The separation was accomplished by means of a time to pulse-height conversion in the fast coincidence circuit. The pulses from the fast coincidence integrator, after amplification, were fed to the 20 channel pulse height analyzer. The events in each channel correspond to a particular range of delay times of the 81 kev gamma relative to the 355 kev gamma. The integrator circuit operates in such a manner that the largest output pulses are generated by input pulses in almost exact time coincidence, which gives the greatest overlap of the two input pulses. As the delay between the input pulses increases, the overlap decreases and a smaller output is generated.

Time Calibration of Analyzer Channels

In order to interpret the delayed correlation data each analyzer channel must be assigned a corresponding delay time. The method employed was essentially the time alignment procedure previously discussed. Coincidences were observed between the 570 kev gamma ray and the 74 kev x-ray in Bi_{207}. An almost symmetrical distribution of events in the analyzer channels was observed (see Fig. 10). The position of the center of the distribution was estimated to within 1/4 of a channel width. Using the
Figure 10: Delayed Triple Coincidences in $^{133}$Cs and $^{207}$Bi
Bi\textsuperscript{207} source, the triple coincidence rate vs. channel number was studied for delay times of 0, 5.3, 8.3, 11.3, 17.3, and 20.6 nanoseconds. The time delays were introduced by insertion of appropriate lengths of cable between one limiter and the coincidence circuit. The zero delay condition was presumed to be attained when the coincidence peak appeared in the highest channel. This condition corresponded to the maximum overlap of the two signals at the fast coincidence integrator. A curve of delay vs. channel in which the coincidence peak was centered is shown in Fig. 11. The error bars correspond to experimental uncertainties in delay time and in channel position of the centers of the distributions.

**Time Resolution Estimates**

The time resolution determination is less certain in this case than in those previously discussed because the resolving time was set here to a smaller value which makes the resolving time more sensitive to the energies of the radiations. The full width in time units at half maximum of the distribution for Bi\textsuperscript{207}, as shown in Fig. 10, provides an estimate of the resolving time $2\tau \approx 11$ ns. This finite resolving time is partially due to a transit time uncertainty in the photomultipliers, which is specified as approximately 4 ns, by the manufacturer, and partially due to an inherent statistical fluctuation in pulse height which is a well known property of NaI(Tl) scintillators (44). These effects are strongly dependent on gamma ray energy for energies below several hundred kev. The Bi\textsuperscript{207} x-ray has an energy of about 74 kev while the low energy Cs\textsuperscript{133} gamma ray has an energy of 81 kev. Thus the value of $2\tau$ when working with the Cs\textsuperscript{133} cascade should be somewhat less than 11 ns.
The pronounced asymmetry that appears in the coincidence distribution for the gamma-gamma cascade in Cs$^{133}$, which is also shown in Fig. 10, is a consequence of the 6 ns. life-time of the 81 kev level. A precise knowledge of the time resolution is not essential for analysis of the present experiments, however, it is necessary to show that the resolution is adequate to demonstrate a finite lifetime for the 81 kev level. Otherwise, a possible attenuation effect on the directional correlation could not be studied very well using the delayed coincidence method.

**Directional Correlation Measurement**

The correlation data were taken in 15 minute intervals by manually recording the coincidence events of the twenty channels and the gamma counts fulfilling pulse height conditions set by the two single channel analyzers. Certain other data were also recorded for purposes of monitoring operation of the circuits. The sequence of counting positions was randomized to eliminate systematic errors. Two runs were made, each of about 15 hours duration. Time alignment was checked before and after each run. No noticeable time shift was observed during the runs. A sample of the data is shown in Table 3. Analysis of the data was generally similar to that of the other directional correlations and is outlined in Appendix C.

The results of the measurement are presented in Fig. 12 as a plot of the asymmetry vs. delay of the 81 kev gamma ray. The coincidence data recorded by the twenty-channel analyzer were combined into three groups with average delay times of about 5, 12, and 18 ns. respectively. The horizontal error bars in Fig. 12 indicate the delay range included in each data group. The accidental coincidence rate was determined by use
Table 3. Sample of Data for the Delayed Directional Correlation Between the 355 kev Gamma and the 81 kev Gamma.

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<td>557</td>
<td>549</td>
<td>588</td>
<td>528</td>
<td>514</td>
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</table>

Note: The numbers represent counts for each channel at different angles and the total number of counts for each category.
Figure 12: Asymmetry Between 355 keV Gamma Rays and Coincident 81 keV Gamma Rays in Cs133 as a Function of Delay Time of the 81 keV Gamma Ray.
of two sources and was found to be approximately 6.5% of the total count rate at the 180° position. The number of coincidences recorded at 180° was 20,631; 14,995; and 19,120 for the groups delayed by 5, 12, and 18 ns., respectively. As shown in Fig. 12, the asymmetry shows no dependence on delay time, hence, the results of this experiment indicate that no attenuation of the directional correlation exists for which the decay time is on the order of 6 nanoseconds.

**Summary of Experimental Results**

The results of the directional correlation experiments are quoted here again for convenience. The $A_2$ coefficient, corrected for the finite solid angles subtended by the detectors, for the directional correlation between the conversion electron from the 355 keV transition and the 81 keV gamma ray is $0.074 \pm 0.008$. The corrected $A_2$ coefficient for the 355 keV gamma-81 keV gamma directional correlation is $0.040 \pm 0.006$. The error limits represent the statistical mean standard deviations based on the number of counts recorded in each experiment. The error limits could possibly be larger due to small instrumental asymmetries. The results of the delayed gamma-gamma directional correlation experiment indicate that no serious attenuation of the directional correlation exists for which the decay time is on the order of 6 nanoseconds.

The present result for the gamma-gamma directional correlation may be compared with $A_2 = 0.042 \pm 0.005$ found by Bodenstedt et al. (27), $A_2 = 0.046 \pm 0.011$ found by Subba Rao (25), $A_2 = 0.031 \pm 0.006$ found by Clikeman and Stewart (42), and $A_2 = 0.042 \pm 0.005$ found by Arya (43). In each of these previous studies, the coefficient $A_4$ was found to be...
zero, within the limits of experimental error. It is to be noted that
the present data were taken at only two independent angles so no determina-
tion of $A_4$ was possible. It was thought that previous work had fixed the
value of $A_4$ with good precision, hence, it was deemed best to devote all
of the counting time in the present work to a determination of the asymmetry.
CHAPTER III

INTERPRETATION OF THE RESULTS

Multipolarity of the 355 kev Transition

The results of the gamma-gamma and conversion electron-gamma directional correlations can be interpreted with respect to the multipolarity of the converted 355 kev transition by use of equations (B-15) and (B-17) of Appendix B. Assuming the 355 kev transition is a pure multipole, the directional correlation of the conversion electron-gamma cascade is given by equation (B-16) of Appendix B. The internal conversion directional correlation particle parameter \( b_2 \) is obtained from the experimental results of the two correlations by the relation

\[
b_2 = \frac{A_2(e^-,\gamma)}{A_2(\gamma,\gamma)}. \tag{3-1}
\]

Using the results of the present investigation \( b_2 \) is found to be 1.9 \( \pm \) 0.5. This value of the particle parameter was compared to the theoretical predictions of Biedenharn and Rose (41) presented in Fig. 13 as plots of \( b_2 \) vs. \( E/mc^2 \) where \( E \) is the total transition energy and \( mc^2 = 511 \) kev. For \( E = 355 \) kev \( b_2 \) is -1.42, 0.40, 1.24, and 1.70 for the transition multipolarities E1, M1, M2, and E2, respectively. In Fig. 13 the curve for the E1 case is not shown since \( b_2 \) is negative for energies below about 2\( mc^2 \). It is concluded that the 355 kev transition is electric quadrupole, E2.

It should be mentioned here that the particle parameters of reference (41) were calculated using the point nucleus approximation with no screening. These parameters are directly related to the internal conversion coefficients.
Figure 13: Internal Conversion Electron-Gamma Directional Correlation Particle Parameter vs E/mc² (From Biedenharn and Rose (41)).
and are subject to the same sources of inaccuracy due to finite nuclear size and penetration effects as are discussed in Appendix B. It has been shown by Sliv and Band (45) that at Z = 55, for electric radiation of this energy, the variation of point nucleus conversion coefficient values from those with finite nuclear size, screening, and penetration corrections included is less than 4%.

The Mixing Ratio of the 81 kev Transition

For the cascade 1/2(355 kev E2 gamma) 5/2(81 kev M1 + E2 gamma)7/2 the second transition has been found to be predominately M1 and the multipole mixing ratio has been determined by several investigators (20), (27), (42), (43) and (29). The results of the present investigation for the conversion electron-gamma directional correlation were used to determine the multipole mixing ratio, δ, of the 81 kev transition by plotting the theoretically predicted curve of $A_2$ vs. δ in Fig. 14 and in more detail in Fig. 15. The experimental value of $A_2$, with the error limits, is also shown in Fig. 15. There are two possible values for δ from the present directional correlation data alone. A selection of one of the two possible values of δ can be made by use of previous results on the gamma-gamma directional correlation. The value of δ was selected by plotting, in Fig. 16, the theoretically predicted curve of $A_4$ vs. δ for the gamma-gamma directional correlation. The experimental limits on $A_4$ (27), (29), (33), (42), (43) are also shown and they agree with the theoretical prediction of $A_4$ only for the smaller possible value of δ. It is concluded that the multipole mixing ratio of the 81 kev transition is $δ = -0.16 \pm 0.01$ which generally agrees with earlier reports (27), (29), (42) and (43) based on the gamma-gamma directional correlation.
Figure 14: $A_2(e^-,\gamma)$ vs the Multipole Mixing Ratio of the 81 kev Transition
Figure 15: $A_2(e^-\gamma)$ vs the Multipole Mixing Ratio of the 81 kev Transition.
Figure 16: $A_4(\gamma,\gamma)$ vs the Absolute Value of the Multipole Mixing Ratio of the 81 kev Transition
This value of $\delta$ is slightly outside the limits for agreement with the experimental value for the conversion coefficient for the 81 kev transition. However, the theoretical conversion coefficient for this 81 kev transition is expected to be only approximately correct due to the probable presence of penetration effects.

**Discussion**

The present results show that the 355 kev gamma-81 kev gamma and the 319 kev conversion electron-81 kev gamma directional correlations are consistent with one another and are consistent with a spin assignment of $1/2$ to the 436 kev level in Cs$^{133}$ and a value for the mixing ratio of the 81 kev transition of $-0.16 \pm 0.01$. These results are in agreement with the conclusions of several previous works based mainly on gamma-gamma directional correlations, as discussed in Chapter I.

The result which disagreed with the present interpretation and with the earlier interpretations was the experiment by Subba Rao (25) based mainly on the directional correlation between the 355 kev gamma and the 81 kev conversion electron. The precise reason for this disagreement cannot be ascertained, however, several ambiguities in the interpretation of Subba Rao's experiment may be noted. The particle parameters used in his analysis were deduced from the tables of Biedenharn and Rose (41). Extrapolation of the particle parameters from the lowest energy shown in the table, $E/mc^2 = 0.3$, to an energy of $E/mc^2 = 0.16$ was necessary and this extrapolation may introduce serious errors. Furthermore, in the case of low energy $\ell$-forbidden transitions it has been shown by Church and Weneser (14) that penetration of the orbital electron into the nucleus can cause the internal conversion coefficients and the directional correlation
particle parameters to depend on the details of the structure of the nucleus. For M1 transitions, such as the 81 kev transition in Cs$^{133}$, this effect may be pronounced (45). There are also experimental difficulties peculiar to Subba Rao's experiment. Scattering of the low energy conversion electrons by the source material may be serious and could result in an erroneous directional correlation determination. For these reasons it may be concluded that the experiment involving the conversion electrons from the higher energy transition should yield more reliable results for purposes of spin assignments and mixing ratio determinations.
CHAPTER IV

RECOMMENDATIONS FOR FURTHER RESEARCH

It is recommended that solid state electron detectors be employed in order to distinguish between the K and L shell conversion electrons. Theoretically the directional correlation involving the K shell conversion electron and that which involves the L or M shell conversion electron from the same transition are generally quite distinct, which makes good electron energy resolution desirable in order that the directional correlation measurements in cases involving conversion electrons may be attributed exclusively to electrons from a particular shell.

The conversion electron-gamma correlations involving K and L shell conversion electrons from the 355 kev transition could be studied again and, if the low electron energy did not lead to serious problems, the correlations involving K and L shell conversion electrons from the 81 kev transition could be studied. Such measurements would serve to compare the K and L shell particle parameters as well as to improve the reliability of the data used in assigning spins, multipolarities, and mixing ratios. Further insight into the nuclear penetration effects in the 81 kev transition could be gained by study of its conversion electrons.

It is further recommended that solid state electron counters be used in coincidence studies of the K to L + M ratio of the conversion electron from the 355 kev transition. This measurement, in conjunction with directional correlation data, would be helpful in verifying the spin assignment of the 436 kev level and the multipolarities and mixing ratios of the 355 kev and 81 kev transitions.
APPENDIX A

FINITE SOLID ANGLE CORRECTION

The directional correlation function is frequently written in the familiar form (41):

\[ W(\theta) = \sum_{k} A_k P_k(\cos \theta) \quad (A-1) \]

where the \( A_k \) are coefficients, with \( A_0 = 1 \), and the \( P_k \) are Legendre polynomials. The summation is over even values of \( k \).

Experimentally \( W(\theta) \) is proportional to the coincidence counting rate when the axes of the counters form an angle \( \theta \), with the radioactive source at the vertex. The \( A_k \) depend on the geometry of counting. In particular they depend on the solid angle subtended by the counters at the source.

A detailed study of this problem is given by Rose (46) whose results are used in correcting the data for finite angular resolution in this experiment.

The attenuated correlation coefficient \( A'_k \) is related to the unattenuated coefficient \( A_k \) by the following relation:

\[ A'_k = A_k \frac{J_k^{(1)} J_k^{(2)}}{J_0^{(1)} J_0^{(2)}} \quad (A-2) \]

where \( J_i^{(1)} \) is the \( i \)th order attenuation coefficient for one detector and \( J_i^{(2)} \) is the attenuation coefficient for the other detector.
The following equation has been derived by Rose (46):

\[ J_\ell = \int_0^\alpha P_\ell (\cos \beta) (1 - \exp[-\mu \cdot x(\beta)]) \sin \beta \, d\beta \quad (A-3) \]

where \( \mu \) is the absorption coefficient of the detecting crystal, \( x(\beta) \) is the distance traversed by the radiation incident upon a right circular cylindrical detector, \( \beta \) is the angle between the detector axis and the trajectory of the radiation, and \( \alpha \) is the half angle subtended by the face of the detector at the source.

**Corrections for the Electron-Gamma Correlation**

**Electron Counter Correction**

For full absorption \((x_\mu \to \infty)\) the result of \((A-3)\) is

\[ \frac{J_\ell}{J_0} = \frac{P_{\ell-1} (\cos \alpha) - \cos \alpha P_\ell (\cos \alpha)}{(1 + \ell)(1 - \cos \alpha)} \quad (A-4) \]

and

\[ \frac{J_2}{J_0} = \frac{P_1 (\cos \alpha) - \cos \alpha P_2 (\cos \alpha)}{3(1 - \cos \alpha)} \quad (A-5) \]

For the electron counter the 1.5 inch diameter by 0.0394 inch thick anthracene crystal was mounted with its face 1.8 inches from the source. The crystal was mounted so that approximately 0.4 inches around the rim was masked in the mount causing the effective radius to be 0.71 inches. For this geometry \( \cos \alpha = 0.930 \)

\[ P_1(0.930) = 0.930 \text{ and } P_2(0.930) = 0.800 \]

Using equation \((A-5)\) we see that \( J_2(e)/J_0(e) = 0.887. \)
Gamma Counter Correction

The results of calculations of $J_2/J_o$ for NaI(Tl) crystals have been tabulated by Stanford and Rivers (47). In the electron-gamma experiment the gamma counter was located so that the front of the 1.5 inch diameter by 1 inch thick NaI(Tl) crystal was located 2.12 inches from the $^{133}$Ba source. The energy of the gamma ray was 81 kev. Fig. 1 of Ref. (47) gives $J_2(\gamma)/J_o(\gamma) = 0.928$.

The finite solid angle correction for both counters is then 0.825.

Corrections for Gamma-Gamma Correlation

During the gamma-gamma directional correlation the moving counter crystal was 4.5 inches from the source and it detected the 355 kev gamma ray. The stationary counter was 3.5 inches from the source and detected the 81 kev gamma ray. Both crystals were 1.5 inch diameter by 1 inch thick NaI(Tl). The finite solid angle correction is then found to be (47) 0.957.
APPENDIX B

THEORETICAL CONSIDERATIONS

Angular Correlation Theory

In this section selected results of angular correlation theory that appear in the literature are outlined. Considerations of interest in the interpretation of the present experiments are included but no comprehensive review of the theory is intended.

For the development of the correlation function, consider two successive nuclear radiations which have propagation directions $\vec{k}_1$ and $\vec{k}_2$, respectively, and which accompany the transitions $a \to b$ and $b \to c$, where $a$, $b$, and $c$ are nuclear states. Let each state be characterized by a total angular momentum quantum number $J$ which is degenerate with respect to the $(2J + 1)$ values of the projection quantum number $m$. The correlation function

$$W(\theta) = W(\vec{k}_1 \cdot \vec{k}_2)$$

is defined as the relative probability that the propagation vector $\vec{k}_2$ will be rotated an angle $\theta$ from the propagation vector $\vec{k}_1$. Hamilton (6) treated $W(\theta)$ for several gamma-gamma correlations by applying second order perturbation theory to an initial system of an excited nucleus and quantized radiation field. $W(\theta)$ was given as

$$W(\theta) = S_1 S_2 \sum_{m_a} \sum_{m_b} \sum_{m_c} \left| \langle m_c | H(\vec{k}_2) | m_b \rangle \langle m_b | H(\vec{k}_1) | m_a \rangle \right|^2$$  \hspace{1cm} (B-1)
where \( S_1 \) and \( S_2 \) denote sums over unobserved polarizations of the radiations.

Falkoff and Uhlenbeck (12) have shown that the interference terms in (B-1) can be eliminated by choosing the z-axis of quantization in the direction of either of the propagation vectors \( \vec{k}_1 \) or \( \vec{k}_2 \). Equation (B-1) then becomes

\[
W(\theta) = S_1 S_2 \sum_{m_b} \left[ \sum_{m_a} \left| \langle m_b | H_1(0) | m_a \rangle \right|^2 \right] \times \left[ \sum_{m_c} \left| \langle m_c | H_2(\theta) | m_b \rangle \right|^2 \right]. \tag{B-2}
\]

The summands of (B-2) have a particularly simple interpretation. To demonstrate this we define

\[
\begin{align*}
S_1 & | \langle m_b | H_1(0) | m_a \rangle |^2 = p_{ba}(\theta) \tag{B-3} \\
S_2 & | \langle m_c | H_2(\theta) | m_b \rangle |^2 = p_{cb}(\theta)
\end{align*}
\]

where \( p_{ba}(\theta) \) is the relative probability that \( \vec{k}_1 \) will be along the quantization axis and \( p_{cb}(\theta) \) is the relative probability that \( \vec{k}_2 \) will be at an angle \( \theta \) with respect to the quantization or z-axis. The correlation can be written in the form

\[
W(\theta) = \sum_{a, b, c} p_{ba}(0) p_{cb}(\theta). \tag{B-4}
\]

Angular Distributions and Intensities

The quantities \( p_{ba} \) and \( p_{cb} \) contain both angular dependence and intensity information of the transition between the appropriate substates. It has been shown (48) that the probability of the component transition between a state with total and z-component of angular momentum characterized
by the quantum number $J, m$ to one characterized by $J', m'$ has the form

$$P_{mm'}(\theta) = G(m, m') F_L^M(\theta)$$  \hspace{1cm} (B-5)$$

where $L, M$ are the quantum numbers characterizing the total and $z$-component of angular momentum carried off by the radiation. Furthermore, Eckart has shown (49) that in general the intensity of the $m, m'$ component

$$G(m, m') = \langle J'Lm'M|Jm^\rangle^2$$  \hspace{1cm} (B-6)$$

where the quantity on the right is the square of a vector addition coefficient. The angular dependent part of (B-5), $F_L^M(\theta)$, is called the angular distribution of the component characterized by the projection quantum number $M$ of the radiation of multipole order $2^L$.

Angular Distribution of Gamma Rays

The calculation of $F_L^M(\theta)$ for electromagnetic radiation can be made on the basis of semi-classical considerations. If an ensemble of oriented radioactive nuclei were located in the vicinity of a point, the angular distribution of gamma radiation would be proportional to the angular dependent factor of the Poynting vector of the radiation. Since the distribution is desired separately for each multipole order of the radiation, the electric and magnetic fields are most conveniently expressed in terms of the vector spherical harmonics (see Blatt and Weisskopf (50)). The electric Field for pure magnetic radiation, for example, is written

$$E = \sum_{L,M} f(r) \{a(LM) \vec{X}_L^M\}$$  \hspace{1cm} (B-7)$$

where
\[ \vec{X}_L^M = \frac{L(\text{operator})}{\sqrt{L(L+1)}} Y_L^M(\theta,\phi). \]  

(B-8)

At field points quite distant from the nuclei the radiation can be considered as being completely transverse radiation resulting in the Poynting vector magnitude

\[ |\vec{S}| \propto E^2 \]  

(B-9)

for which the angular dependence is completely contained in the expression

\[ F_L^M(\theta) = X_L^M(\theta) \cdot X_L^M(\theta). \]  

(B-10)

The calculation of (B-10) is facilitated by expressing the Cartesian components of the angular momentum operator in terms of the ladder operator \((L_x + iL_y)\). The resulting angular distribution for L, M gamma rays is

\[ F_L^M(\theta) = \frac{(L-M)(L+M+1)}{2L(L+1)} |Y_L^{M+1}|^2 + \frac{(L+M)(L-M+1)}{2L(L+1)} |Y_L^{M-1}|^2 + \frac{M^2}{2L(L+1)} |Y_L^M|^2. \]  

(B-11)

The aximuthal angular dependence is lost in the squaring of the spherical harmonics.

A quantized radiation field approach to multipole radiation yields the same results and has been discussed by Moszkowski (51).

**Angular Distributions of Internal Conversion Electrons**

For an internal conversion electron in a final state described by the wave function \( \Psi(\vec{r}, t) \), the angular distribution \( F_L^M(\theta) \) is obtained
from the radial component of the Dirac electron current per unit solid angle, which is given by

\[
F_L^M(\theta) = -r^2 \left\langle \mathbf{\Psi}(\mathbf{r},t) \right| \alpha_\tau \mathbf{\Psi}(\mathbf{r},t) \right\rangle. \tag{3-12}
\]

the calculation of \( F_L^M(\theta) \) has been made by Rose et al. (13).

Gamma-Gamma and Conversion Electron-Gamma Directional Correlations

When the polarization directions of the radiations are summed over, the directional correlation function has the following simple form (41)

\[
W(\theta) = \sum_{k=0}^{k_{\text{max}}} A_k P_k (\cos \theta) \tag{8-13}
\]

where \( k \) is even and \( k_{\text{max}} \) is the minimum of \( 2J_b \), \( 2L_1 \), or \( 2L_2 \). The explicit calculation of the coefficients \( A_k \) is facilitated by the fact that the coefficients can be written as the product of two factors, each of which depends on parameters of only one of the transitions of the cascade. For a gamma-gamma cascade of pure multipole radiations we have

\[
A_k = F_k (L_1 L_2 J_a J_b) F_k (L_2 L_2 J_a J_c). \tag{8-14}
\]

Values for the \( F \) coefficients are tabulated in references (52) and (41).

A<sub>k</sub> for Mixed Multipole Transitions

For any given transition the radiation given off can have any angular momentum consistent with the selection rule \( |J_a - J_c| \leq L \leq (J_a + J_c) \).

This implies that the radiations may be a mixture of many multipole orders. Actually, it is well known that in most instances an appreciable mixture of multipoles occur only in the case of the "parity unfavored" transitions. For example, mixtures of M1 and E2 radiations are common. In other cases
one multipole may be retarded by another selection rule ($\ell$-forbiddenness for example).

For the case of the correlation between a pure multipole transition $L_2$ and a mixed multipole transition of orders $L_1$ and $L'_1$, Rose (53) has shown that

$$W(\theta) = \sum_{k} P_k(\cos \theta) \left[ F_k\left(L_1 L_1 J_a J_b\right) + \frac{1}{2} F_{k'}\left(L'_1 L'_1 J_a J_b\right) \right]$$

$$+ 2\eta (-1)^{J_a - J_b + 1} \frac{1}{(2J_b + 1)(2L'_1 + 1)}$$

$$\times G_k\left(L_1 L'_1 J_a J_b\right) \times F_{k'}\left(L'_2 L_2 J_a J_b\right)/(1 + \eta^2).$$

The coefficients $G_k(L_1 L'_1 J_a J_b)$ are tabulated for $L'_1 = L_1 + 1$ in reference (41) and $\eta$ is the ratio of the reduced matrix element for the multipole of order $L'_1$ to that of order $L_1$ with $L'_1 \gg L$.

Relation Between Conversion Electron-Gamma and Gamma-Gamma Correlations

It has been shown by Rose et al. (13) that the directional correlation between a gamma ray and any particle, or between any two particles, may be found if the directional correlation for the gamma-gamma cascade between the same states is known. This fact is exhibited by the directional correlation function $W(\theta)$ for a conversion electron-gamma cascade

$$W(\theta, e\gamma) = \sum_{k=0}^{K_{\text{max}}} b_k A_k(\gamma\gamma) P_k(\cos \theta).$$

It has been assumed in (B-16) that the converted transition is a pure multipole. The quantity $b_k$ is known as the particle parameter and depends on the multipole order and the electric or magnetic character of the converted transition, the energy of the transition and the atomic number.
of the nucleus. The K shell conversion electron particle parameters are tabulated in reference (41).

For the case of a mixed gamma of multipole orders \( L_1 \) and \( L'_1 \) with \( L'_1 > L_1 \) and a conversion electron from a mixed transition of multipole orders \( L_2 \) and \( L'_2 \), with \( L'_2 > L_2 \), the directional correlation is (41)

\[
W(\theta) = \sum_k \left( \sum_l \left( \begin{array}{c} \delta_{L_1 L'_1} \delta_{J_a J_b} \\ \delta_{L_2 L'_2} \delta_{J_a J_b} \end{array} \right) + \delta_1 \frac{2}{1} \delta_{L_1 L'_1} \delta_{J_a J_b} + 2\delta_1 (-1)^J \right) J_a J_b
\]

\[
x \left[ \frac{1}{(2J_b + 1)(2L_1 + 1)(2L'_1 + 1)} \right]^{1/2} G_{J_a J_b} \]

\[
x \left[ \delta_{L_2 L'_2} \delta_{J_a J_b} + \frac{2}{1} \delta_{L_1 L'_1} \delta_{J_a J_b} \right] \frac{1}{(2J_b + 1)(2L_2 + 1)(2L'_2 + 1)}
\]

\[
x \left( \begin{array}{c} \frac{1}{(2J_b + 1)(2L_2 + 1)(2L'_2 + 1)} \frac{1}{(2J_b + 1)(2L_2 + 1)(2L'_2 + 1)} \end{array} \right) F_{K_a}(L_2 L'_2 c J_b)
\]

\[
J_a J_b \frac{1}{(1 + P^2)(1 + S_1^2)} .
\]

Here \( P = \delta_2 \left[ \frac{\alpha(L_2)}{\alpha(L'_2)} \right] \frac{1}{2} \) where \( \delta_2 \) is the mixing ratio for the gamma transition connecting states \( b \) and \( c \) and \( \alpha(L_2) \) and \( \alpha(L'_2) \) are the internal conversion coefficients, for the appropriate shell, of the transitions of multipolarity \( L_2 \) and \( L'_2 \) respectively. The mixed particle parameters, \( b_K(L_2 L'_2) \), depend on the multipolarities, atomic number, and transition energy, but are independent of \( K \). They are tabulated in reference (41) for the case of a mixed transition with magnetic radiation of multipolarity \( L_2 \) and electric radiation of multipolarity \( L'_2 = L_2 + 1 \).

Church et al. (54) found that the algebraic signs of the tabulated mixed particle parameters (41) were systematically reversed, which was subsequently verified by the original authors (55).
Perturbed Directional Correlations

The previous discussions and resulting formulas are valid only for directional correlations of radiations from nuclei which are not under the influence of extranuclear fields. It is seen by the form of (B-2) that if the intermediate states \( |m_b\rangle \) evolve in time during their existence, the resulting correlation function would be affected. Perturbing interactions may be an external magnetic field interacting with the nuclear magnetic dipole moment or an external electric field gradient interacting with the nuclear electric quadrupole moment. The interactions remove the magnetic substate degeneracies and cause transitions between the substates which change the relative populations of the substates. This effect causes angular correlations to be dependent upon parameters associated with the physical environment of the nucleus.

The interaction between the electric field gradient at the nucleus and the nuclear electric quadrupole moment can be significant even when the lifetime of the intermediate nuclear state is as short as \( 10^{-10} \) second. Some common sources of these extranuclear perturbations are: (a) interaction between the electric field gradient caused by a temporarily vacant electron state during electron capture or internal conversion and the nuclear electric quadrupole moment (b) the magnetic and electric interactions with nuclear moments of strong crystalline magnetic fields and electric field gradients.

The effect of extranuclear perturbations on angular correlations has been treated by many authors (4)(56)(57). It is convenient to write the unperturbed correlation in a more general form than (B-2) by expanding the intermediate state vectors in terms of an arbitrary complete set
spanning the same space

\[ W(\theta) = S_1 S_2 \sum_{m_a, \beta^m} \langle m_a | H_1 | \beta \rangle \langle \beta | H_2 | m_c \rangle \langle m_c | H_2 | \beta' \rangle \langle \beta' | H_1 | m_a \rangle. \]  

(B-18)

A perturbation on the intermediate state can be formulated in terms of a unitary operator \( U \) which describes the evolution of the state vectors \( |\beta\rangle \) in time. We then write

\[ W(\theta) = \sum_{m_a, \beta^m} \langle m_a | H_1 | \beta \rangle \langle \beta | U H_2 | m_c \rangle \]

\[ \times \langle m_c | H_2 U | \beta' \rangle \langle \beta' | H_1 | m_a \rangle. \]  

(B-19)

It is appropriate here to consider only the special case which arises in the present investigation, namely that of a time independent extra-nuclear interaction occurring in a source comprised of an ensemble of randomly oriented microcrystals. The perturbing Hamiltonian \( k \) for this case corresponds to the operator \( U = \exp(-i k t/\hbar) \). The extent of the evolution depends upon the length of time the particular nuclear state exists. The probability that a state will decay during the time interval \( t \) to \( t + dt \) is simply \( \exp(-t/T)(dt/T) \), where \( T \) is the mean life of the intermediate state. Since \( k \) represents a static interaction, the set of eigenvectors \( |\beta\rangle \) may be chosen as the eigenvectors of the perturbing Hamiltonian \( k \). The time dependent correlation function may then be written

\[ W(\theta, t) = 1/T \sum_{m_a, \beta^m} \int_0^t dt' e^{-t'/T} \exp[i(E \beta - E \beta') t'/\hbar] \]

\[ \times \langle m_a | H_1 | \beta \rangle \langle \beta | H_2 | m_c \rangle \langle m_c | H_2 | \beta' \rangle \langle \beta' | H_1 | m_a \rangle. \]  

(B-20)
where all coincidences between radiations of the cascade are observed that occur between times 0 and t. A simplification introduced by Alder (57) allows (B-20) to be written in the particularly simple form

\[ W(0, t) = \sum_{k=0}^{k_{\text{max}}} G_k(t) A_k P_k(\cos \theta). \]  

(B-21)

The time dependent attenuation parameters \( G_k(t) \) can have various forms depending upon the physical configuration of the source. Several cases of interest are discussed in references (4) and (56). For the integral correlation, which is obtained from (B-20) for the upper limit \( t \to \infty \), it has been shown (56) that for an axially symmetric static interaction the attenuation factors have an irreducible minimum or "hard core" limiting value given by \( G_k(\text{min}) = 1/(2k + 1) \).

An interesting case occurs for an axially symmetrical static electric quadrupole interaction in a microcrystalline source. It has been shown (56) that the delayed correlation has an oscillatory behavior. For an intermediate state of spin 5/2, such as the 81 kev level in Cs\(^{133} \),

\[ G_2(t) = 1/5 \left[ 1 + 13/7 \cos \omega t + 10/7 \cos 2\omega t + 5/7 \cos 3\omega t \right]. \]  

(B-22)

Here the correlation is observed at a time \( t \) after emission of the first radiation and \( \omega \) is a "precession" frequency characteristic of the quadrupole interaction.

**The Internal Conversion Process**

During the transition from one level to another a nucleus can give up energy \( k \), angular momentum \( L \) and parity \( \pi \) by the emission of a gamma ray. The transition can also occur with the transfer of energy,
angular momentum and parity to an orbital electron by internal conversion. The ratio of the transition rate of internal conversion to that of gamma emission is defined as the internal conversion coefficient $\alpha$.

The total conversion coefficient is the sum of coefficients corresponding to conversion in the K, L_I, L_{II}, L_{III}, ... etc. electron shells.

The calculation of the internal conversion coefficients depends to some degree on the model adopted to describe nuclear transition currents and charge distributions. With few exceptions, the coefficients calculated with simplifying assumptions, for example, a point nucleus with screening or a finite size static uniformly charged nucleus with the electron always outside and with screening, are not significantly different from those calculated with dynamic penetration effects. One of the exceptions is the case of a low energy magnetic transition which is $\ell$-forbidden (selection rule on the orbital angular momentum of the radiating nucleus).

The interaction between the electron and nuclear charges and transition currents occurs through the mechanism of the electromagnetic field. This interaction can be written as (59)

$$H' = \int d\tau_n d\tau_e (p_e \rho_n - j_e \cdot j_n) \frac{\exp \left[ i k | \vec{\tau}_n - \vec{\tau}_e | \right]}{| \vec{\tau}_n - \vec{\tau}_e |}$$

This represents the case in which the nucleus undergoes a transition of energy $k$, characterized by nuclear states $\phi_n \rightarrow \phi_f$, while an orbital electron is lifted from the state $e_i$ to the state $e_f$. The quantities $p_e$ and $j_e$ are the usual Dirac electron charge and transition current densities given by
\[
\rho_e = -e \left( \Psi_{ef}^* (\vec{r}_e) \sum_{\alpha} \Psi_{ei} (\vec{r}_e) \right) \\
\vec{J}_e = -e \left( \Psi_{ef} (\vec{r}_e) \vec{\alpha} \sum_{\alpha} \Psi_{ei} (\vec{r}_e) \right).
\]

(B-24)

The explicit forms for \(\rho_n\) and \(\vec{J}_n\) depend upon the nuclear model employed in the calculation.

Conversion coefficients have been tabulated for several nuclear models of which the two most common are: Rose (58) employs a uniformly charged sphere of radius \(1.2 A^{1/3} \times 10^{-13}\) cm, with Thomas-Fermi screening and with no penetration of the nucleus by the electrons; Sliv and Band (45) employ a model with the above features but for which the nuclear penetration effect is evaluated by the assumption that the nuclear transition current lies on the nuclear surface.

An expression for the conversion coefficient has been derived by Church and Weneser (59) as the product of the tabulated result of Sliv and Band (45) and a correction factor containing the dynamical penetration effects. The penetration effect may be important in the interpretation of the 81 kev transition in Cs\(^{133}\) since it is a predominantly M1 transition which has been labeled (27) as a transition between single particle levels \(d_{5/2} \rightarrow g_{7/2}\), which is \(\ell\)-forbidden. Observation of a penetration effect in this transition has been reported (25).
APPENDIX C

ANALYSIS OF DIRECTIONAL CORRELATION DATA

Electron-Gamma Correlation

The data for the electron-gamma directional correlation were divided into semi-daily groups and each group was analyzed as a separate experiment in order to detect systematic errors more readily. Each data group was normalized to the average 180° position gamma singles count by multiplying the triple coincidence count at each position by the factor

\[ \Gamma(\theta) = \frac{\overline{N}_1(180^\circ)}{N_2(180^\circ)/\overline{N}_1(\theta) N_2(\theta)} \]

where \(\overline{N}_1(\theta)\) and \(\overline{N}_2(\theta)\) are the average singles counts for position \(\theta\) for the electron counter and the gamma counter, respectively. A sample computation is given below for the data collected during one run.

\begin{table}[h]
\centering
\begin{tabular}{cccccc}
\hline
\(\theta\) & \(\overline{N}_T\) & \(\Gamma(\theta)\) & \(N'_T\) & \(\overline{NA}\) & \(N''\) \\
\hline
180° & 289.4 & 1.000 & 289.4 & 47 & 242.4 \\
90° & 278.0 & 0.981 & 272.8 & 47 & 225.8 \\
270° & 286.6 & 0.970 & 278.0 & 47 & 231.0 \\
\hline
\end{tabular}
\caption{(C-1)}
\end{table}

Here \(\overline{N}_T\) is the average number of triple coincidences, \(N'_T = \overline{N}_T \Gamma(\theta)\), \(\overline{NA}\) is the average number of accidental coincidences, and \(N'' = N'_T - \overline{NA}\). The resulting asymmetries are found to be
\[ \eta(\theta) = N'(180^\circ) / N'(\theta) \]
\[ \eta(90^\circ) = 1.07 \pm 0.03 \]
\[ \eta(270^\circ) = 1.05 \pm 0.03 . \]

The error assigned is the statistical mean standard deviation for this single run.

There were ten such data groups for the electron-gamma correlation. The results of each run were plotted versus run number in chronological order in order to demonstrate undesirable trends in the data. No such trends were observed.

The average asymmetry was computed for both 90° and 270° data by taking a weighted average of the individual data group results in the following fashion

\[ \bar{\eta}(\theta) = \sum_{j=1}^{10} S_j \eta_j(\theta) \]  

(C-3)

where \( S_j \) is the weighting factor defined by

\[ S_j = \left( \frac{\text{number of coinc. at } \theta \text{ for } j\text{th data group}}{\text{Number of coinc. at } \theta \text{ for entire experiment}} \right) . \]  

(C-4)

The resulting average asymmetries are

\[ \bar{\eta}(90^\circ) = 1.10 \pm 0.01 \]
\[ \bar{\eta}(270^\circ) = 1.09 \pm 0.01 \]
\[ \bar{\eta} = (\eta(90^\circ) + \eta(270^\circ)) / 2 = 1.10 \pm 0.01 . \]

A disagreement between the 90° and 270° results would indicate an instrumental asymmetry. No such asymmetry is noted.
Gamma-Gamma Correlation

The data for the gamma-gamma correlation were analyzed in a manner similar to that described in the previous section. Sample data for one run are shown below.

Table (C-2)

<table>
<thead>
<tr>
<th>$\theta$</th>
<th>$\overline{N_T}$</th>
<th>$\Gamma(\theta)$</th>
<th>$N'_T$</th>
<th>$\overline{N_A}$</th>
<th>$N''$</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°</td>
<td>240.8</td>
<td>1.017</td>
<td>244.9</td>
<td>20.3</td>
<td>224.6</td>
</tr>
<tr>
<td>90°</td>
<td>238.5</td>
<td>1.000</td>
<td>238.5</td>
<td>19.0</td>
<td>219.5</td>
</tr>
<tr>
<td>270°</td>
<td>238.5</td>
<td>0.984</td>
<td>234.7</td>
<td>18.2</td>
<td>216.5</td>
</tr>
</tbody>
</table>

In this case, $\Gamma(\theta)$ normalizes the coincidence count to the product of the gamma counter rates at 90°. For the data shown above

$\eta(90°) = 1.02 \pm 0.03$
$\eta(270°) = 1.04 \pm 0.03$

The results of the weighted average, for all data taken are

$\overline{\eta}(90°) = 1.05 \pm 0.008$
$\overline{\eta}(270°) = 1.06 \pm 0.01$
$\overline{\eta} = (\overline{\eta}(90°) + \overline{\eta}(270°))/2 = 1.06 \pm 0.01.$

Delayed Gamma-Gamma Correlation

The data for the delayed gamma-gamma directional correlation were analyzed in three data groups. Group A is comprised of coincidence events representing an average delay of the 81 kev gamma relative to the 355 kev gamma of approximately $5 \pm 5$ nanoseconds. Groups B and C represent coincidence events with delays of approximately $12 \pm 5$ and $18 \pm 5$ nanoseconds, respectively.
The data analysis is shown in Tables (C-3), (C-4) and (C-5) below.

Table (C-3)
Data Group A Corresponding to 5 ± 5 ns. Delay

<table>
<thead>
<tr>
<th>θ</th>
<th>N_T</th>
<th>Γ(θ)</th>
<th>N'_T</th>
<th>NA</th>
<th>N''</th>
<th>( \overline{\eta} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°</td>
<td>18837</td>
<td>1.015</td>
<td>19120</td>
<td>1338</td>
<td>17780</td>
<td>- - - - - - -</td>
</tr>
<tr>
<td>90°</td>
<td>18323</td>
<td>1.000</td>
<td>18323</td>
<td>1338</td>
<td>16985</td>
<td>1.047 ± 0.016</td>
</tr>
<tr>
<td>270°</td>
<td>18677</td>
<td>0.986</td>
<td>18415</td>
<td>1338</td>
<td>17077</td>
<td>1.041 ± 0.016</td>
</tr>
</tbody>
</table>

The average asymmetry for data group A is

\[ \overline{\eta} \text{ (5ns.)} = 1.04 ± 0.02. \]

Table (C-4)
Data Group B Corresponding to 12 ± 5 ns. Delay

<table>
<thead>
<tr>
<th>θ</th>
<th>N_T</th>
<th>Γ(θ)</th>
<th>N'_T</th>
<th>NA</th>
<th>N''</th>
<th>( \overline{\eta} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°</td>
<td>14773</td>
<td>1.015</td>
<td>14995</td>
<td>1050</td>
<td>13945</td>
<td>- - - - - - -</td>
</tr>
<tr>
<td>90°</td>
<td>14322</td>
<td>1.000</td>
<td>14322</td>
<td>1050</td>
<td>13272</td>
<td>1.051 ± .02</td>
</tr>
<tr>
<td>270°</td>
<td>14207</td>
<td>0.986</td>
<td>14008</td>
<td>1050</td>
<td>12958</td>
<td>1.076 ± .02</td>
</tr>
</tbody>
</table>

The average asymmetry for data group B is

\[ \overline{\eta} \text{ (12 ns.)} = 1.06 ± 0.02. \]

Table (C-5)
Data Group C Corresponding to 18 ± 5 ns. Delay

<table>
<thead>
<tr>
<th>θ</th>
<th>N_T</th>
<th>Γ(θ)</th>
<th>N'_T</th>
<th>NA</th>
<th>N''</th>
<th>( \overline{\eta} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°</td>
<td>20326</td>
<td>1.015</td>
<td>20631</td>
<td>1444</td>
<td>19187</td>
<td>- - - - - - -</td>
</tr>
<tr>
<td>90°</td>
<td>19437</td>
<td>1.000</td>
<td>19437</td>
<td>1444</td>
<td>17993</td>
<td>1.066 ± 0.015</td>
</tr>
<tr>
<td>270°</td>
<td>19891</td>
<td>0.986</td>
<td>19613</td>
<td>1444</td>
<td>18169</td>
<td>1.056 ± 0.015</td>
</tr>
</tbody>
</table>
The average asymmetry for data group C is

$$\bar{\eta} \ (18 \text{ ns.}) = 1.06 \pm 0.02.$$  

The time uncertainties are estimated from the prompt distribution of Figure 10. The error in the value of the asymmetry is the mean standard deviation based on the number $N_T$ in each case.
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