AN ABSTRACT OF THE THESIS OF

Raymond Walter Sommerfeldt for the Ph. D. in Physics

Date thesis is presented August 6, 1964

Title PERTURBED DIRECTIONAL CORRELATION IN Ta-181

Abstract approved (Major professor)

The effect of a static quadrupole interaction on the 133-482 Kev gamma-gamma angular correlation in Ta-181 has been studied by the delayed coincidence method. Using a source of Hf-181 in polycrystalline hafnium metal, the anisotropy was measured as a function of time between the formation and decay of the 482 Kev state with a fast-slow coincidence spectrometer. The anisotropy as a function of time revealed clearly the periodic fluctuations arising from the re-orientation of the nuclear spin due to coupling of the quadrupole moment of the 482 Kev state with the electric field gradients of the hafnium crystals. The interaction strength evaluated on the basis of the theory of Abragam and Pound for axially-symmetric gradients was 317 ± 8 Mc/sec. Evidence for the presence of a rhombic (non-axial symmetric) electric interaction was observed in the departure of the results from the predictions of the axially-symmetric theory. The results are, however, in agreement with the theory for rhombic
interactions if the electric field gradient in the hafnium crystals used is assumed to have an asymmetry parameter \( \eta = (V_{xx} - V_{yy})/V_{zz} = 0.3 \). A similar study of the 133-345 KeV gamma-ray cascade did not permit a direct observation of the quadrupole coupling due to interference from the 133-482 and 345-137 KeV cascades; however, when the contributions of the interfering cascades were removed from the composite data, the results were consistent with those of the 133-482 experiment as well as with existing theory. An indirect determination of the anisotropy of the 345-137 KeV cascade gave the value 0.32 ± 0.02 in agreement with a previous measurement. In addition, the half-life of the 482 KeV excited state was remeasured to be 10.4 ± 0.2 nano-seconds consistent with prior determinations.
PERTURBED DIRECTIONAL CORRELATION IN Ta-181

by

RAYMOND WALTER SOMMERFELDT

A THESIS

submitted to

OREGON STATE UNIVERSITY

in partial fulfillment of
the requirements for the
degree of

DOCTOR OF PHILOSOPHY

August 1964
APPROVED:

Professor of Department of Physics

In Charge of Major

Chairman of Department of Physics

Dean of Graduate School

Date thesis is presented August 6, 1964

Typed by Illa W. Atwood
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>THEORY</td>
<td>11</td>
</tr>
<tr>
<td>Qualitative Description of Gamma-Gamma Angular Correlation</td>
<td>11</td>
</tr>
<tr>
<td>Static Electric Quadrupole Interaction</td>
<td>15</td>
</tr>
<tr>
<td>Perturbed Angular Correlation</td>
<td>24</td>
</tr>
<tr>
<td>APPARATUS</td>
<td>44</td>
</tr>
<tr>
<td>TEST AND CALIBRATION OF THE SPECTROMETER</td>
<td>51</td>
</tr>
<tr>
<td>SOURCE PREPARATION</td>
<td>57</td>
</tr>
<tr>
<td>EXPERIMENTAL MEASUREMENTS</td>
<td>61</td>
</tr>
<tr>
<td>Lifetime of the 482 kilovolt Excited State in Ta</td>
<td>61</td>
</tr>
<tr>
<td>The Unperturbed Anisotropy of the 133-482 kilovolt Cascade</td>
<td>65</td>
</tr>
<tr>
<td>The Perturbed Anisotropy of the 133-482 kilovolt Cascade</td>
<td>67</td>
</tr>
<tr>
<td>The Perturbed Anisotropy of the 133-345 kilovolt Cascade</td>
<td>71</td>
</tr>
<tr>
<td>CONCLUSIONS</td>
<td>80</td>
</tr>
<tr>
<td>BIBLIOGRAPHY</td>
<td>87</td>
</tr>
<tr>
<td>APPENDIX</td>
<td>91</td>
</tr>
</tbody>
</table>
## LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Decay scheme of Hf$^{181}$</td>
<td>9</td>
</tr>
<tr>
<td>2</td>
<td>Example of a directional correlation measurement.</td>
<td>13</td>
</tr>
<tr>
<td>3</td>
<td>The definition of the position vectors of the nuclear and extranuclear charges.</td>
<td>16</td>
</tr>
<tr>
<td>4</td>
<td>Precession of a quadrupole nucleus in a non-uniform electric field.</td>
<td>25</td>
</tr>
<tr>
<td>5</td>
<td>The angular coordinates of the propagation directions $k_1$ and $k_2$ of $\gamma_1$ and $\gamma_2$, respectively.</td>
<td>33</td>
</tr>
<tr>
<td>6</td>
<td>Position of the $z'$-axis defined by the Euler angles $\alpha$ and $\beta$.</td>
<td>37</td>
</tr>
<tr>
<td>7</td>
<td>The attenuation coefficient $G_2(t)$ for $I = \frac{5}{2}$ as a function of $\omega_0 t / \pi$.</td>
<td>43</td>
</tr>
<tr>
<td>8</td>
<td>Block diagram of the delayed-coincidence spectrometer.</td>
<td>45</td>
</tr>
<tr>
<td>9</td>
<td>Time-to-pulse-height converter circuit.</td>
<td>49</td>
</tr>
<tr>
<td>10</td>
<td>Overall response of the time-to-pulse-height converter showing the linear relation between the difference in arrival time of the fast pulses at the converter and the output pulse height.</td>
<td>52</td>
</tr>
<tr>
<td>11</td>
<td>Prompt coincidence curves showing the time resolution of the spectrometer.</td>
<td>53</td>
</tr>
<tr>
<td>12</td>
<td>The differential pulse-height spectrum of Ta$^{181}$.</td>
<td>55</td>
</tr>
</tbody>
</table>
### LIST OF FIGURES (Continued)

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>13</td>
<td>Delayed coincidence curves for the 133-482 Kev gamma-ray cascade.</td>
<td>62</td>
</tr>
<tr>
<td>14</td>
<td>The anisotropy of the 133-482 kilovolt gamma cascade of Ta$^{181}$ obtained with a liquid source. The horizontal dotted line represents the value calculated from the angular correlation coefficients assuming no perturbation.</td>
<td>67</td>
</tr>
<tr>
<td>15</td>
<td>Delayed coincidence data at 90° and 180° for the 133-482 kilovolt gamma-ray cascade in Ta$^{181}$.</td>
<td>69</td>
</tr>
<tr>
<td>16</td>
<td>Time dependent anisotropy of the 133-482 kilovolt gamma cascade in polycrystalline hafnium metal.</td>
<td>70</td>
</tr>
<tr>
<td>17</td>
<td>Partial level scheme of Ta$^{181}$.</td>
<td>71</td>
</tr>
<tr>
<td>18</td>
<td>Composite delayed coincidence data for the 133-482, 133-345, and 345-137 kilovolt gamma-ray cascades in Ta$^{181}$.</td>
<td>73</td>
</tr>
<tr>
<td>19</td>
<td>The differential anisotropy of the mixed 133-482, 133-345 cascade as a function of time.</td>
<td>77</td>
</tr>
<tr>
<td>20</td>
<td>Differential attenuation coefficients $G_2(t)$ for rhombic quadrupole interaction in polycrystalline sources.</td>
<td>83</td>
</tr>
<tr>
<td>21</td>
<td>Differential attenuation coefficients $G_2(t)$ for axially-symmetric quadrupole interaction with frequency distribution $\delta$ in polycrystalline sources.</td>
<td>83</td>
</tr>
</tbody>
</table>

**Appendix**

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>The angular coordinates of the propagation vectors $\vec{k}_1$ and $\vec{k}_2$.</td>
<td>92</td>
</tr>
</tbody>
</table>
### LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>Results of studies of the unperturbed angular correlations in Ta$^{181}$.</td>
<td>8</td>
</tr>
<tr>
<td>II</td>
<td>Results of lifetime measurements for the 482 kilovolt excited state in Ta$^{181}$.</td>
<td>10</td>
</tr>
</tbody>
</table>
PERTURBED DIRECTIONAL CORRELATION IN Ta-181

INTRODUCTION

A great deal of information about the properties of low-lying nuclear levels has been accumulated during the past few years. The rapid improvement of the experimental techniques of nuclear spectroscopy in recent years has brought an ever increasing number of levels into the region of observation while the use of nuclear models as a means of correlating and interpreting experimental results has provided a much needed basis for the systematic study of nuclear properties. As a result, there exist today consistent decay schemes for a large number of nuclides and probable schemes for many others. In addition, many of the properties of ground and excited states as well as the properties of the transitions between states are known for many nuclides. These properties include the energies, angular momenta, and parities of ground and excited states and the energies, transition probabilities, and character of the transitions between levels.

Among the excited state properties less well known are the electric quadrupole and magnetic dipole moments. The direct measurement of these quantities is quite difficult owing to the short-lived character of excited states plus other difficulties to be mentioned later. Some twenty or so excited state magnetic moments have been
measured to date, but as yet not a single excited state quadrupole moment has been determined by a model-independent method. Although values for these moments can be derived on the bases of nuclear models which permit their evaluation in terms of more easily measured parameters, this procedure is necessarily limited to those cases for which the validity of the particular model used can be established. In principle, this test can only be provided through a knowledge of the moments themselves, and regardless of the success of the model in predicting other parameters of the nucleus under study, one must still regard model-dependent values as somewhat uncertain.

One of the most valuable tools of nuclear spectroscopy for the investigation of excited state nuclear moments is the study of angular correlations perturbed by extranuclear fields. The angular correlation method in which the "true" or unperturbed directional distribution between two successively emitted radiations from a radioactive nucleus is observed is itself a very useful tool for the determination of angular momenta of excited nuclear levels. In some cases, the angular correlation can be altered by electric and magnetic fields acting on the electric and magnetic moments of the nucleus. These may be externally applied fields or may be internal fields arising from the atomic, molecular or crystalline structure of the source. A measurement of the magnetic dipole and electric
quadrupole moment then becomes possible if the fields are known. Moreover, the dependence of the angular correlation on the electric and magnetic interactions of the nuclear moments with extranuclear fields can be used to study the magnetic and electric fields prevailing at the site of the decaying nucleus, and yield information on the atomic and molecular structure of matter. Thus, extranuclear perturbations which make questionable to some extent all measurements based on the observation of the true angular correlation offer as a natural compensation a method for the investigation of excited state nuclear moments and of certain aspects of the extranuclear structure as well.

The possibility of using perturbed angular correlation studies for the determination of excited state nuclear moments was first suggested in 1950 by Brady and Deutsch (10, p. 559) who pointed out that the g-factor of an excited level intermediate to the member radiations of a gamma-gamma cascade could be measured by observing the effect of an external magnetic field on the angular correlation of the cascade. Alder (5, p. 369-370; 6, p. 235-251) and Lloyd (31, p. 277-278) developed the pertinent theory a short time later; the first successful experiment was performed in 1951 by Aeppli and his co-workers (2, p. 370-371; 3, p. 339-369) who used this technique to determine the magnitude and sign of the g-factor of the first excited state in Cd$^{111}$. Since that time, similar measurements have been
made for excited states of several other nuclides. The method has also been extended to include situations involving alpha-gamma cascades (27, p. 234-237) as well as those in which the angular distribution of the de-excitation gamma-rays following Coulomb excitation is observed (22, p. 701-707).

The determination of the electric quadrupole moment of an excited state is, in principle at least, also possible. That is, an electric field gradient present at the nucleus will interact with the nuclear quadrupole moment and lead to a perturbation of the angular correlation in much the same manner as in the magnetic case. However, one major difference exists here which makes the precise measurement of the nuclear electric quadrupole moment incomparably more difficult than the measurement of the nuclear magnetic dipole moment. In the study of a magnetic interaction, the externally applied magnetic field is known, and the dipole moment can then be calculated from the observed coupling. In the quadrupole case, however, it is impossible to produce synthetic electric field gradients of sufficient strength to perturb the angular correlation measurably, and it becomes necessary to make use of the sizable field gradients existing in crystals to provide the interaction. Since these are generally not known and, moreover, are very difficult to calculate (45, p. 293), only the strength of the quadrupole coupling
which involves the product of the unknown quadrupole moment and the unknown field gradient can be reliably determined.

Several experimental arrangements for the investigation of the effects of excited state quadrupole interactions on angular correlations have been suggested in the theoretical treatments of the subject by Alder and his co-workers at Zurich (7, p. 761-784) and by Abragam and Pound (1, p. 943-962). Again, the first significant experimental work was done by the Zurich group who determined the quadrupole coupling of the 247 kilovolt level in Cd$^{111}$ with the electric field gradient in an indium single crystal by observing the attenuation of the angular correlation as a function of crystal orientation (4, p. 322-323).

An alternate procedure, suggested by Abragam and Pound (1, p. 948), is to use a polycrystalline source and observe the time-differential angular correlation as a function of the time between formation and decay of the intermediate excited state. This method has the feature of displaying the effect of the perturbing interaction in a manner which reflects clearly the nature of the quadrupole precession of the nuclear spin about the direction of the field gradient, and permits a direct measurement of the precession period (and hence the coupling if the spin is known) from the resulting periodic behavior of the angular correlation. However, such an experiment is feasible only if the lifetime of the intermediate state is long.
enough to permit observation over at least one precession period. Of equal importance, is that the apparatus have a resolving time which is short as compared to the precession period in order that the structure of the differential correlation be well resolved. Unfortunately, the strength of the quadrupole interaction is not usually known a priori and this, together with the restrictions on the lifetime of the excited level and the instrumental resolution limits the number of levels known to be suited to studies of this type to but a few cases. The only experiments of this kind reported thus far are those of Lehmann and Miller on the 247 kilovolt, 80 nanosecond level in Cd$^{111}$ (28, p. 298-299; 29, p. 526-527) and that of Wertheim and Pound on the 1.27 Mev, 0.27 microsecond level in Pb$^{204}$ (48, p. 185-189). In the work on Pb$^{204}$ cited, the strength of the interaction in the metallic thallium environment used resulted in a precession period of about 3 microseconds. Because of the comparatively short lifetime of the excited state, it was not possible to obtain a check of the functional dependence of the differential correlation except over the relatively short period of the first few hundred nanoseconds of the decay of the level. In the cadmium sulphate crystals used by Lehmann and Miller in their initial work with Cd$^{111}$, on the other hand, the interaction was sufficiently strong to permit observation over three precession periods with reasonably good statistics. However, here the resolving time was insufficient
to show very detailed structure of the correlation function. Shortly after this, Lehmann and Miller repeated their experiments on Cd\textsuperscript{111} using a metallic indium source rather than CdSO\textsubscript{4}, and observed a precession period sufficiently long to permit a fairly detailed study of the correlation function over one period.

Some recent work on gamma-gamma and electron-gamma angular correlations in Ta\textsuperscript{181} reported by Grabowski et al. (24, p. 256) indicated that the conditions for a differential experiment might also be met in a situation involving the 133-482 kilovolt gamma cascade in this nucleus. An estimate made on the basis of their work with metallic hafnium sources indicated the strength of the interaction between the quadrupole moment of the 10 nanosecond, 482 kilovolt state in Ta\textsuperscript{181} and the electric field gradient in hafnium metal to correspond to a precession period in the interval 10-20 nanoseconds. Since it is possible to attain resolutions of a few nanoseconds through the use of the latest fast coincidence techniques, such an experiment appeared feasible. If a source of polycrystalline hafnium metal containing the radioactive Hf\textsuperscript{181} → Ta\textsuperscript{181} were used, a study of the differential anisotropy of the 133-482 kilovolt gamma-ray cascade should permit a determination of the quadrupole coupling and a detailed check of the functional dependence of the correlation over several precession periods as well. This thesis describes the performance of such an experiment.
The radiations from the decay of $^{181}\text{Hf}$ have been the subject of numerous investigations; the results have been tabulated in the Nuclear Data Sheets (23, p. 60-2-110 to 60-2-117). The main features of the $^{181}\text{Hf}$ decay and the level structure of $^{181}\text{Ta}$ are shown in Figure 1. The lifetime of the 482 kilovolt state and the unperturbed angular correlation coefficients of the 133-482 kilovolt cascade, both pertinent to the proposed work, have been well-established. In addition, both gamma-gamma and electron-gamma angular correlations for the other principal cascades of this nucleus have been measured. The results of specific interest here are summarized in Tables I and II.

Table I. Results of studies of the unperturbed angular correlations in $^{181}\text{Ta}$. The $A_2$ and $A_4$ are the correlation coefficients. (See page 110 of the appendix.)

<table>
<thead>
<tr>
<th>Cascade</th>
<th>Correlation Coeff.</th>
<th>Source Conditions</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>133-482, r-$r$</td>
<td>$-0.280 \pm 0.004$</td>
<td>$-0.058 \pm 0.003$</td>
<td>Hafnium metal in 27N HF</td>
</tr>
<tr>
<td>133-345</td>
<td>$0.110 \pm 0.012$</td>
<td>$0.020 \pm 0.014$</td>
<td>HfF$_4$ in HF</td>
</tr>
<tr>
<td>345-137</td>
<td>$0.202 \pm 0.009$</td>
<td>$-0.053 \pm 0.014$</td>
<td>Hf in HF</td>
</tr>
<tr>
<td>133-482, 345-137</td>
<td>$-0.281 \pm 0.010$</td>
<td>$-0.071 \pm 0.04$</td>
<td>HfF$_4$ in HF</td>
</tr>
<tr>
<td>133-345, 345-137</td>
<td>$0.16 \pm 0.05$</td>
<td>$0.00 \pm 0.04$</td>
<td>HfF$_4$ in HF</td>
</tr>
<tr>
<td>133-482, 345-137</td>
<td>$0.17 \pm 0.04$</td>
<td>$0.01 \pm 0.04$</td>
<td>HfF$_4$ in HF</td>
</tr>
<tr>
<td>133-482</td>
<td>$-0.230 \pm 0.012$</td>
<td>$-0.080 \pm 0.02$</td>
<td>HfO$_2$ in HF</td>
</tr>
</tbody>
</table>

Although a number of angular correlation experiments involving the radiations from transitions through the 482 kilovolt state in $^{181}\text{Ta}$ show evidence for the existence of a strong quadrupole interaction, a detailed study of the interaction which a differential correlation experiment makes possible has not yet been reported. The lack of quantitative information in this particular case, and the fact
(Energies in Kev)

Figure 1. Decay scheme of Hf$^{181}$. 
Table II. Results of lifetime measurements for the 482 kilovolt excited state in Ta$^{181}$.

<table>
<thead>
<tr>
<th>Method</th>
<th>Half-Life, $\tau_{1/2}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>($&lt;0,2$) - ($\sim 0,5$) Mev $\gamma$-$\gamma$ delayed coinc.</td>
<td>$10.8 \pm 0.8$ nanoseconds</td>
<td>(8, p. 332)</td>
</tr>
<tr>
<td>(133) - (482) Kev conversion electron- $\gamma$ delayed coincidence</td>
<td>$10.6 \pm 0.3$</td>
<td>(15, p. 259)</td>
</tr>
<tr>
<td>(133) - (345, 482) conversion electron- conversion electron delayed coinc.</td>
<td>$10.0 \pm 0.8$</td>
<td>(42, p. 942)</td>
</tr>
<tr>
<td>(137) - (482) Kev K conversion electron- $\gamma$ delayed coincidence</td>
<td>$10 \pm 1$</td>
<td>(43, p. 758)</td>
</tr>
<tr>
<td>(0,13) - (hard $\gamma$) Mev conversion electron- $\gamma$ delayed coincidence</td>
<td>$10.6 \pm 0.3$</td>
<td>(12, p. 1151)</td>
</tr>
</tbody>
</table>

that so little data on the detailed behavior of differential angular correlations influenced by extranuclear fields are presently available, is the justification for the work to be described.
THEORY

The literature of angular correlation theory is very complete. Many rigorous and comprehensive treatments of the unperturbed angular correlation of nuclear radiations have been given; in addition, the theory of extranuclear perturbations on angular correlations has been developed during recent years to a high degree of completeness. The following discussion is limited, therefore, to only those portions of angular correlation theory which are pertinent to the present work. Even so, it will be convenient to consider first the qualitative aspects of the angular correlation phenomenon without regard to extranuclear perturbations in order to provide insight into the subject and to introduce the notation. The electric quadrupole perturbation mechanism is discussed next and then the quantum-mechanical formulation of the perturbed angular correlation due to a static quadrupole interaction is presented.

Qualitative Description of the Gamma-Gamma Angular Correlation

The basic problem of all angular correlation calculations may be stated as follows: A nucleus decays from an initial state $A$ by the emission of radiation $R_1$ with direction $\vec{k}_1$ to a short-lived intermediate state $B$ which subsequently decays through the emission of a second radiation $R_2$ having direction $\vec{k}_2$. One then asks
for the relative probability \( W(\theta)d\Omega \) that \( R_2 \) be emitted in a direction \( \vec{k}_2 \) at an angle \( \theta \) with respect to \( \vec{k}_1 \), the direction of the first radiation, into the solid angle \( d\Omega \). The probability \( W(\theta) \) depends in general on the properties of the levels and radiations involved and also on the presence of extranuclear fields.

A consideration of the general features of an experiment designed to measure the angular correlation between successive gamma-rays is useful in providing a qualitative picture of the effect and a basis upon which the question as to why a correlation exists may be discussed. Figure 2 displays the essential ideas involved. The source consists of a large number of randomly oriented nuclei undergoing the transitions shown. It is assumed for the present that the lifetime \( \tau_n \) of the intermediate state is small as compared to the interaction time \( \hbar/\Delta E \) associated with the coupling of the nuclear moments to extranuclear fields so that no reorientation of the nuclear spin occurs during the interval between the formation and decay of level B. The basic apparatus consists of a fixed and a movable counter for the detection of \( \gamma_1 \) and \( \gamma_2' \), respectively, and a coincidence circuit to select \( \gamma_1 - \gamma_2 \) coincidence pairs from common nuclei. \( W(\theta) \) is to be determined by determining the true coincidence counting rate as a function of the angle \( \theta \).

Consider first the single transition \( A \rightarrow B \) via \( \gamma_1 \). Although the probability for emission of a gamma-ray photon depends in
Figure 2. Example of a directional correlation measurement.
general on the angle between the nuclear spin axis and the angle of emission, the random distribution of nuclear spins results in randomly directed propagation vectors so that the total $\gamma_1$ (and $\gamma_2$) radiations are isotropic. Another way of describing the situation is to note that at room temperature and in the absence of strong magnetic fields the Boltzmann factor is approximately unity. Hence, all magnetic substates of a particular level are equally populated and there exists no preferred direction in space for the nuclear spin. In the notation of quantum mechanics, the first transition takes place between states characterized by the quantum numbers $I_a, m_a$ and $I_b, m_b$ with the radiations having angular momenta $L$ and projection quantum numbers $M = m_a - m_b$. One cannot, with present scintillation techniques, resolve the closely spaced energy components distinguished by different $M$ values, and in practice a summation over all transitions from the $m_a$ substates of $A$ to the $m_b$ substates of $B$ is observed. Thus, in spite of the fact that the individual components of $\gamma_1$ are anisotropic, the inability to resolve them results in an observable radiation that represents an average over all components and is in general isotropic. However, this would not be the case if the radiations from a non-uniform population of substates were observed, and it is just this condition which is fulfilled by the coincidence requirement.

Consider now the two-step process in which the nucleus decays by the emission of $\gamma_1$ with $k_1$ directed along the axis of the fixed...
counter followed by the emission of $\gamma_2$ at some angle $\theta$ with respect to this axis. If the axis of the fixed counter be designated as the axis of quantization with respect to which the projection quantum numbers are defined, then the requirement that $\mathbf{k}_1$ be along this axis selects out, in effect, only those components of $\gamma_1$ for which $M = \pm 1$. This follows from the fact that in the plane wave representation of the photon the angular momentum along the propagation direction is that due to the intrinsic spin of the photon only. Thus, transitions characterized by $M = 0$ and $M > |1|$ are not observed, and the $\gamma_2$ radiations in coincidence with this selected set of $\gamma_1$ radiations emanate from a level $B$ whose substates are, in effect, unequally populated. A summation over all the substates of $B$ is no longer observed, and the radiation pattern of $\gamma_2$ with respect to the $\mathbf{k}_1$ direction is in general not isotropic.

Static Electric Quadrupole Interaction

Before turning to the description of the influence of a static quadrupole interaction on the angular correlation process, some introductory remarks concerning the nature of the electric interactions between a nucleus and its extranuclear environment are in order. The general problem is to compute the interaction energy of the nucleus with the charges constituting its environment. More specifically, we are concerned with that part of the electrostatic
interaction energy which depends on the orientation of the nucleus and thus contributes to the spin-dependent effects involved in angular correlations.

The Hamiltonian for the interaction of the nuclear charge with the static charge distribution of its surroundings, e.g., with the charged ions in a crystal lattice, is (33, p. 262)

\[
H_{el} = \sum_{p,c} \frac{e_p e_c}{|\vec{r}_p - \vec{r}_c|}
\]  

(1)

where \(\vec{r}_p\) is the position vector of the point charges \(e_p\) in the nucleus and \(\vec{r}_c\) denotes the position vector of all charges other than those of the nucleus under consideration (see Figure 3).

Figure 3. The definition of the position vectors of the nuclear and extranuclear charges.
Expanding $1/|\vec{r}_p - \vec{r}_c|$ in a Taylor's series about the nuclear center of mass yields the well-known expression for the interaction energy in terms of a series of electric multipoles, i.e.,

$$H_{el} = \sum_{\ell=0}^{\infty} \sum_{p,c} \frac{r_p^\ell e_p e_c}{r_c^{\ell+1}} P_\ell \cos (\vec{r}_p, \vec{r}_c). \quad (2)$$

By applying the spherical harmonic addition theorem to (2), the interaction Hamiltonian can be expressed as a sum of products of two factors, a nuclear factor and an external field factor, that is,

$$H_{el} = \sum_{\ell=0}^{\infty} \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} (-1)^m \left\{ \sum_p e_p^\ell r_p Y_{\ell m}(\theta_p, \phi_p) \right\} \left\{ \sum_c \frac{e_c}{r_c^{\ell+1}} Y_{\ell, -m}(\theta_c, \phi_c) \right\}. \quad (3)$$

Introducing the definitions:

$$T^{(\ell)}_m = \sum_p e_p^\ell r_p Y_{\ell m}(\theta_p, \phi_p),$$

$$V^{(\ell)}_m = \sum_c \frac{e_c}{r_c^{\ell+1}} Y_{\ell, -m}(\theta_c, \phi_c),$$

$$T^{(\ell)}_m \cdot V^{(\ell)}_m = \sum_{m=-\ell}^{\ell} (-1)^m T^{(\ell)}_m V^{(\ell)}_{-m}$$

where $T^{(\ell)}_m$ and $V^{(\ell)}_m$ are the tensor operators of the nuclear moments and the extranuclear field, respectively, and $T^{(\ell)}_m \cdot V^{(\ell)}_m$ is the scalar product ($\ell$ is the rank of the operator as well as the
multipole order permits $H_{el}$ to be written more compactly as:

$$H_{el} = \sum_{\ell=0}^{\infty} \frac{4\pi}{2\ell + 1} T^{(\ell)} \cdot V^{(\ell)}.$$  \hspace{1cm} (4)

Written in greater detail, (4) becomes

$$H_{el} = 4\pi \left( T_0^0 \cdot V_0^0 + \frac{1}{3} \sum_{m=-1}^{1} (-1)^m T_m^{(1)} V_{-m}^{(1)} \right)$$

$$+ \frac{1}{5} \sum_{m=-2}^{2} (-1)^m T_m^{(2)} V_{-m}^{(2)} + \frac{1}{7} \sum_{m=-3}^{3} (-1)^m T_m^{(3)} V_{-m}^{(3)} + \ldots \right)$$  \hspace{1cm} (5)

The first term in (5) represents the electrostatic energy of a point nucleus and is independent of the size, shape, or orientation of the nucleus and is of no significance here. The second term also plays no role here since it can be shown from parity considerations that nuclei possess zero electric dipole moments, in fact, all odd-order electric moments vanish (11, p. 328). Thus, the interaction Hamiltonian pertinent to spin-orientation effects reduces to

$$H_{el} = \frac{4\pi}{5} \sum_{m=-2}^{2} (-1)^m T_m^{(2)} V_{-m}^{(2)} + \text{higher order terms.}$$  \hspace{1cm} (6)

It is interesting to consider the order of magnitude of the successive non-zero terms in the expansion (5). For a nucleus of
atomic number $Z$, the first term is $Z e V_o$ where $V_o$ is the ordinary Coulomb potential. Here $V_o \sim e_p / r_c$ where $r_c$ is a typical atomic dimension, say $10^{-8}$ cm. The first term in (5) is then roughly $Ze^2/r_p \sim 10^5$ cm$^{-1}$, corresponding to an ultraviolet frequency. The quadrupole term ($\ell = 2$) is of the order

$$(Ze^2/r_p)r_p^2/r_c^2$$

where $r_p$ is a typical nuclear dimension, say $10^{-12}$ cm for a heavy nucleus. The quadrupole interaction energy is then of the order $10^{-8}$ times the electrostatic energy of the point nucleus, that is, of the order $10^{-3}$ cm$^{-1}$ or an interaction frequency of roughly 30 Mc/sec. The next non-zero term is about $10^{-8}$ of the quadrupole term and is ordinarily outside of the range of detection. Therefore the electric interaction Hamiltonian of interest here reduces to a single term (hereafter called $H_Q$).

We now turn to the problem of finding the matrix elements of the quadrupole interaction Hamiltonian. In the $m_z$-representation these are

$$<\text{Im}z|H_Q|\text{Im}z^\prime> = <\text{Im}z|\frac{4\pi}{5} \sum_{m=-2}^{2} (-1)^m T_m^{(2)} V_m^{(2)}|\text{Im}z^\prime>.$$  \hspace{1cm} (7)

In order to evaluate (7) we need to know $V_m^{(2)}$ and $T_m^{(2)}$ in the $m_z$-representation. Now the spherical components of the electric field tensor $V^{(2)}$ in an arbitrary coordinate system are given by
\[ V^{(2)}_m = \sum_c \frac{e_c}{(r_c^i)^3} Y_{2,m}(\theta^i_c, \phi^i_c). \]  

The components \( V^{(2)}_m \) can be expressed in terms of the Cartesian gradients \( V^{(2)}_{x^i x^i}, V^{(2)}_{y^i y^i}, \ldots \) in a straightforward way. For example, for \( m = 0 \), one has

\[
V^{(2)}_0 = \sum_c e_c \left. Y_{2,0}(\theta^i_c, \phi^i_c) \right|_{m = 0}
\]

\[
= \sum_c \frac{e_c}{(r_c^i)^3} \left[ \frac{2 \ell + 1}{4\pi} \frac{(\ell - |m|)!}{(\ell + |m|)!} \right]^{1/2} P^m_\ell (\cos \theta^i_c) e^{-i m \phi^i_c} \bigg|_{m = 0}
\]

\[
= \frac{1}{4} \sqrt{\frac{5}{\pi}} \sum_c \frac{e_c}{(r_c^i)^3} (3 \cos^2 \theta - 1).
\]  

The ordinary Coulomb potential due to the extranuclear charge is

\[ V'(r^i_c) = \sum_c \frac{e_c}{r^i_c}. \]

But

\[ V'_{z^i z^i} = \frac{\partial^2 V'}{\partial z^i z^i} = \sum_c \frac{e_c}{(r^i_c)^3} (3 \cos^2 \theta - 1), \]

so that

\[ V^{(2)}_0 = \frac{1}{4} \sqrt{\frac{5}{\pi}} V'_{z^i z^i}. \]  

(10)
In like manner, it is easy to show that the \( V_{\pm 1}^{(2)} \) and \( V_{\pm 2}^{(2)} \) components are

\[
V_{\pm 1}^{(2)} = \pm \frac{1}{2} \sqrt{\frac{5}{6\pi}} \left( V'_{x'z'} \pm i V'_{y'z'} \right) \quad (11)
\]

and

\[
V_{\pm 2}^{(2)} = \frac{1}{4} \sqrt{\frac{5}{6\pi}} \left( V'_{x'x'} - V'_{y'y'} \pm 2i V'_{x'y'} \right). \quad (12)
\]

It is always possible to choose a coordinate system in which the off-diagonal elements which involve mixed derivatives of the potential \( V' \) are zero, i.e., a transformation to the principal axes system is performed. If the heretofore arbitrary \( xyz \)-system of Figure 3 be designated the principal axes system, we have for the components of \( V^{(2)} \)

\[
V_{0}^{(2)} = (1/4) \sqrt{5/\pi} \ V_{zz} \quad (13)
\]

\[
V_{\pm 1}^{(2)} = 0 \quad (14)
\]

\[
V_{\pm 2}^{(2)} = (1/4) \sqrt{5/6\pi} \ (V_{xx} - V_{yy}) = (1/4) \sqrt{5/6\pi} \ \eta V_{zz} \quad (15)
\]

where \( \eta \) is called the asymmetry parameter of the electric field gradient after Matthias, et al. (34, p. 41) and is defined as

\[
\eta = (V_{xx} - V_{yy})/V_{zz}. \quad (16)
\]

It is customary to choose the principal axes in such a way that
\[ |V_{xx}| < |V_{yy}| < |V_{zz}| \] so that \( 0 < \eta < 1 \) because of the Laplace condition: \( V_{xx} + V_{yy} + V_{zz} = 0 \). Thus the gradient tensor in the principal axes system is determined by the parameters \( V_{zz} \) and \( \eta \) only.

In the present work in which the extranuclear field due to the ions of a hafnium metal lattice possessing axial-symmetry gives rise to an axially-symmetric field gradient, the gradient tensor is simply

\[ V^{(2)} = (1/4) \sqrt{5/\pi} \ V_{zz}. \tag{17} \]

Thus, for axially-symmetric gradients, the matrix elements of the interaction Hamiltonian are from (7) and (17)

\[ <\Im_{z} |H_0| \Im_{z}^{t}> = \sqrt{\pi/5} <\Im_{z} |T_{0}^{(2)}| \Im_{z}^{t}> V_{zz}. \tag{18} \]

In writing (18) in this way (with \( V_{zz} \) treated as a constant), it is assumed that the electric field gradient can be described classically. Applying the Wigner-Eckart theorem (see Equation A18 in the appendix) yields

\[ <\Im_{z} |H_0| \Im_{z}^{t}> = \sqrt{\pi/5} (-1)^{I-m_{z}} C(I2I; m_{z}^{t} 0 m_{z}) <I||T_{0}^{(2)}||I > V_{zz}. \tag{19} \]

where \( C(\ , \ ) \) is a Clebsch-Gordon coefficient and \( <I||T_{0}^{(2)}||I > \) is the reduced matrix element of the tensor operator \( T_{0}^{(2)} \). It is significant to note that the reduced matrix element does not involve the
projection quantum numbers $m^z$ and that the conservation of angular momentum is implicit in the Clebsch-Gordon coefficient which vanishes unless the vector triple $(l2l)$ forms a triangle and $m^z = m^z$.

We introduce now the conventional definition of the electric quadrupole moment of the nucleus:

$$eQ = <II|Q_{zz}|II> = <II| \sum_p e \left( \frac{3z^2}{p^2} - \frac{r^2}{p^2} \right) |II>.$$  \hspace{1cm} (20)

That is, the quadrupole moment $Q$ as it is ordinarily defined (11, p. 320) is the expectation value, measured in units of the proton charge $e$, of $Q_{zz}$ in the state in which the component of the nuclear spin $I$ along the $z$-axis is a maximum. We can express $T_0^{(2)}$ in terms of $Q_{zz}$ in just the way in which $V_0^{(2)}$ was written in terms of $V_{zz}$ so that (20) can be rewritten as

$$eQ = 4\sqrt{\pi}/5 \ <II| T_0^{(2)}|II>.$$  \hspace{1cm} (21)

Again, using the Wigner-Eckart theorem yields

$$eQ = 4\sqrt{\pi}/5 \ C(l2l;10I) \ <I|| T^{(2)} || I>.$$  \hspace{1cm} (22)

Thus, the reduced matrix elements which appear in (19) and (22) can be eliminated to obtain for the matrix elements of $H_Q$

$$<Im_z|H_Q|Im_z^1> = \frac{eQ}{4} V_{zz} \ (-1)^{l-m^z} C(l2l;m^z_0m^z_0)/C(l2l;10I).$$  \hspace{1cm} (23)
Evaluating the Clebsch-Gordon coefficients in the equation above (39, p. 39) gives the energy eigenvalues

\[ E_{m_z} = \langle \text{Im}_{z} | H_Q | \text{Im}_{z} \rangle = \frac{eQ V_{zz}}{4} \frac{3m_z^2 - I(I + 1)}{I(2I - 1)} \]  

(24)

For future use, we introduce the angular frequency \( \omega_o \), corresponding to the smallest non-vanishing energy difference for transitions among the \( m_z \) states. For odd half-integer \( I \),

\[ \omega_o = -\frac{3eQ V_{zz}}{2\hbar I(2I - 1)} \]  

(25)

Also useful is the quantity

\[ \Delta \nu_Q = eQ V_{zz}/\hbar, \]  

(26)

which is frequently used to designate the strength of the quadrupole interaction and is known simply as the "quadrupole coupling".

**Perturbed Angular Correlation**

In this section, we consider how the gamma-gamma angular correlation is influenced when the interaction just described involves the quadrupole moment of the intermediate nuclear state. The gross features of the perturbed correlation can be deduced from the following semiclassical argument based on the vector model of the nucleus. This description supplements that given earlier for the unperturbed
correlation and provides a useful introduction to the quantum-mechanical formulation of the problem.

Consider the situation depicted in Figure 4 which illustrates the precession of a quadrupole nucleus in a non-uniform electric field.

Figure 4. Precession of a quadrupole nucleus in a non-uniform electric field

The classical expression for the energy of alignment is obtained from (24) through the application of the Correspondence Principle. That is,

$$E_Q = \lim_{I \to \infty} \left[ \frac{eQ V_{zz}}{4} \frac{3m_z^2 - I(I+1)}{I(2I-1)} \right] = \frac{eQ V_{zz}}{8} (3 \cos^2 \theta - 1).$$

The interaction between $Q$ and $V_{zz}$ results in a torque on the nucleus of magnitude

$$m_z = I \cos \theta$$
\[ C = -\frac{dE_Q}{d\theta} = \frac{3eQ V_{zz}}{4} \cos \theta \sin \theta \]

which causes a precession of the nuclear gyroscope about the z-axis. Since \( \dot{C} = \frac{d\vec{P}}{dt} \), where \( \vec{P} \) is the angular momentum of the gyroscope, the precession frequency \( \omega \) is given by

\[ \omega = \frac{dP}{dt} \frac{1}{P \sin \theta} = \frac{3eQ V_{zz}}{4P} \cos \theta. \quad (27) \]

It is seen from (27) that the classical precession frequency depends on the orientation of \( \vec{P} \) with respect to the z-axis. This is reflected in the quantum-mechanical case by the non-equidistant splitting of the \( 2I+1 \) energy levels caused by the quadrupole coupling, a fact which is well known from pure quadrupole spectra (14, p. 113). The classical precession frequencies are related through the Correspondence Principle to the interaction frequencies associated with the transitions between the magnetic sublevels of a nuclear state of particular I, where the interaction frequencies can be computed directly from equation (24).

This simple picture of the quadrupole interaction is now applied to the intermediate state of a gamma-gamma cascade in order to provide a qualitative description of the perturbed angular correlation. Suppose that the lifetime of the intermediate state is of the same order of magnitude as the quadrupole precession period and that each of these quantities is large compared to the resolving time of
the coincidence apparatus so that the correlation can be studied as a function of time between the emission of the first and second radia-
tions. We consider first the case in which the z-axis, the axis of symmetry of the field gradient, coincides with the quantization axis, the direction of emission $\vec{k}_1$ of the first gamma-ray $\gamma_1$. In this case, the projection quantum numbers $m_b(z)$ and $m_b(\vec{k}_1)$ are the same. Since the $m_b(z)$ population is not affected by the precession about z, the $m_b(\vec{k}_1)$ population is also unchanged and the angular correlation is unperturbed. Thus, an experiment in which the decaying nuclei occupy lattice positions in an axially-symmetric single crystal whose symmetry axis coincides with the direction $\vec{k}_1$ yields the unperturbed correlation, independent of the time-delay between the emission of the first and second gamma-rays of the cascade. If, however, the z-axis does not coincide with $\vec{k}_1$, the population of the $m_b(\vec{k}_1)$ states changes due to the continuous periodic motion of the angular momentum vector about the z-axis. Since it is the $m_b(\vec{k}_1)$ population that is relevant in the angular distribution of the second gamma-ray, the presence of the interaction alters the correlation. However, if only those events in which the second gamma-ray follows the first by exactly one precession period are recorded, the correla-
tion is restored. Thus, the time-differential angular correlation under the influence of a static quadrupole interaction is periodic in time. A study of the differential correlation then permits the
determination of the precession period and, through (25), the determination of the quadrupole coupling.

The analogous quantum-mechanical problem has been treated by Alder (7, p. 761-784) and by Abragam and Pound (1, p. 943-962). In a recent review article, Steffen and Frauenfelder (46, p. 3-89) present the general problem of extranuclear perturbations on angular correlations in a formulation which is readily adaptable to a variety of perturbation mechanisms and which contains the unperturbed correlation as a special case. In what follows, we make use of their formulation for the case of interest in the present work. This case is characterized by the following assumptions: 1) the source consists of a statistically significant number of randomly oriented microcrystals of axial symmetry in which the radioactive nuclei occupy regular lattice sites, 2) the crystalline field is static in the sense that the field gradient does not fluctuate with time but has a constant value from nucleus to nucleus, and 3) the field gradient acting on the nuclear spin, but not the nuclear spin itself, can be described classically. In order to maintain continuity in the succeeding development, much of the mathematical detail has been relegated to the appendix or, in cases where adequate discussion appears elsewhere, omitted entirely. In particular, a fairly complete discussion of the theory of unperturbed angular correlation is given in the appendix.
In the following, we consider the successive emission of $\gamma_1$ and $\gamma_2$ in the directions $\vec{k}_1$ and $\vec{k}_2$, respectively, as a nucleus decays through the states $A \rightarrow B \rightarrow C$ (see Figure 2). The state function describing a particular state is denoted by $|I \text{m} \rangle$ where $I$ and $m$ with appropriate subscripts denote the angular momentum and projection quantum numbers characterizing the state. The problem to be considered is that of finding $W(\theta, t) = W(\vec{k}_1, \vec{k}_2; t)$, the probability that $\gamma_2$ emitted at a time $t$ after $\gamma_1$ be emitted at an angle $\theta$ with respect to the direction of $\gamma_1$. We write first the well-known expression for the unperturbed correlation (equation A8 in the appendix, which is the usual starting point for calculations of explicit correlation functions, e.g., (9, p. 732) and (17, p. 324).

$$W(\vec{k}_1, \vec{k}_2) = S \sum_{m_a, m_c} \sum_{m_b} | \sum_i <I_{mc} | H_2 | I_{mb} > | <I_{mb} | H_1 | I_{ma} > |^2, \quad (28)$$

where $H_1$ is the interaction Hamiltonian for the emission of $\gamma_1$ in the direction $\vec{k}_1$ and $H_2$ is the analog for $\gamma_2$ in the direction $\vec{k}_2$. $S$ denotes an average over the unobserved quantities associated with the radiations, e.g., when only the directional correlation is observed, the average is taken over all quantities except $\vec{k}_1$ and $\vec{k}_2$. The matrix elements $<I_{i} \text{m}_i | H_{ij} | I_{j} \text{m}_j >$ are probability amplitudes for the various possible transitions between the degenerate substates.

Expanding (28) gives
\[ W(\vec{k}_1, \vec{k}_2) = \sum_{m_a, m_c} \sum_{m_b, m_b'} < I_{m_c} | H_2 | I_{m_b} > \langle I_{m_c} | H_2 | I_{m_b'} >^* \]

\[ \times \langle I_{m_b} | H_1 | I_{m_a} > \langle I_{m_b'} | H_1 | I_{m_a} >^* \] (29)

where \(< >^*\) denotes the complex conjugate. If transitions between the \(m_b\) substates occur due to the interaction of the nuclear quadrupole moment \(Q\) with the extranuclear field gradient during the time \(t\) (\(t = 0\) is defined by the emission of the first radiation), the state functions \(|I_{m_b} >\) undergo the evolutions

\[ |I_{m_b} > \xrightarrow{\text{interaction}} |I_{m_b}^* > . \]

Thus in the perturbed correlation, the final states \(|I_{m_b} >\) and \(|I_{m_b'} >\) for emission of the first radiation are no longer the initial states of the second radiation as is the case when the perturbation is absent, e.g., when \(W\) is given by (28). The change in the state functions can be represented by introducing the unitary operator \(\Gamma(t)\) that describes the evolution of the \(|I_{m_b} >\), and the perturbed correlation can be written as
\[
W(k_1, k_2; t) = \sum_{m_a, m_c} \sum_{m_b, m'_b} \langle I c m_c | H_2 \Gamma | I_b m_b \rangle \langle I c m_c | H_2 \Gamma | I_b m'_b \rangle^* \\
\times \langle I_b m_b | H_1 | I m_a \rangle \langle I_b m_b | H_1 | I m_a \rangle^*.
\]

The states \( |I_b m_b^n> \) form a complete set and the state vector \( \Gamma(t) |I_b m_b> \) can be represented as a superposition of the functions \( |I_b m_b> \) with different projection quantum numbers \( m_b^n \) but with the same total angular momentum \( I_b \). Thus, we have

\[
\Gamma(t) |I_b m_b> = \sum_{m_b^n} \langle I_b m_b^n | \Gamma | I_b m_b > | I_b m_b^n >
\]

and a similar expression for \( \Gamma(t) |I_b m_b^i> \). The expansion coefficients are the matrix elements \( \langle I_b m_b^n | \Gamma | I_b m_b > \) of the time evolution operator \( \Gamma(t) \) in the \( |I_b m_b^n> \) representation.

The expression for the perturbed angular correlation can now be written in the form

\[
W(k_1, k_2; t) = \sum_{m_a, m_c} \sum_{m_b, m_b^i, m_b^m} \sum_{m_b^i, m_b^m} \langle I c m_c | H_2 \Gamma | I_b m_b^n> \langle I_b m_b^i | \Gamma | I_b m_b^m > \langle I_b m_b^m | I m_a \rangle \langle I_b m_b^i | I m_a \rangle^* \\
\times \langle I_b m_b | H_1 | I m_a > \langle I_b m_b | H_1 | I m_a \rangle^*.
\]

We now replace the matrix elements for the emission of the \( k \)-th
gamma-ray, i.e., the $\langle I_i m_i | H_k | J_j m_j \rangle$, by the expressions (A10) and (A11) derived in the appendix and perform the sums over $m_a$ and $m_c$. From (A28) we have

$$\Omega^1_{mb m_b} = \sum_{m_a} <I_b m_b | H_1 | I_a m_a > <I_b m_b | H_1 | I_a m_a >^*$$

$$= \sum_{\nu_1} (-1)^{m_b} C(I_b I_b \nu_1 ; m_b, -m_b', s_1) D^{1}_{m_b - m_b'} 0 (a_1 b_1 o) B (1) \nu_1$$

and

$$\Omega^2_{mb m_b} = \sum_{m_c} <I_c m_c | H_2 | I_b m_b > <I_c m_c | H_2 | I_b m_b >^*$$

$$= \sum_{\nu_2} (-1)^{m_b} C(I_b I_b \nu_2 ; m_b, -m_b', s_2) D^{2}_{m_b - m_b'} 0 (a_2 b_2 o) B (2) \nu_2$$

Equations (33) and (34) differ from the corresponding expressions in the appendix in that $m_b$ and $m'_b$ in the latter are replaced by $m''_b$ and $m'''_b$ in the former to account for the changes induced in the magnetic substates of the intermediate state by the extranuclear perturbation. In order to aid in the identification of the factors appearing in the expressions above, Figure A1 is redrawn here. In Figure 5, $\vec{k}_1$ and $\vec{k}_2$ are the propagation vectors of the first and second gamma-rays, respectively, and $\hat{n}$ is the direction of the arbitrarily chosen quantization axis. The rotation matrices
Figure 5. The angular coordinates of the propagation directions \( \vec{k}_1 \) and \( \vec{k}_2 \) of \( \gamma_1 \) and \( \gamma_2 \), respectively.

\[ D(\alpha \beta 0) \] carry the coordinate systems of the radiations into the coordinate system of quantization, i.e., \( \vec{k}_1 \) and \( \vec{k}_2 \) into \( \hat{n} \).

\[ C(I_b^m b^1 v; m_b', -m_b^*, s) \] is a Clebsch-Gordon coefficient which vanishes unless the vector triple \( (I_b, I, v) \) forms a triangle and \( s = m_b - m_b' \).

The parameters \( B_v(1) \) contain quantities which do not depend on the projection quantum numbers and are of no immediate interest here.

A more complete discussion of equations (33) and (34) appears in the formulation given in the appendix for the unperturbed correlation; the above remarks should suffice, however, for the further reduction of (32).

Making the substitutions (33) and (34) in (32) gives
\[ W(\vec{k}_1, \vec{k}_2; t) = \sum_{m_b} \sum_{m_b'} \sum_{m_b''} (-1)^{m_b+m_b'-2I_b} C(I_b I_b \nu_1; m_b', -m_b', s_1) \]
\[ \times C(I_b I_b \nu_2; m_b'' - m_b', s_2) \]
\[ D(\nu_1, \nu_2) D(a_1 \beta_1 0, \nu_1) D(a_2 \beta_2 0, \nu_2) \]
\[ x B(1) B(2) <I_b m_b^n | \Gamma | I_b m_b^m> <I_b m_b^n | \Gamma | I_b m_b^m>^* \] (35)

Replacing \( m_b - m_b' \) by \( s_1 \) and \( m_b'' - m_b' \) by \( s_2 \) and introducing the perturbation factor

\[ s_1 s_2 \]
\[ G(t) = \sum_{m_b, m_b'} (-1)^{m_b+m_b'-2I_b} C(I_b I_b \nu_1; m_b', -m_b', s_1) \]
\[ \times C(I_b I_b \nu_2; m_b'' - m_b', s_2) <I_b m_b^n | \Gamma | I_b m_b^m> \]
\[ \times <I_b m_b^n | \Gamma | I_b m_b^m> \] (36)

yields

\[ W(\vec{k}_1, \vec{k}_2; t) = \sum_{\nu_1, \nu_2} \sum_{s_1, s_2} (-1)^{s_1 s_2} \]
\[ G(t) D(\nu_1, \nu_2) D(a_1 \beta_1 0) \]
\[ D(a_2 \beta_2 0) B(1) B(2). \] (37)

We now proceed to calculate the explicit form taken by the
general perturbation factor (36) for an experiment in which the source consists of a randomly oriented array of microcrystals in which the axially-symmetric electric field gradient interacts with the nuclear quadrupole moment to produce the transitions $|I_b m_b> \rightarrow |I_b m_b''>$ and $|I_b m_b'> \rightarrow |I_b m_b''>$. The evolution operator $\Gamma(t)$ satisfies the time-dependent Schrödinger equation

$$\partial \Gamma(t)/\partial t = (-i/\hbar)H_Q \Gamma(t)$$

(38)

in which $H_Q$ is the quadrupole interaction Hamiltonian discussed previously. For a static field gradient, $H_Q$ does not depend on time, and a solution of (38) is

$$\Gamma(t) = \exp \left[ -(i/\hbar)H_Q t \right].$$

(39)

Designating the unitary operator which diagonalizes the Hamiltonian by $U$, the eigenvalue equation is

$$UH_Q U^{-1} = E$$

(40)

where $E$ is the diagonal matrix whose diagonal elements $E_{m_z}$ are the energy eigenvalues (24). By expanding the exponentials it is easily verified that

$$U e^{-(i/\hbar)H_Q t} U^{-1} = e^{-(i/\hbar)E t}$$
so that $\Gamma(t)$ may be written as

$$\Gamma(t) = U^{-1} e^{-(i/\hbar)Et} U. \quad (41)$$

The matrix elements of $\Gamma(t)$ can be expressed as follows:

$$< I_b m_b^\ast | \Gamma | I_b m_b > = < I_b m_b^\ast | U^{-1} e^{-(i/\hbar)Et} U | I_b m_b >$$

$$= \sum_{m_z} < I_b m_b^\ast | U^{-1} | I_b m_z > < I_b m_z | e^{-(i/\hbar)Et} | I_b m_z >$$

$$\times < I_b m_z | U | I_b m_b >$$

$$= \sum_{m_z} < I_b m_z | U | I_b m_b >^\ast < I_b m_z | U | I_b m_b >$$

$$\times e^{-(i/\hbar)E_m} \quad (42)$$

where the $< I_b m_z | U | I_b m_b >$ are the matrix elements of the unitary operator which diagonalizes $H_Q$. A similar expression exists for the matrix elements $< I_b m_b^\ast \Gamma | I_b m_b^i >$.

If the symmetry axis of the interaction, i.e., the axis of symmetry of the crystal, coincides with the quantization axis (the $z$-axis of Figure 5), the eigenfunctions of $H_Q$ will be the $| I_b m_z >$, and $H_Q$ and $\Gamma(t)$ will both be diagonal in this representation. That is $U = 1$ and the matrix elements of $\Gamma(t)$ become
Equations (43) would apply, for example, when the source consists of a single crystal oriented with the crystal axis along the z-axis. For the randomly oriented microcrystals of a polycrystalline source, the matrix elements of \( \Gamma(t) \) are written for a single microcrystal of arbitrary orientation and the attenuation factor is obtained by averaging over the random directions of the symmetry axes of the ensemble.

Consider a microcrystal whose axis of symmetry \( z' \) is defined by the Euler angles \( \alpha \) and \( \beta \) with respect to the \( xyz \)-coordinates (see Figure 6). The interaction Hamiltonian \( H_Q(z') \) is diagonal in

![Figure 6. Position of \( z' \)-axis defined by the Euler angles \( \alpha \) and \( \beta \).](image-url)
the \( z' \)-system with eigenvalues \( E_{m_z} \), given by (24). The Hamiltonian in the \( z \)-system is obtained by performing the rotation:

\[
D(\alpha \beta 0) H_Q(z) D^{-1}(\alpha \beta 0) = H_Q(z').
\]  

(44)

Now \( H_Q(z') \) being diagonal, the unitary transformation \( D(\alpha \beta 0) \) diagonalizes \( H_Q(z) \), and the matrix elements of \( \Gamma(t) \) found from (42) are

\[
< I_b m^n_b | \Gamma | I_b m^\prime_n_b > = \sum_{m_z m_z'} \frac{I_b^*}{D(\alpha \beta 0)} \frac{I_b}{D(\alpha \beta 0)} e^{-i\hbar E_{m_z} t} \]

(45)

with a similar expression for \( < I_b m^n_b | \Gamma | I_b m^\prime_n_b > \). Replacing the matrix elements appearing in (36) by the results above, puts the perturbation factor in the form

\[
G_s(t) = \sum_{\nu_1 \nu_2} \sum_{m_b^\prime m_b} (-1)^{m_b^\prime + m_b} - 2I_b C(I_b \nu_1 : m_b^\prime, -m_b^\prime, s_1) \\
\times C(I_b \nu_2 : m_b^\prime, -m_b^\prime, s_2) \frac{I_b^*}{D} \frac{I_b}{D} \frac{I_b}{D} \frac{I_b}{D} \\
\times e^{-i\hbar (E_{m_z} - E_{m_z'}) t}
\]

(46)

The summations over \( m_b^\prime \) and \( m_b^\prime \) can be performed with the aid of the general contraction relations (39, p. 58)
\[
\sum_{m_b} C(I_b b_1 \nu_1; m_b, -m_b, l_s) \frac{I_b}{m_z} \frac{I_b}{m_z} = C(I_b b_1 \nu_1; m_z, -m_z, p) \frac{v_1^s}{ps_1}
\]

and

\[
\sum_{m_b} C(I_b b_2 \nu_2; m_b, -m_b, l_s) \frac{I_b^*}{m_z} \frac{I_b^*}{m_z} = C(I_b b_2 \nu_2; m_z, -m_z, p) \frac{v_2^s}{ps_2}
\]

We now average \( \frac{s_1 s_2}{v_1 v_2} \) over all orientations of the symmetry axis \( z' \) by integrating over the Euler angles \( \alpha \) and \( \beta \). Thus,

\[
G = \frac{1}{4\pi} \sum_{m_z, m_z'} C(I_b b_1 \nu_1; m_z, -m_z, p) C(I_b b_2 \nu_2; m_z, -m_z, p)
\]

\[
x e^{-i\phi(E_{m_z} - E_{m_z'})} \int_0^{2\pi} \int_0^\pi \frac{v_1^*}{ps_1} D(a\beta 0)
\]

\[
x \frac{v_2}{ps_2}
\]

Due to the orthogonality of the D-functions, the integral gives

\[
simply \frac{1}{4\pi} \left[ \frac{1}{(2\nu_1 + 1)(2\nu_2 + 1)} \right]^{1/2} \delta_{v_1^s v_2^s} \delta_{s_1 s_2}
\]

and the perturbation factor becomes

\[
\frac{s_1 s_2}{v_1 v_2} G = v_1 v_2 \frac{1}{[(2\nu_1 + 1)(2\nu_2 + 1)]^{1/2}} \sum_{m_z, m_z'} C(I_b b_1 \nu_1; m_z, -m_z, p)
\]

\[
x C(I_b b_2 \nu_2; m_z, -m_z, p) e^{-i(E_{m_z} - E_{m_z'})t/\hbar} \delta_{v_1^s v_2^s} \delta_{s_1 s_2}
\]
Substituting the above result into (37) gives the angular correlation

\[ W(k_1, k_2; t) = \sum_{\nu_1, \nu_2, s_1, s_2} (-1)^{s_2} \frac{\nu_1}{s_1} D (\alpha_1 \beta_1, 0) \frac{\nu_2}{s_2} D (\alpha_2 \beta_2, 0) B_{\nu_1} (1) B_{\nu_2} (2) \]

\[ \times \frac{1}{[(2\nu_1 + 1)(2\nu_2 + 1)]^{1/2}} \sum_{m_z, m_z'} C(I_b \nu_1; m_z, -m_z', p) \]

\[ \times C(I_b \nu_2; m_z, -m_z', p) e^{-i(E_m - E_m') t / \hbar} \delta_{\nu_1 \nu_2} \delta_{s_1 s_2}^{s_1 s_2}. (51) \]

The summations over \( s_1, s_2, \nu_1 \) and \( \nu_2 \) can now be made. Performing the sums and dropping the subscripts on the indices gives

\[ W(k_1, k_2; t) = \sum_{\nu, s} (-1)^s D \nu (\alpha_1 \beta_1, 0) D \nu (\alpha_2 \beta_2, 0) B_{\nu} (1) B_{\nu} (2) \]

\[ \times \frac{1}{2\nu + 1} \sum_{m_z, m_z'} [C(I_b \nu; m_z, -m_z', p)]^2 e^{-i(E_m - E_m') t / \hbar}. (52) \]

A comparison of (52) with (A30) through (A32) of the appendix shows that

\[ \sum_{\nu, s} (-1)^s D \nu (\alpha_1 \beta_1, 0) D \nu (\alpha_2 \beta_2, 0) B_{\nu} (1) B_{\nu} (2) = \sum_{\nu} A_{\nu} P_{\nu} (\cos \theta) \]

where the right side of (53) is just the unperturbed angular correlation. The perturbed correlation is simply
\[ W(k_1, k_2; t) = W(\theta, t) = \sum_\nu G_\nu(t) A_\nu P(\cos \theta) \]  

(54)

with the \( G_\nu(t) \) given by

\[ G_\nu(t) = \frac{1}{2\nu + 1} \sum_{m_z, m'_z} \left[ C(I_b I_b; m_z, -m'_z, p) \right]^2 e^{-i(E_{m_z} - E_{m'_z})t/\hbar}. \]  

(55)

All that remains is to evaluate the \( G_\nu(t) \) for the intermediate state of the gamma-gamma cascade observed in the experiment described in this paper. For the 482 kilovolt level in Ta\(^{181}\), \( I_b = 5/2 \) and, consequently, the \( m_z \) and \( m'_z \) take on the values \( \pm 1/2, \pm 3/2 \) and \( \pm 5/2 \) only. Equation (55) can be written in the form

\[ G_\nu(t) = \frac{1}{2\nu + 1} \sum_{m_z, m'_z} \left[ C(5 5; 2 \nu; m_z, -m'_z, p) \right]^2 \{ \cos[(E_{m_z} - E_{m'_z})t/\hbar] \} \]

\[- i \sin[(E_{m_z} - E_{m'_z})t/\hbar]\}. \]  

(56)

From (24) we note that \( E_{m_z} = E_{-m_z} \). This relation plus the fact that \( [C(I_b I_b; m_z, -m'_z, p)]^2 = [C(I_b I_b; m'_z, -m_z, p)]^2 \) leads to the disappearance of the imaginary terms in the sum and (56) becomes

\[ G_\nu(t) = \frac{1}{2\nu + 1} \sum_{m_z, m'_z} \left[ C(5 5; 2 \nu; m_z, -m'_z, p) \right]^2 \cos[(E_{m_z} - E_{m'_z})t/\hbar]. \]  

(57)

The \( G_\nu(t) \) can be obtained for particular \( \nu \) by evaluating the
Clebsch-Gordon coefficient in (57) and performing the sums. For \(\nu = 0, 2, 4\) we obtain

\[
G_0(t) = 1
\]

\[
G_2(t) = \frac{1}{5} \left[ 1 + \frac{13}{7} \cos \omega_0 t + \frac{10}{7} \cos 2\omega_0 t + \frac{5}{7} \cos 3\omega_0 t \right]
\]

\[
G_4(t) = \frac{1}{9} \left[ 1 + \frac{15}{7} \cos \omega_0 t + \frac{18}{7} \cos 2\omega_0 t + \frac{23}{7} \cos 3\omega_0 t \right]
\]

(58)

where we have used (24) and (25) to write

\[
(E_{m_z} - E_{m'_z})t/\hbar = (m_z^2 - m'_z^2)\omega_0 /2, \quad \{m_z, m'_z\} = \pm \frac{1}{2}, \pm \frac{3}{2}, \pm \frac{5}{2}.
\]

The periodic behavior of the perturbed angular correlation predicted by the semiclassical argument given earlier is clearly evident from the form of the attenuation factors. This is illustrated by the behavior of \(G_2(t)\) shown in Figure 7.

In the appendix it is shown that the index \(\nu\) takes on only even integer values and that \(\nu_{\text{max}} = 4\) for the cascade under consideration here. Thus, (58) includes all the attenuation coefficients of interest.

We conclude by remarking that the attenuation coefficients contain all of the information on the perturbation of the angular correlation. They depend only on the properties of the intermediate state and are completely independent of the properties of the
Figure 7. The attenuation coefficient $G_2(t)$ for $I = 5/2$ as a function of $\omega_0 t/\pi$.

radiations whose directional correlation is observed. The perturbed angular correlation as a function of time between emission of the first and second radiation is given by the unperturbed correlation function multiplied by the time-dependent attenuation factors characterizing the perturbation.
APPARATUS

The basic apparatus used in this investigation consists of a two-counter scintillation spectrometer designed to permit the study of both the directional and time relations between the members of the 133-482 kilovolt gamma-cascade in Ta$^{181}$. In order to do this, the system must provide the following information about the pair of gamma-rays whose members are detected separately in each counter: 1) the angle between their propagation vectors, 2) their genetic relation, i.e., whether or not they emanate from a common nucleus, 3) their time of emission relative to each other, and 4) their energies, which identify the components of the cascade in a general gamma-ray background field.

Without regard to the operational details of the apparatus shown in the block diagram of Figure 8, this information is acquired as follows: corresponding to the detection of a gamma-ray $\gamma_1$ by counter 1 followed (or preceded) at an arbitrary time by the detection of a second gamma-ray $\gamma_2$ by counter 2, there appears at the output of each counter a pair of voltage pulses designated "fast" and "slow". The amplitudes of the slow pulses are proportional to the energies of the corresponding gamma-rays and provide a basis for energy selection while the fast pulses are specially shaped to provide information regarding their time relation. After shaping, the fast
Figure 8. Block diagram of the delayed-coincidence spectrometer.
pulses are sent to a time-to-pulse-height converter circuit which produces an output pulse whenever the members of the fast pulse pair arrive within some predetermined time interval with respect to one another (200 nanoseconds in this experiment). Moreover, the amplitude of the output pulse, if any, is proportional to the difference in arrival times of the fast pulses. In this way, the genetic relation of the two radiations is established and their time relation as well. After amplification, the slow pulses are fed to separate single-channel pulse-height analyzers which either reject or accept the events as being of the proper energy through the analysis of their respective amplitudes. If a slow pulse corresponds to a gamma-ray of the desired energy, a pulse appears at the output of the single-channel analyzer. A 2 microsecond coincidence circuit is fed by the single-channel analyzers and is activated only by the simultaneous appearance of pulses at each of its inputs. Meanwhile, the output pulse of the time-to-pulse-height converter, which has been stored in a delay line of appropriate length to await the processing of the information in the slow channels, arrives at the input of the multichannel pulse-height analyzer. If condition 4) has been met by both slow pulses, the 2 microsecond coincidence provides a gate pulse which activates the multichannel analyzer enabling it to analyze and record the height of the converter pulse. Prior calibration of the time-to-pulse-height converter permits subsequent
reduction of the data stored in the memory of the multichannel analyzer back to a time scale.

The detectors are 1 by 1.5 inch NaI thallium activated crystals optically coupled to type RCA 6810A photomultiplier tubes. The crystal-photomultiplier combination was coaxially positioned inside a high permeability steel cylinder which acted as a mechanical mount as well as a magnetic shield to reduce the effects of external fields on the operation of the tube. The photomultipliers were biased from separate 2500 volt regulated supplies capable of furnishing 15 milliamperes to the dividers from which the individual dynode voltages are obtained. In order to prevent gain fluctuations from lowered bias voltages due to large dynode current drains, the last six stages of the dynode structure are decoupled from the divider through cathode followers. The lower stages draw very small currents so that direct connection to the divider is permissible here. The high voltage distribution system was built from a University of California Radiation Laboratory design drawing No. LE 4437.

The preamplifiers contain a fast limiter circuit fed from the photomultiplier anode and a slow inverter section whose input is taken from the ninth dynode. The anode pulse appearing at the grid of the limiter cuts off the standing plate current of 18 milliamperes in a Western Electric 404-A sharp cutoff pentode yielding an output pulse of 2 volts amplitude, 2 to 4 nanoseconds rise, and several
hundred nanoseconds duration. This pulse is delay-line clipped with a 125 ohm shorted delay cable of such length as to produce a square pulse of 200 nanoseconds duration which drives one input of the time-to pulse-height converter. The output of the inverter is a negative pulse of a few millivolts amplitude, one microsecond rise, and 10 microseconds duration. This pulse is sent to the appropriate slow channel for energy selection through pulse-height analysis.

The time-to-pulse-height converter was built by the author and is a slightly modified form of the converter described in detail by Green and Bell (25, p. 127-132). The circuit used in this work is shown in Figure 9. A 6BN6 gated pentode biased as shown is the active element of the circuit. Fast pulses from the limiters are applied to grids 1 and 3 which are both biased off in the quiescent state. Furthermore, the biasing conditions are such as to prevent the flow of plate current except during the overlap time of the pulses present on both these gate grids. The combination of $R_1$, $R_2$, $C_1$, and $C_2$ in the plate circuit acts as an integrator to produce an output pulse whose amplitude is proportional to the overlap time of the fast pulses. It is clear that the response of this circuit is double-valued in the sense that there are two overlap conditions that produce the same output pulse size. Green and Bell used a supervisory circuit to indicate which pulse arrived first; in the present application, this information is provided by the slow channels which are not
ambivalent in energy, and the supervisory circuit is not needed.

The remainder of the electronic apparatus is conventional. Baird-Atomic Model 215 non-overloading linear amplifiers and Atomic Instrument Model 510 single-channel pulse-height analyzers are used in the slow channels. A Borg-Warner DZ4 coincidence unit is used as the slow coincidence analyzer to provide gating of the Nuclear Data 512-channel pulse-height analyzer.

The spectrometer table was constructed from a radar antenna mount. The detectors are mounted with their axes in a horizontal plane, one detector being fixed and the other capable of rotation.
about a vertical axis through the source at the center of the spectrometer. Adjustments are provided so that the source and the central axes of the detectors can be brought to lie in a horizontal plane throughout a complete rotation of the movable counter. The angular position of the movable counter with respect to the fixed counter provides the last piece of information specified above and is specified within ±0.25 degrees.
TEST AND CALIBRATION OF THE SPECTROMETER

Operational checks of the time-to-pulse-height converter section of the spectrometer included studies of the resolution time, linearity, and time-pulse-height calibration. These studies were made using the 510 kilovolt annihilation photon pairs produced in the decay of the positron emitter Na$^{22}$ as a source of coherent nuclear events. Artificial delay between the members of these prompt pairs was introduced by the insertion of delay cables between the converter and either limiter. The time resolution was obtained by scanning the output of the converter at a fixed value of delay while time calibration and linearity checks were made by repeating this procedure for various delay conditions. The overall response of the system is illustrated in Figure 10. Resolution curves obtained under two different biasing conditions of the single-channel analyzers are presented in Figure 11. Curve (a) was obtained with the single-channel analyzers biased to accept only slow pulses in a limited region about the 510 kilovolt photopeaks which correspond to events in which the total energy of the annihilation photon is lost in the scintillator. The bias conditions under which (b) was obtained were such that events corresponding to losses in the scintillators of roughly 133 and 482 kilovolts, respectively, were recorded. The latter case then closely approximates the situation encountered when the 133-482 kilovolt
Figure 10. Overall response of the time-to-pulse-height converter showing the linear relation between the difference in arrival time of the fast pulses at the converter and the output pulse height.
Figure 11. Prompt coincidence curves showing the time resolution of the spectrometer. Curve (a) is obtained with the single-channel analyzers biased to accept only the 510-Kev photopeaks while (b) is obtained with the analyzers accepting events corresponding to 133-482 Kev pairs.
cascade in $^{181}\text{Ta}$ is observed. The full-width at half-maximum of the prompt curve is a measure of the resolution and is denoted by $2T$ where $T$ is sometimes called the "resolving time".

Single-channel analyzer settings were determined by running a singles spectrum of $^{181}\text{Ta}$ with each analyzer and adjusting the gain of the amplifiers and/or the baseline and window settings of the analyzers to accept the desired photopeak. The singles spectrum of one of the slow channels is given in Figure 12; the window settings of the analyzers are indicated on the diagram.

As discussed earlier, the output pulses of the single-channel analyzers corresponding to the detection of a 133 kilovolt photon in one detector and the detection of a 482 kilovolt photon in the other must, if the photons stem from the same source event, be in coincidence to provide satisfactory operation of the 2 microsecond coincidence unit which gates the multichannel analyzer. The timing of these pulses was checked just prior to the point where the coincidence comparison is made by comparing their arrival time on a dual trace oscilloscope triggered by one of the related fast pulses. Internal adjustment of one or both of the analyzers to advance or retard the triggering point of the baseline discriminator was made to bring the pulses into coincidence. The time relation between the gate pulse of the coincidence unit and output pulse of the converter was observed in much the same way. In this case, however, the pulses were
Figure 12. The differential-pulse-height spectrum of Ta$^{181}$. 
brought into coincidence by utilizing the built-in 2 microsecond delay in the 512-channel analyzer.

Long term stability checks were made by recording prompt curves consecutively every hour for several days. Shifts of the prompt curve on the time-pulse height axis due to drifts in either the converter, the amplifier in the multichannel analyzer, or the analyzer itself were less than $\pm 0.3$ nanosecond per 24 hour period. Amplitude variations in the prompt curves due to changes in the overall detection efficiencies of the slow and fast channels were random in nature and consistent with the expected statistical fluctuations. Reasonable care was taken to insure good stability by the usual precautions of line voltage regulation and the maintenance of a constant ambient temperature by keeping all the apparatus on for several days prior to running.

In order to check the overall performance of the apparatus, the lifetime of the 482 kilovolt state and the anisotropy of the 133-482 kilovolt cascade were measured using a liquid source known to give the unperturbed correlation. These experiments are discussed in the section on experimental measurements.
SOURCE PREPARATION

The Hf$^{181}$ activity was produced by thermal neutron irradiation of spectroscopic pure hafnium metal powder at the General Electric Company reactor facilities at Hanford, Washington. No isotopic enrichment was used so that in addition to the 46 day Hf$^{181}$ activity produced by the Hf$^{180}$ + n → Hf$^{181}$ process a 75 day Hf$^{175}$ activity by Hf$^{174}$ + n → Hf$^{175}$ was also produced. The isotopic abundances of the mass 174 and 180 isotopes are in the ratio 0.18 percent to 35.2 percent while their thermal neutron capture cross-sections are in the ratio roughly 1000 to 10 barns respectively. This results in a Hf$^{175}$ activity initially about one-half as intense as the desired Hf$^{181}$. However, the radiations from the decay of Hf$^{175}$ in no way interfere with those from Hf$^{181}$ in the present experiment due to the stringent generic requirement, and can be ignored. Prior to irradiation, the powder was screened to size between U. S. Series 140 and 200 standard mesh screens. This results in an average microcrystal size such that a physically small counting source can contain a sufficient number of particles to provide a randomly oriented array while each particle is still large enough to minimize surface effects. The powder was irradiated in an otherwise evacuated quartz ampule to produce a Hf$^{181}$ activity of about 3 milli-curies per gram of hafnium. Since finely dispersed hafnium metal
ignites readily in the atmosphere, all operations in the preparation of this material were performed in an argon environment.

Counting sources consisted of about 10 milligrams of powder in thin-walled (~1/32 inch) fluorethelyne holders; the total volume of each source was about $10^{-3}$ cm$^3$. Aside from the geometrical consideration that the counting source approximate a point, the maximum permissible source activity is governed by considerations of the true-to-accidental coincidence rate. In order to keep the chance coincidence rate small and still obtain a sufficient number of true coincidences in a reasonable time, one must choose a source of appropriate activity for the geometry used.

Consider a constant source of strength $N_0$ disintegrations per second which emits two gamma-rays in cascade through an intermediate state of lifetime $T_n$. The true coincidence rate recorded in a given channel of the multichannel analyzer depends upon the following quantities:

\[ N_0 \] = the activity of the source

\[ \omega_i \] = the solid angle subtended at the source by counter $i$

\[ E_i \] = the efficiency of slow channel $i$ for registering quantum $i$

\[ W(\theta, t) \] = the time-dependent directional correlation of the cascade

\[ t \] = the time of emission of quantum 2 with respect to $t = 0$, the time of emission of quantum 1
\[ \Delta t = \text{the time interval represented by each channel of the multichannel analyzer, i.e., the channel width.} \]

If the channels are numbered by the integers \( j = t/\Delta t = 1, 2, 3, \ldots \), the true coincidence counting rate in the \( j^{\text{th}} \) channel is given by the expression

\[
N_{\text{true}}(j\Delta t) = N_0 \omega_1 \omega_2 E_1 E_2 W(\theta, j\Delta t) e^{-j\Delta t/\tau_n} \left(1 - e^{-\Delta t/\tau_n}\right). \tag{59}
\]

The chance coincidence rate is the same for each channel and is simply

\[
N_{\text{chance}} = 2\Delta t N_1 N_2 = N_0^2 \omega_1 \omega_2 E_1 E_2 \cdot 2\Delta t \tag{60}
\]

where \( N_1 \) and \( N_2 \) are the slow singles counting rates. In writing (60) in this way, it is assumed that only converter pulses which stem from the same events that produce the slow coincidences and, hence, the gating of the multichannel analyzer are accepted by the analyzer.

That is, only one pulse is accepted per gate pulse. This is a reasonable assumption in that the analyzer, after accepting a pulse, is internally biased off until the analyzing process is completed. This requires a minimum time of 15 microseconds as compared to the 4 microsecond gate so that the assumption of one accidental pulse per gate pulse is valid. The "true-to-chance ratio" of the \( j^{\text{th}} \) channel is then

\[
\frac{N_{\text{true}}(j\Delta t)}{N_{\text{chance}}} = \frac{W(\theta, j\Delta t) e^{-j\Delta t/\tau_n} \left(1 - e^{-\Delta t/\tau_n}\right)}{2N_0 \Delta t}. \tag{61}
\]
From (61) it is seen that the lifetime of the intermediate state, the channel width, and the activity of the source all play roles in determining the true-to-chance ratio. In practice, however, $T_n$ is fixed and $\Delta t$ is dictated by the structure of $W(\theta, t)e^{-t/T_n}$, which is to be well resolved, so that $N_o$ is the only controllable variable. The sources used in the measurements described in the next section were chosen to yield true-to-chance ratios of about 80 at the peak of the delayed coincidence curve. These activities permit data to be obtained over a period representing about six half-lives of the intermediate state with true-to-chance ratios greater than unity.
EXPERIMENTAL MEASUREMENTS

Lifetime of the 482 kilovolt Excited State in Ta$^{181}$

As a check on the overall performance of the delayed coincidence portion of the apparatus, the well-established lifetime of the 482 kilovolt state in Ta$^{181}$ was measured. A source of Hf$^{181}$ in 27N hydrofluoric acid, known to yield the unperturbed angular correlation (35, p. 472), was used in order to eliminate the time dependence due to the factor $W(\theta, t)$ appearing in equation (59). The source and the counters were arranged in 180 degree geometry with a source to counter distance of about 5 mm. Delayed coincidences between the members of the 133-482 kilovolt gamma cascade were recorded over a range of about seven half-life periods of the 482 kilovolt state. The single-channel analyzers were biased to accept only the photopeaks of the 133-482 kilovolt radiations. The resulting data are shown in (a) of Figure 13. The horizontal regions at the extremities of the curve represent accidental coincidences. Correction for these was made by averaging the counts in these and adjoining channels (not shown) and subtracting the result from each channel. An alternate method is to use the expression given by (60) to calculate the accidental rate from the slow singles rates recorded. In order to validate the assumption discussed in connection with equation (60), this calculation was done and was found to yield the same result as
Figure 13. Delayed coincidence curves for the 133-482 Kev gamma-ray cascade.

Time calibration: 1.25 ns/ch
\[ \tau_{1/2} = 10.4 \pm 0.2 \text{ nanoseconds} \]
the procedure described above. The delayed coincidence curve, corrected for accidentals, together with a prompt curve obtained under identical bias conditions is shown in (b) of Figure 13. The prompt curve, also corrected for accidentals, has been normalized to the same area as the delayed curve.

The lifetime of the 482 kilovolt state was evaluated from the slope of the straight line obtained from a least squares fit to that portion of the data sufficiently far removed from zero time to be independent of the resolution function represented by the prompt curve. Newton (36, p. 490) has given a simple method for the interpretation of delayed coincidence data which is useful here. Let \( F(x) \) and \( P(x) \) represent the delayed and prompt curves, respectively, and let \( f(t) \) be the probability for the decay of the 482 kilovolt state by the emission of a 482 kilovolt gamma-ray between \( t \) and \( t + \Delta t \) after the state is populated by the emission of a 133 kilovolt gamma-ray. If \( P(x) \), \( F(x) \), and \( f(t) \) are all normalized as differential probability amplitudes, i.e., are normalized to unit area, then

\[
F(x) = \int_{-\infty}^{\infty} f(t) P(x-t) \, dt.
\]

Now \( f(t) = 0 \) for \( t < 0 \), and \( f(t) = \lambda e^{-\lambda t} = 1/\tau_n e^{-t/\tau_n} \) for \( t \geq 0 \). Then with \( y = x - t \), (62) becomes
\[ F(x) = \frac{1}{\tau_n} e^{-x/\tau_n} \int_{-\infty}^{x} e^{y/\tau_n} P(y)\,dy \]  \tag{63}

Differentiating (63) gives

\[ \frac{dF(x)}{dx} = \frac{1}{\tau_n} [P(x) - F(x)] \]  \tag{64}

and

\[ \frac{d[\ln F(x)]}{dx} = -\frac{1}{\tau_n} \left[ 1 - \frac{P(x)}{F(x)} \right]. \]  \tag{65}

Equation (65) shows that in the region where \( F(x) \gg P(x) \), the data is not influenced by the finite time resolution of the apparatus, and the negative slope of the delayed coincidence curve yields \( \lambda = 1/\tau_n \) directly. Furthermore, equation (64) indicates that the intersection of \( F(x) \) and \( P(x) \) occurs at the maximum of \( F(x) \), and this information provides a further check on the operation of the delayed coincidence apparatus.

The lifetime of the 482 kilovolt state evaluated from the data in the manner just described is

\[ \tau_n = (15.0 \pm 0.3) \text{ ns}, \]

which corresponds to a half-life

\[ \tau_{1/2} = (10.4 \pm 0.2) \text{ ns}. \]
This result is in excellent agreement with the results of previous measurements listed in Table II. The excellent linearity of the delayed coincidence curve in Figure 13b is also a further confirmation of the linear response of the time-to-pulse-height converter.

The Unperturbed Anisotropy of the 133-482 kilovolt Cascade

It has already been noted that a source of $^{181}$Hf in hydrofluoric acid yields the unperturbed angular correlation of the 133-482 kilovolt gamma cascade in $^{181}$Ta. Moreover, the correlation observed under these conditions should be a constant, independent of the time interval between emission of the 133 and 482 kilovolt radiations, since there is no static interaction with a crystalline field gradient. As a further check on the operation of the spectrometer, the anisotropy of this cascade was measured as a function of time over a period of about 40 nanoseconds, using a source prepared by dissolving a few milligrams of the irradiated hafnium powder in 27N hydrofluoric acid. The solution was contained in a cylindrical cavity 3/16 inch long and 1/16 inch in diameter in a fluorethylene rod; the walls of the cavity were 1/32 inch thick. The source was positioned with its axis of symmetry vertical at the intersection of the axes of the cylindrical detectors five centimeters from the face of each detector. Delayed coincidence data were obtained alternately at 90 and 180 degrees and at 270 and 180 degrees in a series of
two-hour runs made at each angle. As before, the single-channel analyzers accepted only the photopeaks of the 133 and 482 kilovolt radiations. After correction for accidentals, the anisotropy was calculated from the true coincidence counting rates from the expression

\[ A_{\text{exp}} = \frac{N_{\text{true}}(180^\circ) - N_{\text{true}}(90^\circ, 270^\circ)}{N_{\text{true}}(90^\circ, 270^\circ)} . \]

These values were then corrected for the finite angular resolution of the detectors using the correction factors derived by Stanford and Rivers (44, p. 719-721) which have been shown to be applicable to the particular geometry used here by Glasgow, et al. (20, p. 683-684).

The anisotropy as a function of time is shown in Figure 14. As expected, the results are essentially constant within the statistical precision of each point. Although the magnitude of the observed anisotropy agrees well with the results of McGowan (35, p. 473) who reports the value \( -0.38 \pm 0.02 \) obtained under similar source conditions, the results are, nonetheless, consistently too low compared to the value \( -0.407 \pm 0.006 \) computed from the unperturbed angular correlation coefficients. Aside from gross misalignment of the spectrometer, the most likely causes of the observed discrepancy are scattering in the source or surrounding material and nuclear
reorientation due to interaction of the nuclear moments with local fields in the source. However, in the present application of the angular correlation technique the magnitude of the anisotropy need not be known precisely and no extensive effort was made to resolve the discrepancy noted here.

![Graph](image)

Figure 14. The anisotropy of the 133-482 kilovolt gamma cascade of Ta\textsuperscript{181} obtained with a liquid source. The horizontal dotted line represents the value calculated from the angular correlation coefficients assuming no perturbation.

The Perturbed Anisotropy of the 133-482 kilovolt Cascade

The interaction of the quadrupole moment of the 482 kilovolt state of Ta-181 with the crystalline electric field gradient in hafnium metal was observed through its effect on the differential anisotropy obtained with a powder source. The source preparation has already been described; the source volume and activity used in these measurements were about $10^{-3}$ cm\(^3\) and 20 microcuries, respectively.
In order to reduce the effect of a possible non-random distribution of microcrystal orientations, provision was made to permit continuous rotation of the source holder at a few revolutions per minute about an axis perpendicular to the plane of the counters. However, rotation of the source in this way was found to have no observable effect on the results; the degree of preferential orientation in the sources tested, so far as the results of this experiment are affected, is apparently negligible. Aside from the nature of the source, the experimental conditions and procedure here were identical to those described earlier for the liquid source measurements.

Delayed coincidence spectra recorded at 90 and 180 degrees are shown in Figure 15. These data, corrected for accidental counts, represent those collected in six days of two-hour runs made alternately at each angle. In order to establish "zero-time" and to check temporal stability, prompt coincidence curves were obtained at regular intervals throughout the experiment. In no case was the temporal drift greater than ±0.3 channel units in any twenty-four hour period, nor did the zero-time point shift more than this figure over the entire time of the experiment.

The differential anisotropy computed from the data shown in Figure 15 is presented in Figure 16. Correction for the finite angular resolution of the detectors has been made here, but the data are not corrected for the finite time resolution of the coincidence
Figure 15. Delayed coincidence data at 90° and 180° for the 133-482 Kilovolt gamma-ray cascade in Ta181.
Figure 16. Time dependent anisotropy of the 133-482 kilovolt gamma cascade in polycrystalline hafnium metal. The dashed curve represents the theory for an axially-symmetric field gradient and the solid curve represents the theory for a non-axial gradient with an asymmetry parameter $\eta = 0.3$. 
apparatus. The quadrupole precession period evaluated from the position of the centroids of those portions of the second and third peaks above the continuum, assuming for the moment an axially symmetric field gradient, is $21.0 \pm 0.5$ nanoseconds and corresponds to a quadrupole coupling

$$\Delta v_Q = eQ V_{zz} / h = 317 \pm 8 \text{ Mc/sec.}$$

The Perturbed Anisotropy of the 133-345 kilovolt Cascade

Although the decay of the 482 kilovolt state occurs mainly via the 482 kilovolt gamma-ray transition to the ground state, there exists, in addition to the competing internal conversion process, the alternate mode of decay via the 133-345 kilovolt cascade through the short-lived 137 kilovolt state. This portion of the complete level scheme of $^{181}\text{Ta}$ shown in Figure 1 is reproduced here for convenience.

![Partial level scheme of $^{181}\text{Ta}$](image)

Figure 17. Partial level scheme of $^{181}\text{Ta}$.
According to the theory, the perturbation of the angular correlation of all cascades proceeding through the same intermediate state is the same, i.e. the nature of the perturbation depends only on the properties of the intermediate level. The validity of this feature of the theory has been demonstrated by the experiments of Gerber, et al. (19, p. 180-182) in which the attenuation factors for both gamma-gamma and gamma-conversion electron cascades proceeding through the 248 kilovolt state of Cd\textsuperscript{111} were measured for several different source conditions. When the attenuation factors obtained under a given source condition were compared, they were found to be equal within the small experimental uncertainties. A comparison of the differential anisotropies of the 133-482 and 133-345 kilovolt gamma-ray cascades in Ta\textsuperscript{181} should also provide a test of this aspect of the theory. On the other hand, taking this point to be well-established, a study of the differential anisotropy of the 133-345 kilovolt cascade similar to that already described for the 133-482 kilovolt cascade offers the possibility of making an additional, independent determination of the quadrupole coupling.

To obtain the anisotropy of the 133-345 cascade, the single-channel analyzers were biased to accept only the photopeaks of the 133 and 345 kilovolt gamma-rays. The window of the analyzer accepting the 133 kilovolt photopeak also accepted the photopeak of the 137 kilovolt gamma-ray and since the latter is emitted in prompt
coincidence with the 345 kilovolt gamma-ray an anomalously large prompt component appears in the data shown in Figure 18. Another difficulty is that the analyzer biased to record the photopeak of the 345 kilovolt gamma-ray also recorded a portion of the Compton recoil pulse distribution of the 482 kilovolt gamma-ray. Thus, the data collected in the region of zero-time represent a composite correlation function of the 133-482, 133-345, and 345-137 kilovolt
gamma-ray cascades while those collected at points far removed from the prompt peak represent a composite function of the 133-482 and 133-345 cascades.

Aside from the large prompt component, the data in Figure 18 do not exhibit the periodic fluctuations so apparent in the corresponding data for the pure 133-482 cascade. Moreover, the difference in the number of counts between corresponding channels of the 90 and 180 degree runs is not significant over the counting statistics except in the region of zero time. Thus, it appears that the composite anisotropy of the 133-482 and 133-345 cascades is very nearly zero; since the time-dependent attenuation factors never exceed unity, the composite anisotropy is effectively very insensitive as an indicator of the quadrupole induced perturbation. This does not mean, however, that the 133-345 anisotropy is unperturbed by the quadrupole interaction. A comparison of the experimental result for the composite anisotropy with the result calculated from the known angular correlation coefficients and measured relative intensities of the 133-482 and 133-345 kilovolt contributions shows in fact that just such a perturbation is required for agreement.

The relative intensities of the 133-482 and 133-345 kilovolt contributions to the composite data were obtained in the following way. Pulse-height spectra of the pure 510 kilovolt gamma-ray in the decay of Cu$^{64}$ and the 345 and 482 kilovolt gamma-rays in the decay
of Ta$^{181}$ were obtained under the experimental conditions of the delayed coincidence experiment. Numerical integrations performed on each normalized spectrum over the region spanned by the window settings formerly used yielded the number of 510 kilovolt Compton events in one case and the number of 345 photopeak plus 482 kilovolt Compton events in the other. In view of the small energy difference between the 482 and 510 kilovolt gamma-rays, it appeared reasonable to take their Compton pulse distributions to be equivalent. This permitted the relative contributions of the two cascades to be computed from the results of the numerical integrations. In this way, the relative intensities of the 133-482 and 133-345 kilovolt contributions to the composite anisotropy were found to be $0.30 \pm 0.03$ and $0.70 \pm 0.03$, respectively. The uncertainty introduced through the assumed equivalence of the 482 and 510 kilovolt Compton recoil pulse distributions precludes assigning a meaningful probable error to these intensities and the uncertainties cited are those due to counting statistics only. However, it seems unlikely that the intensity factors should be off by more than 0.1 in either direction and, as it turns out, the composite anisotropy is simply not very sensitive to variations in this range.

By definition, the composite anisotropy of the mixed 133-482 and 133-345 kilovolt gamma-ray cascades is
\[ A_{\text{comp}} = \frac{W_{\text{comp}}(\pi)}{W_{\text{comp}}(\pi/2)} - 1 \]  

(66)

where \( W_{\text{comp}} \) is the probability for the emission of either a 482 or 345 kilovolt gamma-ray at the indicated angle with respect to the direction of the coincident 133 kilovolt radiation. Introducing the relative intensities \( I_{345} \) and \( I_{482} \) and the probability factors for the pure cascades into (66) yields

\[ A_{\text{comp}} = \frac{I_{482} W_{482}(\pi) + I_{345} W_{345}(\pi)}{I_{482} W_{482}(\pi/2) + I_{345} W_{345}(\pi/2)} - 1. \]  

(67)

The individual probabilities for emission at a given angle \( \theta \) may be expressed in terms of the angular correlation coefficients as the sum

\[ W(\theta) = 1 + A_2 P_2(\cos \theta) + A_4 P_4(\cos \theta) \]  

(68)

Where the \( A_i \)'s are the angular correlation coefficients associated with a particular cascade and the \( P_i \)'s are the Legendre polynomials as before. Using the values of the \( A_i \)'s measured by McGowan (35, p. 474) and the relative intensity factors measured here, the composite anisotropy was calculated to be

\[ A_{\text{comp}} = -0.01 \pm 0.01. \]

It is perhaps worthwhile to mention that \( A_2 \) and \( A_4 \) are both
negative for the 133-482 cascade while the corresponding quantities for the case of the 133-345 cascade are both positive. This fact, together with the magnitudes of the coefficients and relative intensities, results in an effective cancellation of the rather large anisotropy characterizing each pure cascade.

The degree to which this prediction fits the experimental results is illustrated in Figure 19 in which the horizontal line represents the anisotropy computed from the correlation coefficients and relative intensities and the plotted points are the values of the anisotropy computed from the data of Figure 18 for the region of time where the contribution of the prompt events is negligible. When these

![Figure 19](image)

**Figure 19.** The differential anisotropy of the mixed 133-482, 345 cascade as a function of time. The solid line represents the value calculated from the angular correlation coefficients and relative intensities.
results are compared to those of the pure 133-482 kilovolt cascade in which the anisotropy exhibits the pronounced fluctuations due to the quadrupole perturbation, it must be concluded that the 133-345 anisotropy is similarly affected. The data of Figure 18 could of course be corrected for the 133-482 contribution and the resulting pure 133-345 anisotropy would then show the periodic fluctuations expected. However, this yields no new information and these calculations were not done.

In principle, the separation of the 133-482, 133-345, and 345-137 kilovolt contributions to the coincidence counting rate in the region of zero-time could be done in the same way. Again, this yields no new information about the quadrupole interaction and no attempt was made to actually measure the relative intensities of these contributions. However, a further check on the interpretation of the behavior of the mixed 133-482, 133-345 kilovolt data was made by the following rather simple procedure. If one assumes that the composite anisotropy of the 133-482 and 133-345 kilovolt cascades is constant with time (as indeed the results over the region from 10 to 40 nanoseconds indicate), the shape of the delayed coincidence curves for 90 and 180 degrees for the composite data should be identical to the curve obtained in the lifetime measurements. Thus, the number of counts occurring at zero-time due to 133-482 and 133-345 kilovolt events can be obtained from a comparison of the lifetime data of
Figure 13 with the data of Figure 18. The number of 345-137 kilovolt prompt events in both the 90 and 180 degree data of Figure 18 can then be calculated by subtracting the 133-482 and 133-345 kilovolt contributions found above. Thus, the anisotropy of the pure 345-137 kilovolt cascade can be calculated. This procedure yielded a value of 0.32 ± 0.02 which is in excellent agreement with the result of McGowan (35, p. 474) who obtained the value 0.305 ± 0.023 for a liquid source in which the perturbation was known to be absent.
CONCLUSIONS

The interaction between the quadrupole moment of the 482 kilovolt excited state in Ta\textsuperscript{181} and the electric field gradient in hafnium metal has been studied through its effect on the delayed angular correlation of the 133-482 kilovolt gamma-ray cascade in the decay of the beta emitter Hf\textsuperscript{181}. The quadrupole precession period evaluated from the periodic behavior of the differential anisotropy on the basis of the theory for axially-symmetric gradients is 21.0 ± 0.5 nanoseconds which corresponds to an interaction strength

\[ \Delta \nu_Q = 317 \pm 8 \text{ Mc/sec}. \]

This result has been confirmed by the more recent experiments of Ouseph and Canavan (37, p. 144) and Salomon, et al. (40, p. 14). A similar study of the 133-345 kilovolt gamma-ray cascade did not permit a direct observation of the quadrupole precession period due to interference from the 133-482 and 345-137 kilovolt cascades. The observed composite anisotropy is, however, consistent with the result obtained for the pure 133-482 kilovolt cascade as well as with existing theory. An indirect determination of the anisotropy of the 345-137 kilovolt cascade yielded the value 0.32 ± 0.02 in agreement with the result obtained by McGowan (35, p. 474). In addition, the half-life of the 482 kilovolt state in Ta\textsuperscript{181} was remeasured to be...
10.4 ± 0.2 nanoseconds in agreement with the prior determinations listed in Table II.

Although the differential anisotropy of the 133-482 kilovolt cascade displays the nearly periodic structure predicted by the axially-symmetric theory, its detailed behavior with time (see Figure 16) is not in good agreement with this simple description. The dashed curve in the figure represents the theory for axially-symmetric field gradients and has been modified here to take into account the finite time resolution of the coincidence apparatus. Departure from the dashed curve is characterized by the time-dependent background plus an attenuation and broadening of the peaks with increasing time. Most apparent is the well-defined knee which appears between the initial and second maximum.

Some recent work reported by Matthias, et al. (34, p. 41-43) suggests two mechanisms which might account for these effects. They have calculated the differential attenuation factors of equation (36) for the cases in which 1) the electric interaction is non-axial, the degree of asymmetry being given by the parameter

$$\eta = \frac{(V_{xx} - V_{yy})}{V_{zz}}$$

and 2) the electric field gradient, although possessing axial symmetry, is not unique in the sense that there exists some dispersion in its value from nucleus to nucleus of the sample which leads to a quadrupole frequency distribution \(P(\omega)d\omega\) of finite width. Thus, the assumption of an ideal crystal system
having axial symmetry and a field gradient of the same magnitude at each nuclear site is relaxed to include the more realistic situation in which such effects as nuclear recoil and imperfect crystal structure can play a role. The behavior of the attenuation factor $G_2(t)$ for an intermediate state of spin $I = 5/2$ is shown in Figure 20 for various values of the asymmetry parameter; a similar plot in Figure 21 shows the effect of a normal frequency distribution of various widths on $G_2(t)$.

Hafnium metal is known to be of the hexagonal close packed (hcp) structure at temperatures below 1760 degrees Centigrade (47, p. 212) and its crystalline field gradient should, therefore, be axially symmetric. Nonetheless, the striking similarity between the behavior of the experimental anisotropy in the region of the initial and second maximum and the attenuation factor of Figure 20 in this region suggests that asymmetric field gradients may indeed be present in the hafnium crystals used. The evidence is even more pronounced when one compares the data of Figure 16 with the solid curve representing the anisotropy computed from the known angular correlation coefficients, the measured value of the precession period and the differential attenuation factors of Matthias, et al. for the asymmetry parameter $\eta = 0.3$. The agreement is quite good over the first precession period and although the data between the succeeding maxima are consistently too low, the double-hump
Figure 20. Differential attenuation coefficients $G_2(t)$ for rhombic quadrupole interaction in polycrystalline sources. The parameter $\eta$ is the asymmetry parameter $\eta = (V_{xx} - V_{yy})/V_{zz}$. (34, p. 42)

Figure 21. Differential attenuation coefficients $G_2(t)$ for axially-symmetric quadrupole interaction with frequency distribution $\delta$ in polycrystalline sources. (34, p. 42)
structure of the third and even the fourth peak is evident in the experimental results. Fits made for other values of $\eta$ for which the attenuation factors were available (0.2 and 0.4) were not nearly so good in the region where the $\eta = 0.3$ curve fits well nor was the agreement in the regions between the peaks significantly better. Further comparison of the data with the attenuation factors of Figure 21 suggests that a frequency distribution might account for at least part of the disagreement with the simple theory. The latter effects, however, appear to be much less pronounced than the effects of an asymmetry. Unfortunately, the attenuation factors for frequency distributions of various widths for the region of time over the second and third precession period where the effects become large were not readily available and a fit was not attempted for this case.

Although the finite time resolution of the coincidence apparatus makes it somewhat difficult to distinguish between these two static interaction effects, i.e., between a non-axial field gradient and a frequency distribution, the data are not inconsistent with the presence of either or both of them. In view of the hcp structure of hafnium, it is difficult to understand the apparently large degree of asymmetry in the field gradient. One might expect that the recoil of the nucleus due to neutron capture in the irradiation of the sample or to the beta emission preceding the gamma cascade might move the atoms into anomalous lattice positions in which the local field gradient has
neither axial symmetry nor a constant magnitude from nucleus to nucleus. In this connection, it is significant to note that the experiments of Salomon, et al. (40, p. 14) in which polycrystalline sources were used show the same deviations from the axially-symmetric theory as observed here; the results of this group's work with hafnium single crystals, on the other hand, show no evidence whatsoever for the presence of asymmetric gradients and only a slight indication of the presence of a frequency distribution (41, p. 114).

It is clear that further work is needed if all of the aspects of the internal perturbation mechanism are to be well understood. For example, a study of the differential anisotropy with annealed or recrystallized sources and as a function of beta energy as well might provide information about the origin of the effects associated with asymmetry and frequency distributions. Similar studies as a function of microcrystal size could show whether or not surface effects are important. In an introductory statement, it was remarked that the limitation that all results based on a knowledge of the true angular correlation are certain only to the extent that the influences of extranuclear perturbations can be accounted for has the redeeming feature of providing information about the perturbing interactions themselves. It seems appropriate to end this dissertation with the similar remark that the perturbation of the pure quadrupole
precession by inhomogeneities in the local environment of the
decaying nucleus, although making the measurement of the quadru-
pole coupling somewhat uncertain, offers as a natural compensation
a sensitive probe for the study of certain microscopic properties of
solids.


APPENDIX
APPENDIX

Theory of Unperturbed Angular Correlation

In this section we discuss the problem of angular correlations in the absence of any extranuclear perturbation. Although the presentation here is fairly complete, at a few places in the development where a complete discussion would become prohibitively involved detailed arguments are replaced by simple plausibility arguments with reference made to the original literature for further justification. Also, the discussion here is limited to gamma-gamma cascades in which each member is a pure transition in the sense that each radiation is either pure electric or pure magnetic in character with no admixtures present.

The general problem of angular correlation was first considered by Hamilton in 1940 (25, p. 122-131) who applied second-order time-dependent perturbation theory to the initial system of the excited nucleus and quantized radiation field to derive the general correlation function (1). Progress in the theory since that time has been due largely to the work of Alder (6, p. 235-251), Falkoff (17, p. 323-333), Goertzel (21, p. 897-909), Lloyd (31, p. 716-720) and Racah (38, p. 910-912). Biedenharn and Rose (9, p. 729-777) have presented a detailed review article on the subject; also, a comprehensive review has been given by Frauenfelder (18, p. 531-599). The following
discussion is essentially that given by Rose (39, p. 168-176) in still another review of the subject.

In the following we consider a nucleus in an initial excited state $A$ which decays by the emission of a gamma-ray $\gamma_1$ to a short-lived intermediate state $B$ which subsequently decays to a final state $C$ by the emission of a second gamma-ray $\gamma_2$. The problem to be solved is that of finding the directional correlation between $\gamma_1$ and $\gamma_2$, whose propagation directions $\mathbf{k}_1$ and $\mathbf{k}_2$ with respect to the arbitrarily chosen quantization axis $z$ are shown in Figure A1.

![Figure A1](image)

Figure A1. The angular coordinates of the propagation vectors $\mathbf{k}_1$ and $\mathbf{k}_2$.

We now begin a derivation of the angular correlation function using time-dependent perturbation theory. In order to describe the system of nucleus plus radiation field, we introduce the wave function
\[\psi = \psi_a A_a(t) e^{-iE_a t} + \psi_b A_b(t) e^{-iE_b t} + \psi_c A_c(t) e^{-iE_c t}\]

\[= \sum_n \psi_n A_n(t) e^{-iE_n t}\]  \hspace{1cm} (A1)

where \(n\) takes on the values \(a, b,\) and \(c\) and \(A_a, A_b,\) and \(A_c\) represent the amplitudes of the initial, intermediate, and final states of the cascade under consideration. The \(\psi_a, \psi_b,\) and \(\psi_c\) are the stationary state wave functions for the states \(A, B,\) and \(C\) and the \(E\)'s are the total energy values, i.e., the energy of the nucleus plus emitted radiation. We assume that the interaction Hamiltonian \(H\) can be written as the sum of an unperturbed Hamiltonian \(H_0\) (the Hamiltonian of the nucleus alone) and a perturbation term \(\lambda H_r\) where \(\lambda\) is a real parameter and \(H_r\) is the interaction Hamiltonian responsible for the emission or absorption of radiation. In addition, the following expansion of the amplitudes is introduced:

\[A_n(t) = A_n^{(0)}(t) + \lambda A_n^{(1)}(t) + \lambda^2 A_n^{(2)}(t) + \ldots = \sum_k \lambda^k A_n^{(k)}(t)\]  \hspace{1cm} (A2)

The total wave function \(\psi\) satisfies the time-dependent Schrödinger equation which can be written as:
\[ H\Psi = (H_0 + \lambda H_r)\Psi = i\partial\Psi/\partial t \]

\[ = (H_0 + \lambda H_r) \sum_n A_n(t)\psi_n e^{-iE_nt} \]

\[ = i \sum_n \frac{\partial A_n}{\partial t} \psi_n e^{-iE_nt} + \sum_n A_n E_n \psi_n e^{-iE_nt}. \quad (A3) \]

The eigenvalue equations

\[ H_{0a}\psi_a = E_a\psi_a, \quad H_{0b}\psi_b = E_b\psi_b, \quad \text{and} \quad H_{0c}\psi_c = E_c\psi_c \]

can be used to eliminate the \( H_{0n} \) terms in \( (A3) \) to obtain

\[ \lambda H_r \sum_n A_n\psi_n e^{-iE_nt} = i \sum_n \frac{\partial A_n}{\partial t} \psi_n e^{-iE_nt}. \quad (A4) \]

Taking the inner product \( \langle \psi_k | \psi_n \rangle \) yields

\[ i \frac{\partial A_k}{\partial t} = \sum_n \langle \psi_k | \lambda H_r | \psi_n \rangle A_n e^{i(E_k - E_n)t} \]

\[ = \sum_n \langle \psi_k | \lambda H_r | \psi_n \rangle \left[ A_n^{(0)} + \lambda A_n^{(1)} + \lambda^2 A_n^{(2)} + \ldots \right] e^{i(E_k - E_n)t}. \quad (A5) \]

By equating the coefficients of like power of \( \lambda \) on each side of \( (A5) \), the different orders \( j \) of the perturbation \( \lambda^j H_r \) can be extracted.
Zero-order:

\[ i \frac{\partial A_k^{(0)}}{\partial t} = 0 \text{ or } A_k^{(0)} = \text{a constant} \]

The initial conditions are: \( A_a(t) = 1 \) and \( A_b(t) = A_c(t) = 0 \). Since the \( A_k^{(0)} \) are constant in time, this means that \( A_a^{(0)} = 1 \) and \( A_b^{(0)} = A_c^{(0)} = 0 \), or, more simply: \( A_k^{(0)} = \delta_{ka} \).

First-order:

\[ i \frac{\partial A_k^{(1)}}{\partial t} = \sum_n <\psi_k | H_r | \psi_n> e^{i(E_k - E_n)t} A_n^{(0)} \]

\[ = <\psi_k | H_r | \psi_n> e^{i(E_k - E_n)t}, \quad (A6) \]

since \( A_n^{(0)} = \delta_{na} \). Now the matrix element \( <\psi_c | H_r | \psi_a> \) is zero since we rule out transitions which go from level \( A \) to \( C \) directly without passing through the intermediate state \( B \). Also,

\( <\psi_a | H_r | \psi_a> = 0 \) for the reason that, \( H_r \) being the interaction Hamiltonian for emission and absorption, if a transition does not occur \( H_r = 0 \). Thus, first-order theory yields

\[ \frac{\partial A_a^{(1)}}{\partial t} = \frac{\partial A_c^{(1)}}{\partial t} = 0 \text{ and } A_a^{(1)} = A_c^{(1)} = 0, \text{ the initial values}, \]

and the only non-zero first-order amplitude is \( A_b^{(1)} \) found by integrating (A6).
Thus, 

\[ A^{(1)}_b = -<\psi_b | H_r | \psi_a> \frac{e^{i(E_b - E_a)t}}{E_b - E_a} - 1 = <\psi_b | H_r | \psi_a> \frac{e^{-i\omega_1 t}}{\omega_1} \]  

(A7)

where \( \omega_1 = E_a - E_b \). In order to write the angular correlation function \( W(\theta) \), we need to determine the amplitude \( A_c \) and then \( |A_c|^2 \).

It is noted in passing that since in first-order theory \( A^{(1)}_c = 0 \), it is necessary to include second-order terms. Hence, angular correlation theory is a second-order theory.

**Second-order:**

We are interested in the second-order term \( A^{(2)}_c \) only. From (A5) we find,

\[ i \frac{\partial A^{(2)}_c}{\partial t} = \sum_n <\psi_c | H_r | \psi_n> e^{i(E_c - E_n)t} A^{(1)}_n \]

\[ = <\psi_c | H_r | \psi_b> e^{i(E_c - E_b)t} A^{(1)}_b \]

\[ = <\psi_c | H_r | \psi_b> <\psi_b | H_r | \psi_a> \frac{e^{-1(\omega_1 + \omega_2)t}}{\omega_1} e^{-i\omega_2 t} \]

which, when integrated, gives

\[ A^{(2)}_c = \frac{<\psi_c | H_r | \psi_b> <\psi_b | H_r | \psi_a>}{\omega_1} \left[ e^{-(\omega_1 + \omega_2)t} - 1 \right] \frac{e^{-\omega_2 t}}{\omega_2} \]

where \( \omega_2 = E_b - E_c \).
From (A6) it is seen that $A_c$, the final state probability amplitude, contains one matrix element for each of the two transitions. The operator $H_r$ depends on the properties of the radiation emitted and is not, as the notation implies, the same in each matrix element. To emphasize this point we replace the $H_r$ in all succeeding expressions by $H_1$ and $H_2$, the interaction Hamiltonians for the emission of $\gamma_1$ and $\gamma_2$, respectively. Now $A_c$ is the probability amplitude that the nucleus be in the final state $C$ after having undergone the transitions $A \rightarrow B \rightarrow C$ by the radiation of $\gamma_1$ and $\gamma_2$ in the specified directions $\vec{k}_1$ and $\vec{k}_2$. Hence, we are interested in $|A_c|^2$ summed over all final energy states. The summation can be shown (21, p. 900) to convert the time-dependent factor in (A6) to a factor which is linear in time so that the transition rate, i.e., the time-derivative of $\Sigma |A_2|^2$, is proportional to

$$W(\vec{k}_1, \vec{k}_2) = \sum_{m_a, m_c} |<I_m c | H_2 | I_m b > <I_m b | H_1 | I_m a >|^2.$$  \hspace{1cm} (A7)

In (A7) the $|\psi>$ have been replaced by the $|I m>$ to indicate more clearly that the nuclear states involved are characterized by a particular value of spin $I$ and projection quantum number $m$. Also, an average over the initial substates is implied in (A7). There are $2I_b + 1$ intermediate substates through which the cascade may
proceed; in order to include all the ways in which a transition can
occur from an initial substate of A to a final substate of C via the
different intermediate substates \( m_b \), (A7) is rewritten as

\[
W(k_1, k_2) = S_1 S_2 \sum_{m_c}^{m_a} \sum_{m_b} < I_m | H_2 | I_{m_b} > < I_{m_b} | H_1 | I_{m_a} >^2
\]

where \( S_1 \) and \( S_2 \) denote an average over all unobserved properties
of the radiations.

We now begin the calculation of the explicit correlation function
\( W(k_1, k_2) \) for the \( \gamma_1 - \gamma_2 \) cascade proceeding through the levels
\( A \rightarrow B \rightarrow C \). The general correlation function (A8) can be rewritten
as

\[
W(k_1, k_2) = S_1 S_2 \sum_{m_c}^{m_a} \sum_{m_b} < I_{m_b} | H_1 | I_{m_a} > < I_{m_b} | H_1 | I_{m_a} >^* \\
\times < I_{m_c} | H_2 | I_{m_b} > < I_{m_c} | H_2 | I_{m_b} >^*
\]

where the asterisk denotes the complex conjugate. It is convenient
to break up the correlation function into two parts by introducing the
matrices

\[
\Omega^{(1)}_{m_b m'_b} = S_1 \sum_{m_a} < I_{m_b} | H_1 | I_{m_a} > < I_{m_b} | H_1 | I_{m_a} >^* \quad (A10)
\]

and

\[
\Omega^{(2)}_{m_b m'_b} = S_2 \sum_{m_c} < I_{m_c} | H_2 | I_{m_b} > < I_{m_c} | H_2 | I_{m_b} >^* \quad (A11)
\]
which are characteristic of the first and second transitions, respectively. Thus, the correlation function becomes

\[ W(k_1, k_2) = \sum_{m_b', m_b} \Omega_{m_b, m_b}^{(1)} \Omega_{m_b', m_b}^{(2)}. \]  

(A12)

It has already been mentioned that the direction \( \hat{n} \) of the quantization axis (see Figure A1) is arbitrary. It is perhaps worthwhile to mention that if the quantization axis is taken along \( k_1 \), the propagation direction of the first radiation, the interference terms in (A9) do not appear, i.e., the sum over \( m_b \) is performed incoherently, so that

\[ W(k_1, k_2) = W(0) = S_1 S_2 \sum_{m_a, m_b, m_c} \left| <I_b m_b | H_0 | I_a m_a > <I_c m_c | H_0 | I_b m_b > \right|^2. \]  

(A13)

In this restricted form, involving only squares of matrix elements, the terms in the sum have a particularly simple interpretation, namely that

\[ S_1 \left| <I_b m_b | H_0 | I_a m_a > \right|^2 = \Pi_{m_a, m_b} (0) \]

is the probability for the emission of \( \gamma_1 \) along the direction \( \theta = 0 \) during a nuclear transition between the sublevels \( m_a \rightarrow m_b \). Similarly,
\[ S_2 \langle I_m^c | H_\theta | I_m^b \rangle \|^2 = \Pi_{m_b m_c} (\theta) \]

gives the probability for \( \gamma_2 \) to be emitted at an angle \( \theta \) in the transition \( m_b \rightarrow m_c \). The correlation function is then

\[ W(\theta) = \sum_{m_a, m_b, m_c} \Pi_{m_a m_b} (0) \Pi_{m_b m_c} (0) \]

or

\[ W(\theta) = \sum_{m_b} \Omega_{m_b m_b}^{(1)} \Omega_{m_b m_b}^{(2)} \tag{A14} \]

Although the special choice of the quantization axis along the \( k_1 \) direction gives \( W(\theta) \) in a form which allows a clear physical interpretation of the correlation process in terms of a sum of probability products (which, incidentally, closely corresponds to the simple qualitative argument given on pages 11-15), the restriction on the direction of the quantization axis makes the further reduction of the correlation function somewhat more difficult than in the general case of (A12) where this direction is arbitrary. We proceed, therefore, with the reduction of the latter.

We proceed now to determine the form of the \( \Omega \)-matrices from which we will obtain the general correlation function through (A12).

The development will be outlined for the first radiation only, the corresponding treatment for the second gamma-ray being entirely
equivalent. Since the interaction Hamiltonian $H_1$ is an invariant, it has the general form (39, p. 80)

$$H_1 = \sum_{M_1=-L_1}^{L_1} (-1)^{M_1} A_{L_1, -M_1} T_{L_1 M_1}$$  \hspace{1cm} (A15)

where $T_{L_1 M_1}$ is an irreducible tensor of rank $L_1$ in the coordinates of the nucleus. The tensor components $A_{L_1, -M_1}$ refer to the radiation field and would be obtained from the vector potential of the plane wave associated with the radiation $\gamma_1$ when $T_{L_1 M_1}$ is obtained from the charge and current densities of the source, i.e., the nucleus in this case. Rose (39, p. 171) makes the important point that the precise nature of the operator $T_{L_1 M_1}$ is not known and, in fact, is irrelevant in the present discussion; its properties as an irreducible tensor are all that need be of concern here.

We now perform a transformation from the coordinate system of the first radiation in which $\vec{k}_1$ is the quantization axis to a system in which the $z$-axis becomes the axis of quantization, that is, a rotation is performed which takes $\vec{k}_1$ into $\hat{\alpha}$. The tensors in the new frame are related to those in the old frame by

$$T_{L_1 M_1}^{\prime} = \sum_{\mu_1} T_{L_1 M_1} D^{L_1}_{\mu_1 M_1} (a_1 b_1 0)$$  \hspace{1cm} (A16)
where $D(a, \beta, 0)$ is the rotation matrix discussed in detail by Rose (39, p. 48-75). The $\Omega^{(1)}$-matrix becomes

$$
\Omega^{(1)}_{m_b m'_b} = S L \sum_{m_a} \sum_{L_1, L'_1} \sum_{M_1, M'_1} \sum_{\mu_1, \mu'_1} \frac{(-1)^{M_1 + M'_1}}{A_{L_1, M_1} A_{L'_1, -M'_1}} A_{L_1}^{\mu_1 m_a} A_{L'_1}^{\mu'_1 m'_a} \\
\times D(a, \beta, 0) \frac{L_1 L'_1^*}{\mu_1 M_1 \mu'_1 M'_1} \frac{D(a, \beta, 0)}{<I_b m_b | T_{L_1 \mu_1} | I_a m_a>} \\
\times <I_b m_b | T_{L'_1 \mu'_1} | I_a m_a>.
$$

We now make use of the Wigner-Eckart theorem which permits one to express a matrix element of the operator $T_{L \mu}$ as a product of a factor which contains the entire dependence on the projection quantum numbers and a reduced matrix element which is independent of the $m$. That is, we have (39, p. 85)

$$
<I_b m_b | T_{L_1 \mu_1} | I_a m_a> = C(I_a L_1 I_b; m_a \mu m_b) <I_b \parallel T_{L_1} \parallel I_a>
$$

where the quantity $<I_b \parallel T_{L_1} \parallel I_a>$ is called the reduced matrix element of the tensor $T_{L \mu}$ and $C(\ ; \ )$ is a Clebsch-Gordon coefficient. For future use, we mention the important property that the Clebsch-Gordon coefficient vanishes unless the vector triple $(I_a L_1 I_b)$ forms a triangle and $m_b = m_a + \mu$. Applying the Wigner-Eckart theorem and using the property of the rotation matrices that
\[ L_1 = (-1)^{\mu^*} M_1^* D, \text{ equation (A17) becomes} \]

\[ \Omega^{(1)}_{\mathbf{m}_b, \mathbf{m}_b} = S_1 \sum_{\mathbf{m}_a} \sum_{L_1, L_1'} \sum_{M_1, M_1'} \sum_{\mu_1, \mu_1'} (-1)^{M_1 \pm \mu_1} \left< I_b \parallel T_{L_1} \parallel I_a > \right> \]

\[ x \left< I_b \parallel T_{L_1} \parallel I_a > \right>^* A_{L_1, -M_1} A_{L_1', -M_1'} D \]

\[ x \left< I_b \parallel T_{L_1} \parallel I_a > \right>^* A_{L_1, -M_1} A_{L_1', -M_1'} D \]

\[ x \left< C(L_1 L_1' \mathbf{m}_b; \mathbf{m}_a \mu_1 \mathbf{m}_b) C(L_1' L_1 \mathbf{m}_b; \mathbf{m}_a \mu_1^* \mathbf{m}_b') \right> \]

\[ x \delta \mathbf{m}_a + \mu_1, \mathbf{m}_b \delta \mathbf{m}_b + \mu_1^* \mathbf{m}_b'. \]  

\[(A19)\]

The product of the D-matrices can be expressed as the series

\[ L_1 \]
\[ D \]
\[ D \]
\[ \mu_1 M_1 \]
\[ -\mu_1^*, -M_1' \]
\[ \sum \]
\[ C(L_1 L_1' \nu_1; \mu_1, -\mu_1', \mu_1 -\mu_1') C(L_1 L_1' \nu_1; M_1, -M_1', \mu_1 -\mu_1') \]
\[ D \]
\[ \nu_1 \]
\[ \mu_1 -\mu_1', M_1, -M_1' \]

which provides the connection between the uncoupled representation on the left and the coupled representation on the right and is known as the coupling rule for the rotation matrices. The sum is to be taken over those values of \( \nu_1 \) for which the triangle rule for the vector triple \((L_1 L_1 \nu_1)\) holds. Replacing the product of the D-matrices in (A19) by the expression above and performing the sums over \( \mu_1 \) and \( \mu_1' \) gives
\[ \Omega_{m_b m_b'}^{(1)} = \sum_{m_a} \sum_{L_1, L_1'} \sum_{M_1, M_1'} (-1)^{M_1 - m_a + m_b'} \left< b \parallel T_{L_1} \parallel I \right> \left< I \parallel I \right>_{T_{L_1}}^{*} \]

\[ \times A_{L_1', -M_1} A_{L_1}^{*} C(I_a L_1 I_b; m_a, m_b - m_a, m_b) \]

\[ \times C(I_a L_1' I_b; m_a, m_b' - m_a, m_b') \sum_{\nu_1} C(L_1 L_1' \nu_1; m_b' - m_a, \nu_1 \nu_1) \]

\[ m_a - m_b', m_b - m_b' \quad C(L_1 L_1' \nu_1; M_1', -M_1', M_1 - M_1') \]

\[ \times D \]

\[ m_b - m_b', M_1 - M_1' \]

(A20)

The sum over \( m_a \) can now be carried out. This is done by making use of the symmetry relations and orthogonal property of the Clebsch-Gordon coefficients and the relation between Racah and Clebsch-Gordon coefficients expressed in simplified notation in (A21) through (A24).

\[ C(abc; def) = (-1)^{a+b-c} C(abc; -d, -e, -f) \quad (A21) \]

\[ C(abc; def) = (-1)^{a+b-c} C(bac; edf) \quad (A22) \]

\[ \sum_{m_1} C(I_1 I_2; m_1, m - m_1, m) C(I_1 I_2'; m_1, m - m_1, m) = \delta_{I I'} \quad (A23) \]
\[ \sum_{f} \left[ (2e+1)(2f+1) \right]^{1/2} W(\text{abcd};\text{ef}) C(\text{bdf};\beta, \delta, \beta + \delta) C(\text{afc};\alpha, \beta + \delta, \alpha + \beta + \delta) \]

\[ = C(\text{abe};\alpha, \beta, \alpha + \beta) C(\text{edc}; \alpha + \beta, \delta, \alpha + \beta + \delta) \]  \hspace{1cm} (A24)

In (A24), \( W(\quad ; \quad) \) is a Racah coefficient and the lower-case Latin letters denote the angular momenta while the Greek letters stand for the magnetic quantum numbers associated with the \( a, b, \) and \( d. \)

After considerable manipulation, involving the application of the above formul\( i \), rearrangement of arguments, etc., the sum over \( m_a \) in (A20) can be brought to the form

\[ \sum_{m_a} (-1)^{M_1-m_a+m_b'} C(L_{1}\text{L}_{1}^{1}\nu_{1}; m_{a}, m_{a}^{'}-m_{a}, m_{b}^{'}-m_{b}) C(L_{2}\text{L}_{2}^{2}; m_{a}, m_{a}^{'}-m_{a}, m_{b}^{'}-m_{b}) \]

\[ \times C(L_{3}\text{L}_{3}^{3}; m_{b}^{'}-m_{a}, m_{a}^{'}-m_{a}, m_{b}^{'}-m_{b}) \]

\[ = (-1)^{M_1+m_b'-L_1-L_1^{'}+\nu+I_a} (2I_b+1) W(L_{b}\text{L}_{b}^{b}\nu_{1}; L_{1}\nu_{1}; I_a) \]

\[ \times C(L_{b}\text{L}_{b}^{b}\nu_{1}; m_{b}^{'}-m_{a}, m_{a}^{'}-m_{a}, m_{b}^{'}-m_{b}). \]  \hspace{1cm} (A25)

Following Rose (39, p. 173), we now set \( M_{1}' = M_1 + T_1 \) and define the parameter

\[ c_{\nu_1 T_1} = S_1 \sum_{M_1} (-1)^{L_1-M_1} A_{L_1, -M_1} A^*_{L_1', -M_1'} x C(L_{1}\text{L}_{1}^{1}\nu_{1}; M_{1}', -M_{1} -T_{1}, -T_{1}) \]  \hspace{1cm} (A26)
We note that the $c_{\nu T}$ do not involve the projection quantum numbers associated with the angular momenta of the nuclear states involved in the transitions, but depend only on the properties of the emitted radiation ($\gamma_1$ in the present discussion). In terms of the $c_{\nu T}$ (A20) becomes

$$
\Omega_{m_b m'_b}^{(1)} = (2I_b + 1) \sum_{L_1, L'_1, \nu_1, \tau_1} (-1)^{m'_b - L'_1 + I_a + \nu_1} \langle L_1 || T_{L_1} || I_a \rangle \langle I_b || T_{L_1} || I_a \rangle^* 
\times c_{\nu_1 \tau_1} W(I_b L_1 L'_1 \nu_1 I_a) C(L_1 I_b \nu_1 ; m_b, -m'_b, m_b - m'_b) D_{m_b m'_b \tau_1}^{(\nu_1 \tau_1)} 
$$

(A27)

With regard to the parameter $\tau_1 = M_1 - M'_1$, it is seen that $\tau_1 = 0$ when only the propagation direction of the radiation is observed. This follows from the fact that the rotation which takes $k_1$ into $\hat{n}$ involves only the specification of the two Euler angles needed to specify a direction in space. Only when the complete orientation of the reference frame (including both the propagation direction $k_1$ and a polarization direction) is required, will the third Euler angle enter. Since we do not observe the polarization direction here, the sum over the polarization states $M_1$ can be performed incoherently. That is, in (A26) the sum over $M_1$ will not contain any interference terms involving $M_1$ and $M'_1$. Thus, (A27) becomes
\[
\Omega^{(1)}_{m_b m_b'} = \sum_{\nu_1} (-1)^{m_b'} C(I_{b b}; m_b', -m_b', m_b, -m_b') \cdot \sum_{\nu_1} D_{m_b - m_b', 0} B_{\nu_1}^{(1)}
\]

where we have introduced the factor \( B_{\nu_1}^{(1)} \) to include all of the factors in (A27) which are independent of the projection quantum numbers.

Finally, the general angular correlation function is obtained by substituting for \( \Omega^{(1)}_{m_b m_b'} \) in (A12) the right side of (A28) and making a similar substitution for \( \Omega^{(2)}_{m_b m_b'} \). The result is

\[
W(k_1, k_2) = \sum_{\nu_1, \nu_2} \sum_{m_b} \sum_{s} (-1)^{s} B_{\nu_1}^{(1)} B_{\nu_2}^{(2)} C(I_{b b}; m_b, s - m_b, s) \cdot \sum_{\nu_1} D_{a_1, 0}^{(1)} D_{a_2, 0}^{(2)}
\]

with \( s = m_b - m_b' \). Keeping \( s \) fixed and performing the sum over \( m_b \) gives simply \( \delta_{\nu_1, \nu_2} \) so that

\[
W(k_1, k_2) = \sum_{\nu} \sum_{s} (-1)^{s} B_{\nu}^{(1)} B_{\nu}^{(2)} D_{a_1, 0}^{(1)} D_{a_2, 0}^{(2)}
\]

Consider now the sum over \( s \). Each rotation matrix can be written in terms of a spherical-harmonic (39, p. 60), i.e.,
\[
\nu 
\begin{align*}
D\left(\alpha_1, \beta_1, 0\right) &= \left(\frac{4\pi}{2\nu + 1}\right)^{1/2} Y^*_{\nu, \beta_1, \alpha_1} \\
D\left(\alpha_2, \beta_2, 0\right) &= \left(\frac{4\pi}{2\nu + 1}\right)^{1/2} Y^*_{\nu, -s, \beta_2, \alpha_2} = \left(\frac{4\pi}{2\nu + 1}\right)^{1/2} (-1)^{-s} Y_{\nu, \beta_2, \alpha_2}
\end{align*}
\]

and the sum over \(s\) becomes

\[
\sum_s (-1)^s \nu D\left(\alpha_1, \beta_1, 0\right) \nu D\left(\alpha_2, \beta_2, 0\right) = \frac{4\pi}{2\nu + 1} \sum_s Y^*_{\nu, \beta_1, \alpha_1} Y_{\nu, \beta_2, \alpha_2} = P(\cos \theta)
\]

from the spherical-harmonic addition theorem. This shows, as expected, that the angular correlation is independent of the direction of the arbitrarily chosen quantization axis and depends only on \(\theta\), the angle between the propagation directions \(\vec{k}_1\) and \(\vec{k}_2\). This result can be obtained in a slightly different way which provides more insight into the significance of the rotations. Denoting the Euler angles \(\alpha_1, \beta_1, 0\) of the first rotation collectively by \(R_1\) and those of the second rotation by \(R_2\), and using the unitary property of the \(D\)-matrices, we have

\[
\sum_s (-1)^s \nu D\left(R_1\right) \nu D\left(R_2\right) = \sum_s \nu D\left(R_2^{-1}\right) \nu D\left(R_1\right) = \sum_s \nu D\left(R_2^{-1} R_1\right)
\]

\[
= P(\cos \theta)
\]

from (39, p. 74). Now \((R_2^{-1} R_1)\) is the rotation which first carries
\( \vec{k}_1 \) into \( \vec{n} \) and then \( \vec{n} \) into \( \vec{k}_2 \). The net effect is just the rotation of \( \vec{k}_1 \) through the angle \( \theta \) into \( \vec{k}_2 \).

The angular correlation function is now reduced to the simple expression

\[
W(\vec{k}_1, \vec{k}_2) = W(\theta) = \sum_{\nu} B_{\nu}(1) B_{\nu}(2) P_{\nu}(\cos \theta).
\]

We recall that the factors \( B_{\nu} \) do not depend upon the projection quantum numbers and for any \( \gamma_1 - \gamma_2 \) pair are independent of the propagation directions \( \vec{k}_1 \) and \( \vec{k}_2 \). That is, for a particular gamma-ray cascade, the \( B_{\nu} \)'s are constants. We are, of course, still interested in these factors since they contain the \( L \) and \( I \) dependence that characterizes the gamma-ray transitions, e.g., the angular momentum quantum number \( L \) defines the multipolarity of the gamma-ray. For pure multipole transitions \( (L_1 = L_1' \) and \( L_2 = L_2') \), the \( B_{\nu} \)'s are proportional to

\[
B_{\nu}(1) = F_{\nu}(L_1 I_a I_b) \quad \text{and} \quad B_{\nu}(2) = F_{\nu}(L_2 I_c I_b)
\]

where

\[
F_{\nu}(L_1 I_a I_b) = (-1)^{I_a - I_b - 1}(2I_b + 1)^{1/2}(2L_1 + 1)C(LL_1; 1, -1, 0) W(L_1 I_a L_1 I_b; \nu I_a)
\]

and similar for \( F_{\nu}(L_2 I_c I_b) \). The index \( \nu \) takes on even-integer
values and \( \nu_{\text{max}} \) is the smallest of the integers \( 2l_b, 2l_1, 2l_2 \). The final expression for the angular correlation is simply

\[
W(\theta) = \sum \nu A_{\nu} P_{\nu}(\cos \theta) \quad \text{(A32)}
\]

where \( A_{\nu} = F(L, I_a, I_b) F(L, I_c, I_b) \).

Tabulations of the \( F \)-factors are available so that the coefficients \( A_{\nu} \) can be calculated from a knowledge of the spins of the levels and the multipolarities of the radiations. Conversely, an experiment in which the correlation function is measured can yield information about these parameters.

A quantity of frequent value in experimental work is the anisotropy \( A \) defined as

\[
A = \frac{W(180^\circ) - W(90^\circ)}{W(90^\circ)} \quad \text{(A33)}
\]