AN ABSTRACT OF THE THESIS OF

Lamar William Coleman for the Ph. D. in Physics

(Date thesis is presented: July 31, 1963)

Title: GAMMA-GAMMA DIRECTIONAL CORRELATION STUDY OF BARIUM-134

Abstract approved by (Major professor):

Gamma-gamma directional correlations were measured in Ba$^{134}$ at ten angles between 90 degrees and 180 degrees in ten degree increments using a scintillation spectrometer. The spectrometer which used NaI crystals, differential energy selection, and standard coincidence techniques was tested by measuring the directional correlation of the two cascade gamma rays in Ni$^{60}$ to be in satisfactory agreement with published results and with theoretical predictions. Cs$^{134}$ decays by beta emission to form several excited states below 2 Mev in Ba$^{134}$ which subsequently decay to the ground state by the emission of gamma rays. Two correlation measurements were performed in Ba$^{134}$. The first, the "overall" correlation, consists of the 797 Kev-605 Kev, 802 Kev-563 Kev, 569 Kev-797 Kev, and the 802 Kev-[563 Kev]-605 Kev correlations.
The second, the "separated" correlation, contains the 797 Kev-605 Kev and the 802 Kev-[563 Kev]-605 Kev correlations. The experimental correlation coefficients are consistent with angular momentum assignments of 2, 2, 4, 4 to the 605 Kev, 1168 Kev, 1401 Kev, and 1970 Kev excited states respectively. These states are assumed to have positive parity. An analysis of the data in terms of the electric quadrupole to magnetic dipole mixing ratios of the 569 Kev and the 563 Kev radiations reveals that, within the limits of experimental error, each radiation contains at least a 50 percent magnetic dipole admixture. The other gamma rays involved are assumed to be essentially pure electric quadrupole. This result contradicts the asymmetric rotor model of the nucleus, which, although it accurately predicts the measured energies of the states with these angular momentum and parity assignments in $^{134}$Ba, does not allow magnetic dipole transitions between the rotational states.
GAMMA-GAMMA DIRECTIONAL CORRELATION
STUDY OF BARIUM-134

by

LAMAR WILLIAM COLEMAN

A THESIS
submitted to
OREGON STATE UNIVERSITY

in partial fulfillment of
the requirements for the
degree of
DOCTOR OF PHILOSOPHY

August 1963
ACKNOWLEDGMENTS

The author wishes to express his gratitude to his research director, Dr. Larry Schecter, for proposing the problem, for many enlightening discussions concerning all aspects of this work, and for continued and conscientious help and encouragement while on leave in Europe. He would like to thank Dr. E. A. Yunker for his continued interest and support, Drs. Harry Easterday and David Nicodemus and Mr. Raymond Sommerfeldt for many helpful discussions about the work, Dr. George Trigg and Mr. Timothy Kelley for interesting conversations regarding the theory, Mr. Richard Siemens for his help in reducing the data and checking calculations, and Mr. Jack McKenzie of the staff of the Oregon State University cyclotron for his help in the production of calibration and testing sources. This work was performed under the auspices of the United States Atomic Energy Commission.
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INTRODUCTION

The atomic nucleus is a collection of nucleons which can exist in various discrete states. Each of these states can be described by a set of parameters which includes the energy, angular momentum, parity, mean life, and electric and magnetic moments. A primary concern in low energy nuclear physics is the determination of these parameters. The experimentalist attempts to measure these quantities to aid the theorist in constructing a useful mathematical description of nuclear levels and their properties, that is, a nuclear model, which agrees in its predictions with measured quantities and which can hopefully be used to predict information about other parameters and other nuclei. The electric and magnetic moments of the ground states of many nuclei have been measured with high precision whereas the determination of the moments of excited states is more difficult. Knowledge of the angular momenta and parities of these states is necessary for the development of the theory of the structure of the states and their electric and magnetic properties or, for the testing of models.

It is generally accepted that the ground states of even-even nuclei have zero angular momentum and even parity. The even parity is a definite consequence of the shell model of nuclei in that
an even number of nucleons in any state results in a configuration of even parity. The absence of measurable magnetic moments in the ground states of even-even nuclei can be considered as evidence of zero nuclear angular momentum. Ground state static electric quadrupole moments have not been observed in even-even nuclei, a further consequence of zero angular momentum in the ground state.

Four methods that have been used to determine the angular momenta and parities of excited nuclear states are indicated below:

1. A nucleus may de-excite by internal conversion or if sufficient energy is available by internal pair production. The magnitudes of the pair production and internal conversion coefficients depend upon the changes in angular momentum \((\Delta J)\) and parity \((\Delta \pi)\) in the transition. The ground state of an even-even nucleus has spin zero and even parity so a careful measurement of one or both of these coefficients allows a definite assignment of angular momentum and parity to each of the excited states.

2. The angular momenta and parities of nuclear states can also be inferred from beta decay studies. The shape of the beta spectrum and its log ft value depend upon the angular momentum and parity change in the decay. If the spin and parity of the decaying state of the parent nucleus is known, selection rules may be used to assign a possible angular momentum and parity to the resulting state of the daughter nucleus produced by the transition.

3. If two or more gamma rays are emitted in cascade a measurement of the directional correlation between them determines the multipolarities of the radiations and places restrictions on the possible values of the changes of angular momenta between the nuclear states involved.
4. Polarization-correlation measurements with the gamma rays emitted during a nuclear de-excitation determine the magnetic or electric nature of the radiation. Together with a directional correlation experiment this fixes the parity of the gamma ray which in turn fixes the change in parity of the nuclear levels.

2.3 year $^{134}$Cs decays by beta-ray emission to form excited levels of the even-even $^{134}$Ba nucleus. Several levels exist below 2.0 Mev in $^{134}$Ba resulting in complicated beta- and gamma-ray spectra which make the task of determining the level scheme difficult. Some of the gamma rays are very close in energy to one another creating an experimental problem in resolution in attempting to determine what energies are present. A number of workers have investigated the beta- and gamma-ray spectrum of $^{134}$Cs. Gamma-ray yield and energy determinations have been made from spectrographic studies of the internally and externally converted electrons and by scintillation techniques. The beta-ray spectrum has been studied with magnetic spectrometers in attempts to determine the number of branches, their end point energies and log ft values, and relative intensities. The results of a number of these studies are presented in Table 1. The references to the various investigators are listed across the top using the number by which they appear in the bibliography. The energies of the gamma- and beta-ray branches that are
<table>
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<th>(30)</th>
<th>(31)</th>
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<td>X(6)</td>
<td>X(5)</td>
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<td>X(6)</td>
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<td>X(5)</td>
<td>X(5)</td>
<td>X(3)</td>
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<td>X(75)</td>
<td>X(75)</td>
<td>X(75)</td>
<td>X(81)</td>
<td>X(50)</td>
<td>X(72)</td>
<td>X(56)</td>
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believed to be present are listed in the left hand column. An entry X indicates that a beta ray or gamma ray of that energy was observed by the workers under whose reference number the entry appears. A bracket connecting two or more energies indicates that the separate components were not resolved. The relative intensities when measured appear in parentheses after the entries. For the beta ray components the log ft values when measured appear just below the entries. The results indicate two principal beta ray branches and at least nine well identified gamma ray energies.

On the basis of these measurements together with coincidence studies and correlation data various level schemes have been proposed for Ba$^{134}$. One of the earliest was suggested by Elliott and Bell (17, p. 980) who placed excited levels at 794 Kev, 1396 Kev, and 1964 Kev which de-excited by the emission of four gamma rays. Keister, Lee, and Schmidt (31, p. 453) were able to set an upper limit of one percent for the decay of Cs$^{134}$ by K-capture from the absence of measurable Auger electrons. Bertolini (5, p. 280) arrived at the same conclusion from the absence of detectable Xenon X-rays. Further work on the de-excitation spectrum of Cs$^{134}$ led to the proposal of other decay schemes (13, p. 445), (30, p. 1022), (5, p. 280), (31, p. 455), (20, p. 855)
none of which was completely compatible with all of the experimental information. Most workers agree, however, on the main features of the decay by placing levels at 605 Kev, 1168 Kev, 1401 Kev, and 1970 Kev. Bashilov et al. (3, p. 60) proposed a level sequence and decay scheme which was supported by the work of Girgis and Van Lieshout (22, p. 672) with one additional level added at 1570 Kev and also by the recent work of Segaert et al. (48, p. 76). It is this level scheme that is currently accepted as being the most satisfactory. Figure 1 displays the order of the levels and the proposed gamma ray transitions of interest in this work according to this scheme.

The angular momentum and parity assignments shown on the level diagram (Figure 1) have been made on the basis of gamma ray correlation measurements, beta decay studies, and measured internal conversion coefficients. The ground state angular momentum and parity \((J^\pi)\) is taken to be \(0^+\) as \(^{134}\text{Ba}\) is an even-even nucleus. The internal conversion coefficient for a given gamma ray energy depends on the electric or magnetic nature of the radiation and its multipolarity. Table 2 summarizes some of the measured internal conversion coefficients and the \(K/L\) ratios for \(^{134}\text{Ba}\) gamma rays. The gamma ray energies are listed across the top of the table and the numbers listed in the left column
Figure 1. LEVEL AND DECAY SCHEME OF Ba$^{134}$.
<table>
<thead>
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<th>Gamma Energy (Kev)</th>
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<th>563</th>
<th>569</th>
<th>605</th>
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<td>$a_K \times 10^3$</td>
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<td>42.0</td>
<td>26.0</td>
<td>25.5</td>
<td>20.5</td>
<td>9.50</td>
<td>9.45</td>
<td>4.90</td>
<td>3.75</td>
<td>2.60</td>
</tr>
<tr>
<td>Assignment</td>
<td>E(2)</td>
<td>E(2)</td>
<td>M(1)</td>
<td>E(2)</td>
<td>E(2)</td>
<td>E(2)</td>
<td>E(2)</td>
<td>E(2)</td>
<td>E(2)</td>
</tr>
</tbody>
</table>
refer to bibliography references. The number in parenthesis below an entry is the experimental uncertainty in the entry. The theoretical values given in the table are due to Rose (45, p. 64) and include the effects of finite nuclear size and screening. Possible multipole assignments on the basis of the conversion coefficients are given.

The angular momentum of the ground state of Cs$^{134}$ has been measured to be $J = 4$ by an atomic beam magnetic resonance experiment by Bellamy and Smith (4, p. 33) and by Jaccarino et al. (29, p. 676). The angular momentum and the magnitude of the magnetic moment are consistent with a $g_{7/2}$ - $d_{3/2}$ proton-neutron configuration which predicts positive parity for the ground state of Cs$^{134}$.

The beta ray branches indicated in Figure 1 are the ones that are required from gamma ray studies so that other apparently observed beta groups are not included. There is no mechanism for including in the scheme the 683 Kev beta branch reported by some workers. At the high energy end the internal conversion lines mask the character of the continuous spectrum. As indicated in Table 1 the 86 Kev branch has a log ft value of about 6.4 and in addition appears to have a linear Fermi plot. This branch has an intensity of 27 percent and corresponds to an allowed transition.
(ΔJ=0,±1, no) allowing an assignment of 3, 4, or 5 with positive parity to the 1970 Kev level in Ba$^{134}$. The other intense component, the 658 Kev branch, has a log ft value of 9 and an allowed shape permitting possible assignments of 3, 4, or 5 with either parity to the 1401 Kev level. The identification of other much less intense beta branches depends on being able to remove successive straight line components from the Fermi plot. This is difficult in cases where more than one weak component may be present and could be misleading if one or more of the components did not have an allowed shape. On the basis of the reported log ft values alone, these components could be as much as second forbidden (ΔJ=±2, ±3, no). Waggoner (55, p. 425) has reported a slight deviation from allowed shape for the 658 Kev beta group. If the shape of the beta spectrum is allowed theory predicts a symmetric beta-gamma correlation. Stevenson and Deutsch (53, p. 1203) found no beta-gamma anisotropy in Cs$^{134}$ with the 658 Kev beta group.

Further information about the angular momentum of the levels and the multipolarity of the radiations is available from directional and polarization correlation experiments. Brady and Deutsch (10, p. 870), in an early experiment, were unable to detect a non-isotropic directional correlation with the gamma rays from Cs$^{134}$. They were able to measure a correlation with Co$^{60}$.
which has a simpler decay scheme. In a following experiment with scintillation counters Brady and Deutsch (11, p. 1541) detected a directional correlation at three angles. Using improved scintillation counters these workers (12, p. 558) remeasured the correlation at seven angles and from the similarity in the shape of the curve to that for $^{60}\text{Co}$ suggested that the angular momentum change in the main sequence (the 797 Kev-605 Kev cascade) may be 4, 2, 0, with both radiations being quadrupole. They were unable to detect any change in the correlation using a solid or a liquid source with or without a $10^4$ gauss magnetic field and concluded that the life time of the intermediate state of the cascade must be less than $10^{-8}$ sec. In a similar experiment Beyster and Wiedenbeck (6, p. 411), again in comparison with the $^{60}\text{Co}$ curve, interpreted their data on the basis of a simple decay scheme containing just three gamma rays and suggested a quadrupole-quadrupole correlation for the 605 Kev-797 Kev cascade. The 569 Kev radiation was also assumed to be quadrupole allowing an angular momentum of 4, 5, or 6 for the 1970 Kev state. None of these experiments employed energy discrimination so that all gamma rays were detected in each counter. Using NaI(Tl) scintillation counters and integral energy selection Robinson and Madansky (43, p. 604) measured the directional correlation by requiring a
coincidence between one of the gamma rays and a 660 Kev beta
and found agreement with a basic quadrupole-quadrupole correla-
tion with an angular momentum sequence 4, 2, 0 in the 605 Kev-
797 Kev cascade. They assumed a decay scheme in which the 797
Kev radiation leads to the ground state. From a measurement of
the "overall" correlation, that is, accepting all gamma ray ener-
gies, they assign an angular momentum of five to the 1970 Kev
level with the 569 Kev radiation being dipole. Kloepper and Len-
nox (33, p. 696) also indicate a probable 4(2)2(2)0 correlation for
the main decay sequence of Cs$^{134}$, but it is not clear what gamma
ray energies were accepted in the experiment from which this con-
clusion was drawn. Klema (32, p. 66) used differential energy
discrimination to measure the 1365 Kev-605 Kev directional cor-
relation. Interpreting his data on the basis of a different decay
scheme (31, p. 455), Klema assigns 3$^-$ to the 1970 Kev level by
allowing an E1 + M2 mixture in the 1365 Kev gamma ray to fit his
data. The measured correlation function however, agrees best
with an assignment of $J = 4$ to that level with the 1365 Kev gam-
ma being quadrupole. Everett and Glaubman (18, p. 955) report
measuring the 1365 Kev-605 Kev, 802 Kev-1168 Kev, and 797
Kev-605 Kev correlations and find that they are all consistent with
a quadrupole-quadrupole decay and allow assignments of $2^+, 2^+$,
$4^+, 4^+$ to the 605 Kev, 1168 Kev, 1401 Kev, and 1970 Kev states respectively. It is pointed out that by allowing mixed multipoles for the first radiation the data can also be interpreted as 3-2-0, 3-1-0, or 2-2-0. The effects of the 569 Kev and the 563 Kev radiations which contribute to the 605 Kev photopeak are not mentioned nor is the contribution of the 797 Kev gamma ray to the 802 Kev range of the spectrum. On the basis of some proposed decay schemes these energy overlaps would not contribute to the correlations. Stewart, Scharenberg, and Wiedenbeck (54, p. 691) performed a detailed directional correlation experiment with Cs$^{134}$. They report no observable difference in the measured function for solid or liquid sources. They measured an "overall" correlation, the 797 Kev-605 Kev, the 1365 Kev-605 Kev, and the 570 Kev-605 Kev correlations. On the basis of their results they assign $J = 2$ to the 605 Kev level, $J = 4$ to the 1402 Kev level and $J = 4$ to the 1970 Kev level and report that the 569 Kev radiation is 94 percent quadrupole and 6 percent dipole. These workers interpret their data on the basis of a simplified decay scheme which contains only the 569 Kev, 797 Kev, 605 Kev, and 1365 Kev gamma rays and omits the level at 1168 Kev so that the effects of other cascades which contribute to each of their measurements and which may affect the interpretation have been neglected.
Wintersteiger (57, p. 79) has measured the life time of the first excited state in \( {\text{Ba}}^{134} \) to be \(< 5 \times 10^{-10} \) sec. by the delayed coincidence method.

Metzger and Deutsch (38, p. 557) measured the polarization-direction correlation in \( {\text{Ba}}^{134} \) at the average gamma ray energy. The results are of the form for an E2-E2 cascade which may be characteristic of the intense 797 Kev-605 Kev cascade which contributes most heavily to the counting rate. Williams and Wiedenbeck (56, p. 822), from a similar experiment, concluded that if both the 797 Kev and the 605 Kev radiations are quadrupole, one is E2 and the other is M2. Kloepper (33, p. 697) reported a polarization correlation result that supports the assignment of E2 to both the 797 Kev and 605 Kev gamma rays.

In summary, it is quite well established from previous work that the 605 Kev and 797 Kev radiations are electric quadrupole and that the angular momenta and parities of the 605 Kev and 1401 Kev levels are \( 2^+ \) and \( 4^+ \) respectively. The angular momenta and parities of the 1168 Kev and 1970 Kev levels and the multipo- larities of the 802 Kev, 569 Kev, and 563 Kev radiations are less certain although the 1970 Kev level probably has angular momentum 4. The conclusions drawn from many previous correlation measurements can be questioned because of the omission of some
contributing cascades in the interpretation and because, in most cases, of large statistical errors. A nuclear model has been proposed which explains some of the features of the Ba\textsuperscript{134} level scheme. It is felt that a reinvestigation of the gamma-gamma directional correlation and its interpretation on the basis of a more complete decay scheme will be useful in clarifying the uncertainties in angular momentum and multipole assignments to the levels and radiations, and in strengthening some assignments. The comparison of the experimental results with those calculated on the basis of the decay scheme can be applied to test the predictions of the model.
SINGLE PARTICLE AND COLLECTIVE NUCLEAR MODELS

A complete understanding of nuclear phenomena involves a description of the nucleus as a system of nucleons interacting with a common meson field. The nucleus and some of its properties can, however, be rather well described in terms of a system of well-defined protons and neutrons with certain forces between them. The meson origin of these forces does not play an essential part in the behavior of nuclei at low energy so that the theory of nuclear structure is not concerned with the theory of the force itself but accepts its existence and its properties as experimental fact. Once an expression is given for the nuclear forces all problems of nuclear structure are those of solving a Schrödinger equation for A particles. This equation cannot be solved in general so a resort to approximations, that is, to nuclear models is necessary.

Because of this limitation an attempt is made to use as little detailed information of the forces as possible and to concentrate on qualitative features. This procedure introduces concepts such as radius, shape, and potential well which can be defined and measured and which are useful for a description of the observed facts but whose connection with the fundamental forces is vague.
This emphasis brings about the introduction of many kinds of nuclear models. A model is simply a stressing of certain features which can account for the phenomenon under consideration. From a model it must be possible without prohibitively lengthy calculation to predict various observable properties of nuclides in a systematic way.

The simplest nuclear models have their basis in trying to understand the experimentally inferred shell structure and magic numbers in analogy with the structure of atoms. Whenever the number of protons or neutrons reaches any of certain values called magic numbers the nucleons form a closed and exceptionally stable system. The extreme single particle model of nuclei describes the nucleus as a spherical potential well in which the nucleons move as noninteracting particles and thus ignores any nucleon correlations. This potential is the average effect of all the other nucleons on a single one, has a certain size and depth, and includes a spin-orbit interaction. Such a potential with a suitably chosen form produces a shell structure and predicts the correct location of the magic numbers. This model describes the ground states of nuclei by specifying the number of nucleons in each quantum state assuming that the proton and neutron states fill independently. In the ground state the nucleons are thought of
as being paired off to zero angular momentum by a strong pairing force so that the values of the nuclear parameters are determined solely by a single unpaired nucleon in odd A nuclei. The nucleus can be imagined then to be made up of a core which contains all the particles in closed shells and a cloud which consists of those nucleons in unfilled shells. The simplest case would be a nucleus in which the cloud consisted of a single particle. Then the model allows a determination of the nuclear angular momentum and parity for nuclei with one odd particle as being those of the odd particle. The angular momentum of odd-odd nuclei cannot be predicted because it cannot be determined what coupling of the particles' angular momenta gives the lowest energy state. This single particle approximation gives the Schmidt limits for the ground state nuclear magnetic moments. In general the experimental values lie between these limits but differ greatly from them. The quadrupole moments predicted by this model are completely erroneous (41, p. 145).

In attempting to interpret the large amount of data available on energies, angular momenta, parities, and transition probabilities, it becomes clear that the extreme single particle model is inadequate. The next higher approximation, which will be termed the shell model, is to consider a nucleus with a larger number of
nucleons in the cloud. Low energy nuclear properties are attributed to the particles outside of closed shells. The assumption of a strong pairing force is relaxed to allow recoupling of the angular momenta of the nucleons beyond closed shells. This is necessary in order to obtain a larger number of states as required by experiment than are available in the single particle model. It is also assumed that those nucleons outside of closed shells do not perturb each other very much from single particle states. The interaction is strong enough to remove degeneracy but not so strong (compared to the spin-orbit interaction) that $j$ ceases to be a good quantum number. This assumption can also be relaxed and a superposition of the wave functions of two or three pure single particle states taken to describe a state. This process is termed configuration mixing. In the few places where detailed calculations have been made the model has been found to give an excellent description of the spins and parities of low-lying excited states. In trying to understand the energies and angular momenta of these states it is necessary to stay in the vicinity of closed shells or else the number of competing configurations is overwhelming. The interpretation of excited states by means of this type of coupling scheme has been tested only for cases involving two or three identical nucleons (or holes) outside of (or within) closed shells.
Adding more particles presents a huge amount of computational labor compounded by the fact that configuration mixing, including configuration interactions between protons and neutrons, and with the core, has a strong effect on the quantitative spacing of the states and is not a feasible approach. The Ba$^{134}$ nucleus with $Z = 56$ and $N = 78$ has six protons outside of a closed shell and four holes in a closed neutron shell producing a fairly complicated system.

Proper configuration mixing is able to bring the theoretical and experimental magnetic moments into agreement. In general the measured quadrupole moments agree very poorly with the shell model predictions. Near closed shells some nuclear quadrupole moments can be accounted for with the aid of suitable configuration mixing.

In the rare earth region the quadrupole moments become as much as 30 times the single particle values. This is a difficulty which leads to the conclusion that collective effects are important and that it is not a valid approximation to consider nucleons as moving independently in a spherically symmetric potential. Nuclear moments are sensitive to configuration mixing and collective effects so that an independent particle model does not give very good values for gamma ray decay probabilities. The
formulas for these single particle transition rates are simple, however, and are often used as reference values for comparing experimental data. The results apply quite well for low A. Electric quadrupole transitions are usually strongly enhanced over the single particle values in both light and heavy nuclei which again leads to the collective notion and the large collective quadrupole moments. It is not possible then to attribute all nuclear properties to the nucleons in unfilled shells.

It is necessary to consider correlations in nucleon motion as exemplified by configuration mixings, and an interaction with the closed-shell configurations in the core may also be important. The shell model has its basis in an average static field generated collectively by all the particles but a more complete description must include the variations of the field associated with collective oscillations. Relatively small residual interactions introduce important correlations in the motion of the particles outside of closed shells. The energy spectra of nuclei with many-particle configurations show features which vary in a systematic way from nucleus to nucleus. These regularities are associated with the fact that a major part of the correlations between particles may be described in terms of ordered collective motion of the nucleons corresponding to variations in the shape of the nucleus, for example.
This leads to a generalization of the shell model in which the binding field is no longer considered as a static isotropic potential, but rather as a variable field which may take shapes differing from spherical symmetry.

The first problem is to determine the nuclear equilibrium shape. The equilibrium shape and the character of the collective modes of motion of the nucleons may be understood as the result of the competition between the deforming tendency of the individual nucleons and the effect of the pairing forces. The pairing forces tend to couple two equivalent nucleons to a state of zero total angular momentum, that is, a spherically symmetric state. In the region of closed shells these forces dominate and the nuclear equilibrium shape is spherical. The addition of nucleons in unfilled shells increases the importance of the tendency to deformation, a coherent effect of all these nucleons. The nucleus can acquire a nonspherical equilibrium shape and possesses a large quadrupole moment. The quadrupole moments are small in the region of closed shells and become larger with the addition of more particles. These trends can be accounted for then in terms of the tendency of the particle structure to produce collective deformation of the nucleus. For such nuclei the collective motion separates into rotational and vibrational parts. The former
corresponds to a rotation of the nuclear orientation at constant shape, while the latter corresponds to oscillations about the anisotropic equilibrium shape. The rotational concept arises from the fact that the correlated particle motion may be such that the pattern of particles changes slowly maintaining a fixed shape but with the orientation in space altering. Looked at from the outside, the nucleus of constant shape is rotating. It is natural to attribute kinetic energy and angular momentum to this rotating mass and to consider the rest of the nuclear kinetic energy and angular momentum to be internal. The equilibrium shape of nuclei can be obtained by calculating the energy of the nucleons as a function of the shape of the field and finding the shape that gives minimum energy.

For the description of nuclear structure then, the dynamics of the nucleus is considered in terms of collective and intrinsic modes of excitation. The collective modes are associated with deformations and the lowest modes of this type correspond, in the case of nuclei with sufficiently many particles outside of closed shells, to rotations of the spheroidal shape. The states produced by collective vibrations are at an energy of 1 or 2 Mev in heavy strongly deformed nuclei ($A \approx 250$) and increase in energy as $A$ decreases. For nuclei near closed shells the collective motion
corresponds to vibrations about the spherical equilibrium shape. Experimental evidence indicates the absence of rotational states in spherical nuclei at least for low excitations. The intrinsic modes represent the motion of the nucleons in a fixed field corresponding to the nuclear equilibrium shape and have an average spacing of ~100 Kev. This motion is subjected to the influence of the pairing forces and in the case of odd-A nuclei the low lying intrinsic states may often be described in terms of excitations of the last unpaired nucleon. For configurations sufficiently far removed from closed shells the nucleus acquires a large equilibrium deformation resulting from the deforming effects of many nucleons and an approximate solution is obtained by considering first the relatively fast motion of the particles with respect to the deformed nuclear field considered as fixed in space, and then the relatively slow vibration and rotation of the entire system. If the coupling of the intrinsic and collective motion is small, the problem is usually treated by perturbation theory. A considerable body of data reveals the existence of a rotational structure in the nuclear excitation spectrum. The simple rotational motion is characteristic of the strongly deformed nuclei and is thus especially well defined in regions far removed from closed shells. The rotational structure has been studied most extensively for $A > 140$ where the
distances between shell closings are great and where the deformations are especially large. In this region this approach has met with good success in describing the low lying states of even-even nuclei. Mention will be made of three such models which attempt to describe the low lying states in even-even nuclei with the particular aim of finding a suitable description of the excited states of Ba$^{134}$.

One of the first collective models of this type useful in describing nuclear structure was the Bohr-Mottelson model (9, p. 1). This model assumes that most of the nuclear deformation resides in the cloud of nucleons outside of closed shells although its influence tends to deform the core slightly. In addition only nuclear shapes which have axial symmetry are considered. This ellipsoidal nucleus can then, in addition to vibrating, also perform rotations producing rotational spectra. These collective motions produce excited states at energies lower than the particle excitation energies. The moments of inertia of the nucleus which appear in the rotational Hamiltonian are calculated in terms of one parameter assuming that the nucleus behaves as an incompressible rigid body. For two equivalent particles both the particle forces and the coupling to the deformation form a ground state particle configuration of zero angular momentum so that in even-even
nuclei the ground state particle configuration has angular momentum zero. If this configuration is not excited the low lying rotational levels of a symmetric nucleus rotating about an axis perpendicular to the nuclear symmetry axis are pure and are characterized by a rotational quantum number $J$. The possible rotational quantum states of the nucleus are restricted by the reflection symmetry of the deformation which restricts the collective states to even parity and excludes odd values of $J$. Since the ground state particle configuration in even-even nuclei possesses even parity the collective states for this particle configuration will have even parity. For even-even nuclei the lowest rotational band has spins and parities $J^\pi = 0^+, 2^+, 4^+, 6^+, \ldots$ at energies of

$$E_{\text{rot}} = \frac{\hbar^2}{2I} J(J + 1)$$

where $I = (2/5)MA(\Delta R)^2$ is the moment of inertia for a spheroidal deformation of the incompressible model, $M$ is the nucleon mass, $A$ is the nuclear mass number, and $\Delta R$ is the difference between the major and minor semi-axes of the spheroid. Such a rotational band can be built on an excited particle state or a vibrational state. Very high excitations are thought of as vibrational excitations of the core.

For highly deformed nuclei which possess rotational states the gamma ray transition probabilities to states of the same
family obey simple relations. For electromagnetic transitions of multipole order \( L \) the ratio of the reduced transition probabilities is the square of the ratio of two Clebsch-Gordan coefficients. These probabilities depend on the static moments of the nuclear states. For low energy transitions on the particle model M1 radiation will strongly predominate over the E2 when both are possible. The E2 transition probability depends upon the square of the quadrupole moment so the collective deformation leads to an enhancement of the E2 probability which may lead to an appreciable admixture of E2 in \( |\Delta J| = 1 \) transitions. The E2 transition probabilities from the excited \( 2^+ \) state to the ground state in some even-even nuclei have been determined. The quadrupole moments calculated from these show the expected trend, decreasing regularly with the approach to closed shell configurations.

This type of rotational description works well in the region \( 160 < A < 185 \) and \( A > 225 \). Many even-even nuclei show evidence of a rotational structure but with a more complicated collection of states than the simple \( 0^+, 2^+, 4^+, 6^+, \ldots \) rotational sequence predicted by Bohr and Mottelson. For example, there are often two \( 2^+ \) states, two \( 4^+ \) states, excited \( 0^+ \) states, and states with odd \( J \). These states can sometimes be fitted by building a rotational band on a suitable vibrational or particle excitation.
This can be done assuming a rotational band of an axially symmetric nucleus built on a vibrational state if the ratio of the energy of the first $4^+$ state to that of the first $2^+$ state is greater than 3.27. In Ba$^{134}$ this ratio is 2.31 and such a fit cannot be made. It would be convenient to develop these states naturally as members of the ground state rotational band and also to be able to fit those states which cannot be explained with this model.

Davydov and Filippov (14, p. 237) describe the energy levels of even-even nuclei in terms of rotations of a deformed nucleus which do not involve changes of its internal state. The restriction of axial symmetry is removed and it is found that this affects the rotational spectrum of the axial nucleus slightly although additional rotational states of $2^+, 3^+, 4^+, \ldots$ appear. If the deviations from axial symmetry are small these levels lie very high but become lower as the deviations from symmetry are increased. For example, two $2^+$ states appear and the ratio of their energies varies from infinity to two depending on this asymmetry. The three moments of inertia are not independent but are determined from two parameters, one of which measures the deviation from axial symmetry, and the other characterizes the deformation. For this reason the approach is sometimes referred to as the restricted asymmetric rotor model. The positions of the energy
levels are also determined in terms of these parameters. The value of the parameter describing the deviation from axial symmetry is restricted to a certain range. In other language this restriction allows fitting of energy levels with this model only when the ratio of the energy of the first $4^+$ state to that of the first $2^+$ state is greater than 2.67 (36, p. 553). When this condition is met the Davydov-Filippov model explains the data on energy levels quite well for even-even nuclei far from closed shells with a few exceptions. In Ba$^{134}$ the above condition is not met, since the ratio is 2.31, so that a consistent set of parameters cannot be found with which to describe the known nuclear energy levels.

Ratios of gamma ray reduced transition probabilities for E2 and M1 transitions between some of these rotational states have been calculated in terms of the parameters of the model. The agreement with experimental ratios of E2 reduced transition probabilities is very good in a few cases. The general trends of the ratios of gamma ray transition probabilities as a function of the ratio of the energies of the states are predicted correctly. The results predicted by this model are, in general, in better agreement with experimental results for even-even nuclei than are those given by the Bohr-Mottelson model.

For not too strongly deformed nuclei the rapid rotational
motion will tend to distort the shape of the nucleus, that is produce a centrifugal distortion. This effect is termed a rotation-vibration interaction. Such a rotation-vibration interaction has been introduced in the Davydov-Filippov model and results in improved agreement with experimental data for nuclei far away from closed shells (36, p. 536).

An attempt to extend the validity of this rotational description has been presented by Mallmann (36, p. 535). In this model also, rotational modes of excitation of the nucleus are considered. The nucleus is again considered to be asymmetric but in a more general way in that the three moments of inertia are treated as independent parameters to be determined by comparison with experiment and no attempt is made to determine them from first principles. A rotation-vibration interaction is included in the Mallmann model as a perturbation. It is also assumed that the rotational motion of the even-even nucleus can be treated quasi-adiabatically. That is, it is assumed that the excited states due to other degrees of freedom have energies appreciably higher than the energies due to the rotational motion even though the particle structure will be perturbed because of the inability of the particles to follow the rotations completely adiabatically. The departure of actual nuclei from this behavior is assumed to be small and can be accounted
for in terms of a small perturbation. This model is then referred to as the quasi-adiabatic general asymmetric rotor model. A general asymmetric rotor model without the rotation-vibration correction has been considered but did not result in improved agreement with experiment over the restricted asymmetric rotor model (36, p. 536). The Mallmann model has as parameters the three nuclear moments of inertia and the strength of the rotation-vibration perturbation which is added as a first order correction to the pure asymmetric rotor energy levels. With this approach the comparison of the predicted levels with experimentally known energy levels in even-even nuclei in the range $40 < A < 250$ shows very good agreement, usually within experimental error. It must be pointed out however that in almost every case other experimental levels exist which do not fit well in this or other rotational schemes. This suggests that the general asymmetric rotor Hamiltonian is an essential part of a more complete Hamiltonian which includes other degrees of freedom. The energies of the two excited $2^+$ levels and the two $4^+$ levels in Ba$^{134}$ shown in Figure 1 are fitted very well as rotational levels with this model. The 1570 Kev, 1641 Kev, and 1770 Kev levels in Ba$^{134}$ cannot be unambiguously accounted for in the ground state rotational band. The gamma ray transition probabilities are
calculated by Mallmann with the rotation-vibration interaction omitted. The rotational states all have the same parity so that the normal selection rules forbid E1, E3, ..., M2, M4, ... transitions. Additionally now the model forbids M1 transitions between the rotational states. A survey of the experimental E2 to M1 mixing ratio in the $^2_2 \rightarrow ^1_2$ transition in some even-even nuclei (36, p. 559) lends support to this selection rule except near closed shells. This leaves E2, M3, E4, M5, ... transitions as possibilities of which only the E2 transitions are considered as the others are too slow to compete favorably. The calculated gamma ray transition probabilities and ratios of gamma ray intensities between the rotational levels of even-even nuclei assuming pure E2 radiation are compared to experimentally determined values for $110 \leq A \leq 250$ (36, p. 566). The agreement of the theory with experiment is within experimental error with a few exceptions. Although the energy levels of "known" angular momentum and parity are well described by the model the calculated intensity ratios in Ba$^{134}$ do not, in general, agree well with the experimental values. Table 3 displays the comparison. The notation $^2_2$, for example, means the second state of angular momentum 2 reached in order of ascending energy. It is observed that the two ratios departing the most from the measured values are both low and are those which contain the 569 Kev and 563 Kev radiations
Table 3. THEORETICAL AND MEASURED GAMMA RAY INTENSITY RATIOS FROM ROTATIONAL STATES IN Ba$^{134}$:

<table>
<thead>
<tr>
<th>Transition ratio</th>
<th>Energy ratio (Kev)</th>
<th>Model prediction</th>
<th>Measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{2_2 \rightarrow 1_2}{2_2 \rightarrow 1_0}$</td>
<td>$\frac{563}{1168}$</td>
<td>1.5</td>
<td>7.0</td>
</tr>
<tr>
<td>$\frac{2_4 \rightarrow 2_2}{2_4 \rightarrow 1_2}$</td>
<td>$\frac{802}{1365}$</td>
<td>2.7</td>
<td>2.1</td>
</tr>
<tr>
<td>$\frac{2_4 \rightarrow 1_4}{2_4 \rightarrow 1_2}$</td>
<td>$\frac{569}{1365}$</td>
<td>0.35</td>
<td>2.3</td>
</tr>
</tbody>
</table>

each of which is a transition between states of the same angular momentum.

Of the various models discussed it appears that the model proposed by Mallmann is the one best able to describe the states of known angular momentum and parity in the Ba$^{134}$ nucleus and its predictions will be compared with the experimental results of the directional correlation experiments as a further test of its validity for this nucleus, particularly with respect to the additional selection rule imposed on gamma ray transitions.
THEORY OF DIRECTIONAL CORRELATION

The probability of emission of a particle or quantum from a nucleus depends, in general, on the angle between the nuclear spin (angular momentum) axis and the direction of emission. Under ordinary circumstances, however, the total radiation from a collection of radioactive nuclei is isotropic because the nuclei are randomly oriented in space. An anisotropic radiation pattern can be observed only from a collection of nuclei which are not randomly oriented. The nuclei can be oriented in space by placing the sample at low temperature in a strong magnetic field. As a result of the interaction between the magnetic field and the nuclear magnetic moment alignment will take place. The angular distribution of the emitted radiation is then measured with respect to the direction of the applied field. The interaction of an electric field gradient with the nuclear quadrupole moment can also be employed to produce an orientation of the nuclei in space.

Another method consists in picking out only those nuclei whose spins happen to lie in a preferred direction. This case can be realized experimentally if the nuclei decay by successive emission of two radiations, 1 and 2, through a sufficiently short lived intermediate state. The observation of 1 in a specified direction
then selects an ensemble of nuclei which has a nonisotropic distribution of spin orientations. The following radiation, 2, then shows, in the usual case, a directional correlation, or nonisotropic radiation pattern, with respect to radiation 1. This is the technique employed in the present experiment.

Figure 2 shows the essential aspects of a directional correlation experiment. The angular correlation function $W(\phi)$ is the probability that if radiation 1 is emitted in a direction $\mathbf{K}_1$ that radiation 2 will be emitted in a direction $\mathbf{K}_2$ at an angle $\phi$ with respect to $\mathbf{K}_1$. Experimentally the number of coincidences between radiations 1 and 2, $N(\theta)$, is determined as a function of $\theta$, the angle included by the axes of the two counters. Because of the finite solid angles of the detectors these numbers $N(\theta)$ are averages of the true correlation $W(\phi)$ over a range of angles $\Delta\phi$ distributed around $\theta$. As a result $N(\theta)$ must be properly corrected and normalized to yield $W_{\exp}(\phi)$. The comparison of $W_{\exp}(\phi)$ with the theory finally gives the desired information about the properties of the nuclear levels and the radiations.

It is necessary to understand why there is a correlation at all, that is, why the coincidence counting rate depends on $\phi$. Imagine a transition in which radiation is emitted from a state characterized by quantum numbers $j, m$ leading to a state $j'$,
Figure 2. THE ASPECTS OF A DIRECTIONAL CORRELATION EXPERIMENT
m'. The radiation will have angular momentum L, projection quantum number \( M = m - m' \), and will or will not result in a parity change between the levels depending upon the type of radiation. The parity change is determined by a factor \((-1)^L\) for electric radiation carrying angular momentum L, and \((-1)^{L+1}\) for magnetic radiation carrying angular momentum L. Several L values ranging from \( |j-j'| \) to \( j+j' \) may be possible but the multiplicity of L values is of no importance for this argument so the discussion will be in terms of a single value of L. In practice the magnetic substates of the level \( j \) are equally populated so that transitions are observed from all substates \( m \) to all substates \( m' \).

The angular distribution of the emitted radiation, for given M, depends on \( \phi \), but the summation over all substates (or summation over M) is independent of \( \phi \). Therefore, the radiation observed in a single transition from a non-oriented source is isotropic.

For the observation of the two radiations in coincidence the situation is changed. For a fixed M the radiation is not isotropic because a particular direction in space is being singled out. This is the quantization axis with reference to which \( M \), as well as \( m \) and \( m' \), is measured. If of all the radiation emitted in all directions only that proceeding in a specified direction is
observed, then this is equivalent to selecting a particular set of \( M \) values. Further, if the axis of quantization is chosen to lie along the direction of emission of the first radiation, \( M \) will be \( \pm 1 \) with \( M = 0 \) omitted. This is a result of the fact that a transverse wave corresponds to a photon which can have intrinsic spin only parallel or antiparallel to the propagation direction and that only the zero component of the "orbital" angular momentum is radiated in the \( z \)-direction. That the same conclusion can be reached from classical field theory is demonstrated later on although the separation of the angular momentum into an "intrinsic spin" and an "orbital" part is not made. The result is then that not all values of \( M \) are summed over and in the final state not all \( m' \) values are equally populated. If this final state for the first transition is also the initial state for the next transition, then the ensuing radiation is anisotropic by virtue of the fact that it originates from unequally populated substates.

The general theory of angular correlation was established by Hamilton (26, p. 122). Further development and refinement of the theory has resulted mainly from the work of Hamilton (27, p. 782), Lloyd (34, p. 716), Alder (2, p. 235), Racah (42, p. 910), Falkoff (19, p. 98) and Goertzel (23, p. 897). Biedenharn and Rose have presented a detailed review article of the subject
The theory of directional correlation can be summarized in the following way. Let the initial nuclear state be described by a state function \( |j_1 m_1\rangle\), the intermediate state by \( |j m\rangle\), and the final state by \( |j_2 m_2\rangle\). The \( j\)'s are the angular momenta of the various nuclear levels, and the \( m\)'s are their projections on the quantization axis. It is assumed that the perturbation of the intermediate state by extra nuclear fields is negligible. This will be true if the lifetime of the intermediate state is small compared to the nuclear precession period produced by an external coupling. An application of second-order perturbation theory (44, p. 168) gives the probability that a nucleus decaying through the states \( 1 \to i \to 2 \) will emit radiation \( l \) in direction \( \vec{K}_1 \) followed by radiation \( Z \) in the direction \( \vec{K}_2 \). This probability is the correlation function \( W(\vec{K}_1 \vec{K}_2) \) given by

\[
W(\vec{K}_1 \vec{K}_2) = S \sum_{m_1 m_2 m} \left| (j_2 m_2 | H_2 | j m)(j m | H_1 | j_1 m_1) \right|^2
\]

where constant factors have been omitted. \( H_1 \) is the interaction Hamiltonian responsible for the emission or absorption of radiation \( l \) and \( H_2 \) is that for the emission or absorption of radiation \( Z \). The factor \( S \) indicates that a summation is also made over unobserved quantities, for example, the state of polarization of the radiation.
Equation (1) may be brought into a more usable form by carrying out some simplifying manipulations. First the matrix elements are split into multipole components according to the L, M, and parity of the radiation. A transformation is then performed rotating the coordinate system of quantization over into the coordinate system of the radiation. The matrix elements of distinct multipole orders are then split into geometrical factors and reduced matrix elements using the Wigner-Eckart theorem (44, p. 85). This gives the dependence on the magnetic quantum numbers and hence on geometrical factors separately by means of Clebsch-Gordan coefficients. Finally, with the help of relations among the Clebsch-Gordan coefficients (44, p. 35) the total expression can be reduced by means of Racah algebra (44, p. 173). The result for the correlation function for pure multipole transitions is

\[ W(\phi) = \sum_{v = 0}^{v_{\text{max}}} A_v P_v(\cos \phi) \]  

(2)

where \( v \) takes on only even values and \( v_{\text{max}} \) is given by the smallest of \( 2j, 2L_1, 2L_2 \). \( P_v(\cos \phi) \) is the Legendre polynomial of order \( v \). The coefficients \( A_v \) are given by

\[ A_v = F_v(L_{11j}) F_v(L_{22j}) \]
where

\[ F_v(L_j;j) = (-)^{j_2-j_1-1} (2j + 1)^{1/2} (2L + 1) C(LLv; 1, -1) W(jjLL; vj_1). \]

\[ C(LLv; 1, -1) \] is a Clebsch-Gordan coefficient with \( \vec{v} = \vec{L} + \vec{L} \) and \( W(jjLL; vj_1) \) is a Racah coefficient. The Racah coefficient \( W(abcd;ef) \) is zero unless each of the three triangle rules \( \Delta(abe), \Delta(cde), \Delta(acf), \) and \( \Delta(bdf) \) are satisfied. It is this fact that determines the restrictions on \( v_{\text{max}} \) in Equation (2). Tabulations of the functions \( F_v \) are available (7, p. 746) for various \( L, j_1 \), and \( j \) values. Thus, the constants \( A_v \) in the expansion depend on five parameters: \( L_1, L_2, j_1, j_2 \), so that a comparison between the theoretical and experimental correlation functions can give information about these parameters.

If one of the radiations in the cascade consists of a mixture of different multipole orders, \( [ j_1(L_1^1; j_1) j_2(L_2; j_2) ] \), the correlation function has the form (7, p. 747)

\[ W(\phi) = W_1(\phi) + \delta^2 W_2(\phi) + 2\delta W_3(\phi). \]  

\( \delta^2 \) is the ratio of the total angle-integrated intensity of the \( L_1^1 \)-pole to that of the \( L_1 \)-pole radiation. In general, the real mixing ratio \( \delta \) is defined as the ratio of the reduced matrix elements for the \( L_1^1 \) pole to the \( L \) pole radiation,
\[ \delta = \frac{(j|L'|j_1)}{(j|L|j_1)}. \]

\( W_I(\phi) \) and \( W_{II}(\phi) \) are given by Equation (2).

\[ W_I(\phi) = \sum_v F_v(L_1,j_1,j) F_v(L_2',j') P_v(\cos \phi) \]

\[ W_{II}(\phi) = \sum_v F_v(L_1',j_1,j) F_v(L_2',j') P_v(\cos \phi) \]

\( W_{III}(\phi) \) is the contribution due to the interference between \( L_1 \) and \( L_1' \) and is given by

\[ W_{III}(\phi) = \sum_v F_v(L_1 L_1' j_1 j) F_v(L_2 j_2') P_v(\cos \phi) \]

with

\[ F_v(L_1 L_1' j_1 j) = (-)^{j-j_1-1} \left( \frac{(2j+1)(2L+1)(2L'+1)}{(2j_1+1)(2L_1'+1)} \right)^{1/2} \]

\[ \times G_v(L_1 L_1' j_1 j) \]

and

\[ G_v(L_1 L_1' j_1 j) = C(L_1 L_1' v; 1, -1) W(jj L_1 L_1'; v j_1). \]

The function \( G_v(LL' j_1 j) \) is tabulated by Biedenharn and Rose (7, p. 749) for the case \( L' = L + 1 \).

The explicit form of \( W(\phi) \) for the case in which both radiations of the cascade are mixtures of two multipoles is given by Rose (46, p. 477).
It is possible, because of the finite energy resolution of the detecting system, that two or more correlations from different cascades consisting of radiations of nearly the same energy may contribute to the experimentally measured function. The theoretical expression for such a composite correlation may be found by incoherently combining the separate contributing functions with proper weights. These weights will be determined by the relative gamma ray intensities of the radiations involved.

In the event of a cascade of the form \( j_0(L_1)j_1(L_2)j_2(L_3)j_3 \) a directional correlation between the first and third radiations, the "cross over" correlation, can be measured with the intermediate radiation, \( L_2 \), unobserved. The correlation function is given by Equation (2) with

\[
A_v = F_v(L_1,j_0,j_1)F_v(L_3,j_3,j_2) W(vL_2) W(j_1,j_2,j_3) W_0(L_2),
\]

so that the coefficients depend upon the angular momenta of the states and upon the multipolarities of the radiations, including that of the unobserved radiation. If either of the observed radiations is mixed, the correlation function is calculated as indicated above for the ordinary correlation, Equation (3). If the intermediate unobserved radiation is mixed the correlation function is
given by

\[ W(\phi) = W(L_2^\prime) + \delta^2 W(L_2) \]

where \( \delta^2 \) is the ratio of the intensity of the \( L^\prime \)-pole to that of the \( L \)-pole in the intermediate radiation.

Another quantity often used in angular correlation work is the anisotropy \( A \) defined as

\[ A \equiv \frac{W(180) - W(90)}{W(90)}. \]

If the highest coefficient in the expansion for \( W(\phi) \) is \( A_4 \) this becomes

\[ A = \frac{12A_2 + 5A_4}{8 - 4A_2 + 3A_4}. \]
THE ANGULAR MOMENTUM OF ELECTROMAGNETIC RADIATION

In order to calculate the angular momentum carried by the electromagnetic field it is convenient to use the spherical wave solutions for the field (28, p. 113). The solution of the scalar wave equation (the Helmholtz equation) in spherical coordinates can be written

$$\psi(x) = \sum_{\ell, m} \left[ A_{\ell m}^{(1)} h_{\ell}^{(1)}(kr) + A_{\ell m}^{(2)} h_{\ell}^{(2)}(kr) \right] Y_{\ell m}(\theta, \phi)$$  \hspace{1cm} (1)

where the $h_{\ell}^{(1,2)}$ are spherical Hankel functions and $k = \omega/c$. The coefficients $A_{\ell m}$ will be determined by the boundary conditions. The $Y_{\ell m}(\theta, \phi)$ are the spherical harmonics which are solutions of the angular part of the wave equation in spherical coordinates. In operator notation this can be written

$$L^2 Y_{\ell m} = \ell (\ell + 1) Y_{\ell m}$$

where $L^2 = L_x^2 + L_y^2 + L_z^2$ and $L = -i(r \times \nabla)$. Defining

$$L_{\pm} = L_x \pm i L_y, \hspace{1cm} L_z$$

the following equations hold:

$$L_{\pm} Y_{\ell m} = \sqrt{(\ell \pm m)(\ell \pm 1)} \hspace{0.2cm} Y_{\ell, m \pm 1}$$

$$L_z Y_{\ell m} = m Y_{\ell m}.$$  \hspace{1cm} (2)

The orthogonality of the spherical harmonics is expressed by

$$\int Y_{\ell m}^* Y_{\ell' m'} d\Omega = \delta_{ll'} \delta_{mm'}.$$  \hspace{1cm} (3)
In a source free region, assuming harmonic fields, Maxwell's equations are

\[ \nabla \times \mathbf{E} = ik \mathbf{B} \quad \nabla \times \mathbf{B} = -ik \mathbf{E} \]  
\[ \nabla \cdot \mathbf{E} = 0 \quad \nabla \cdot \mathbf{B} = 0 \]  

(4)

From these equations it can be shown that \( \mathbf{E} \) and \( \mathbf{B} \) separately satisfy the vector Helmholtz equation

\[ (\nabla^2 + k^2) \mathbf{F} = 0. \]  

(5)

The spherical wave multipole solutions for \( \mathbf{E} \) and \( \mathbf{B} \) can be obtained from Equation (5) which indicates that each rectangular component of \( \mathbf{E} \) or \( \mathbf{B} \) satisfies the scalar Helmholtz equation, so each component can be represented by an expression of the form Equation (1). These can be combined to yield the vectorial result, for example, for \( \mathbf{E} \).

\[ \mathbf{E} = \sum_{l,m} \left[ A_{lm}^{(1)} h_l^{(1)}(kr) + A_{lm}^{(2)} h_l^{(2)}(kr) \right] Y_{lm}(\theta, \phi) \]  

(6)

In evaluating the coefficients \( A_{lm} \) the divergence condition on \( \mathbf{B} \) must be satisfied. This leads to a condition that the \( \mathbf{B} \) field is transverse to the radius vector, resulting in a special set of electromagnetic fields,

\[ B_{lm} = f_l(kr) \mathbf{L} Y_{lm}(\theta, \phi) \]

\[ E_{lm} = \frac{i}{k} \nabla \times B_{lm} \]  

(7)
where
\[ f_\ell(kr) = A_\ell(1) h_\ell^{(1)}(kr) + A_\ell(2) h_\ell^{(2)}(kr). \]  
(8)

These represent solutions in which the magnetic field is perpendicular to the radius vector and are called electric multipole fields.

The calculation for \( \mathbf{E} \), instead of \( \mathbf{B} \), gives an alternative set of multipole fields in which \( \mathbf{E} \) is perpendicular to the radius vector.

\[ \mathbf{E}_{\ell m} = f_\ell(kr) \mathbf{L} Y_{\ell m}(\theta, \phi) \]
\[ \mathbf{B}_{\ell m} = \frac{i}{k} \nabla \times \mathbf{E}_{\ell m} \]  
(9)

These are called magnetic multipole fields. These two sets of fields form a complete set of vector solutions to Maxwell's equations (28, p. 120).

The vector spherical harmonics are defined by
\[ \mathbf{X}_{\ell m}(\theta, \phi) = \frac{1}{\sqrt{\ell (\ell + 1)}} \mathbf{L} Y_{\ell m}(\theta, \phi) \]  
(10)

with \( \mathbf{X}_0 = 0 \) and the orthogonality property
\[ \int \mathbf{X}^*_{\ell m} \cdot \mathbf{X}_{\ell' m'} \, d\Omega = \delta_{\ell \ell'} \delta_{mm'} . \]  
(11)

The general solution of Maxwell's equations will be a combination of the two types of fields.

\[ \mathbf{B} = \sum_{\ell, m} \left[ a_E(\ell, m) f_\ell(kr) \mathbf{X}_{\ell m} - \frac{i}{k} a_M(\ell, m) \mathbf{\nabla} \times g_\ell(kr) \mathbf{X}_{\ell m} \right] \]
\[ \mathbf{E} = \sum_{\ell, m} \left[ \frac{i}{k} a_E(\ell, m) \mathbf{\nabla} \times f_\ell(kr) \mathbf{X}_{\ell m} + a_M(\ell, m) g_\ell(kr) \mathbf{X}_{\ell m} \right] \]  
(12)
The coefficients $a_{E}(\ell, m)$ and $a_{M}(\ell, m)$ specify the amounts of the electric and magnetic fields, respectively, and will be determined by the sources and boundary conditions. The radial functions $f_{\ell}(kr)$ and $g_{\ell}(kr)$ are of the form Equation (8).

The multipole fields of a radiating source can be used to calculate the energy and angular momentum carried off by the radiation. For example, consider an electric multipole field.

From Equation (12), the fields which satisfy the boundary conditions of finiteness at the origin and outgoing waves at infinity are

$$
\vec{B}_{\ell m} = a_{E}(\ell, m)h^{(1)}_{\ell}(kr)\vec{X}_{\ell m} e^{-i\omega t}
$$

$$
\vec{E}_{\ell m} = \frac{i}{k} \nabla \times \vec{B}_{\ell m}.
$$

The time-averaged energy is

$$
U = \frac{1}{16\pi} \int (\vec{E} \cdot \vec{E}^*) + (\vec{B} \cdot \vec{B}^*) \, dV.
$$

Considering the radiation zone where the two terms in the integrand are equal on the average and using Equation (13) for $\vec{B}$,

$$
U = \frac{1}{8\pi} \int |a_{E}(\ell, m)|^2 |h^{(1)}_{\ell}(kr)|^2 \vec{X}_{\ell m} \cdot \vec{X}_* \, dV.
$$

(14a)

With the orthogonality relation for the vector spherical harmonics, Equation (11), this becomes

$$
U = \frac{1}{8\pi} \int |a_{E}(\ell, m)|^2 |h^{(1)}_{\ell}(kr)|^2 r^2 \, dr.
$$

(14b)
The time-average of the Poynting vector, giving the energy flux in the field is

$$\overline{S} = \frac{c}{8\pi} \text{Re}(\mathbf{E} \times \mathbf{B}^*).$$

The average momentum density is then $\overline{S}/c^2$, so the time-averaged angular momentum carried by the field is

$$\overline{M} = \frac{1}{8\pi c} \int \text{Re}[\mathbf{r} \times (\mathbf{E} \times \mathbf{B}^*)] \, dV.$$ 

Expanding the triple vector product and recalling that $\mathbf{r} \cdot \mathbf{B} = 0$ for this electric multipole field, the integral becomes

$$\overline{M} = \frac{1}{8\pi} \int \text{Re}[\mathbf{B}^*(\mathbf{r} \times -i \nabla) \cdot \mathbf{B}] \, dV.$$ 

Substituting for $\mathbf{E}$ from Equation (13) this becomes

$$\overline{M} = \frac{1}{8\pi} \int \text{Re}[\mathbf{B}^*(\mathbf{r} \times -i \nabla) \cdot \mathbf{B}] \, dV.$$ 

Using the operator identity $\mathbf{L} = -i\mathbf{r} \times \nabla$ the integral can be written

$$\overline{M} = \frac{1}{8\pi} \int \text{Re}[\mathbf{B}^*(\mathbf{L} \cdot \mathbf{B})] \, dV.$$ 

Using the expression for $\mathbf{B}$ from Equation (13),

$$\overline{M} = \frac{1}{8\pi} \int |a_E(\ell, m)|^2 |h_1^{(1)}(kr)|^2 \text{Re}(\mathbf{X}^*_{\ell m} \mathbf{L} \cdot \mathbf{X}^*_{\ell m}) \, dV.$$ 

From the definition of $\mathbf{X}^*_{\ell m}$, $(\mathbf{X}^*_{\ell m} \mathbf{L} \cdot \mathbf{X}^*_{\ell m}) = (Y_{\ell m}^* \mathbf{L} \cdot Y_{\ell m})$, so

$$\overline{M} = \frac{1}{8\pi} \int |a_E(\ell, m)|^2 |h_1^{(1)}(kr)|^2 \text{Re}(Y_{\ell m}^* \mathbf{L} \cdot Y_{\ell m}) r^2 \, dr \, d\Omega.$$ 

Equations (2) and (3) show that only the $z$-component of $\overline{M}$ exists.
For a magnetic \((\ell, m)\) multipole, \(a_E(\ell, m)\) is replaced by \(a_M(\ell, m)\). Comparing Equation (14b) for the energy with Equation (15),

\[
\frac{M_z}{U} = \frac{\frac{m}{\omega}}{\frac{\omega}{\hbar}} = \frac{m\hbar}{\hbar\omega}, \quad m = -\ell, -\ell + 1, \ldots, \ell.
\]

This is interpreted quantum mechanically to mean that radiation from a multipole of order \((\ell, m)\) carries off the \(z\)-component of angular momentum \(m\hbar\) per photon of energy \(\hbar\omega\). Furthermore, since according to this semiclassical calculation, only the \(z\)-component of the angular momentum exists, the ratio of the square of the total angular momentum to the square of the energy is

\[
\frac{M^2}{U^2} = \frac{m^2}{\omega^2}.
\]

This result arises from the above calculation using classical fields. Quantum mechanically it might be expected that \(L^2 = \ell(\ell+1)\hbar^2\), as is indeed the result given by a quantum electrodynamical calculation. A quantized field can be introduced by treating the amplitudes \(a_E(\ell, m)\) (or \(a_M(\ell, m)\)) as quantum mechanical operators.
which create and destroy photons of type \((\ell, m)\) (41, p. 297). The calculation is then performed in the same general way as in the classical case. DeWitt and Jensen (16, p. 268) have performed the calculation for a quantized multipole field containing \(N\) photons and find,

\[
L_z = Nm \hbar, \quad \vec{L}^2 = \{N^2 m^2 + N[\ell(\ell+1) - m^2]\} \hbar^2
\]

and

\[
\frac{U^2}{N} = (\hbar N \omega)^2 \quad m = -\ell, -\ell + 1, \ldots, \ell
\]

where \(\ell\) is the multipole order of the radiation.

For large \(N\) (the classical case) this result gives

\[
\lim_{N \to \infty} \frac{\vec{L}^2}{U^2} = \frac{m^2}{\omega^2}
\]

agreeing with the classical calculation. For a single photon, however, the rigorous treatment shows that

\[
\vec{L}^2 = \ell(\ell + 1)\hbar^2
\]

\[
L_z = m \hbar
\]

for the square and \(z\)-component of the angular momentum carried off by a photon of the multipole field \((\ell, m)\).

The question to be answered in connection with a directional correlation experiment concerns the \(z\)-component of the angular momentum carried off by the first gamma ray. From the form of the fields, Equation (12), the angular dependence of the Poynting
vector will be given by $|\vec{X}_{lm}(\theta, \phi)|^2$. The direction of propagation of the first radiation is defined as the z-axis, that is, the direction $\theta = 0$. $\vec{X}_{lm}(\theta, \phi)$ has the property that $\vec{X}_{lm}(0, \phi) = \delta_{m, \pm 1}$ for all $l$. Therefore, energy will be transmitted in the z-direction only when $m = \pm 1$ and will thus carry z-component of angular momentum of $\pm \hbar$ and no other value. Therefore, even this semi-classical argument shows clearly that the choice of the direction of propagation of the first radiation to be the z-axis fulfills the conditions which are required to see an anisotropy, namely, that the populations of the substates of the intermediate state will not be equal.
SOURCE PREPARATION

In order to obtain good statistics in the true coincidence rate, the chance coincidence rate should be kept as small as possible compared to the true coincidence rate. The counters must be positioned so as to subtend a large solid angle and a source of appropriate activity employed in order to meet this condition and still get a sufficient number of true coincidence counts in a reasonable time. Consider a source which emits two gamma rays in cascade and whose strength is $N_0$ disintegrations per second. The number of single counts recorded in each channel per second may be written as $N_i = N_0 w_i e_i$ where $w_i$ is the solid angle subtended by counter $i$ at the source and $e_i$ is the detection efficiency of channel $i$ for quantum $i$. The number of chance coincidence counts per second is given by

$$N_{\text{ch}} = 2TN_0N_2 = 2TN_0^2w_1w_2e_1e_2$$

where $2T$ is the experimentally determined resolving time of the apparatus. The number of true coincidence counts per second is

$$N_{\text{true}} = N_0 w_1 w_2 W(\phi)e_1 e_2$$

where $W(\phi)$ is the correlation function. The ratio of true coincidences to chance coincidences is then
\[ \frac{N_{\text{true}}}{N_{\text{ch}}} = \frac{W(\phi)}{2TN_o} \]

From this equation then, for a given \(2T\), \(N_o\) must be as small as possible (consistent with a sufficiently high coincidence counting rate) in order to make the true-to-chance ratio as large as possible.

The size and surroundings of the source are very important in angular correlation work. If a quantum is scattered in material near the source it may lose its original direction and smear out the correlation function, an effect which may be reduced by decreasing the thickness of the source and of the walls of the source holder. If the source is not cylindrically symmetric about the rotation axis of the movable counter the absorption can be angularly dependent. On the basis of expressions presented by Aeppli, et al. (1, p. 339) for a thin cylindrical source surrounded by scattering material, Frauenfelder (21, p. 149) gives the critical source and absorber thicknesses for various materials. For a lucite container for 600 Kev gamma rays, the critical wall thickness is about 2.00 mm. The walls of the lucite containers used in this experiment were 0.79 mm thick. The critical source thickness is on the order of 1.00 mm. The thickness of the source used in this experiment was 0.79 mm. Scattering in sources of less than critical thickness reduces the anisotropy by less than a factor of 0.98 (21, p. 150).
The symmetry of the source was confirmed by observing the singles counting rates at various angles.

The 2.3 year $^{134}\text{Cs}$ sources used in this experiment were prepared from material obtained from the Oak Ridge National Laboratory. This was in the form of high specific activity $\text{CsCl}$ dissolved in HCl. A small amount of this material was dropped into a small cavity in the lucite source holder, the liquid was allowed to evaporate and the process was repeated until a source of the desired activity was obtained. The cavity was then sealed with a lucite cap to contain the source material and to minimize evaporation.

Figure 3 shows the gamma ray pulse height distribution in the region of interest for the $^{134}\text{Cs}$. 

Commercial sealed $^{60}\text{Co}$ sources of 0.1 and 1.1 millicuries were used for testing the spectrometer by performing a $^{60}\text{Ni}$ directional correlation.
Figure 3. PULSE HEIGHT SPECTRUM OF Cs\textsuperscript{134}. 

Backscatter peak

605, 569, and 563 Kev

797 and 802 Kev
EXPERIMENTAL APPARATUS

The arrangement of the electronic equipment used in this investigation was similar to the conventional fast-slow delayed coincidence system in which the functions of timing and energy selection were performed separately and then combined in a triple coincidence circuit.

A pair of scintillation counters was used to detect the nuclear gamma rays. The voltage pulses generated by each counter were proportional to the energy of the gamma rays detected in that counter. These output pulses were fed into pre-amplifiers and then into linear amplifiers, after which the signals were separated into timing and energy selection channels. Constant amplitude output pulses from each amplifier were generated by selected amplified counter pulses and fed into a fast coincidence circuit. Additionally, unselected amplified counter pulses were fed into single channel differential pulse height analyzers. Each pulse height analyzer generated a constant amplitude output pulse whenever the size of an input pulse fell between two voltage levels, \( V \) and \( V + dV \), thus acting as an energy selector. The output pulses from the analyzers were fed into a slow coincidence circuit. The output of the fast coincidence circuit was delayed before being fed
to the triple coincidence stage. This was necessary in order to compensate for the delay introduced by the pulse height analyzers. By this technique only those triple coincidences were formed corresponding to signals from detectors 1 and 2 whose amplitudes fell within the respective windows of the analyzers. Figure 4 shows a block diagram of the scintillation spectrometer.

The gamma-ray detectors consisted of NaI(Tl) crystals 1.5 inches in diameter and 1.0 inches thick optically coupled to Dumont K-1719 photomultiplier tubes. This assembly was positioned in a steel cylinder which acted as a mechanical support and a partial electromagnetic shield. Cylindrical mu-metal magnetic shields were installed around the steel cylinders to eliminate effects produced by the earth's magnetic field.

The preamplifiers were stacked cathode followers which provided a fast rising, negative output pulse. The amplifiers were R-C coupled non-overloading types designed for a rise time of 0.2 microsecond. The pulse height selector output pulse used for the fast coincidence was a 30 volt negative pulse of 0.7 microsecond duration. Each differential pulse height analyzer was fed by a positive output pulse of 1.6 microseconds duration followed by a negative undershoot with an exponential decay.

The differential pulse height analyzers were of a
Figure 4. BLOCK DIAGRAM OF THE SCINTILLATION SPECTROMETER.
conventional design. Each unit produced a negative output pulse only when a positive input signal had an amplitude falling within two preset voltage levels. The instrument determined whether or not the height of the input pulse exceeded the lower level of the channel and then whether or not the pulse size exceeded the upper level. When the input pulse size exceeded the lower level a negative output pulse was generated which would then be vetoed by an anticoincidence circuit if the pulse also exceeded the upper level. In this way output pulses were generated only for those pulses whose sizes fell between the upper and lower levels set on the instrument.

The coincidence circuit consisted of two fast and two slow channels. The fast pulse height selector pulses from the amplifiers were fed directly to the fast coincidence circuit. If these two pulses arrived within the resolving time of the fast coincidence circuit, a coincidence occurred producing a large pulse at a common plate junction. This pulse, after shaping, was fed to the triple coincidence bus after having been delayed about 5.5 microseconds in order to compensate for the extra time needed to carry out the energy selection in the slow channel pulse height analyzers. Output pulses from the analyzers were fed to the two slow channels of the coincidence circuit. The outputs of the two
fast and the two slow channels, all positive pulses, were fed to the junction of four diodes and a load resistor on the triple coincidence bus. The bias across each diode was set so that any one of the diodes could hold the voltage down on the load resistor even though one or more of the other diodes was cut off. This condition could be attained by applying a positive pulse to the cathode of the diode or by opening the circuit through the diode by a manual switch.

When a triple coincidence occurred (fast and two slows) a positive pulse was generated on the triple coincidence bus. This pulse was fed to the output stage which in turn fed a scaler of the conventional type.

The photomultiplier high voltage power supplies were designed to provide a very stable (0.02 percent per day) d-c voltage source for precision scintillation counting. The output voltage changes by less than 0.35 percent for a current increase from zero to maximum load (1 milliampere) and by less than 0.00035 percent per volt change of line from 100-130 volts. The ripple was less than 0.01 percent of the output voltage. Each unit consisted of an input regulating transformer followed by a high voltage transformer, a rectifier, and an R-C filter network. The filtered d-c voltage was applied to a bank of 17 cold cathode tubes from which the stabilized output voltage was obtained.
One detector was fixed on the spectrometer table while the other could be rotated about the central axis, on which the source was placed, and set at various angular positions to within 0.25 degree. The distance of each detector from the central axis was adjustable. The alignment of the counters and the source was done with the aid of a telescope.
EXPERIMENTAL MEASUREMENTS

Before measurements on Ba\(^{134}\) were begun an angular correlation measurement was performed with the gamma rays of Ni\(^{60}\). The data were taken at five angles using a 0.1 millicurie Co\(^{60}\) source. The Ni\(^{60}\) correlation is well known and serves as a convenient means of testing the experimental methods and equipment. The experiment was done in the same way as is described below in the case of Ba\(^{134}\). The Ni\(^{60}\) measurement yielded results in agreement with the published (50, p. 553) directional correlation coefficients for Ni\(^{60}\) and with the theory. With this assurance that the equipment was functioning properly and that the method of data treatment was satisfactory the directional correlation measurements with Ba\(^{134}\) were undertaken.

The Cs\(^{134}\) source was prepared as described in the section on source preparation and was mounted in the source holder on the angular correlation table where it remained for the duration of the entire experiment. This served to eliminate fatigue effects in the photomultiplier tubes that are observed when the tubes are first exposed to a source. The source strength was about 80 microcuries. This gave a ratio of true to chance coincidence of about two. All runs were made at a source to detector distance
of 7.0 cm.

Two different angular correlations were measured in the gamma decay of Ba$^{134}$. The decay scheme is quite complicated and includes some gamma rays of nearly the same energy. The energy resolution of the detecting system was measured to be about nine percent in the geometry used, so that gamma rays differing in energy by less than this amount could not be resolved.

The first correlation that was measured was an "overall" correlation which is the composite correlation made up of the 797-605 Kev, the 802-563 Kev, the 569-797 Kev correlations, and the 802-[563]-605 Kev cross-over correlation. Because of the small energy differences the low energy photopeak will be composed of the 605 Kev, 569 Kev, and 563 Kev gamma rays. The higher energy photopeak will be made up of the 797 Kev and 802 Kev gamma rays. For simplicity the lower peak will be termed the 605 Kev peak and the upper one the 797 Kev peak.

In order to measure the "overall" correlation the gains of the amplifiers were adjusted so that the pulses in the 797 Kev gamma ray peak in channel 2 had the same amplitude as the pulses from the 605 Kev gamma ray peak in channel 1. The amplifier and analyzer discriminators of each channel were then set at point L in Figure 5. The windows of the analyzers were then
set so as to bracket the peaks. The discriminator on amplifier 1 was then varied slightly to optimize the delay and pulse shape and maximize the coincidence counting rate. The background level with no source in place was found to be negligible. The counting rates in each channel were kept to about $10^3$ per second so that counting losses were negligible.

In order to be able to correct the total coincidence counting rate for chance coincidences the effective resolving time of the fast-slow coincidence circuit had to be determined. It was measured by the incoherent source method during which the settings of the electronic equipment were identical to those used in a run. The directional correlation source was left in position where it was viewed by the fixed detector and the movable detector was removed from the table and inserted into a thick-walled lead cave. A second source of the material under investigation was placed in the cave and its distance from the counter was varied until the singles counting rate of that channel was identical to its rate during a correlation run. The two detectors and their sources were completely shielded from one another so that the coincidences that were recorded were due only to chance. From this measurement the resolving time was given by
\[ 2T = \frac{N_{\text{ch aux}}}{N_{1 \text{ aux}} N_{2 \text{ aux}}} \]

where \( N_{1 \text{ aux}}, N_{2 \text{ aux}}, \) and \( N_{\text{ch aux}} \) are the analyzed singles of channels 1 and 2 and the coincidence rate, respectively, of this auxiliary experiment. The resolving time was measured during each run and was found to be about \( 12 \times 10^{-8} \) sec remaining essentially constant from run to run.

The second correlation measurement that was attempted, the "separated" correlation, was that composed of two cascades only; the 797-605 Kev and the 802-[563]-605 Kev correlations. With an energy resolution of nine percent the half widths at half maximum of the photopeaks of the 569 Kev and 563 Kev gamma rays would be about 25 Kev. The amplifier gains were readjusted and the amplifier and analyzer discriminators were set as shown in Figure 6 at L' with the analyzer windows, \( W, \) bracketing the peaks as shown. In this way, setting the discriminator levels of channel 1 at about 605 Kev, the contributions of the 563 Kev and 569 Kev gamma rays were largely eliminated. From this point the same procedures were followed as in measuring the "overall" correlation.

The triple coincidence and slow singles rates were determined at 19 angles between 90 degrees and 270 degrees in ten
Figure 5. PULSE HEIGHT SPECTRUM OF Cs$^{134}$ FOR CHANNELS 1 AND 2 IN THE "OVERALL" CORRELATION.

Figure 6. PULSE HEIGHT SPECTRUM OF Cs$^{134}$ FOR CHANNELS 1 AND 2 IN THE "SEPARATED" CORRELATION.
degree increments for each correlation. The fast singles were observed as a stability monitor. In each case the data were collected in a series of half-hour runs at each angle alternating with half-hour runs at 90 degrees (or 270 degrees) for normalization purposes. In the overall correlation at least $10^5$ true coincidence counts were accumulated at each angle, except for 90 (or 270) degrees, where approximately $10^6$ were accumulated. For the $(797 \pm 802)-605$ Kev ("separated") correlation about $4 \times 10^4$ true coincidences were obtained at each angle except 90 (or 270) degrees where approximately $4 \times 10^5$ counts were obtained. A slow electronic drift was apparent during an eight or ten hour run so the equipment was reset whenever the singles counting rate in either channel changed by more than one percent. The data were reduced according to the treatment given in the appendix. The results for the two correlation measurements are given in Figures 7 and 8 and in Table 4. In the figures the solid curve is the theoretical directional correlation function for the case $4(2)2(2)0$ modified to correct for the finite solid angles subtended by the two detectors. The experimental coefficients presented in Table 4 have been corrected for solid angle. The uncertainties indicated on the experimental points are due to statistical effects only.
Figure 7. Ba$^{134}$ "OVERALL" CORRELATION.
Figure 8. $^{134}$Ba "SEPARATED" CORRELATION.
Table 4. DIRECTIONAL CORRELATION COEFFICIENTS AND ANISOTROPY OF $^{134}$Ba.

<table>
<thead>
<tr>
<th></th>
<th>$A_0$</th>
<th>$A_2$</th>
<th>$A_4$</th>
<th>Anisotropy</th>
</tr>
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<td>$^{134}$Ba</td>
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<td>$+0.1478 \pm 0.0067$</td>
</tr>
<tr>
<td>least squares</td>
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<td>$+0.0142 \pm 0.0086$</td>
<td>$+0.1610 \pm 0.0114$</td>
</tr>
<tr>
<td>&quot;Overall&quot; correlation</td>
<td>1</td>
<td>$+0.0904 \pm 0.0026$</td>
<td>$+0.0097 \pm 0.0052$</td>
<td>$+0.1478 \pm 0.0067$</td>
</tr>
<tr>
<td>&quot;Separated&quot; correlation</td>
<td>1</td>
<td>$+0.0968 \pm 0.0043$</td>
<td>$+0.0142 \pm 0.0086$</td>
<td>$+0.1610 \pm 0.0114$</td>
</tr>
</tbody>
</table>
CONCLUSIONS

The results of the gamma ray directional correlation measurements in Ba$^{134}$ are summarized in Figures 7 and 8 and in Table 4.

In the decay of a state with angular momentum \( j \) to a state with angular momentum \( j' \) by the emission of radiation with angular momentum \( L \), the possible values of \( L \) are determined by the conservation law \( |j - j'| \leq L \leq j + j' \), so that angular momentum sequences commensurate with this selection rule must be considered. Any gamma ray to the ground state in Ba$^{134}$ must be a pure multipole since the ground state angular momentum is zero. On the basis of reported work the 605 Kev and 1401 Kev states are taken to be \( 2^+ \) and \( 4^+ \) respectively with the 797 Kev and 605 Kev radiations both electric quadrupole.

The "separated" correlation will be composed of two components, the 797 Kev-605 Kev cascade, and the 802 Kev-[563 Kev] - 605 Kev cascade in which the intermediate 563 Kev radiation is unobserved. The expected correlation function can be calculated on the basis of the decay scheme by weighting the contributions according to the gamma ray intensities. On the basis of lifetime considerations and reported internal conversion coefficients some
combinations of angular momenta for the 1970 Kev and 1168 Kev levels can be ruled out. The remaining reasonable pairs of values of the angular momenta of the 1970 Kev and 1168 Kev states, respectively, are 3 and 1, 3 and 2, and 4 and 2. Table 5 contains the calculated directional correlation coefficients for the "separated" correlation for the possible pure multipole orders of the radiations for these angular momenta. The contributions have been weighted using the experimentally observed gamma ray intensities given in Figure 1 and the weights are 72.5/81.7 and 9.2/81.7 respectively in the order in which the contributions appear in Table 5. None of these possibilities can be ruled out on the basis of the experimental coefficients, particularly since multipole mixing is possible and suitable mixing can bring the calculations into agreement with the experiment within the experimental error.

The "overall" correlation is a composite of four cascades, the 797 Kev-605 Kev (72.5/102.4), the 802 Kev-563 Kev (9.2/102.4), the 569 Kev-797 Kev (11.5/102.4), and the 802 Kev-[563 Kev]-605 Kev (9.2/102.4) in which the 563 Kev radiation is again unobserved. Expected coefficients have again been calculated for the possible angular momenta and multipolarities and are shown in Table 6 where measured intensities have again been used to determine the weights of the contributions. The weight of each cascade is given
<table>
<thead>
<tr>
<th>J</th>
<th>J</th>
<th>797-605</th>
<th>(A_2)</th>
<th>(A_4)</th>
<th>802- [563] -605</th>
<th>(A_2)</th>
<th>(A_4)</th>
<th>Combined (A_2)</th>
<th>(A_4)</th>
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<td>+.0026</td>
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"Separated" Experimental Coefficients

\[ A_2 = +0.0968 \pm 0.0043 \]
\[ A_4 = +0.0142 \pm 0.0086 \]
**TABLE 6.** CALCULATED COEFFICIENTS FOR THE "OVERALL" CORRELATION IN Ba$^{134}$.

<table>
<thead>
<tr>
<th>J</th>
<th>J</th>
<th>802-563</th>
<th></th>
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<th>569-797</th>
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<td>+.1516</td>
<td>+.0550</td>
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</table>

"Overall" Experimental Coefficients

\[
A_2 = +0.0904 \pm 0.0026 \\
A_4 = +0.0097 \pm 0.0052
\]
above in parentheses after the cascade. The constant contribution of the 797 Kev-605 Kev 4(2)2(2)0 cascade has been omitted from the table for simplicity but its effect is included in the calculated coefficients. The coefficients for the 802 Kev-[563 Kev]-605 Kev cascade have not been listed explicitly since they are the same as those given in Table 5. In this case multipole mixing is possible in at least two of the radiations for each pair of angular momenta and by the choice of proper mixing ratios the calculations in each case can be brought into agreement with the experimental data within their errors. None of the three possible angular momentum sequences can be ruled out on the basis of these experimental results alone. Since it is possible to include mixing in at least two radiations the values of the mixing ratios cannot be assigned unambiguously.

The interpretation of the results of directional correlation measurements between the 1365 Kev and the 605 Kev radiations are not complicated by interfering radiations on a more complete decay scheme and indicate the assignment of angular momentum 4 to the 1970 Kev level (32, p. 66), (18, p. 955), (54, p. 691). Taking the 1970 Kev level to have angular momentum 4 and, from the allowed nature of the beta branch, to have positive parity leaves angular momentum 2 as the probable assignment for the
1168 Kev level. This angular momentum sequence is supported by the recent work of Segaert, et al. (48, p. 87). The parity of this state is taken to be positive so that the 802 Kev gamma ray is electric quadrupole. Otherwise the 802 Kev radiation would be magnetic quadrupole, electric octupole, or a higher order multipole and would be very weak. Comparing Tables 5 and 6 for these angular momenta it appears that the experimental $A_2$ of the "separated" correlation is best fitted by taking the 563 Kev radiation to be magnetic dipole whereas the coefficient for the "overall" correlation is best fitted by taking the 563 Kev radiation to be electric quadrupole and the 569 Kev gamma ray to be magnetic dipole. This apparent inconsistency can be removed by considering both the 563 Kev and the 569 Kev radiations to be magnetic dipole-electric quadrupole mixtures. The other possible mixtures would involve magnetic octupole or higher order multipoles and can be neglected. Figure 9 is a plot of the experimentally allowed values of $\delta_{563}$ versus those of $\delta_{569}$, where the mixing ratio $\delta$ is, in each case, the ratio of the electric quadrupole to magnetic dipole intensity in the radiation. In the "separated" correlation only $\delta_{563}$ appears. By requiring the theoretical mixed $A_2$ for this case to equal the experimental $A_2$ within its standard error, the restriction $0 \leq |\delta_{563}| \leq 0.92$ is obtained. The horizontal
lines labeled 1 at $\delta_{563} = \pm 0.92$ then define the boundaries of the allowed values of the mixing ratio to fit the separated correlation. The experimental $A_4$ for the "separated" case places no restrictions on $\delta_{563}$. The same procedure was applied to the overall correlation in which both mixing ratios appear. The solid "vertical" lines labeled 2 in Figure 9 represent the boundaries of the region of allowed mixing in order for the theoretical mixed $A_4$ of the "overall" correlation to agree with the experimental "overall" $A_4$ within its standard error. Finally, the areas within the pairs of curves labeled 3 and 4 represent the allowed values of mixing so that the theoretical mixed $A_2$ for the "overall" correlation agrees with the experimental "overall" $A_2$ within its standard error. The areas of overlap of these three regions are shaded and represent the allowed range of values of $\delta_{563}$ and $\delta_{569}$ on the basis of these experiments. The arrow at $\delta_{563} = 0$, $\delta_{569} = -0.38$ indicates the closest fit to the "separated" and "overall" experimental A's. At this point the calculated "separated" $A_2$ is $+0.0963$ and the calculated "overall" $A_2$ is $+0.0904$. From these curves it is seen that either or both of the 563 Kev or 569 Kev radiations can be at most 50 percent electric quadrupole, corresponding to $\delta = \pm 1$. This is in disagreement with the results reported by Stewart et al. (54, p. 694) who report the 569 Kev radiation to be
Figure 9. EXPERIMENTALLY ALLOWED $\delta_{563}$ VERSUS EXPERIMENTALLY ALLOWED $\delta_{569}$.
94 percent electric quadrupole. If the condition on the "overall
A_4 (curves 2) is ignored in Figure 9, however, it is seen that a
large positive \( \delta_{569} \), corresponding to that radiation being pre-
dominantly electric quadrupole in agreement with Stewart et al.,
is not in disagreement with the \( A_2 \)'s of the present work pro-
vided that the 563 Kev radiation is taken to be a nearly pure mag-
netic dipole. Generally, the results of the present experiment
support the assignment of large magnetic dipole admixtures to
both the 569 Kev and the 563 Kev gamma rays.

If the angular momenta and parities of the levels being con-
sidered are assumed to be \( 2^+, 2^+, 4^+, 4^+ \) in order of ascending
energy then the asymmetric rotor model of Mallmann is able to
correctly predict the energies of these levels. One result of the
model is that magnetic dipole transitions are forbidden. Tables
of reduced electric quadrupole transition probabilities have been
prepared by Day and Mallmann (15, p. 1). Assuming the experi-
mental beta branching ratios and neglecting the presence of the
three states of unknown angular momenta, the gamma ray intensi-
ties were calculated using these reduced transition probabilities.
The probability per unit time for an electric quadrupole transition
is proportional to \( E^5 \) (\( E \) is the gamma ray energy) times the re-
duced transition probability. The results are given in Table 7.
Table 7. THEORETICAL AND MEASURED GAMMA RAY INTENSITIES IN Ba$^{134}$.

<table>
<thead>
<tr>
<th>Gamma Energy (Kev)</th>
<th>Asymmetric Rotor Reduced Transition Probability</th>
<th>Gamma Rays per 100 Decays of Cs$^{134}$</th>
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<td>563</td>
<td>0.3196</td>
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</tr>
<tr>
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<td>2.2</td>
</tr>
<tr>
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<td>0.2553</td>
<td>78.6</td>
</tr>
<tr>
<td>797</td>
<td>0.3635</td>
<td>63.2</td>
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<tr>
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<td>18.2</td>
</tr>
<tr>
<td>1168</td>
<td>0.0108</td>
<td>11.1</td>
</tr>
<tr>
<td>1365</td>
<td>0.0029</td>
<td>6.6</td>
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</table>

The comparison for the radiations from the 1970 Kev level is most meaningful and is not good. On the basis of the model all these radiations must be electric quadrupole or higher order multipoles so mixing can be neglected. Using this fact and the calculated intensities, the expected correlation coefficients were computed. The predictions for the "separated" correlation are $A_2 = +0.0882$, $A_4 = +0.0078$, and those for the "overall" correlation are $A_2 = +0.0718$, $A_4 = +0.0117$. These coefficients are not in agreement with the experimental results.

In summary, the results of these directional correlation measurements are in agreement with the "accepted" assignments of angular momenta and parities of $2^+$, $2^+$, $4^+$, $4^+$ to the 605 Kev, 1168 Kev, 1401 Kev, and 1970 Kev excited states respectively, but they are not in disagreement with angular momenta.
assignments of 2, 1, 4, 3 or 2, 2, 4, 3 to these states respectively. For the assignments \( 2^+, 2^+, 4^+, 4^+ \) the experimental results indicate, however, the predominant magnetic dipole character of two of the radiations in contradiction to the selection rule imposed by the asymmetric rotor model, a model which successfully predicts the energies of these excited states in \(^{134}\text{Ba}\).
BIBLIOGRAPHY


13. Cork, J. M. et al. The radioactive decay of Cs\textsuperscript{134}, Os\textsuperscript{185}, Os\textsuperscript{191}, and Os\textsuperscript{193}. Physical Review 90:444-447. 1953.


APPENDICES
APPENDIX I

Geometrical Correction Factors to $W(\phi)$

The results of an angular correlation experiment are most conveniently expressed in terms of an expansion in Legendre polynomials. That is, the corrected true coincidence counting rate per unit solid angle is proportional to

$$W(\theta)_{\text{exp}} = \sum_{n} a_n P_n(\cos \theta)$$

where $n$ takes the values 0, 2, and 4, and the $a_n$ are determined from a least-squares fit to the data. $W(\theta)_{\text{exp}}$ is related to the theoretical correlation function $W(\phi)$. Since the scintillation crystals subtend finite solid angles at the source, the theoretical correlation function must be modified. This smeared (or averaged) correlation function $\overline{W(\phi)}$ is then compared with the $W(\theta)_{\text{exp}}$ determined from the measurements (47, p. 610). The object here is to find the relationship between $\overline{W(\phi)}$ and $W(\theta)_{\text{exp}}$. The geometry under consideration is shown in Figure 10, where

- $x(\beta_i)$ are the interaction paths for the gamma rays in the crystals,
- $\phi_i$ are the azimuth angles of the radiations measured with respect to the crystal axes,
- $\theta$ is the angle between the crystal axes,
Figure 10. GEOMETRY OF THE CRYSTALS FOR THE FINITE DETECTOR SIZE CORRECTION.
\( \phi \) is the angle between the directions of propagation of the cascade gamma rays,

\( \alpha \) is the angle between the axis of crystal 1 and the propagation direction of radiation 2,

\( h \) is the distance from the source to the face of either crystal,

\( t \) is the thickness of each crystal, and

\( r \) is the radius of each crystal.

The crystals are identical right circular cylinders with the bases oriented toward the source. The source is situated at the origin of the coordinate system at the intersection of the crystal axes. In this case, as will be seen, each coefficient in the expansion will be multiplied by an easily determined attenuation factor.

\( x(\beta_1) \) is the interaction path of gamma 1 at an angle \( \beta_1 \) so that the absorption is proportional to \( (1 - e^{-\tau_1(E_1)x(\beta_1)}) \), where \( \tau_1(E_1) \) is the energy-dependent absorption coefficient. The smeared out, or measured, correlation function is given by

\[
\overline{W(\phi)} = \frac{\int W(\phi)(1 - e^{-\tau_1 x(\beta_1)})(1 - e^{-\tau_2 x(\beta_2)})dw_1 dw_2}{\int (1 - e^{-\tau_1 x(\beta_1)})(1 - e^{-\tau_2 x(\beta_2)})dw_1 dw_2}
\]

(2)

where the theoretical function is given by

\[
W(\phi) = \sum_v A_v P_v(\cos \phi)
\]

with \( v = 0, 2, 4 \). Substituting this into Equation (2) it can be
seen that the integrals of interest are of the form

\[ I_v = \int P_v(\cos \phi)(1 - e^{-\tau_1 x(\beta_1)}) (1 - e^{-\tau_2 x(\beta_2)}) \sin \beta_1 d\beta_1 d\phi_1 \]

\[ \times \sin \beta_2 d\beta_2 d\phi_2 \]  

(3)

with

\[ x(\beta_1) = \text{tsec} \beta_1 \quad \text{for} \quad 0 < \beta_1 < \tan^{-1} \frac{r}{h + t} = \beta' \]

and

\[ x(\beta_2) = \text{t csc} \beta_2 - \text{h sec} \beta_1 \quad \text{for} \quad \beta' < \beta_1 < \tan^{-1} \frac{r}{h} = \gamma \]

The evaluation of Equation (3) is accomplished by using the addition theorem for spherical harmonics

\[ P_v(\cos \phi) = \frac{4 \pi}{2v+1} \sum_m Y_{v}^{m*} (\beta_1 \phi_1) Y_{v}^{m} (\alpha \phi_2). \]

A double application of this theorem and the realization that \( x(\beta_1) \) and \( x(\beta_2) \) are independent of \( \phi_1 \) and \( \phi_2 \), respectively, and that \( \theta \) is independent of \( \phi_1 \) and \( \phi_2 \) so that the integration over these angles can be performed easily leads to

\[ P_v(\cos \phi) = P_v(\cos \beta_1) P_v(\cos \alpha) \]

and

\[ P_v(\cos \alpha) = P_v(\cos \beta_2) P_v(\cos \theta). \]

Recalling Equation (2), the use of these expressions in Equation (3) gives

\[ W(\phi) = \sum_v A_v P_v(\cos \theta) \frac{J_v(1) J_v(2)}{J_o(1) J_o(2)} \]  

(4)
where

\[ J_v(i) = \int_0^\gamma P_v(\cos \beta_i) (1 - e^{-\tau_i x(\beta_i)}) \sin \beta_i d\beta_i \]

and again \( v = 0, 2, 4 \). Since Equations (1) and (4) are supposed to represent the same thing, then

\[ a_n = A_v \frac{J_v(1)}{J_o(1)} \frac{J_v(2)}{J_o(2)} \]

relates the experimental \( a_n \) and the theoretical \( A_v \) coefficients.

The \( J_v \) integrals have been evaluated by Stanford and Rivers for crystals of various \( r \) and \( t \) for values of \( h \) from 7 to 50 cm. using the absorption coefficients for NaI corresponding to energies from 0.05 Mev to 5.0 Mev (52, p. 719).

The \( J_v(i) \) integrals can be approximated by

\[ J_v(i) = \int_0^\gamma P_v(\cos \beta_i) \epsilon(\beta_i) \sin \beta_i d\beta_i \]

where \( \epsilon(\beta_i) \) is the experimentally determined angular resolution.

\( J_0 \), \( J_2 \), and \( J_4 \) can then be evaluated by graphical integration of this integral. The \( J_v \) values have been experimentally determined in this way for the detectors used in this experiment by Glasgow et al. (25, p. 683) and by Mansfield and Schecter (37, p. 574).

The results support the use of Stanford and Rivers' calculated values for the correction factors.
APPENDIX II

Treatment of the Data

The dependence of the coincidence counting rate on the angular position of the movable detector corresponds to the theoretical correlation function only under assumptions of point detectors, centered point sources, and the absence of scattered or disturbing radiations. The comparison of the experimental and theoretical results then follows after corrections have been made to the experimental data for deviations from such an ideal arrangement. The source and its container were made small (active source volume about $7 \times 10^{-4}$ cm$^3$) so that no corrections were necessary for the source size or scattering in the source or its container. The experiments were performed with a scintillation spectrometer with its energy selection protection against scattered and other disturbing radiations. In addition the sides of the scintillation crystals were shielded with lead for further protection against radiation scattered from one crystal into the other.

The true coincidence rate $N_{i1}(\theta_k)$ of the $i^{th}$ run at the angle $\theta_k$ was determined in each case by subtracting the chance (or accidental) coincidence rate $N_{i\text{ch}}(\theta_k) = 2T N_{i1}(\theta_k) N_{i2}(\theta_k)$ from the total coincidence rate $N_{i\text{tot}}(\theta_k)$. $N_{i1}(\theta_k)$ and $N_{i2}(\theta_k)$
are the analyzed singles rates of channels one and two and $2T$ is
the resolving time of the system.

The true coincidence rate was then divided by the product
of the slow singles rates of channels one and two. This division
by the singles rates will correct to first order small errors in the
centering of the source (50, p. 597), and should also be independ-
ent, to first order, of any changes in the efficiencies of the two
channels. The result is the true coincidence ratio

$$D_i^1(\theta_k) = \frac{N_i^{it}(\theta_k)}{N_i^{i1}(\theta_k)N_i^{i2}(\theta_k)} \cdot \frac{N_i^{itot}(\theta_k) - 2TN_i^{i1}(\theta_k)N_i^{i2}(\theta_k)}{N_i^{i1}(\theta_k)N_i^{i2}(\theta_k)}.$$

The standard error in a function $F(x_i)$ due to errors in
the variables $x_i$ is given by

$$\sigma_{F(x_i)} = \left[ \sum_i \left( \frac{dF}{dx_i} \right)^2 \sigma_i^2 \right]^{1/2}$$

(8, p. 313). Then the standard error associated with $D_i^1(\theta_k)$ is

$$\sigma_{D_i^1(\theta_k)} = \frac{D_i^1(\theta_k)}{N_i^{it}(\theta_k)} \left[ N_i^{itot}(\theta_k) + N_i^{ich}(\theta_k) \right]^{1/2}$$

where $N_{ch\ aux}$ is the coincidence rate of the auxiliary two source
experiment.

The true coincidence ratio at $\theta_k$ was divided by the true
coincidence ratio at 90 degrees to give a normalized ratio

$$R_i^1(\theta_k) = D_i^1(\theta_k)/D_i^1(90)$$

independent to first order of instrumental
drift. The error associated with this normalized ratio is

\[
\frac{\sigma}{R_i(\theta_k)} = R_i(\theta_k) \left[ \frac{\sigma^2 D_i(\theta_k)}{D_i^2(\theta_k)} + \frac{\sigma^2 D_i(90)}{D_i^2(90)} \right]^{1/2}
\]

The average weighted ratio for all the \(i\) runs at angle \(\theta_k\) will be

\[
R(\theta_k) = \frac{\sum_i w_i R_i(\theta_k)}{\sum_i w_i}
\]

where the weight \(w_i\) associated with the \(i^{th}\) run is the ratio of a normalizing factor \(b\), chosen for convenience, and the square of the standard error associated with the \(i^{th}\) ratio, \(R_i(\theta_k)\). The standard error of the weighted average ratio is given by

\[
\sigma_{R(\theta_k)} = \frac{b^{1/2}}{\left(\sum_i w_i\right)^{1/2}} \text{ and } \left[ \frac{\sum_i w_i d_i^2}{(n-1)\sum_i w_i} \right]^{1/2}
\]

for internal and external consistency respectively. \(n\) is the number of pieces of data and each \(d_i\) is the deviation of each \(R_i(\theta_k)\) from \(R(\theta_k)\). No data were rejected. These two expressions were found to give nearly the same value for the standard error \(\sigma_{R(\theta_k)}\) of \(R(\theta_k)\), though the larger of the two values was used in each case. The weight assigned to \(R(\theta_k)\) in the least-squares analysis was taken as the ratio of the normalizing factor \(b\) and \(\sigma^2_{R(\theta_k)}\).
Table A-1 shows a sample calculation of $w_i R_i(\theta_k)$, $d_i$, $d_i^2$, and $w_i d_i^2$. As an example, at $\theta_k = 140$ degrees, 14 experimental determinations of $R_i(140)$ were made for the case of the "overall" correlation, yielding a weighted mean value of $1.0693 \pm 0.0059$.

Table A-1. Sample calculation of $w_i R_i(140)$, $d_i$, $d_i^2$, and $w_i d_i^2$.

<table>
<thead>
<tr>
<th>$R_i(140)$</th>
<th>$\sigma_{R_i}^2$</th>
<th>$w_i = \frac{b}{\sigma_{R_i}^2} = 10^{-3}$</th>
<th>$w_i R_i(140)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0631±0.0215</td>
<td>.000462</td>
<td>2.1645</td>
<td>2.3011</td>
</tr>
</tbody>
</table>

\[
d_i = R(140) - R_i(140) \quad d_i^2 \quad w_i d_i^2
\]

\[
\begin{array}{ccc}
& .0062 & .000038 & .000082 \\
\end{array}
\]

The total reduction of the 14 pieces of data gave:

1. The mean weighted normalized ratio

\[
R(140) = \frac{\sum w_i R_i(140)}{\sum w_i} = \frac{31.0716}{29.0567} = 1.0693 \pm 0.0059
\]

2. The standard error associated with $R(140)$

\[
\sigma_{in} = \sqrt{\frac{b^{1/2}}{\left(\sum w_i\right)^{1/2}}} = \frac{3.163 \times 10^{-2}}{(29.06)^{1/2}} = 0.0059
\]
3. The least-squares weight of $R(140)$

$$w_i \ell. s. = \frac{b^2}{\sigma^2} = \frac{10^{-4}}{0.0000348} = 2.8727$$

The identical procedure was carried out at each angle.

**Least Squares Analysis of the Data**

A least-squares analysis was performed to determine the coefficients describing $W(\theta)_{\text{exp}}$. A series of Legendre polynomials was used to fit the $R(\theta_k)$ as closely as possible (47, p. 613):

$$W(\theta_k)_{\text{exp}} = R(\theta_k) = \sum_{j=0}^{4} a_j P_j(\cos \theta_k) = \sum_j a_j A_{kj}. \quad \text{for } j \text{ even}$$

The most probable coefficients $a_j$ for the given data were obtained by minimizing the function

$$\sum_k w_k \left[ R(\theta_k) - \sum_j a_j A_{kj} \right]^2$$

yielding the normal equations

$$\sum_k w_k \left[ R(\theta_k) - \sum_j a_j A_{kj} \right] A_{ki} = 0$$
A symmetric square matrix is now defined so that

\[ C = \tilde{A} W A \]

with elements

\[ C_{ji} = \sum_k w_i A_{kj} A_{ki} = C_{ij} \]

where \( W \) is a diagonal matrix with elements \( w_k \), \( A \) is a matrix of Legendre polynomials, and \( \tilde{A} \) is its transpose. Now define

\[ B = \tilde{A} W R \]

where \( R \) is a matrix with elements \( R(\theta_k) \). From the normal equations it follows that \( C a = B \), or multiplying by the inverse of \( C \), \( a = C^{-1} B \).

In detail,

\[ a_j = \sum_i C^{-1}_{ji} B_i \]

gives the desired coefficients.

\( a \) is a linear homogenous function of the counting rates \( R(\theta_k) \). Hence, due to the existence of a variation in \( R(\theta_k) \) expressed by \( \sigma^2_{R(\theta_k)} \) there will be corresponding mean square deviations in the coefficients. Writing out the expression for \( a_j \),

\[ a_j = \sum_{k,\ell} w_k R(\theta_k) C^{-1}_{\ell j} A_{k\ell} \]

and

\[ \sigma^2_{a_j} = \sum_{k,\ell} \left( C^{-1}_{\ell j} A_{k\ell} \right)^2 \sigma^2_{R(\theta_k)} w_k^2 \sigma^2_{R(\theta_k)} . \]
This can be written

\[ \sigma^2_{a_j} = \sum_{k,l,i} C^{-1}_{kj} C^{-1}_{ij} A_{kl} A_{ki} w_k \frac{b'}{R(\theta_k)} \sigma^2_{R(\theta_k)} \]

\[ = b' \sum_{\ell,i} C^{-1}_{\ell j} C^{-1}_{ij} \sum_k A_{\ell k} A_{ki} w_k \]

\[ = b' \sum_{\ell,i} C^{-1}_{\ell j} C^{-1}_{ij} C_{\ell i} . \]

Now let \( i = j \) and the sum over \( \ell \) and \( j \) is performed giving

\[ \sigma^2_{a_j} = b'C^{-1}_{jj} . \]

The square of the standard error in the coefficients is given by the diagonal elements of \( C^{-1} \) multiplied by \( b' \). After determining the coefficients and their errors they were corrected for the finite size of the detectors using the \( J_0, J_2, \) and \( J_4 \) discussed in Appendix I, assuming that the errors in the \( J_i \)'s were negligible. Tables A-2 and A-3 show the calculation of the \( a_j \), their errors, and \( W(\theta)_{\text{exp}} \) for the two cases measured.
**TABLE A-2.** CALCULATION SHEET FOR THE LEAST SQUARES ANALYSIS OF THE EXPERIMENTAL COEFFICIENTS FOR THE "OVERALL" CORRELATION OF $B_{\alpha}^{134}$

<table>
<thead>
<tr>
<th>$\theta_k$</th>
<th>$R(\theta_k)$</th>
<th>$w_k$</th>
<th>$A_{k2}$</th>
<th>$w_k A_{k2}$</th>
<th>$A_{k2}^2$</th>
<th>$w_k A_{k2}^2$</th>
<th>$A_{k4}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>0.9998</td>
<td>39.0625</td>
<td>-0.50000</td>
<td>-19.5313</td>
<td>+0.25000</td>
<td>+9.7656</td>
<td>+0.37500</td>
</tr>
<tr>
<td>100</td>
<td>0.9979</td>
<td>3.4294</td>
<td>-0.45478</td>
<td>-1.5596</td>
<td>+0.20683</td>
<td>+0.7093</td>
<td>+0.26592</td>
</tr>
<tr>
<td>110</td>
<td>1.0164</td>
<td>2.0408</td>
<td>-0.32453</td>
<td>-0.6623</td>
<td>+0.10532</td>
<td>+0.2149</td>
<td>-0.00383</td>
</tr>
<tr>
<td>120</td>
<td>1.0230</td>
<td>2.0408</td>
<td>-0.12500</td>
<td>-0.2551</td>
<td>+0.01563</td>
<td>+0.0319</td>
<td>-0.28906</td>
</tr>
<tr>
<td>130</td>
<td>1.0542</td>
<td>3.1888</td>
<td>+0.11977</td>
<td>+0.3819</td>
<td>+0.01435</td>
<td>+0.0457</td>
<td>-0.42753</td>
</tr>
<tr>
<td>140</td>
<td>1.0693</td>
<td>2.8727</td>
<td>+0.38023</td>
<td>+1.0923</td>
<td>+0.14458</td>
<td>+0.4153</td>
<td>-0.31904</td>
</tr>
<tr>
<td>150</td>
<td>1.0889</td>
<td>2.2957</td>
<td>+0.62500</td>
<td>+1.4348</td>
<td>+0.39063</td>
<td>+0.8968</td>
<td>+0.02344</td>
</tr>
<tr>
<td>160</td>
<td>1.1139</td>
<td>2.0408</td>
<td>+0.82453</td>
<td>+1.6827</td>
<td>+0.67985</td>
<td>+1.3874</td>
<td>+0.47495</td>
</tr>
<tr>
<td>170</td>
<td>1.1270</td>
<td>2.7778</td>
<td>+0.95478</td>
<td>+2.6522</td>
<td>+0.91161</td>
<td>+2.5322</td>
<td>+0.85323</td>
</tr>
<tr>
<td>180</td>
<td>1.1416</td>
<td>2.1626</td>
<td>+1.00000</td>
<td>+2.1626</td>
<td>+1.00000</td>
<td>+2.1626</td>
<td>+1.00000</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$w_k A_{k4}$</th>
<th>$A_{k4}^2$</th>
<th>$w_k A_{k4}^2$</th>
<th>$w_k R(\theta_k)$</th>
<th>$w_k A_{k4}^2 R(\theta_k)$</th>
<th>$w_k A_{k4}^2 R(\theta_k)$</th>
</tr>
</thead>
</table>
Table A-2 (Continued)

\[
\sum_{k} w_k = +61.9119 \quad \sum_{k} w_k A_{k2} A_{k4} = -2.9161
\]

\[
\sum_{k} w_k A_{k2} = -12.6017 \quad \sum_{k} w_k R(\theta_k) = +63.4448
\]

\[
\sum_{k} w_k A_{k2}^2 = +18.1619 \quad \sum_{k} w_k A_{k2} R(\theta_k) = -11.5526
\]

\[
\sum_{k} w_k A_{k4} = +18.2386 \quad \sum_{k} w_k A_{k4} R(\theta_k) = +18.8051
\]

\[
\sum_{k} w_k A_{k4}^2 = +11.4280
\]

\[
|C| = \begin{vmatrix}
\sum_{k} w_k & \sum_{k} w_k A_{k2} & \sum_{k} w_k A_{k4} \\
\sum_{k} w_k A_{k2} & \sum_{k} w_k A_{k2}^2 & \sum_{k} w_k A_{k2} A_{k4} \\
\sum_{k} w_k A_{k4} & \sum_{k} w_k A_{k2} A_{k4} & \sum_{k} w_k A_{k4}^2
\end{vmatrix}
\]

\[
\begin{array}{c|c|c|c|c}
\hline
a_0 & a_2 & a_4 & +63.4448 & -11.5526 & +18.8051 \\
+0.0343 & +0.156 & -0.0507 & x & \\
+0.0156 & +0.0645 & -0.0085 & \times & -11.5526 \\
-0.0507 & -0.0085 & +0.1663 & \times & +18.8051 \\
\hline
\end{array}
\]
Table A-2 (Continued)

<table>
<thead>
<tr>
<th>(a_n)'s normalized to 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_0) = 1.0402</td>
</tr>
<tr>
<td>(a_2) = 0.0869</td>
</tr>
<tr>
<td>(a_4) = 0.0076</td>
</tr>
</tbody>
</table>

Error in the \(a_n\)

\[
\sigma^2_{a_0} = b^t C^{-1} b = 10^{-4} \times 3.427 \times 10^{-2}
\]

\[
\sigma_{a_0} = 0.0019
\]

\[
\sigma_{a_2} = 0.0025
\]

\[
\sigma_{a_4} = 0.0041
\]

Error in the \(a_n\) normalized and compounded.

\[
\sigma_{a_0} = 0.0026
\]

\[
\sigma_{a_2} = 0.0024
\]

\[
\sigma_{a_4} = 0.0039
\]

Solid angle correction of the \(a_n\)

\[
A_2 = a_2 \frac{J_0(1)J_0(2)}{J_2(1)J_2(2)} = \frac{0.0835}{0.9610 \times 0.9614} = 0.0904
\]

\[
\sigma_{A_2} = 0.0026
\]
Table A-2 (Continued)

\[
A_4 = a_4 \frac{J_0(1)J_0(2)}{J_4(1)J_4(2)} = \frac{0.0073}{0.8660 \times 0.8675} = 0.0097
\]

\[
\sigma_{A_4} = 0.0052
\]

\[
W(\theta)_{\text{exp}} = 1 + (0.0904 \pm 0.0026)P_2(\cos \theta)
\]

\[
+ (0.0097 \pm 0.0052)P_4(\cos \theta)
\]

\[
A_{\text{exp}} = \frac{W(180) - W(90)}{W(90)} = 0.1478 \pm 0.0067
\]
TABLE A-3. CALCULATION SHEET FOR THE LEAST SQUARES ANALYSIS OF THE EXPERIMENTAL COEFFICIENTS FOR THE "SEPARATED" CORRELATION OF Ba$^{134}$.

<table>
<thead>
<tr>
<th>$\theta_k$</th>
<th>$R(\theta_k)$</th>
<th>$w_k$</th>
<th>$A_{k2}$</th>
<th>$w_k A_{k2}$</th>
<th>$A_{k2}^2$</th>
<th>$w_k A_{k4}^2$</th>
<th>$A_{k4}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>0.9992</td>
<td>16.000</td>
<td>-0.5000</td>
<td>-8.0000</td>
<td>+0.2500</td>
<td>+4.0000</td>
<td>+0.3750</td>
</tr>
<tr>
<td>100</td>
<td>0.9939</td>
<td>1.1317</td>
<td>-0.4547</td>
<td>-0.5147</td>
<td>+0.2068</td>
<td>+0.2341</td>
<td>+0.2659</td>
</tr>
<tr>
<td>110</td>
<td>1.0011</td>
<td>0.9070</td>
<td>-0.3245</td>
<td>-0.2944</td>
<td>+0.1053</td>
<td>+0.0955</td>
<td>-0.0038</td>
</tr>
<tr>
<td>120</td>
<td>1.0265</td>
<td>1.1080</td>
<td>-0.1250</td>
<td>-0.1385</td>
<td>+0.0156</td>
<td>+0.0173</td>
<td>-0.2890</td>
</tr>
<tr>
<td>130</td>
<td>1.0461</td>
<td>1.0000</td>
<td>+0.1197</td>
<td>+0.1198</td>
<td>+0.0145</td>
<td>+0.0143</td>
<td>-0.4275</td>
</tr>
<tr>
<td>140</td>
<td>1.0714</td>
<td>1.1815</td>
<td>+0.3803</td>
<td>+0.4492</td>
<td>+0.1445</td>
<td>+0.1708</td>
<td>-0.3190</td>
</tr>
<tr>
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<td>1.1125</td>
<td>0.7432</td>
<td>+0.6250</td>
<td>+0.4645</td>
<td>+0.3906</td>
<td>+0.2903</td>
<td>+0.0234</td>
</tr>
<tr>
<td>160</td>
<td>1.1212</td>
<td>1.1080</td>
<td>+0.8245</td>
<td>+0.9136</td>
<td>+0.6798</td>
<td>+0.7533</td>
<td>+0.4749</td>
</tr>
<tr>
<td>170</td>
<td>1.1256</td>
<td>1.1317</td>
<td>+0.9547</td>
<td>+1.0806</td>
<td>+0.9161</td>
<td>+1.0317</td>
<td>+0.8532</td>
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<tr>
<td>180</td>
<td>1.1450</td>
<td>0.5102</td>
<td>+1.0000</td>
<td>+0.5102</td>
<td>+1.0000</td>
<td>+0.5102</td>
<td>+1.0000</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$w_k A_{k4}^2$</th>
<th>$A_{k4}^2$</th>
<th>$w_k A_{k2} A_{k4}$</th>
<th>$w_k A_{k2}^2 A_{k4}$</th>
<th>$w_k R(\theta_k)$</th>
<th>$A_{k2}^2 R(\theta_k)$</th>
<th>$w_k A_{k4} R(\theta_k)$</th>
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<tbody>
<tr>
<td>+6.0000</td>
<td>+0.14063</td>
<td>+2.2500</td>
<td>-3.0000</td>
<td>+15.9872</td>
<td>-7.9936</td>
<td>+5.9952</td>
</tr>
<tr>
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<td>+0.07071</td>
<td>+0.0800</td>
<td>-0.1369</td>
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<td>-0.5116</td>
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<td>+0.00015</td>
<td>+0.0001</td>
<td>+0.0011</td>
<td>+0.9080</td>
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<td>+0.0926</td>
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<tr>
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<td>+0.1828</td>
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<td>-0.1433</td>
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<td>+0.4813</td>
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<td>+0.2258</td>
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<td>+1.0966</td>
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<td>+1.00000</td>
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<td>+0.5842</td>
<td>+0.5842</td>
<td>+0.5842</td>
<td>+0.5842</td>
</tr>
</tbody>
</table>
Table A-3 (Continued)

\[
\begin{align*}
\sum_{k} w_k &= +24.8214 & \sum_{k} w_k A_{k2} A_{k4} &= -1.4133 \\
\sum_{k} w_k A_{k2} &= -5.4097 & \sum_{k} w_k R(\theta_k) &= +25.4079 \\
\sum_{k} w_k A_{k2}^2 &= +7.1176 & \sum_{k} w_k A_{k2} R(\theta_k) &= -4.9831 \\
\sum_{k} w_k A_{k4} &= +7.1922 & \sum_{k} w_k A_{k4} R(\theta_k) &= +7.4011 \\
\sum_{k} w_k A_{k4}^2 &= +4.3101
\end{align*}
\]

\[
|C| = \begin{bmatrix}
\sum_{k} w_k & \sum_{k} w_k A_{k2} & \sum_{k} w_k A_{k4} \\
\sum_{k} w_k A_{k2} & \sum_{k} w_k A_{k2}^2 & \sum_{k} w_k A_{k2} A_{k4} \\
\sum_{k} w_k A_{k4} & \sum_{k} w_k A_{k2} A_{k4} & \sum_{k} w_k A_{k4}^2
\end{bmatrix}
\]

\[
\begin{bmatrix}
a_0 \\ a_2 \\ a_4
\end{bmatrix} = \begin{bmatrix}
+0.0876 \\ +0.0402 \\ -0.1329
\end{bmatrix} \begin{bmatrix}
+0.0402 \\ +0.1687 \\ -0.0117
\end{bmatrix} \begin{bmatrix}
-0.1329 \\ -0.0117 \\ +0.4500
\end{bmatrix} \begin{bmatrix}
+25.4079 \\ -4.9831 \\ +7.4011
\end{bmatrix}
\]
Table A-3 (Continued)

<table>
<thead>
<tr>
<th>( a_0 )</th>
<th>( a_0 )</th>
<th>( a_2 )</th>
<th>( a_2 )</th>
<th>( a_4 )</th>
<th>( a_4 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0407</td>
<td>1.0000</td>
<td>0.0931</td>
<td>0.0894</td>
<td>0.0111</td>
<td>0.0106</td>
</tr>
</tbody>
</table>

**Error in the \( a_n \)\textsuperscript{'}s normalized to 1**

\[
\sigma^2_{a_0} = b_iC^{-1}_{jj} = 10^{-4} \times 8.756 \times 10^{-2}
\]

\[
\sigma_{a_0} = 0.0030
\]

\[
\sigma_{a_2} = 0.0041
\]

\[
\sigma_{a_4} = 0.0067
\]

**Error in the \( a_n \) normalized and compounded.**

\[
\sigma_{a_0} = 0.0041
\]

\[
\sigma_{a_2} = 0.0039
\]

\[
\sigma_{a_4} = 0.0064
\]

**Solid angle correction of the \( a_n \)**

\[
A_2 = a \frac{J_0(1)J_0(2)}{2J_2(1)J_2(2)} = \frac{0.0894}{0.9610 \times 0.9614} = 0.0968
\]

\[
\sigma_{A_2} = 0.0043
\]
Table A-3 (Continued)

\[ A_4 = a_4 \frac{J_0(1)J_0(2)}{J_4(1)J_4(2)} = \frac{0.0106}{0.8660 \times 0.8675} = 0.0142 \]

\[ \sigma_{A_4} = 0.0086 \]

\[ W(\theta)_{\text{exp}} = 1 + (0.0968 \pm 0.0043)P_2(\cos \theta) \]

\[ + (0.0142 \pm 0.0086)P_4(\cos \theta) \]

\[ A_{\exp} = \frac{W(180) - W(90)}{W(90)} = 0.1610 \pm 0.0114 \]