ANGULAR CORRELATION OF CASCADED
GAMMA RADIATIONS FROM ORIENTED NUCLEI

DISSERTATION

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By

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PREFACE

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Richard C. Sapp

Columbus, Ohio
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INTRODUCTION

Application of the techniques for producing very low temperatures to the problem of achieving nuclear orientation yields information of current interest in both solid state and nuclear physics. This dissertation is intended to demonstrate how orientation may be achieved, how it may be observed, what information may be reasonably deduced, and the limitations of this approach, by applying it to a specific example.

Adiabatic demagnetization of a suitable paramagnetic salt containing the nucleus of interest makes it possible to achieve a considerable degree of orientation at the extremely low temperatures thus reached. It is necessary that the parent nucleus possess an electric or magnetic moment, which can interact with local electric or magnetic fields. If the nuclear properties are known, details of the orientation mechanism may be deduced. A cryostat has been assembled in which single crystals of cerium magnesium nitrate may be mounted so that small heat leaks are achieved. Adiabatic demagnetizations may be made from initial conditions of 24 kilogauss and 10K.

Properties of the gamma rays emitted by the excited states of the daughter nucleus produced by beta-decay or electron capture give knowledge about the spins and parities
of the nuclear states involved, as well as indirectly about the ground state of the parent nucleus and the beta-decay. Angular correlation and polarization-direction correlation, as well as internal conversion coefficients, depend on the same nuclear parameters. Also, the nuclear spin and moment of the parent state may be evaluated if the orientation mechanism is understood; this can also be obtained from magnetic resonance and atomic beam experiments. The g-factor may be obtained from the dependence of angular correlation on applied magnetic field.

Observation of the gamma radiations from excited states of nickel 60 formed by the beta-decay of the 5.2-year cobalt 60 ground state (5.2-year half life) permits the study of the mechanism of nuclear orientation, as well as the testing of the nuclear theory applicable to this case. For this purpose, high resolution scintillation counters have been employed with associated equipment capable of measuring the angular correlation as well as the angular distributions of the gamma rays.

In the succeeding chapters, methods of producing nuclear orientation are reviewed (Chapter I) and the theory of gamma emissions from oriented nuclear systems is summarized for the specific case of cobalt 60 (Chapter II). After the details of the apparatus are given in Chapter III, experimental techniques and data on the angular distributions and correlation of the nickel 60 gamma rays.
are presented (Chapter IV). Then the results are discussed in relation to existing information on cobalt 60 in the final chapter. A complete bibliography on nuclear orientation is also included, as well as some of the most important references on cobalt 60.
CHAPTER I

PRODUCTION OF NUCLEAR ORIENTATION

1. Introduction of Concepts and Nomenclature.

Certain terms are used in a technical sense in the following discussion. "Orientation" is the general term referring to any situation in which the directions of the nuclear spins are not randomly distributed in space. As special cases there are "polarization," which occurs when a single preferred direction in space exists and a resultant magnetic moment is imparted to the nuclei, and "alignment," in which case the nuclei tend to parallel a preferred axis without regard to direction along this axis. In the latter circumstance a two-fold degeneracy remains with respect to the magnetic substates of the nuclei, corresponding to positive and negative directions being equally favored energetically.

The degree of orientation achieved by the application of any of the methods discussed in the succeeding sections may be readily deduced, in the case of radioactive nuclei, from the departure from spherical symmetry of the pattern of gamma emissions by the daughter nucleus; the specific relationships will be taken up in Chapter II.

All methods of producing nuclear orientation depend on the existence of a splitting $\Delta E$ of the degenerate
energy level of a nucleus with spin quantum number $I$ into magnetic substates characterised by quantum numbers $m_I = I, I-1, \ldots, -I$. Then at sufficiently low temperatures these sublevels are unequally populated according to the quantum Maxwell-Boltzmann statistics. For an ensemble of $N$ nuclei in equilibrium at temperature $T$, the number in state $m$ (with degeneracy $w_m$) is

$$N(m) = \left( \frac{N}{Z} \right) w_m \exp \left[ -\frac{E(m)}{kT} \right] ,$$

where the partition function

$$Z = \sum_{m} w_m \exp \left[ -\frac{E(m)}{kT} \right] .$$

The relative populations of neighboring substates is

$$\frac{N(m)}{N(m+1)} = \frac{w_m}{w_{m+1}} \exp \left[ \frac{E(m+1) - E(m)}{kT} \right]$$

and the requirement for appreciable nuclear orientation is that $\Delta E = |E(m) - E(m+1)| \approx kT$; this corresponds to an ordering of the nuclei with respect to some axis in space.

Stationary states with non-equilibrium nuclear populations, such as are produced by resonance methods, will be discussed at the end of this chapter.

For discussing the orientation mechanisms in cases of interest it is useful to consider the Hamiltonian for a paramagnetic ion with hyperfine structure in a crystalline field of axial symmetry, which is shown by Abragam and Pryce\(^1\) to be, with the $z$-axis as the axis of symmetry,

\[ H = g_{\parallel} B_z + g_{\perp} B_y \] 
\[ + D \left[ S_z^2 - (1/3)S(S+1) \right] + a S_z I_z + b (S_x I_x + S_y I_y) \] 
\[ + Q \left[ I_z^2 - (1/3)I(I+1) \right] - g_n \mu_B \mathbf{B} \cdot \mathbf{I} + G(SS'). \]

Here the electronic \( g \)-factor is a tensor with principal values \( g_{\parallel} \) and \( g_{\perp} \); \( B_x, B_y, B_z \) are components of the magnetic field acting on the electronic system; the D-term results from the interaction between the electrons and the crystal field. An effective electron spin \( S \) is defined by setting the multiplicity of the electronic energy levels equal to \( 2S+1 \); for example, \( S = \frac{1}{2} \) for an ion exhibiting Kramers degeneracy at low temperatures.

The terms with coefficients \( a \) and \( b \) are due to the hyperfine coupling between the nucleus and the magnetic field of an unfilled electron shell, while nuclear electric quadrupole coupling with the electric field gradient at the nucleus is represented by the Q-term. The direct effect of an external field \( B \) on the nuclear magnetic moment is also included, with \( g_n \) the nuclear gyromagnetic ratio (\( g \)-factor) equal to \( \mu_n / I \) and \( \mu_n \) the nuclear magneton; \( I \) is the spin angular momentum, in units \( \hbar \), with \( I_z \) equal to \( I \).

Finally, a term \( G(SS') \) is included to account for electronic coupling between neighboring ions, such as magnetic dipole-dipole and exchange interactions; these are usually negligible at higher temperatures, but would become the dominant terms in the Curie region of the salt where
ordering of the electronic moments takes place.

2. **External Field Polarization.**

The first method of orienting nuclei to be proposed arises from discussions of nuclear paramagnetism by Gorter\(^2\) and by Kurti and Simon.\(^3\) A two-stage adiabatic demagnetization with the nuclei as the second stage was proposed, the success of which would depend on the attainment of a sufficiently low temperature and strong external field at the nuclear stage; the final low temperature reached would then be limited by the small internuclear interactions, and should be of the order of \(10^{-5}\) to \(10^{-6}\) degrees Kelvin. Whether the ordering at the lowest temperature would be ferromagnetic or antiferromagnetic in nature is not yet certain; this question has been discussed by Frohlich and Nabarro\(^4\) and more recently by Ruderman and Kittel\(^5\) for the case of metals.

Quantitative considerations of the possibility of producing nuclear polarization in a metal by a strong static magnetic field have been given by Simon.\(^6\)

\(^6\) F.E.Simon, "Le Magnetisme" **3**, 1 (Strasbourg Conference, 1939).
The amount of nuclear entropy which can be removed by magnetizing at temperature $T$ is
\[ \frac{\Delta S_n}{R} \approx 2.15 \times 10^{-16} g_n \mu_n^2 \frac{(I+1)}{I} \left( \frac{B}{T} \right)^2 + \ldots \ldots \; (5) \]

Thus for practicable external fields of 50 KG, say, a temperature of about $0.01^\circ K$ would be needed for a 20-30\% entropy reduction; see Figure 1.

The important term in the hamiltonian is $-g_n \mu_n \vec{B} \cdot \vec{I}$. The $(2I+1)$ nuclear energy levels are given by
\[ E(m_I) = -\mu \cdot \vec{B} = -g_n \mu_n B m_I \; . \; (6) \]

They are equally spaced by an amount
\[ \Delta E = \left| E(m_I) - E(m_{I+1}) \right| = g_n \mu_n B \; . \; (7) \]

The populations are
\[ \frac{N(m_I)}{N} = \frac{1}{Z} \exp \left[ \frac{g_n \mu_n B}{kT} m_I \right] \; . \; (8) \]
These levels are sketched in Fig. 2. The $m_I = +I$ or $-I$ level lies lowest depending on whether the algebraic sign of the nuclear magnetic moment is positive or negative. These levels and their corresponding space orientations are given in the accompanying diagrams for $I = 7/2$, as in cobalt 59.

The degree of nuclear polarization is specified by the first moment of the population distribution over the magnetic substates:

$$f_N = (1/I) \sum_{m_I} m_I \exp \left[ -E(m_I)/kT \right]/\sum_{m_I} \exp \left[ -E(m_I)/kT \right].$$

The leading term for small degrees of polarization is

$$f_N \approx (I + 1) g n \mu B / 3kT.$$  \quad (9)

It is possible to define an orientation parameter

$$\beta = \Delta E/kT = \mu B/kTI = g n \mu B / kT$$  \quad (10)

for all cases of equally spaced magnetic energy levels.

Then the relative populations are proportional to $\exp(\beta m)$, and the condition for an appreciable amount of nuclear orientation is that $\beta$ be of the order of unity.
Now

\[ \beta = 3.65 \times 10^{-8} g_n B/T \]  \hspace{1cm} (11)

and with \( g_n \) approximately 1, the requirement becomes

\( B/T \) of the order of \( 10^7 \) to \( 10^8 \) gauss/degree. At \( .01^\circ K \),

external magnetic fields of at least 100 kilogauss would be required, in agreement with Simon's considerations.

Due to these stringent field and temperature requirements, the external field polarization method has not been experimentally successful as yet. Attempts by the Oxford group have been reported by Kurti\(^7\), in which 500 fine copper wires were embedded in chrome alum to provide thermal contact with a cobalt salt; the latter was in a field of 50 kilogauss supplied by a water-cooled solenoid. The lowest temperature achieved at the nuclear stage was \(.05^\circ K\), after demagnetization of the chrome alum.

It is interesting to note that if the technical difficulties could be overcome and considerable nuclear polarization produced by this method, then a sufficiently reversible nuclear demagnetization might lead to a nuclear ordering at the very low Curie point where nuclear interactions become important. A distinct advantage of the Simon method is that it may be applied to any nucleus.

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8) N. Kurti, International Conference on Low Temperature Physics, Houston, Texas (December, 1953).
3. **Magnetic Hyperfine Structure Polarization.**

It was proposed independently by Gorter\(^9\) and by Rose\(^10\) that the very strong magnetic field \(10^5\) to \(10^6\) gauss) at the nucleus due to the unpaired electrons of a paramagnetic ion exhibiting considerable hyperfine structure (HFS) would be sufficient to polarize the nuclei if the electronic moments are first oriented by a small external magnetic field \(100\) to \(1000\) gauss) applied at a very low temperature. The method's most obvious limitation is that the nucleus to be studied must belong to a paramagnetic ion.

Gorter's suggestion was to use a mixed crystal or two salts in thermal contact for cooling the nuclear spin system to low temperatures near \(0.1^\circ\)K. Rose put forth the idea of using a paramagnetic salt containing the nucleus considered, hence making the system "self-cooling;" that is, demagnetization of the electronic system from \(1^\circ\)K and strong field to a relatively small final field provides sufficient cooling, the residual field and the HFS limiting the final temperature but at the same time supplying the orientation mechanism.

The appropriate terms in the Hamiltonian (4), if it is assumed that the external field is applied along the

\(^9\) C. J. Gorter, Physica 14, 504 (1948).
crystal axis and the HFS is isotropic, are $g \mu_B S_z + a \vec{I} \cdot \vec{S}$. The energy levels depend on the strength of the applied field, and in the usual cases approximate those for a weak field Zeeman effect. These are shown in Figure 3 for the case of $S = \frac{3}{2}$, $I = 3/2$, to show the behavior.

Figure 3

Energy Levels of a Paramagnetic Salt
in an External Field

Steenberg\textsuperscript{11)\textsuperscript{11}} has calculated the populations for the Gorter-Rose method and finds

\begin{equation}
N(m_I)/N = D \exp \left[ \frac{(1+q)a}{4kT} \right] + D' \exp \left[ \frac{(1-q')a}{4kT} \right] + E \exp \left[ \frac{(1-q)a}{4kT} \right] + E' \exp \left[ \frac{(1+q')a}{4kT} \right],
\end{equation}

where \( q(m_I) = \left[ \left( \frac{2g_{ij} \mu_B}{a} B \right)^2 + \frac{4g_{ij} \mu_B B}{a} \left( 2m_I - 1 \right) + (2I+1)^2 \right]^{1/2} \)

\( q'(m_I) = q(m_I + 1) \quad \text{(12)} \)

\[ D(m_I) = \frac{2g_{ij} \mu_B}{a} (2m_I + 1) / 4q \]

\[ D'(m_I) = D(m_I + 1) \]

\[ E(m_I) = \left[ (I+m_I)(I-m_I+1) \right] \left/ \left[ q \left( \frac{2g_{ij} \mu_B}{a} + 2m_I - 1 + q \right) \right] \right. \]

\[ E'(m_I) = E(m_I + 1) \]

If the electronic structure were indeed saturated, so that all the individual HFS fields were parallel to the external field, equations (7)-(10) could be applied directly with \( B \) determined from the HFS (\( \cdot 0.1 \text{ cm}^{-1} \)). If \( f_e \) is the degree of electronic polarization, defined analogously to \( f_N \), it can be shown\(^{12}\) that for \( S = \frac{1}{2} \) and \( kT \sim a/2 \),

\[ f_N \approx f_e \left[ \frac{(I+1)/3}{(a/2kT)} \right] + \ldots \quad \text{(13)} \]

This approximate formula gives, for \( kT \approx a/2 \) and \( f_e \approx 1 \), \( f_N \approx 0.2 \). Simon et al\(^{12}\) give correction terms in the next order of approximation.

First attempts by the Leiden group to apply the method to iron-59 in iron ammonium alum gave inconclusive results;\(^{13}\) probably due to insufficient cooling of the salt in the partial demagnetization. However, the feasibility of the method has been amply demonstrated since; first by the Oxford group in a study of cobalt-60 (and recently manganese).


\(^{13}\) Gorter, De Klerk, Poppema, Steenland, De Vries, Physica 15, 679 (1949).
54), and currently in the present work. In each case the radioactive nuclei are incorporated in single crystals of cerium magnesium nitrate.\textsuperscript{14,15} In addition to a lack of hyperfine structure in the cerium ions, this salt has the very important advantage of a highly anisotropic electronic $g$-factor, which permits the application of a polarizing field along the direction of minimum susceptibility with little consequent heating of the crystal. Although the ionic interactions play a by-no-means negligible part in these experiments, impressive degrees of orientation were attained by use of rather moderate externally applied fields. Some questions raised by the results of Ambler et al.\textsuperscript{14} concerning the orientation mechanism will be taken up in connection with the present results in Chapter V. Magnetic HFS polarization has also been successfully used at Oak Ridge for the orientation of stable manganese-55 in manganeseous ammonium sulfate\textsuperscript{16} to study the spin orientation dependence of neutron absorption\textsuperscript{17}.

It may be possible to produce Gorter-Rose orientation through use of the fields due to impurity atoms or color centers having sufficient HFS coupling with adjacent nuclei.

\textsuperscript{14} Ambler, Grace, Halban, Kurti, Durand, Johnson, Lemmer, Phil. Mag. 44, 216 (1953).
\textsuperscript{15} Grace, Johnson, Kurti, Lemmer, Robinson, Phil. Mag. 45, 1192 (1954).
\textsuperscript{17} Bernstein, Roberts, Stanford, Dabbs, Stephenson, Phys. Rev. 94, 1243 (1954).
Daunt mentioned this possibility specifically with regard to F-centers, which show an $\vec{I} \cdot \vec{S}$ coupling with their six neighboring alkali nuclei. Two possible procedures exist; one might cool the color centers in a magnetic field to very low temperatures by contact with a paramagnetic salt, in which case the trapped electrons would become polarized. Alternatively, it might be feasible to partially demagnetize the F-centers themselves to a final low temperature determined mainly by the HFS. In either case severe technical difficulties exist, as the centers are very dilute and the couplings are rather small. Also, it would be difficult to predict the lowest energy configuration of the nuclear spins; the application to the study of radioactive nuclei introduces the further complication that the nucleus of interest must be in the neighborhood of the defect (these latter difficulties are somewhat less restrictive in the case of an electron trapped at an impurity, e.g. a donor impurity in a semiconductor at low temperatures).

4. **Electric Hyperfine Structure Alignment.**

A method which avoids the necessity of a magnetic field or of a paramagnetic salt has been put forward by Pound.

It makes use of the interaction between the nuclear electric quadrupole moment and the local gradient of the crystalline electric field. If the crystal field has axial symmetry, \( m_I \) is a good quantum number, but levels \( \pm |m_I| \) are not split.

The Hamiltonian term here is \( Q \left[ I_z^2 - (1/3)I(I+1) \right] \), where \( Q = \frac{3(eg)}{4I(2I-1)} \left( \frac{\partial E_z}{\partial z} \right) \) with \( (eg) \) the quadrupole moment and \( (\partial E_z/\partial z) \) the axial electric field gradient. Then

\[
E(\pm m_I) = -Q \left[ m_I^2 - (1/3)I(I+1) \right]
\]

and

\[
\Delta E(|m_I|) = |E(|m_I|) - E(|m_I|+1)| = Q (2|m_I|+1).
\]

\[
N(|m_I|)/N = \frac{2}{Z} \exp \left[ Q m_I^2 / kT \right]
\]

\( m_I = \pm 1/2, \pm 3/2 \)

\( I = 7/2 \)

\( \pm 5/2 \)

\( 6Q \)

\( \pm 7/2 \)

The energy level of largest \( |m_I| \) is lowest if the quadrupole coupling \((eg) (\partial E_z/\partial z)\) is positive, and highest if it is negative. An example is drawn in Figure 4 for positive coupling and \( I = 7/2 \). The expected degree of nuclear
alignment $\Delta N$ is given for $\Delta E < kT$ by an approximate expression:

$$N = \frac{1}{I^2} \left[ \sum_m m^2 \frac{N(m)}{N} / \sum_m \frac{N(m)}{N} - \frac{1}{3} I(I+1) \right]$$

$$\approx \frac{4(I+1)}{15 I} \left( \frac{q}{kT} \right) [I(I+1) - 3/4] + \ldots$$

For the maximum quadrupole splittings expected $(10^2 - 10^3$ Mc/sec), the single crystal must be cooled in contact with a paramagnetic salt or with paramagnetic ions in a mixed crystal. Molecular crystals with $p$-type covalent bonds are predicted to have a very large gradient at the nucleus. Hence the biggest limitation to the method is finding a favorable combination of nucleus and crystal with sufficient quadrupole splitting.

Attempts to effect a Pound alignment were reported by Daniels and Kurti. The iodine-131 nucleus, $I=7/2$, is believed to have a large quadrupole moment. Such crystals as $p$-C$_6$H$_4$I$_2$ and $(p$-IC$_6$H$_4$SO$_3$)(Co,Zn)$\cdot$6H$_2$O were employed, but no alignment was detected for temperatures of $0.05^\circ$K. Failure was attributed to insufficient thermal contact between the nuclear spin system, the lattice, and the paramagnetic ions.

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Existence of highly anisotropic hyperfine structure in certain paramagnetic salts, notably those of copper and cobalt, led Bleaney to suggest a different mechanism for alignment.\(^26\) The terms \(aS_x I_z + b (S_x I_x + S_y I_y)\) lead to \((2I+1)\) doublets as shown in Figure 5 for the two limiting cases \(a \gg b\) and \(a \ll b\) for \(S = \frac{3}{2}, I = \frac{7}{2} \).\(^27\) The former circumstance is seen to cause axial alignment, while the latter leads to alignment more or less in a plane perpendicular to the symmetry axis of the ion. The level spacings and demagnetization properties of a salt with \(a \ll b\) are considerably less favorable for the production of high degrees of alignment. For \(a > b\), the relative population of any two-fold degenerate sublevel has been calculated by Steenberg.\(^28\) In case \(a \gg b\),

\[
N(\frac{|m|}{N}) = \frac{2}{Z} \cosh \left[\frac{a|m|}{2kT}\right]
\]

(18)

With \(S = \frac{3}{2}\), no crystal field effects exist, and the degree of alignment is given by \(^29\)

\[
\Delta N \approx \frac{2}{45} \left(\frac{I+1}{I}\right) \left(\frac{a}{2kT}\right)^2 \left[I(I+1)-\frac{3}{4}\right] + \ldots \ldots
\]

(19)

If \(b < a\) but not negligible, mixing of adjacent levels shifts the energies slightly and causes \((a/2kT)^2\) to be

\(^{27}\)B. Bleaney, Phil. Mag. 42, 441 (1951).
\(^{29}\)Simon, Rose, Jauch, Phys. Rev. 84, 1155 (1951).
Figure 5
Energy Levels in a Paramagnetic Ion with Anisotropic HFS (Bleaney).

\[ a \gg b \]

\[ \pm \frac{1}{2} \]

\[ \pm \frac{3}{2} \]

\[ \pm \frac{1}{2} \]

\[ \pm \frac{1}{2} \]

\[ \pm \frac{3}{2} \]

\[ \pm \frac{5}{2} \]

\[ \pm \frac{7}{2} \]

\[ a \ll b \]

\[ \pm \frac{1}{2} \]

\[ \pm \left( \frac{1}{2}, \frac{3}{2} \right) \]

\[ \pm \left( \frac{3}{2}, \frac{5}{2} \right) \]

\[ \pm \left( \frac{5}{2}, \frac{7}{2} \right) \]

\[ \pm \left( \frac{3}{2}, \frac{5}{2} \right) \]

\[ \pm \left( \frac{5}{2}, \frac{7}{2} \right) \]
replaced by \((a^2-b^2)/(2kT)^2\) in equation (19).

If \(|D| \gg a, b\) and \(S > \frac{1}{2}\), it is still possible to produce alignment, since the crystal field splitting will make one of the two-fold degenerate electronic levels lie lowest, HFS producing a set of \(2(2I+1)\) nuclear levels arranged as before, with splitting \(a/2\).

When a small external magnetic field is applied along the crystal axis, nuclear polarization may be obtained even with \(a \gg b\). Then the doublets are split by an amount \(g_\|/\mu_B B\), and a small net moment can be imparted to the nuclear system; the degree of polarization is

\[
f_N = \frac{1}{I} \left[ \sum_{|m_I|=0,1} m_I \sinh \left( \frac{a|m_I|}{2kT} \right) \right] \frac{\sum_{|m_I|} \cosh \left( \frac{a|m_I|}{2kT} \right)}{\sum_{|m_I|}} \tanh \left( \frac{g_\|/\mu_B B}{2kT} \right). \tag{20}
\]

The effective mechanism in Bleaney's method may be pictured as a two-stage process, in which the crystalline field gradient causes the electronic moments to become aligned; then the hyperfine fields of the ions in the crystal are no longer randomly oriented with respect to the z-axis. This requires a single crystal, but has the advantage that the demagnetization is made to zero field.

In terms of entropy, it is easily seen that the initial electronic entropy of \(\text{Rln2}\) may be almost completely removed at \(1^\circ\text{K}\) by a strong field; the nuclear entropy of \(\text{Rln (2I+1)}\) is practically unchanged. After an adiabatic demagnetization to zero field, the electronic entropy

\[30\)B. Bleaney, Phil. Mag. 42, 441 (1951).\]
is $R \ln 2$ and the nuclear entropy will be reduced by just this amount at the final low temperature.

Magnetic HFS alignment yielded the first experimental success, being applied at Oxford to a mixed Tutton salt, $(\text{Co }1\%, \text{ Cu }12\%, \text{ Zn }87\%) \text{Rb}_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ containing about 50 microcuries of cobalt 60.$^{31)}$ A similar experiment was performed at Leiden using diluted cobalt ammonium sulfate containing 90 microcuries of cobalt 60.$^{32),33)}$ Alignment of cobalt 58 nuclei has also been reported using the same experimental techniques.$^{34)}$ The entire nuclear orientation programs of the Oxford and the Leiden laboratories have been reviewed.$^{35),36)}$ Recent work at the National Bureau of Standards with cerium-141 and neodymium-147 in cerium magnesium nitrate involved Bleaney alignment with $b \gg a.$$^{37)}$

6. **Antiferromagnetic Alignment.**

It was suggested by Daunt$^{38)}$ and by Gorter$^{39)}$ that the nuclei in an antiferromagnetic crystal might be aligned

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$^{32)}$Gorter, Poppema, Steenland, Beun, Physica 17, 1050 (1951).
$^{33)}$Gorter, Tolhoek, Poppema, Steenland, Beun, Physica 18, 357 (1952).
$^{34)}$Daniels, Grace, Halban, Kurti, Robinson, Phil. Mag. 43, 1297 (1952).
by HFS coupling with the ionic moments when the salt was cooled below its Neel point. The model of the antiferromagnetic state is one of interpenetrating sublattices arranged in such a way that the macroscopic ionic moment vanishes. A distinction needs to be made between this method and that of Bleaney. Here the aligned state is two-fold degenerate in direction but ordered with respect to lattice position, rather than randomly distributed.

An attempt at Leiden to observe such an alignment using a concentrated cobalt ammonium sulfate crystal having a Neel point near 0.1°K \(^{40}\) was inconclusive. The null result was tentatively attributed to the model of an antiferromagnet being a non-static one, rapid interchanges taking place among the sublattices. If the time for a nuclear spin to come to equilibrium in the HFS field were much longer than the time between ionic reversals, no nuclear alignment would result on the average. \(^{41}\)

7. **Resonance Methods for Producing Nuclear Orientation**.

Important methods of orienting nuclei have been proposed, in which a stationary state which is not a state of thermodynamic equilibrium is produced by electronic spin resonance of systems in which I·S coupling exists. These will be mentioned only briefly here, along with the

\(^{41}\)O. J. Poppema, Thesis (Groningen, 1954).
references of interest, since they do not necessarily involve the use of low temperatures.

A mechanism by which both angular momentum and energy can be conserved in the simultaneous reversal of an electron spin and a nuclear spin which are coupled has been studied in detail by Overhauser.\(^\text{42), 43, 44}\) Considerations of nuclear relaxation mechanisms led him to predict that a nuclear polarization would be produced by saturating the electronic resonance in a metal. Further, the nuclear spin substate populations would be enhanced as if the nuclear g-factor were equal to that of the electrons.\(^\text{44, 45}\) The Overhauser effect has been experimentally confirmed by Carver and Schlicter.\(^\text{46}\)

Korringa\(^\text{47}\) and Bloch\(^\text{48}\) independently suggested that the Overhauser effect might also be observed in paramagnetic salts, and this, too, was soon observed.\(^\text{49}\) In addition, an application of the idea to the saturation of the electron resonance in F-centers was made by Korringa.\(^\text{50}\)

Another experiment to polarize nuclei has been reported

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\(^{44}\)A. W. Overhauser, Phys. Rev. 92, 411 (1953).
\(^{45}\)C. Kittel, Phys. Rev. 95, 589 (1953).
\(^{48}\)F. Bloch, Physics Today 7, No. 4, 30 (April, 1954).
\(^{49}\)Beljers, Kint, Wieringen, Phys. Rev. 95, 1683 (1954).
\(^{50}\)J. Korringa, Phys. Rev. 94, 1388-9 (1954).
by Honig\textsuperscript{51}) and interpreted by Kaplan\textsuperscript{52}) Resonance at one of the hyperfine components of the electronic spin resonance of an arsenic-75 impurity in semiconducting silicon produced an enhancement of a neighboring HFS line at the expense of the line being resonated; hence by sweeping all but one of the HFS components a large nuclear polarization approaching 100\% could be expected. The method here depends on the existence of resolved HFS.

Kastler has considered nuclear orientation by resonance methods,\textsuperscript{53}) but his most interesting comments concern the possibility of orienting atoms by optical resonance with circularly polarized light.\textsuperscript{54}) In an atom the ground state of which is \textit{^1S_0}, \textit{F = I}, this would lead to nuclear polarization. Bitter, Lacy, and Richter have attempted this experiment with mercury-199, \textit{I = \frac{1}{2}}; a lack of positive results was attributed by them to experimental difficulties involving the imprisonment of the resonance radiation.\textsuperscript{55})

Resonance techniques seem promising as a means for producing oriented radioactive nuclei. Some advantage for studying excited states of these nuclei are (a) a new class of nuclei is opened by these experiments; (b) the degree of nuclear orientation may be held constant during the experi-

ment by fixing the incident power and/or the rate of sweeping the magnetic field, enabling one to obtain very good counting statistics if a gamma-ray pattern is being observed, for instance; (c) it may be possible to achieve very high degrees of polarization approaching nuclear saturation, depending on the detailed behavior of the nuclear spin reorientation processes; (d) some of the experiments may be done at room temperature, which simplifies the experimental apparatus in these cases. A possible disadvantage for the study of nuclear properties is the presence of the resonance magnet, but this may be overcome by using Helmholtz coils or by employing a light-pipe embedded in the pole-piece. A second disadvantage is that inconveniently large activities would be present when resonating nuclei with relatively short half-lives. To circumvent this problem, one might use a small amount of activity and detect the resonance by the anisotropy of gamma emissions, having first studied the electronic resonance in the ion containing the stable isotope.
CHAPTER II

GAMMA RADIATIONS FROM ORIENTED NUCLEI

1. **Theory of the Angular Distribution.**

After the first suggestion by Spiers\(^1\) that directional effects would be produced in the patterns of radiations from ensembles of oriented nuclei, the theory was rapidly developed by Steenberg \(^2\),\(^3\),\(^4\) and independently by Cox and Tolhoek.\(^5\),\(^6\),\(^7\) Both methods of approach are essentially the same; they involve a summation over the emissions from each of the nuclear magnetic energy substates, each term being weighted by the relative nuclear population of the initial level in the transition. If there is a series of emissions, e.g. the negatron decay of cobalt 60 followed by two gamma rays in cascade, the calculation may proceed step by step, with the substate populations at each level being determined from the previous transitions.

Since Cox and Tolhoek have made the only explicit calculation of angular correlation,\(^7\) their results will

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be employed consistently throughout and specialized for quadrupole radiations. The treatment they use is elegant in form, and the results may be stated in simple formulas. Steenberg's work can be extended to cover correlation in cascades; it may be more readily modified by the introduction of perturbations to the magnetic substates.

A simple expression can be given for the angular distribution of gamma rays under the following assumptions:

(a) Each nuclear state involved may be characterized by a definite spin (I) and parity (±);
(b) the radiation is a pure multipole of order \( L \); (c) the nuclei are oriented with respect to some axis of symmetry, which is taken as the axis of quantization (\( z \)-axis), and the relative nuclear populations of the initial states \( N(m_1)/N \) may be calculated. Then the probability of observing a gamma emission at an angle \( \Theta \) with respect to the axis of orientation is given by

\[
F(\Theta) = \sum_{m_1=-I_1}^{+I_1} \left[ N(m_1)/N \right] F_{I_1,m_1}(\Theta) \tag{1}
\]

where \( F_{I_1,m_1}(\Theta) \) is the distribution function for a single initial state.
The following normalizations are assumed:
\[ \frac{1}{4\pi} \int F(\theta) d\Omega = 1 \quad \sum_{m_i} \frac{N(m_i)}{N} = 1. \]

Now equation (1) can be expanded in a series of Legendre polynomials:

\[ F(\theta) = \sum_{k=0}^{k_{\text{max}}} A_{2k} P_{2k}(\cos \theta). \quad (2) \]

The coefficients \( A_{2k} \) are determined by the spins and multipole order involved in the transition and the relative populations of the nuclear sublevels. Maximum \( k \) is equal to \( L \) or to \( I_i \), whichever is smaller.

For a quadrupole transition \((L=2)\) with \( I_i \rightarrow I_i - 2 \), one obtains

\[ F(\theta) = 1 - \frac{15}{7} N_2 f_2 P_2 - 5 N_4 f_4 P_4, \quad (3) \]

where \( N_{2k} = 2^k \frac{I_i^{2k} (2I_i - 2k)!}{(2I_i)!} \quad (4) \)

and \( f_{2k} \) is the degree of orientation of order \( 2k \) (only even orders appear in directional distributions); it is a measure of the departure of the \( 2k \)th moment of the:

\[ 8) \text{H.A. Tolhoek and J.A.M. Cox, Physics 19, 101 (1953).} \]

\[ 9) \text{S. R. de Groot, Physica 18, 1201 (1952).} \]
nuclear ensemble from its value for random orientation.

Explicitly, \( f_0 = 1 \),

\[
f_1 = f_N = \frac{1}{1} \left[ \sum_m m N(m/N) \right],
\]

\[
f_2 = f_N = \frac{1}{1} \left[ \sum_m m^2 N(m)/N - (1/3)I(I+1) \right],
\]

\[
f_3 = \frac{1}{3^3} \left[ \sum_m m^3 N(m)/N - 1/5(3I^2+3I-1) \sum_m mN(m)/N \right],
\]

\[
f_4 = \frac{1}{4^4} \left[ \sum_m m^4 N(m)/N - \frac{1}{7}(6I^2+6I-5) \sum_m m^2 N(m)/N \right.
\]
\[
\left. - \frac{3}{35} I(I-1)(I+1)(I-2) \right].
\]

Figure 8

Behavior of the \( f_k \) (Poppema)

Poppema\(^{10}\) gives curves for \( f_1, f_2, f_3, \) and \( f_4 \) as functions of \( B = \mu B/kT_I \) and also of \( I \) (see Figure 8) for the case of equally spaced levels.

In the limit of complete orientation by any method, equation (3) becomes

\[
F(\theta) = \left( \frac{5}{4} \right)(1 - \cos^4 \theta)
\]

The polar diagram and pattern are shown for this case in Figure 9.

For comparison with experiment, it is convenient to use ratio \( F(0)/F(\pi/2) \) as a measure of the orientation.

The anisotropy of the radiation pattern, usually called \( \epsilon \) in the literature, is \( 1 - F(0)/F(\pi/2) \). The behavior of these quantities is sketched in Figure 10.

Figure 9

Radiation Pattern of Gamma Transition $I_1 \rightarrow I_{1-2}$

Figure 10

Typical Temperature-dependent Behavior of the Radiation Pattern Anisotropy
If the state of spin $I_i$ is formed by a preceding decay, it is possible to calculate the product $N^{\text{ft}}_{2k} f_{2k}$ from the product of the corresponding quantities for the preceding state\cite{11}, computed by equations (3), (4), (5) to (8). If the preceding transition is a beta-emission, the effect of the type of beta-decay is introduced in this step\cite{11}. See Figure 11.

Figure 11

Decay Scheme Involving Beta Decay

\[
\begin{array}{c}
I_o \\
\text{\gamma} \\
I_i \\
\text{\gamma} \\
I_f
\end{array}
\]

In order to specialize the radiation pattern formulae further, it is necessary to present the specific decay scheme to be considered; that of cobalt 60 is shown in Figure 12. Its present status is such that the details of the beta-decay are not completely established\cite{12},\cite{13}. It is sufficient to consider the following possible cascades: $5 \rightarrow 4 \rightarrow 2 \rightarrow 0$; $4 \rightarrow 2 \rightarrow 2 \rightarrow 0$.

The small branching of the beta-decay ($0.15\%$) will be neglected for the present; the cross-over transition is completely negligible. The angular distribution may be

\begin{itemize}
  \item \cite{11} J. A. M. Cox and H. A. Tolhoek, Physica 19, 673 (1953).
  \item \cite{12} M. Deutsch and G. Scharff-Goldhaber, Phys. Rev. 83, 1059 (1951).
  \item \cite{13} G. L. Keister and F. H. Schmidt, Phys. Rev. 93, 140 (1954).
\end{itemize}
Figure 12

Decay Scheme of Cobalt 60
FIGURE 12
COBALT 60 DECAY SCHEME
characterized by

\[ F(0) = 1 - 2.14 N_2 f_2 - 5.00 N_4 f_4 = F(n), \]  
\[ F(n/2) = 1 + 1.07 N_2 f_2 - 1.88 N_4 f_4. \]  

(10)

(11)

It is convenient to make use of the numerical calculations of Cox, De Groot, and Hartogh \(^{14}\) for cobalt 60 to plot \(F(n)/F(n/2)\) versus \(\beta\) for the various decay schemes (see Table I); the results are shown in Figure 13.

2. Gamma-Gamma Angular Correlation.

In case the nuclei emitting a gamma cascade are partially oriented along some axis in space, Cox and Tolhoek \(^{15}\) have shown that the gamma angular correlation function depends in general on three independent angles. These are the angles \(\theta_1, \theta_2\) between the orientation axis and the directions of the first and second emissions, respectively, and the angle \(\theta\) between the two emissions (see Figures 14 and 15). These angles need not lie in a common plane,

---

Table I

Theoretical $N\gamma f_2$ Coefficients for Angular Distribution and Correlation (Cox, de Groot, Hartogh)

<table>
<thead>
<tr>
<th>$\beta$</th>
<th>$N_2 f_2$</th>
<th>$I_0 = 5$</th>
<th>$I_0 = 4, I_\beta = 0$</th>
<th>$I_0 = 4, I_\beta = 1$</th>
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<table>
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<th>$N_4 f_4$</th>
<th>$I_0 = 5$</th>
<th>$I_0 = 4, I_\beta = 0$</th>
<th>$I_0 = 4, I_\beta = 1$</th>
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Figure 13

Theoretical Anisotropy of the Gamma Radiation Patterns for Nickel 60
FIGURE 13
THEORETICAL ANISOTROPY OF THE GAMMA RADIATION PATTERN
of course. A fundamental assumption is that the nuclear orientation is not disturbed during the lifetime of the intermediate state in the cascade.

Their very general results are reduced to a simple form, suitable for application to cobalt-60, in the case of a quadrupole-quadrupole cascade with stretched configuration $I_1 \rightarrow I_1 - 2 \rightarrow I_1 - 4$. Then the joint relative probability per unit solid angle that $\gamma_a$ and $\gamma_b$ are emitted in directions $\theta_1$ and $\theta_2$, respectively, is given by

$$W(\theta_1, \theta_2, \theta) = \frac{25}{4} \sum_{k=0}^{4} N_{2k} f_{2k} g_{2k}(\theta_1, \theta_2, \theta).$$

Here the coefficients $N_{2k}$ are as given in Section 1. The temperature dependence is contained in the orientation functions $f_{2k}$; in addition to those previously given, one needs

$$f_6 = \frac{1}{2} \left[ \sum_m m^6 N(m)/N - \frac{5}{11} (3I^2 + 3I - 7) \sum_m m^4 N(m)/N + \frac{1}{11} (5I^4 + 10I^3 - 20I^2 - 25I + 14) \sum m^2 N(m)/N - \frac{5}{231} I(I+1)(I-1)(I+2)(I-2)(I+3) \right],$$

$$f_8 = \frac{1}{2} \left[ \sum_m m^8 N(m)/N - \frac{14}{15} (2I^2 + 2I - 9) \sum_m m^6 N(m)/N + \frac{7}{39} (6I^4 + 12I^3 - 50I^2 - 56I + 81) \sum_m m^4 N(m)/N - \frac{7}{2145} (60I^6 + 180I^5 - 690I^4 - 1680I^3 + 1958I^2 + 2828I + \frac{9132}{7}) \sum m^2 N(m)/N + \frac{7}{1287} I(I+1)(I-1)(I+2)(I-2)(I+3)(I-3)(I+4) \right].$$
Summations of products of the cosines of angles $\theta_1$, $\theta_2$, $\theta$ are involved in the geometrical functions $G_{2k}$; these are symmetric in $\theta_1$ and $\theta_2$ for the stretched configuration. For this case it may be seen that they are never of higher than fourth degree in $\cos \theta$. The explicit expressions are\(^\text{16)}\)

$$G_0(\theta) = \frac{48}{215}\left[1 + \frac{1}{8} \cos^2 \theta + \frac{1}{24} \cos^4 \theta\right], \quad (15)$$

$$G_2(\theta_1, \theta_2, \theta) = \frac{1}{231}\left[96 - 144(\cos^2 \theta_1 + \cos^2 \theta_2) + 36 \cos \theta_1 \cos \theta_2 \cos \theta + 18 \cos \theta_1 \cos \theta_2 \cos^2 \theta_1 + \cos \theta_2 \right], \quad (16)$$

$$G_4(\theta_1, \theta_2, \theta) = \frac{1}{238}\left[-201 + 1005(\cos^2 \theta_1 + \cos^2 \theta_2) - 1225(\cos^4 \theta_1 + \cos^4 \theta_2) + 315 \cos^2 \theta_1 \cos \theta_2 - 540 \cos \theta_1 \cos \theta_2 \cos \theta + 420 \cos \theta_1 \cos \theta_2 \cos \theta(\cos^2 \theta_1 + \cos^2 \theta_2) - 270 \cos \theta_1 \cos \theta_2 \cos \theta(\cos^2 \theta_1 + \cos^2 \theta_2) + 630 \cos \theta_1 \cos \theta_2 \cos \theta - 360 \cos \theta_1 \cos \theta_2 \cos \theta + 108 \cos \theta + 36 \cos^4 \theta\right], \quad (17)$$

$$G_6(\theta_1, \theta_2, \theta) = \frac{1}{60}\left[-6 + 63(\cos^2 \theta_1 + \cos^2 \theta_2) - 756 \cos \theta_1 \cos \theta_2 - 63(\cos^4 \theta_1 + \cos^4 \theta_2) + 693 \cos \theta_1 \cos \theta_2 (\cos^2 \theta_1 + \cos^2 \theta_2) + 504 \cos \theta_1 \cos \theta_2 \cos \theta - 1005 \cos \theta_1 \cos \theta_2 \cos \theta (\cos^2 \theta_1 + \cos^2 \theta_2) + 1848 \cos \theta_1 \cos \theta_2 \cos \theta + 2(\cos^2 \theta_1 + \cos^2 \theta_2) + 1512 \cos \theta_1 \cos \theta_2 \cos \theta + 336 \cos \theta_1 \cos \theta_2 \cos \theta^3 \right], \quad (18)$$

\[ G_0(\theta_1, \theta_2, \theta) = \frac{1}{32} \left[ 3 - 54(\cos^2 \theta_1 \cos^2 \theta_2) + 1188 \cos^2 \theta_1 \cos^2 \theta_2 
+ 99(\cos^4 \theta_1 + \cos^4 \theta_2) - 2574 \cos^2 \theta_1 \cos^2 \theta_2 (\cos^2 \theta_1 + \cos^2 \theta_2) 
+ 6435 \cos \theta_1 \cos^4 \theta_2 - 432 \cos \theta_1 \cos \theta \cos \theta 
+ 1584 \cos \theta_1 \cos \theta_2 \cos \theta (\cos \theta_1 + \cos \theta_2) 
- 6864 \cos^3 \theta_1 \cos \theta_2 \cos \theta - 216 \cos^2 \theta (\cos^2 \theta_1 + \cos^2 \theta_2) 
+ 2376 \cos^2 \theta_1 \cos^2 \theta_2 \cos \theta - 238 \cos \theta_1 \cos \theta_2 \cos \theta 
+ 24 \cos^2 \theta + 8 \cos^4 \theta \right]. \] (19)

The normalization of (12) is such that
\[ \frac{1}{(4\pi)^2} \int W(\theta_1, \theta_2, \theta) \, d\Omega_1 \, d\Omega_2 = 1. \]

When the nuclei are randomly oriented, \( f_{2k} = 0 \) for \( k > 0 \); \( N_0 = 1, f_0 = 1 \), and it is seen that \( G_0(\theta) \) is just the ordinary room-temperature angular correlation function for nickel 60.

In the limit of complete orientation, Cox and Tolhoek show\(^{17} \) that
\[ W(\theta_1, \theta_2, \theta) = F(\theta_1) \cdot F(\theta_2), \] (20)
that is, the distribution of the second gamma-ray is independent of the first. This should be true generally for any stretched configuration. Then for \( \theta_1 = 0 \), \( W(\pi)/W(\frac{\pi}{2}) \to 0 \) while for \( \theta_1 = \frac{\pi}{2} \), the ratio approaches (both for point geometry) \( \infty \).

Now a choice needs to be made of the angles of observation. The correlation obtained may be characterized by the choice of the direction of the first emission, and two cases of interest will be discussed.

---

(a) $\theta_1 = 0$

Now take $\theta_2 = \pi/2, \pi$ and hence $\theta = \theta_2 = \pi/2, \pi$. The functions are, apart from a normalizing constant,

$$W(0, \pi/2, \pi/2) = 0.1525 - 0.208N_2^2 - 1.47N_4^4 - 1.0N_6^6 + 1.5N_8^8$$  \hspace{1cm} (21)

$$W(0, \pi, \pi) = 0.1780 - 0.554N_2^2 - 0.532N_4^4 + 3.7N_6^6 + 4.0N_8^8$$  \hspace{1cm} (22)

The ratio of $W(0, \pi, \pi)/W(0, \pi/2)$, which shall be abbreviated simply as $W(\pi)/W(\pi/2)$, is a measure of the anisotropy of the correlation pattern; its variation with increasing orientation is sketched in Figure 16. The region of $\beta < 1.0$ corresponds to experimentally observed radiation pattern anisotropies.

(b) $\theta_1 = \pi/2$

With $\theta_2 = \pi, 3\pi/2, \theta = \theta_2 - \theta_1 = \pi/2, \pi$; note that all angles are measured in the same plane. Now
\[ W(\pi/2, \pi, \pi/2) = 0.1525 - 0.208N_2f_2 - 1.47N_4f_4 - 1.0N_6f_6 + 1.5N_8f_8 \]

\[ \Xi W(0, \pi/2, \pi/2) \]

\[ W(\pi/2, 3\pi/2, \pi) = 0.1780 + 0.277N_2f_2 - 0.200N_4f_4 - 1.2N_6f_6 + 1.1N_8f_8 \]  

Dependence of \( W(\pi/2, 3\pi/2, \pi)/W(\pi/2, \pi, \pi/2) = W(\pi)/W(\pi/2) \) on orientation is illustrated in Figure 17.

Figure 17

Variation of the Angular Correlation with \( \beta \) for \( \theta_1 = \pi/2 \).

As in the case of the directional distribution (Section 1), dependence on the beta-decay is introduced through the products \( N_{2k}^2 f_{2k}^2 \). Again using numerical values of Cox et al.\(^{18}\) and noting that the contributions from \( f_6 \) and \( f_8 \) terms in equation (12) are very small for \( \beta < 1.0 \), the theoretical angular correlation is plotted in Fig. 18.

Figure 18

Anisotropy of the Gamma-Gamma Angular Correlation of Nickel 60.
FIGURE 18
ANISOTROPY OF THE GAMMA-GAMMA ANGULAR CORRELATION
It should be noted that the computations of both the radiation patterns and angular correlation are based on the assumption that it is possible to express the degree of orientation by means of a single parameter $\beta$ associated with a Boltzmann distribution of the magnetic sublevels. In the Gorter-Rose method this assumption is not expected to hold. However, when the correlation is expressed as a function of the anisotropy, as in the present work, it appears likely that, for the comparatively small degrees of orientation obtained, the relationship is insensitive to the specific manner in which the magnetic substates are populated.

3. Finite Geometry Corrections.

The observed angular distribution and correlation functions will be smeared by the finite size of the detectors and source; this effect may be conveniently introduced into the theoretical expressions in the form of attenuation factors, $J_1$: \(^{19}\)

$$W(\theta_1, \theta_2, \theta) = \sum_{k=0}^{2} A_{2k} J_{2k}^2 P_{2k}(\cos \theta), \quad (25)$$

and

$$F(\theta) = \sum_{k=0}^{2} B_{2k} J_{2k} P_{2k}(\cos \theta). \quad (26)$$

\(^{19}\) M. E. Rose, Phys. Rev. 91, 610 (1953).
The coefficients $J_{2k}$ can be calculated in a manner which includes the dependence of detection efficiency on gamma energy as well as on the geometry of the detector. These results have been employed, for example, by Klema and McGowan in a study of the anisotropy of the angular correlation function for randomly oriented nickel 60 nuclei.

Lawson and Frauenfelder use the empirically determined angular resolution of their counters to study the nickel 60 correlation; this is necessary for highly precise comparisons between experiment and theory in looking for extranuclear effects.

The statistical precision of the data to be presented later is not sufficient to justify such a procedure. Instead, Rose's results can be interpolated for a geometrical solid angle determined by the distance from center of sample to center of scintillation crystal (2 inches) and the diameter of the cylindrical crystal (1 inch); the planar half-angle is $13^\circ 46'$, and $\frac{\omega}{4\pi} = .0144$. The resulting estimates for the solid-angle attenuation factors are $J_2 = .96$, $J_4 = .68$, $J_0 = 1$.

Walter, Huber, and Zunti included the effect of a finite source, and found the source and detector correc-

---

tions additive to first approximation. In the present experiments the source size is such that the first approximation is insufficient; the second order correction involves some computational difficulty for cases of interest, so it appears preferable (and sufficient) to estimate the effect by approximate geometrical considerations.

Consider the extended source to be roughly one inch high and \( \frac{1}{2} \) by \( \frac{1}{3} \) inch in cross-section. For point detectors at 2 inches, the angular uncertainty defined by the extremities of the source is somewhat less than that due to the finite extension of the detectors. The vertical extension is especially ineffective in smearing either the directional distribution or the angular correlation.

Assuming these corrections are approximately additive, we take \( J_2 = .93, J_4 = .80 \) as reasonable estimates. When these are introduced into equations (10), (11), (21), (22), (23), (24) one obtains

\[
\begin{align*}
F(0) &= 1 - 2.02N_2f_2 - 4.00N_4f_4 = F(\pi) \\
F(\pi/2) &= 1 + 1.01N_2f_2 - 1.50N_4f_4 \\
W(0, \pi/2, \pi/2) &= .1533 - .225N_2f_2 - 1.34N_4f_4 - .26N_6f_6 + .97N_8f_8 \\
W(0, \pi, \pi) &= .1755 - .544N_2f_2 - .421N_4f_4 - .26N_6f_6 + .88N_8f_8 \\
W(\pi/2, \pi/2, \pi/2) &= W(0, \pi/2, \pi/2) \\
W(\pi/2, 3\pi/2, \pi) &= .1755 + .267N_2f_2 - .158N_4f_4 - .88N_6f_6 + .71N_8f_8
\end{align*}
\]
The net effect on the ratios $W(\pi)/W(\pi/2)$ and $F(\pi)/F(\pi/2)$ are 1-2% and less than 1%, respectively.

A small error in centering the source will be corrected in first approximation by dividing by the appropriate single counting rates, as is done for example in Eq. (30) below.

4. Relationship between Theoretical and Experimental Quantities.

A word should be said here about the correspondence between the theoretical distributions and the experimental observables. Consider a rather general type of decay (Fig. 19) in which the gamma-gamma correlation is to be

![Gamma-Gamma Correlation Diagram](image)

measured. Efficiency-solid angle factors \((E\Omega)\) will in general be different for each counter and for each gamma ray at a particular counter. In nickel-60 the gamma rays are very close in energy and the two cannot be distinguished; in other decays they may be so widely different that each counter may be set so as to receive only one particular gamma ray, or the method of delayed coincidences may be used if appropriate.

In the sample decay scheme shown in Fig. 19, let \(\gamma_a\) and \(\gamma_b\) denote the rates of emission of \(\gamma_a\) and \(\gamma_b\). These rates will in general be unequal if there is appreciable beta-decay to the first excited state, or cross-over gamma transitions between the second excited state and the ground state. In a coincidence detector circuit which cannot distinguish the gamma-rays by energy or time discrimination, the number of true coincidences per second for counter 1 at angle \(\phi_1\) and counter 2 at angle \(\phi_2\) is

\[
C_{12} = \frac{\gamma_a}{(4\pi)^2} \left[ W(a_1, b_2) \varepsilon_{1a} \varepsilon_{2b} \Omega_2 + W(a_2, b_1) \varepsilon_{2a} \varepsilon_{1b} \Omega_1 \right]
\]

(27)

Here the notation \(W(a_1, b_2)\), etc. signifies the correlation which applies when \(\gamma_a\) enters counter 1 and \(\gamma_b\) enters counter 2; the correlation function is assumed to be corrected as outlined in Section 3 for finite geometry.

Now \(W\) is symmetric in \((1,2)\) for stretched configurations; \(C_{12}\) is symmetric in \((a,b)\) if the detectors are unable to distinguish the gamma rays. Both conditions are
true for nickel 60; therefore in this case

$$W(a_1,b_2) = W(a_2,b_1).$$

the corresponding singles rates are

$$C_1 = \left[ \nu_a F(a_1) \varepsilon_{1a} + \nu_b F(b_1) \varepsilon_{1b} \right] \frac{\Omega_1}{4\pi}, \quad (28)$$

$$C_2 = \left[ \nu_a F(a_2) \varepsilon_{2a} + \nu_b F(b_2) \varepsilon_{2b} \right] \frac{\Omega_2}{4\pi}, \quad (29)$$

with $F(a_1)$, etc., denoting the angular distribution function of a particular gamma in a given direction, corrected for solid angle. Now in the special case of nickel 60, both gamma rays are quadrupole and correspond to spin changes of two units; hence they have the same radiation pattern, and the designation of a or b on $F(\theta)$ can be dropped.

In the absence of orientation, the singles rates are

$$C_1^0 = \left[ \nu_a \varepsilon_{1a} + \nu_b \varepsilon_{1b} \right] \frac{\Omega_1}{4\pi}, \quad (28')$$

$$C_2^0 = \left[ \nu_a \varepsilon_{2a} + \nu_b \varepsilon_{2b} \right] \frac{\Omega_2}{4\pi}. \quad (29')$$

In case $\theta_1$ equals either 0 or 90°, counter 1 will be taken as the "fixed" counter in the coincidences, and counters 2, 3 represent the successive positions $\theta = \pi/2, \pi,$ relative to counter 1, of the "movable" counter. Hence for the purposes of comparing theory with experiment we determine the ratios

$$\frac{C_1^0}{C_1^0} = \frac{W(\pi/2) \left[ \varepsilon_{1a} \varepsilon_{3b} + \varepsilon_{3a} \varepsilon_{1b} \right]}{W(\pi) \left[ \varepsilon_{2a} \varepsilon_{3b} + \varepsilon_{3a} \varepsilon_{2b} \right]} \frac{\left[ \nu_a \varepsilon_{2a} + \nu_b \varepsilon_{2b} \right]}{\left[ \nu_a \varepsilon_{3a} + \nu_b \varepsilon_{3b} \right]}, \quad (30)$$

$$\frac{C_3^0}{C_2^0} = \frac{F(\pi/2)}{F(\pi/2)} \quad (31)$$

The ratio $C_1/C_1^0$ will be denoted by $N_1$ and called the "normalized (to case of random orientation) singles rate"; similarly, let $C_{11}/C_1^0$ be abbreviated to $CR_{11}$ and called the "normalized coincidence rate."

It should be noted that normalizing factors in $F(\theta)$ and $W(\theta_1, \theta_2, \theta)$ are cancelled by using equations (30) and (31). Also, efficiency factors do not appear in (31). They may be cancelled in (30) under the further assumptions that the branching of the beta-decay is negligible ($\nu'_a = \nu'_b$) and that the detectors have equal efficiencies for the two gamma rays ($\varepsilon_{1a} = \varepsilon_{1b}, \varepsilon_{2a} = \varepsilon_{2b}, \varepsilon_{3a} = \varepsilon_{3b}$), both of which appear to be good approximations in the special case of nickel 60. Then

$$\frac{CR_{13}}{CR_{12}} = \frac{W(\pi)}{W(\pi/2)} \quad (30)$$

$$\frac{N_3}{N_2} = \frac{F(\pi)}{F(\pi/2)} \quad (31')$$

The same cancellation of efficiency factors would occur in (30) if the counters could discriminate completely between the gammas.

It should be emphasized that one must start with the general expressions (27), (28), (29) if correlations other than the special case of nickel 60 are involved.
5. Influence of Extranuclear Fields.

The presence of a strong magnetic field at the nucleus, such as the HFS field used in producing nuclear orientation by the Gorter-Rose and Bleaney methods, may be expected to alter the "free nucleus" theory of directional distribution and correlation if the lifetimes of the intermediate nuclear states of a cascade of radiations are sufficiently long. This problem has been discussed by several authors; 25), 26), 27), 28), 29), 30) a possible reorientation effect on the angular distribution of nickel 60 gamma rays in cerium magnesium nitrate has been considered by Steenberg. 31) No specific treatment of the effect on correlation for the case of oriented nuclei has been given.

Alder 26) showed that the correlation function for nuclei in a paramagnetic ion with isotropic HFS coupling can be written as

\[ W(\theta) = \sum_{k=0}^{k_{\text{max}}} A_{2k} Q_{2k} P_{2k}(\cos\theta). \]  

(52)

The effect of reorientation is represented by an attenuation coefficient \( Q_{2k} \) which depends on the value of \( J \), the spin of the intermediate state \( I_g \), the lifetime of the

intermediate state $T_e$, the HFS splitting ($\Delta \nu$), and the order $2k$ of the term. In the case of present interest, $J=\frac{3}{2}$,

$$Q_{2k} = 1 - \frac{2k(2k+1)}{(2I_e+1)^2} \cdot \frac{2\pi (\Delta \nu) T_e}{1 + [2\pi (\Delta \nu) T_e]^2}.$$  \hspace{1cm} (33)

It is seen that the correlation can never be completely destroyed: Even in the limit of large ($\Delta \nu$), according to equation (33), there remains a "hard core" correlation given by

$$Q_{2k} \lim = 1 - \frac{2k(2k+1)}{(2I_e+1)^2}.$$  \hspace{1cm} (33')

This result is based on the assumptions that the interactions are time-independent and that no preferred direction exists as far as the nuclei are concerned. 32) These considerations, therefore, are not applicable to single crystals containing nuclei which are partially oriented. Nevertheless, they are useful in the limit of random orientation. Fig. 20 illustrates the behavior of $Q_e$. 33) The order of magnitude

---

**Figure 20**

Attenuation of Angular Correlation by Isotropic Magnetic HFS (Biedenharn and Rose)

---

of the effect is quite striking; for example, it $I_e=1$, $Q_2^{\text{lim}}=0.33$; when $I_e=2$, $Q_2^{\text{lim}}=0.76$, $Q_4^{\text{lim}}=0.20$. If the assumptions are violated, attenuation may be complete.

For a single nucleus, the influence of the extranuclear field can be pictured semi-classically as a tendency to cause the nuclear spins to precess about the field direction, thereby inducing transitions between the magnetic substates of the intermediate state (taking the $Z$-axis along the direction of either emission) if its lifetime is of the order of magnitude of the period $\tau_L$ of Larmor precession about the field $B$ at the nucleus. If $B$ arises from an isotropic HFS field of about $10^5$ gauss, say, then with $g_n \simeq 2$, we have

$$\tau'_L = \frac{2\pi}{\alpha_L} = \frac{h}{E_n \mu_B B} \simeq 3 \times 10^{-9} \text{ sec.} \quad (34)$$

The result of equation (34) is to be compared with Alder's definition of a "critical time" $\tau'_{\text{crit}}$ which is, for .01 cm$^{-1}$ HFS,

$$\tau'_{\text{crit}} = \left[ \frac{2\pi c(\Delta \nu)}{\nu} \right]^{-1} \simeq 5 \times 10^{-10} \text{ sec.} \quad (35)$$

In principle it should be possible to extend Alder's argument to a system of partially oriented nuclei by taking into account the unequal populations of the initial magnetic substates. So far these calculations have not been carried out.

---


So far it has been assumed that the radiation detectors are insensitive to the state of transverse or circular polarization. If such is not the case, anisotropic distributions of the gamma polarizations will be observed as the degree of nuclear orientation is increased.\(^{35,36}\)

As polarization has not been detected in the experiments to be described here, only two comments will be given in this section, pertaining to a quadrupole emission (\(L=2\)) in which \(I_1 \rightarrow I_1 - 2\), e.g. cobalt 60. First, a maximum of transverse polarization of gamma rays will be observed in the equatorial plane (\(\theta=90^\circ\)), with a degree of polarization parallel to the orientation axis given by

\[
P_\parallel = k \epsilon
\]

(36)

See Figure 21. Here \(\epsilon = \left[ F(\pi/2) - F(0) \right] / F(\pi/2) \) is the Figure 21

Geometry for Observation of Transverse Gamma Polarization

radiation pattern anisotropy and $k = -1 (+1)$ corresponding to electric (magnetic) radiation. Steenberg has discussed the detection of gamma polarization by the Compton process \(^{37}\); the method has been successfully applied to cobalt $58$ and $60^{38}$ and to manganese $54^{39}$.

Second, if and only if the nuclei are polarized, a net circular polarization of the radiations will be observed, having its maximum effect along the orientation axis. Tolhoek and Cox show\(^{35}\) that this effect is proportional to $f_k$ of odd $k$, and hence depend on the algebraic sign of the nuclear magnetic moment. Experimental observation of this effect is difficult, as has been pointed out by several authors,\(^{37},38,40,41\) and so far has not been carried out.

7. Information from Gamma Radiations of Oriented Nuclei.

Knowledge to be gained by observing the gamma radiations from oriented nuclear systems has been dealt with by Cox and Tolhoek;\(^{42}\) it will be summarized in this section.

(A) For nuclear physical data, assume the mechanism of orientation to be known.

(1) If considerable information on the nucleus

\(^{39}\)Bishop, Daniels, Durand, Johnson, Perez, Phil. Mag. **45**, 1197 (1954).
involved exists, so that the decay scheme may be constructed and the possible spin assignments reduced to a relatively few combinations, one can in favorable circumstances establish unambiguously the spins and multipole orders involved in a gamma cascade from the shape and variation with temperature of the radiation pattern. This is true even if the mechanism is understood only qualitatively, but not in detail.

(2) When in addition the quantitative picture of the orientation mechanism can be considered known, the magnitude of the nuclear g-factor may be deduced from the anisotropy of the radiation pattern. This implies that the field B at the nucleus is known, that the Kelvin temperature T is accurately known, and that the orientation proceeds in a manner which can be calculated. Under these stringent conditions a choice may be made between the possible ground state spins and modes of decay of the parent nucleus assuming sufficient counting statistics are obtained. With the spin and g-factor known, the nuclear

---


* For example, the radiation pattern anisotropy of photons which follow beta decay varies with the relative contribution of Fermi and Gamow-Teller matrix elements, and affords in principle a method of distinguishing between them. There are, however, experimental difficulties; cf. (44): M. A. Grace and H. Halban, Physica, 18 1227 (1952).
magnetic moment follows immediately.

(3) In case the nuclei of interest can be polarized and the circular polarization of the gamma rays detected, the sign of the magnetic moment is obtained.

(4) Observation of the transverse gamma polarization determines whether radiation of known multipolarity is electric or magnetic. This established the relative parity of the initial and final states involved.

(5) Although in principle angular correlation for oriented nuclei does not give information about nuclear states which could not be deduced from individual radiation patterns, it can be of advantage when the decay scheme is complex, e.g. cobalt 56. It may also be more sensitive to perturbation of the intermediate state by extranuclear fields arising from the surrounding lattice; see paragraph (6) below.

(B) Assuming the nuclear properties to be well established, data about the solid state may be deduced.

(6) If the gamma radiation pattern agrees with one calculated from a model for the orientation mechanism, the assumed picture is confirmed. With T known, the field B at the nucleus will be given. Deviations from ideal behavior may be due to ionic interactions or to nuclear precession; in cases where one or the other is predominant an idea of the local fields can result.

(7) Since orientation occurs in paramagnetic salts with HFS for temperatures in the range 0.001 to 0.1°K, the anisotropic radiation pattern is a thermometric parameter, if the nuclear and solid state properties are well known.

(8) Gamma-ray observations would give an independent demonstration of the amount of nuclear orientation achieved by resonance methods (see Chapter I, Section 7). In addition, it has been suggested that the disappearance of the gamma pattern anisotropy might be a sensitive detector of nuclear resonance in small quantities of radio-isotopes. 47)

CHAPTER III

EXPERIMENTAL APPARATUS

Design and construction of equipment for producing and measuring low temperatures and for observing gamma radiations from radioactive nuclei will be described in the succeeding sections. Most details of sample preparation and operational procedures will be reserved for the following chapter.

1. Low Temperature Cryostat.

An apparatus has been constructed for the cooling of paramagnetic substances by adiabatic demagnetization from liquid helium temperatures. Noteworthy design features of this cryostat consist of the following:

(a) A large capacity oil ejector pump evacuates the space above the liquid helium bath, permitting initial temperatures for demagnetization of below 1°K. Actual bath temperature is very close to 1°K because of an evaporation rate of about 40 cm$^3$/hour of liquid helium. The KB-150 pump (Consolidated Vacuum Corporation) has a speed of about 150 liters/second at 100 microns pressure; the required forepressure of 400-1000 microns is supplied by a Kinney VSD8811 rotary pump (15 liters/second at 1000
microns). The pumping tube is six-inch i.d. pipe to give minimum resistance to flow; a large vacuum valve (Crane 1650 Y-pattern diaphragm type) permits this line to be shut off when desired.

(b) Rapid evacuation of the sample chamber can be achieved by use of high speed oil diffusion pumps (Consolidated Vacuum Corp. MCF-300 (260 liters/second at 10^{-5} mm Hg) backed by an MB-100 booster and a Welch 1405B Duoseal rotary pump. Use of a liquid air trap prevents the back diffusion of oil molecules. A high vacuum (of the order 1 \times 10^{-5} \text{ mm Hg} as read by an ionization gage at the top of the cryostat) is established in a few minutes with this system; the pressure is one or two orders of magnitude lower at the low temperature end of the sample chamber.\(^1\)

(c) Suppression of vibration is provided for by mounting the entire cryostat on a 1000-lb. concrete base which is insulated from the floor by rubber shock mountings; the assembly is also insulated by rubber washers from its two ceiling supports. The Kinney pump is shock mounted at a distance from the apparatus, and flexible bellows sections have been inserted in the vacuum line, while the Welch pump, at the end of several feet of Cenco vacuum hose, is set on foam rubber.

\(^1\)Roberts, Sartain, Dabbs, ORNL Report 1164 (Oak Ridge, 1952).
Small residual vibrations leaking through the flexible connections, the circulation of oil in the diffusion pumps, turbulent flow of cooling water, and the boiling of the liquid nitrogen bath establish the minimum vibration which could be attained by these precautions. These are probably important factors in setting the lower limit on evaporation rate of the bath (40 cm$^3$/hour) and heat leak to the specimen (20-30 ergs/minute). The large mass of the cryostat helps to reduce these effects.

A view of the cryostat with counters in place is shown in Fig. 22.

(d) A glassware system of considerable adaptability was constructed of 10 mm Pyrex tubing; it is mounted at the back of the cryostat, connections being made through $\frac{1}{2}$-inch copper tubing. Bath pressure can be read by a mercury manometer with an oil manometer (butyl phthalate or Octoil-S) connected in parallel for use as an absolute or differential pressure indicator. The oil manometer can be used to measure pressures below the lambda-point of liquid helium (38 mm Hg, 2.18°K), with a manometric equivalent of about 15 to 1.

A layout of the glassware is included in Fig. 23. The numbers beside the stopcocks refer to Eck and Krebs' types of 4 mm bore high vacuum Pyrex stopcocks—the arrows indicate how they are to be oriented so that the back of the hollow plug may be pumped out.
Figure 22

View of Cryostat with Counters in Place
(e) The dewar system was designed for rigidity; this was accomplished mainly by the use of a glass flange on top of the helium dewar. Ground flat on top, the flange is seated in a brass plate and held by a split retainer ring; see Fig. 24.
The nitrogen dewar is externally supported, and is fitted with a cap to prevent solid air from dropping into it.

Both dewar tails were made of very thin Pyrex tubing (0.8—1.0 mm wall) to allow maximum space for the specimen chamber and bridge coils inside the dewars. Dimensions may be found on Figure 25, which shows a cross-section of the cryostat head.

For greater versatility, an alternate set of dewars, of greater length and helium capacity, can be attached by removing the lower extension of the head. In this way the cryostat can be modified for studies of the demagnetization properties of paramagnetic salts, two-stage demagnetizations, etc.

(f) The general arrangement of the sample chamber is given in Fig. 25. It is removable through the top for ease of changing samples, and has all bridge leads and coils attached to it. Liquid helium is introduced through a hole in the top flange plate, which is sealed by a tapered plug.

At the bottom of a ½" diameter nickel silver tube the sample space itself is located; its construction is detailed in Fig. 26. In these experiments the sample consisted of eight single crystals of Ce₂M₂₃(NO₃)₁₂·24H₂O, of total mass 2.5 grams and containing 10–20 microcuries of cobalt 60 activity (preparation of the specimen is outlined in
Figure 25

Cross-sectional View of Cryostat Head
Figure 25
CROSS-SECTIONAL VIEW OF CRYOSTAT HEAD
Figure 26
Sample Chamber
Figure 26
SAMPLE CHAMBER
Chapter IV). These were glued to a mica sheet with nail lacquer and wrapped with lens tissue. The support was a thin-walled Pyrex tube which was attached to a guard salt of about 12 grams of iron ammonium alum. Araldite "101" (Ciba Co., Inc.) was used to fasten the assembly to the glass pedestal, and was found to be a satisfactory adhesive in the low temperature range.

Considerable effort was made to reduce the most important sources of heat leaks to a specimen at low temperature. First, no metal was used in the support, so that the possibility of induced eddy currents was eliminated.

Second, conduction of heat through the sample support can be greatly reduced by the use of a guard salt, which is cooled by demagnetization at the same time as the sample. It serves as a good insulator when very cold, and its high specific heat allows it to absorb such heat as does flow into it. Use of glass tubing with very thin walls reduces the thermal conductivity and cross-sectional area factors in conduction through the supports.

In the third place, radiation piping from the room temperature end of the sample chamber tube is inhibited by the radiation trap, which acts to reflect it or to provide a long path of cold walls on which it may be absorbed.

The role of condensation of the gas phase on a cold object in an enclosure with walls at bath temperature has
been studies at Oak Ridge, 2) and a heat leak proportional to the residual gas pressure was obtained. Kinetic theory applied to a dynamic system in which a pressure \( p_r \) is read at room temperature (300°K) and the pressure \( p \) in the sample space at 1°K leads to the relation 3)

\[
p = \sqrt{\frac{1}{300}} \ p_r
\]

and hence values of \( p \) of the order of \( 10^{-7} \) mm Hg. Condensation could cause extremely large heat leaks, which are not observed; the Oak Ridge workers concluded that much lower pressures of about \( 10^{-9} \) mm actually exist due to "getting" by the paramagnetic salts. This contributes to the irreversibility of the demagnetization, of course, but the guard salt is of assistance in reducing the effect.

Importance of eliminating vibration in low temperature cryostats is at present difficult to estimate. However, experience indicates a possible correlation between the achievement of small heat leaks and the reduction of mechanical vibration. 4) Accordingly, the precautions mentioned in part (c) above were undertaken. Quite manageable heat leaks (20-30 ergs/minute) were estimated to be present.

(g) Mutual inductance bridge coils were designed to measure the magnetic moment of the sample, hence indicating its temperature. These coils form part of a Hartshorn

---

a.c. bridge circuit,\textsuperscript{5} the use of which has been amply discussed by others.\textsuperscript{6,7} The bridge used in the experiments is described in unpublished work by Hayes\textsuperscript{8} and by Nicol.\textsuperscript{9}

Because of the presence of the guard salt, precautions had to be taken in order that only the contribution of the cerium magnesium nitrate crystals were observed. A set of two concentric primary coils were wound on Dielecto (Continental-Diamond Fiber Co.) forms in such a way that their dipole moments cancel, resulting in a large reduction of the stray field.\textsuperscript{10} A two-section secondary arrangement kept the guard salt as far away as possible. Positions of the coils are shown in Fig. 26, and Table I summarizes the coil data. The same current (1-2ma) traverses each primary coil, as they are connected in series opposing. The number of turns on the outer coil \(N_0\) is determined from the condition \(N_0 = (r_1/r_0)^2 N_i\), where \(N_i\) is the number of turns on the inside coil and \(r_0, r_1\) are the coil radii.

All three coils are mounted directly on the sample chamber, so that there is no possibility of axial shifts

\textsuperscript{6}Casimir, de Haas, de Klerk, Physica 5, 241 (1939).
\textsuperscript{7}D. Bijl, Thesis, Groningen (1950).
\textsuperscript{8}W. B. Hayes, Thesis, The Ohio State University (1950).
\textsuperscript{9}J. Nicol, Dissertation, The Ohio State University (1952).
\textsuperscript{10}Casimir, Bijl, duPre, Physica 8, 449 (1941).
Table I

Design Data for Bridge Coils

<table>
<thead>
<tr>
<th>Coil</th>
<th>Inside Diameter (in.)</th>
<th>Length (in.)</th>
<th>Wire Size (A.W.G.)</th>
<th>Total Turns</th>
<th>No. Layers</th>
<th>Resistance at 20°C (ohms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Secondary</td>
<td>1.0</td>
<td>4</td>
<td>40</td>
<td>4000</td>
<td>4</td>
<td>1250</td>
</tr>
<tr>
<td>Inside</td>
<td>1.120</td>
<td>6</td>
<td>36</td>
<td>2940</td>
<td>3</td>
<td>365</td>
</tr>
<tr>
<td>Outside</td>
<td>1.220</td>
<td>6</td>
<td>36</td>
<td>2475</td>
<td>3</td>
<td>330</td>
</tr>
</tbody>
</table>

relative to one another during the pumping down of the bath and calibration of mutual inductance against vapor pressure.

2. Electromagnet.

A 3½-ton Bitter-type magnet providing up to 24 kilogauss for demagnetization work has been constructed. Fig. 27 is a view of it in place at the cryostat. The yoke is entirely enclosed except for four openings to admit the dewars and provide access to the experimental space; consequently the stray field is negligibly small. Moving laterally on rails, the magnet can serve two cryostats. Screw drives and cranks allow the halves to be separated quickly.

In Fig. 28 the magnet housing is detailed, while Fig. 29 illustrates the parts that make up the coil assembly. A total of 960 turns of G. E. rectangular Formex magnet wire (.270 x .115 inch cross-section) were
Figure 27

View of Electromagnet in Position at the Cryostat
Figure 28

Magnet Housing
Figure 29
Magnet Coil Assembly
Figure 29

Oil Seal Plate
- E GRADE MATERIAL: ALUMINUM
- Scale: 1/2" = 1'-0"
- 5/16" drill holes on 1" square, for oil cooling.
- Threads, drill, and tap, at rear of solid stud, to sky split, O.D.
- Install 1/4" drill, 0.250" deep, at center of each stud, to clear stud.
- 5/16" to 18 UNC, 3/4" deep, at center of each stud, to clear stud.
- To receive OIL SEAL, 0.624" O.D.
- Spigot seal, equally spaced, in 0.64".

Retainer Ring
- E GRADE MATERIAL: BRASS
- Scale: 1/2" = 1'-0"
- 5/16" drill, 0.250" deep, at center of each stud, to clear stud.
- 5/16" to 18 UNC, 3/4" deep, at center of each stud, to clear stud.
- To tighten OIL SEAL, 0.624" O.D.
- Spigot seal, equally spaced, in 0.64".

Spacer
- E GRADE MATERIAL: ALUMINUM
- Scale: 1/2" = 1'-0"
- 5/16" drill, 0.250" deep, at center of each stud, to clear stud.
- 5/16" to 18 UNC, 3/4" deep, at center of each stud, to clear stud.

Coil Reel
- E GRADE MATERIAL: "Stem-Steel" and Phillips Aluminum
- Scale: 1/4" = 1'-0"
- Install 1/4" drill, 0.250" deep, at center of each stud, to clear stud.
- 5/16" to 18 UNC, 3/4" deep, at center of each stud, to clear stud.
- Spigot seal, equally spaced, in 0.64".
wound in the reels, 450 in each half, in 30 layers of 16
turns each. About 600 lbs. (5000 feet) of wire were used.
The series resistance under full power is 1.62 ohms.
Two views of the winding arrangement are shown in Fig. 30.
Clearance for the flow of cooling oil is supplied

Figure 30
Detail of the Magnet Windings

by melamine strips, 4-5/8 x 1/4 x 1/32" in size, laid be-
tween layers of Formex. Spacer rings and 3/8" holes in
the end plates of the coil reel permit entrance and exit
of the oil, which was circulated by a rotary pump and cooled
by passing through an interhanger with water from a 55,000
gallon reservoir.

An oil seal at the front plate is effected by means
of a large "O-ring" under the oil seal plate being squeezed
down by 12 Allen set screws in the retainer ring. A simi-
lar seal was made at the center. Oil flowed out at about
10 lbs. pressure through four 1" holes into a square col-
lector ring formed of pipe and fittings, having entered
at the back at 50 lbs. The exit holes also have "O-ring"
seals. The electrical leads were brought out through two
of the oil inlets using a neoprene-melamine oil tight insulated gasket assembly.

Figure 31

Magnet Pole Pieces

The pole tips of Armco iron, have a face diameter of 5-3/4" and a separation of 2-1/8". See Fig. 31. They are held to the housing core by 3/4" diameter bolts. The central field intensity and the profiles achieved are graphed in Figs. 32 and 33. The magnetic intensity B was measured with a Cambridge fluxmeter (sensitivity 15,000 maxwell-turns per division) and a pull out coil with 30 turns of wire on a 1/2" diameter; some of the lower fields were checked with a proton resonance magnetometer. The latter instrument indicated homogeneity of about 1 part in 5000 at 5000 gauss over 1 cm² at the center of the gap. The considerable self-inductance of the
Figure 32
Central Field Characteristic of 24 KG Electromagnet
COILS IN SERIES
960 TURNS
5 3/4" POLE DIAMETER
2 1/8" POLE GAP
RESISTANCE = 1.3 - 1.6 Ω
HOMOGENEITY = 1:5000
INITIAL SLOPE = 235 GAUSS/AMP.

FIGURE 32
CENTRAL FIELD CHARACTERISTIC OF
24 KG ELECTROMAGNET
Figure 33
Field Profiles of 24 KG Electromagnet
FIELD PROFILES OF 24 KG ELECTROMAGNET

FIELD INTENSITY (KILOGAUSS)

RADIAL DISTANCE FROM CENTER (INCHES)

200 AMPS (60 KW)

45 AMPS (2.5 KW)

POLE FACE

FIGURE 33
FIELD PROFILES OF 24 KG ELECTROMAGNET
magnet (roughly 25 henrys) provides and efficient ripple remover for the D.C. power, field fluctuations being less than $10^{-2}$ gauss at full power.

Power is supplied by a 150-KW General Electric motor-generator set. Due to the resistance of the magnet windings and control rheostats, maximum current was about 190 amps at 310 volts, giving 24 KG. By putting the magnet windings in parallel, the full current capacity of 600 amps could be drawn from the generator.

Certain safety requirements need to be considered in operating such high-current magnets. First of all, the output circuit of the generator should be solidly connected at all points and never opened. In fact, it is extremely difficult, if not practically impossible, to break circuits with such large inductance without damage to the magnet windings or to the generator. All switching and current controls were accordingly placed in the field exciter circuit, which carries only about 10 amps maximum and has a smaller (but not negligible) inductance.

Besides the regular breaker and overload relay supplied with the motor, several auxiliary safety items were installed. A Minneapolis-Honeywell temperature controller was mounted on each magnet-half, with bulb immersed in the cooling oil; if the oil temperature indicates overheating, a mercury switch turns off the motor.
Another very desirable feature is a limit switch set so that it is closed only when the magnet halves are together. The switch is in the motor starting relay circuit, so that the motor-generator cannot be started unless the magnet is closed.

A commercial overcurrent breaker was rigged so that the breaker stops the motor if the rated current is exceeded.

Control boards at the generator panel and at the magnet enable the operator to monitor the voltage drop across the magnet for signs of too rapidly increasing resistance. Push button controls are provided for stopping the motor, as well as the oil and water circulating pumps if desired.

Finally, in the event of dire emergency, a large red "panic button" is added which overrides all other controls and stops everything in the minimum time (magnet current decay time is about 20 seconds, and it takes two minutes for the motor to run down). The current in the exciter circuit is broken immediately by a plunger relay operating a knife switch. This emergency stop circuit is diagrammed in Fig. 34. Resistances are 100-watt power resistors; a 15 mfd. condenser across the relay contacts reduces arcing. (The 6 volt battery is for bucking out any small voltages developed by remanence in the exciter core.)
3. Counting Equipment.

All electronic circuits except the commercial scalers and wide-band amplifiers were constructed from designs furnished by P. S. Jastram, who developed them to their present form at Washington University.

Before dealing with the individual components, one may describe the gamma ray detection process briefly as follows (see the block diagram, Fig. 35): Gamma rays entering the scintillation crystal NaI(Tl) produce light flashes which strike the cathode of an RCA 6199 photomultiplier tube. The cascade-amplified impulses are taken off the plate as negative pulses and into a linear amplifier. Then the amplified pulses appear at the input of a pulse height analyzer, where they are sorted as to voltage. The output is a multivibrator pulse whenever an input of
Figure 35

Block Diagram of Counting Equipment
Figure 35
DIAGRAM OF COINCIDENCE CIRCUIT FOR 2 CHANNELS
the right amplitude is registered. Either a scaler or a "slow" coincidence circuit may be driven directly with the discriminator output.

The coincidence circuit puts out a pulse whenever two (or more) inputs reach it within its "resolving time" $\tau_{\text{res}}$. Combining linear amplification of NaI(Tl) pulses with a one micro-second coincidence circuit enables one to make a precise energy selection, at some expense to time resolution. Now the number of accidental coincidences due to random singles counts arriving at the coincidence circuit within time interval $\tau_{\text{res}}$ is given by

$$C_{12}^{\text{acc}} = 2 \tau_{\text{res}} C_1 C_2$$

Hence a parallel channel of fast amplifiers and a "fast" coincidence circuit ($\tau_{\text{res}}$ of the order of 10 millimicroseconds) is employed to reduce the accidental background. Its output then enters the slow coincidence circuit, so that a simultaneous time and energy resolution is effected.

Fig. 36 is a view of the counting apparatus mounted in relay racks. Power supplies are at the bottom of the panels. Fast amplifiers are at the upper left, coincidence circuits in the center, and linear amplifiers on the right. Scalers are set up on the bench.

Use of scintillation counters to detect gamma rays has the advantage of greater efficiency, so that relatively weak sources can be employed. This is important for very low temperature work, as a source of heat leak
Figure 36
View of the Counting Apparatus
will be the beta-rays absorbed in the sample. It is advantageous to use weak sources and large solid angles for the detectors, since the number of true coincidences is proportional to the source strength while the number of accidentals varies as the square of the activity.

In addition, with crystals such as thallium-activated sodium iodide the strong photoelectric cross-section is available to make energy selection. It is necessary to set the discriminator threshold above the Compton edge and accept only the photopeak (Fig. 37).

Figure 37

Typical Scintillation Spectrum of NaI(Tl) for Co\textsuperscript{60} Gamma Rays

![Typical Scintillation Spectrum of NaI(Tl) for Co\textsuperscript{60}Gamma Rays](image)

In Fig. 38 is shown a quarter-section through the counter assembly; Fig. 39 is an "exploded" view of the components. The base connections consist of a resistor network (Fig. 40) to provide the proper dynode voltages on the phototube. A high voltage power supply (double unit) gives about 900-1200 volts for normal operation (Fig. 41).
Figure 38
SCINTILLATION COUNTER
LIGHT PIPE & SHIELD ASSEMBLY

SCALE - FULL

NOTE
PARTS A, B, D, E, F ARE ALUMINUM.
PART C IS STEEL.
D. & T. DENOTES HOLES DRILLED & TAPPED TO ACCOMMODATE 2-56 MACH. SCREWS.
D. & C. DENOTES HOLES DRILLED & COUNTERBORED TO ACCOMMODATE SHANK & HEAD OF 2-56 MACH. SCREWS.
ALL HOLES EQUALLY SPACED ON BOLT CIRCLE - INCLUDES 6 HOLES.
Figure 39

Exploded View of Counter Components
Figure 40

Photomultiplier Base Connections
HIGH VOLTAGE (1000-1200 V DC)

PHOTOCATHODE

12

100 K

56 K

1

56 K

2

56 K

10

56 K

3

56 K

9

56 K

4

56 K

8

56 K

5

68 K

7

82 K

6

10 K

OUTPUT

ANODE

99

DYNODES

FIGURE 40

RCA 6199 PHOTOMULTIPLIER BASE CONNECTIONS
HIGH VOLTAGE POWER SUPPLY

**Figure 41**
Because an external magnetic field is applied to polarize the electrons, it is necessary to shield the phototubes of the scintillation detectors. This was accomplished in three ways. First, the NaI(Tl) crystals were mounted on Lucite light pipes 7 inches long. These were in turn attached to the photo-multipliers, which were protected by thick iron cases. Finally, two coaxial opposed pairs of Helmholtz coils with cancelling dipole moments (described in the next section) were used to give a polarizing field of 200 gauss with minimum fringing. Thus it was possible to eliminate any field effects on the counting rates outside of the statistics. For stronger fields, the fringing was not negligible but may be tolerated so long as the field is held constant.

Circuits for the linear amplifiers and pulse height discriminators are given in Figs. 42 and 43. One linear amplifier and one pulse height discriminator are mounted on a single chassis. Three views of the layout are shown (Figs. 44, 45, 46). Some precautions to be observed in the construction are as follows:

(a) keep plate load resistors and signal-carrying components as close to the chassis as possible to minimize inductive pickup;

(b) make signal and r-f by-pass condenser (.01 mfd) leads as short as possible;

(c) for maximum stability, operate all parts at a
Figure 42
Linear Amplifier Circuit
Figure 43

Pulse Height Discriminator
Figure 44

Layout of the Linear Amplifier and Discriminator (Top View)
Figure 45

Layout of the Linear Amplifier and Discriminator (Bottom View)
Figure 46

Front View of the Amplifier-Discriminator
Figure 47
Linear Amplifier Power Supply
FIGURE 47
LINEAR AMPLIFIER POWER SUPPLY
Figure 48

Slow Coincidence Circuit
Figure 48
SLOW 4-CHANNEL COINCIDENCE CIRCUIT
Figure 49

Chassis Layout of Slow Coincidence Channels
Figure 50

Power Distribution Circuits for Coincidence Channels
**Figure 50**

DISTRIBUTION CIRCUIT FOR FAST & SLOW COINCIDENCE CIRCUITS
small fraction of its rated value;

(d) make every solder joint well, and keep acid fluxes away;

(e) lay out circuits so as to avoid stray couplings between parts of the circuit.

Feedback features in the amplifiers result in good linearity and stability properties. An amplification of $10^4$ can be achieved; it is designed to amplify pulses only.

Filament power is supplied from a transformer on the chassis. A separate power supply (Fig. 47) delivers regulated B-plus (300 V.D.C.). All power supplies were operated on a 5KVA "stabiline" (Superior Electric Co.) line voltage regulator.

Slow coincidence circuits ($\tau_{res}$ about 1 $\mu$sec.) are diagrammed in Fig. 48; a view of the chassis layout is also shown (Fig. 49). Filament and B-plus power for all coincidence channels was distributed by the circuit in Fig. 50.

At least two stages of fast amplification, using wide-band "chain" amplifiers (Hewlett-Packard 460A or Spencer-Kennedy 202C), are required to drive the fast coincidence channels (refer to Figs. 51 and 52). The latter are designed for quick response and recovery times. Use is made of a length of RG-62U coaxial cable with an r-f short at one end to shape the pulse and determine resolving time. A cable length of 121 cm introduces
Figure 51

Fast Coincidence Circuit
Figure 51
FAST COINCIDENCE CIRCUIT (3 CHANNEL)
Figure 52

Chassis Layout of
Fast Coincidence Circuits
5 mμsec delay (1 mμsec = 24.5 cm of RG62U cable), so that the pulse is cut off after 10 mμsec. Resolving time can be measured by observing coincidences from a source with only one gamma ray (e.g. Na$^{22}$) and applying equation (2); or by observing the half-width of the coincidence-delay peak (Fig. 53). Both methods yield $\tau_{res} \approx 10^{-8}$ sec.

**Figure 53**

Dependence of Fast Coincidences on Relative Delay in the Fast Channels

Berkeley 410 electronic decade counters were used for scaling, with electric clocks (Standard Electric Time Co. S-100) for timing purposes. The counters could be stopped and started simultaneously by the electronic switch of Fig. 54, or by a simple "ganging" of mechanical switches. The latter must be positive acting with no "bounce" when making contact; toggle switches are not suitable for this purpose.

4. **Polarizing Field.**

For polarizing nuclei by the Gorter-Rose method, a
Figure 54

Electronic Switch for Scalers
FIGURE 54
SWITCHING CIRCUIT

1/2 OF 12AU7

6AS6
small external field is applied. This was provided by a coaxial set of two Helmholtz pairs, which could be connected in parallel opposing (see Fig. 55). By making the dipole moments of the pairs equal and opposite, a very small fringing field results. The condition is that \( N_1 I_1 r_1^2 = N_2 I_2 r_2^2 \), where \( N \) is the number of turns, \( I \) the current, and \( r \) the radius of a single coil. As constructed, \( N_2 = 0.54 N_1 \), and \( I_2 = (2/3)I_1 \). For higher fields either pair can be used, or both, connected aiding. Power was supplied by eight 200 amp-hour storage batteries giving 100 V.D.C. and about 12 amps total. A certain amount of cooling was obtained by passing water through tubing soldered to the coil frame. Current was monitored by hand or with an electronic current regulator.

A quarter-section of the coil assembly is given in Fig. 56. The geometry is such that both pairs are at their respective Helmholtz conditions and subtend a rela-
Figure 56

Section Through Helmholtz Coils
1 3 0

1/64" MELAMINE INSULATION

DEWAR CLEARANCE

1/8" BRASS

SOLDERED JOINT

ROLLED BRASS

1/4" O.D. Cu TUBING

SOFT SOLDERED

TO BRASS.

THREADED JOINT

114 TURNS OF #18

G.E. FORMEX

MAGNET WIRE.

211 TURNS OF #16

G.E. FORMEX

MAGNET WIRE.

FIGURE 56

CROSS-SECTION OF HELMHOLTZ COIL PAIR
tively small solid angle of 20° at the sample position; this permits the radiation pattern to be observed if desired. Some construction data are given in Table III.

Fields achieved for coils opposed, aiding, and by the inner coil alone are plotted in Fig. 57. About 1200 watts was the maximum power dissipation which could be tolerated.

Table III
Design Data for Polarizing Field Coils

<table>
<thead>
<tr>
<th>Coil</th>
<th>Mean Radius (in.)</th>
<th>Wire Size (A.W.G.)</th>
<th>Total Turns</th>
<th>Layers</th>
<th>Turns per Layer</th>
<th>Resistance at 20°C (ohms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inside</td>
<td>3</td>
<td>16</td>
<td>211</td>
<td>16</td>
<td>13</td>
<td>4</td>
</tr>
<tr>
<td>Outside</td>
<td>5</td>
<td>18</td>
<td>114</td>
<td>11</td>
<td>10</td>
<td>5</td>
</tr>
</tbody>
</table>

The counters and field coils could be rolled into position around the dewar tail (Fig. 58) on a movable table. Either the counters on the coils could be rotated independently through 90° around a vertical axis.
Figure 57
Polarizing Field Curves
Polarizing field from coaxial Helmholtz coil pairs for varying current through inner coils.

Figure 57

Graph showing the relationship between $I_1$ (amps) and $H_p$ (oersteds) for

- Inner coil only ($I_2 = 0$)
- In parallel, aiding ($I_2 = 2/3 I_1$)
- In parallel, opposing ($I_2 = 2/3 I_1$)
Figure 58

Closeup of the Counters and Field Coils in Position around the Tail of the Cryostat
1. Sample Preparation.

A pile-irradiated cobalt sample (obtained from Oak Ridge National Laboratory) in the form of cobalt chloride was used. It contained about one millicurie of cobalt 60, giving 20,000 counts per minute at 20 inches from the window of a Geiger tube. After being evaporated to dryness under a heat lamp, the sample was taken up in a drop of concentrated nitric acid.

Next, 20 grams of magnesium nitrate hexahydrate and 22.6 grams of cerous nitrate hexahydrate crystals were dissolved in 50 ml of distilled water, and the radioactivity was added to the solution. At this point, the pH of the mixture could be adjusted in the range 2-6, say, by the addition of magnesium oxide and water. Excess magnesium oxide can be removed by centrifuging and decanting. The rate of crystal production depends on the pH of the solution, which of course is rather on the acid side and changes with volume of the solution. It was found that, by starting with a pH in the range mentioned above, crystals of fairly good quality were produced in a reasonable time.

By using a heat lamp to reduce the volume of the
solution to about 30 ml, very small crystals will begin to form at the bottom of the beaker. Then the solution should be transferred to a desiccator for growth of the crystals by evaporation over a desiccant such as sulfuric acid. A Styrofoam (Dow expanded polystyrene) box was constructed to contain the desiccator, in order that temperature changes would be minimized. Evaporation from open containers is too rapid to be practicable, as crusts tend to form on the surface.

When some crystalline cerium magnesium nitrate has formed on the bottom of the beaker, it should be culled for seed crystals. These are small, clear crystals exhibiting the characteristic hexagonal flat plate shape. They should be placed in the solution on a wire screen support. Copper screen is usable but is attacked by the acid solution; platinum or plastic screen is preferred.

Growing paramagnetic crystals need to be turned over and rotated every day to insure symmetric accumulation of material without "barnacles." Of course, slow growth leads to the best quality crystals. Eight single crystals may ordinarily be produced in two to four weeks by these methods. They should be used as soon as possible, since the large amount of hydration which provides the g-factor anisotropy is unstable. Dehydration tends to take place rather quickly, even when crystals are
stored over a saturated solution. It is better to keep the crystals in solution until needed.

On being removed for use, such a specimen usually contains about 10-40 microcuries of cobalt 60. No cobalt nitrate carrier solution was used, as it would hinder the introduction of the radioactivity into the sample.

In general one wants small specific activity to reduce beta-ray heating of the crystals, which have only about 500 ergs per gram of heat capacity between their Curie point and 1°K. As a rule-of-thumb one may expect roughly one-half erg per minute heating per microcurie of cobalt 60, if it is assumed that all betas and no gammas are absorbed in the source. Natural heat leaks such as are discussed in Chapter III will be present in addition to the radioactive heat leak.


Introducing cobalt 60 ions as divalent impurities in single crystals of Ce₂Mg₃(NO₃)₂₄·2₄H₂O is a technique already employed by Ambler et al.¹ to obtain oriented nuclei. Anisotropies of the gamma ray patterns of up to 50% were reported, depending on the strength of the small external magnetic field applied (0-430 gauss). Relevant thermal and magnetic properties of this salt

Figure 59

Entropy of Cerium Magnesium Nitrate

The zero-field curve is from Daniels and Robinson. Curves in strong fields were computed from the tables of J. R. Hull and R. A. Hull, J. Chem. Phys. 2, 465 (1941).
FIGURE 59

MAGNETIC ENTROPY OF CERIUM MAGNESIUM NITRATE
Figure 60

Entropy of Cerium Magnesium Nitrate Near 1°K as a Function of Field

The curves were computed using tables of J. R. Hull and R. A. Hull, J. Chem. Phys. 9, 465 (1941)
FIGURE 60
MAGNETIC ENTROPY OF CERIUM MAGNESIUM NITRATE AT 1°K

\[
\frac{S}{R} \quad \text{vs} \quad \frac{H}{T} \quad (\text{kilogauss/degree})
\]
Figure 61

Properties of Cerium Magnesium Nitrate in the Region of the Curie Point

Data is taken from the paper of Daniels and Robinson.
FIGURE 61
PROPERTIES OF CERIUM MAGNESIUM NITRATE
AT THE CURIE POINT.
at low temperatures have been investigated by Cooke, Duffus, and Wolfe,2) and by Daniels and Robinson.3) Cooke and his co-workers find a Curie law susceptibility in the helium region given by

\[ \chi = \left( \frac{4.1}{T} \right) + 0.32 \times 10^{-4} \text{ per gram.} \]  

(1)

The crystalline density is 1.87 grams per cm\(^3\). Further, Daniels and Robinson found this Curie law to be obeyed within 2% down to six millidegrees, attainable by a demagnetization from 7,000 gauss at 1\(^\circ\)K. The minimum temperature attainable is about three millidegrees, determined by small ionic interactions believed to be of dipole-dipole nature; stable cerium isotopes have no magnetic moment and hence give no HFS. Above .01\(^\circ\)K, Daniels and Robinson found a specific heat

\[ \frac{C}{R} = \left( \frac{6.4}{T^2} \right) \times 10^{-6}. \]  

(2)

Entropy curves are given in Figs. 59 and 60, and the Curie point properties deduced by Daniels and Robinson are graphed in Fig. 61.

The success of magnetic hyperfine structure polarization is made possible by the very low temperature attained by adiabatic demagnetization of this salt, as well as by the highly anisotropic electronic g-factor of the cerium ions (g =0.25, g =1.84 as measured by Cooke

et. al.). The latter property enables one to apply a small magnetic field along the crystal axis immediately following demagnetizing without serious heating. Note that the final temperature is determined almost entirely by the cerium ions, rather than the cobalt ions which are in high dilution. Another advantage of the salt is that there is a single axis of orientation.

Paramagnetic resonance measurements on cobalt ions in bismuth magnesium nitrate suggest the complication that two magnetically non-equivalent ions may be present in the crystal. Type "X" ions have a practically isotropic g-factor and hfs, while type "Y" ions possess extremely anisotropic hfs and g-tensor with axial symmetry along the crystalline axis. The former outnumber the latter by about two to one. These measurements are summarized in Table IV. It should be pointed out that it is desirable to have these measurements extended to 4°K., and to the cerium salt itself if this be feasible. However, one needs to apply the existing results with caution, since the additional strong ionic couplings present in the cerium salt undoubtedly change the local fields experienced by a cobalt ion and its nucleus.

Table IV

Paramagnetic Resonance Parameters of Cobalt 59 in Bismuth Magnesium Nitrate at 20°C (Trenam).

<table>
<thead>
<tr>
<th>Ion</th>
<th>$E_{ll}$</th>
<th>$E_{lt}$</th>
<th>$a$ (cm$^{-1}$)</th>
<th>$b$ (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{111}X^{m}$</td>
<td>4.108</td>
<td>4.385</td>
<td>.0085</td>
<td>.0103</td>
</tr>
<tr>
<td>$^{111}Y^{m}$</td>
<td>7.29</td>
<td>2.338</td>
<td>.0283</td>
<td>&lt;10$^{-4}$</td>
</tr>
</tbody>
</table>

Adiabatically demagnetizing from 24KG in the plane of the crystals (perpendicular to the axis) and 1°C assures a final temperature in the Curie region even after a 400-gauss polarizing field is applied. The counters and field coils could be put into place in 45-60 seconds from the time of demagnetization, and with heat leaks estimated to be 20-30 ergs/min. at the lower temperatures a useful orientation time of 20-25 minutes was achieved in the earlier experiments.

Coincidences were recorded continuously, with readings being taken every 20 seconds; singles counts, on the other hand, were integrated over a 15-second interval, with a 5 second off-period for recording and resetting the scalers (mechanical registers were not reset). An advantage of the time resolution plus energy selection afforded by the circuitry described in Chapter III was that the observed coincidence rates were practically all true.
coincidences, the accidental and background corrections being small. High discriminator settings selected only the photopeaks of the gamma distribution, so that the effect of scattering in the source and cryostat, in the detectors, and between pairs of detectors was eliminated. Typical counting rates were about one coincidence per second and 1500 singles per second in the initial runs.

Before a run, several checks must be made on the counting equipment. First, the electronic circuits should be turned on at least 48 hours in advance in order to reach an equilibrium temperature. A differential spectrum of the pulse distribution must then be run in order to locate the proper discriminator settings.

The coincidence-delay peak of the fast coincidence circuits needs to be checked by inserting various lengths of RG62-U cable before the fast amplifiers. When 50 millimicroseconds of delay is inserted, the "background" coincidence rate is obtained, and this should correspond roughly to the random background computed from the singles rate and $2\tau_{\text{res}}$. Width of the coincidence-delay peak (Fig. 53, Chapter III) gives the quantity $2\tau_{\text{res}}$.

Finally, a curve of coincidence rate versus pulse height selector setting of the fast coincidence circuit is traced. This has a small plateau between regions of
steeply ascending and descending rates representing pulses leaking through and biasing a
above cutoff of the output circuit, respectively. The setting should be on this plateau (see Fig. 62), which may be narrow unless sufficient fast amplification is available.

Success of the experiment depends finally on gaining sufficient counting statistics, in view of the relatively short times of orientation. This is possible if a series of demagnetizations is made to build up the number of true coincidences corresponding to any chosen range of anisotropies. Ten demagnetizations yielded roughly 3% statistics in the correlation.

After orientation had disappeared, longer counting intervals could be taken, and these counting rates $C_i^o$ were used to normalize both singles and coincidence rates (see Chapter II) to the situation of random orientation, so that only temperature-dependent effects are exhibited (the counting apparatus being sufficiently stable over the periods of time involved). Fluctuations in the counting time intervals are corrected for by
taking the ratios of counting rates for corresponding intervals.

It should be noted that the source strength is cancelled by the normalization of singles and coincidence counts (see Chapter II). Hence it is in general unnecessary to correct for source decay during the experiment. The half life of cobalt 60 is of course so great that no measurable decay was observed. In fact, the half-life limitation is not too stringent, since one may start with enough activity so that there is still a measurable amount left when the experiments are performed. A practical lower limit of about one day is set by the time necessary for crystal growth. Useful counting rates may then be achieved by varying the detector solid angle. In general, for coincidence experiments relatively weak sources and large solid angles are desirable to decrease the accidental background.

3. Temperature Measurement.

A mutual inductance bridge measuring the magnetic moment of the sample is conventionally used to define a magnetic temperature parameter $T^\circ$ by means of Curie's Law. Although interference from the guard salt can be virtually eliminated by proper coil design, other effects which complicate the conversion of $T^\circ$ to the true
thermodynamic temperature $T$ are the presence of the polarizing field, the question of the demagnetizing factor of the crystals, and the problem of the local fields at the ions.

Suppose a small alternating field $h$ is applied perpendicular to the crystal axis, and a static field $H$ is maintained along the axis (refer to Fig. 63). Assume that $h \ll H$ and that the interionic fields may be neglected, excluding the Curie region. Then one writes

$$m_h = \frac{N q_i^2 \mu_B^2}{4kT} = \frac{C_{\text{meas}}}{T_{\text{meas}}} = (\chi_L)_{\text{meas}}. \quad (3)$$

a Curie law defining $T_{\text{meas}}$. We have taken $J = S_{\text{eff}} = \frac{1}{2}$ for cerium ions at low temperatures. 5)

For correcting the temperature $T_{\text{meas}}$ in their results, the Oxford group used the expression 6)

$$m_h = \frac{N q_i^2 \mu_B^2}{2g_i H} \tanh \frac{g_i \mu_B H}{2kT} = \frac{N q_i^2 \mu_B^2}{2kT} \tanh \frac{x}{x}. \quad (4)$$

where $x = \frac{g_i \mu_B H}{2kT}$.

Now equation (4) gives a modified Curie Law defining $T_\text{\tiny{\ominus}}$ which is expected to be a valid approximation in the region where $T_\text{\tiny{\ominus}} = T$. Combining equations (3) and (4) yields the relation

$$T_\text{\tiny{\ominus}} = \frac{g_i \mu_B H}{2kT} \left[ \tanh \frac{g_i \mu_B H}{2kT_{\text{meas}}} \right]^{-1}. \quad (5)$$

Note that $T_\text{\tiny{\ominus}} < T_{\text{meas}}$, i.e. presence of a static field

6) E. Ambler, private communication.
causes the $^\circ{T}$ thermometer to read high.

In practice, $T_{\text{meas}}$ is not observed because the single crystals are not spherical, nor even ellipsoidal. Hence the internal field is not uniform and a correction for the demagnetizing effect of induced surface magnetization is not strictly applicable. If the demagnetizing factor $D$ is given by

$$H_{\text{internal}} = H_{\text{applied}} - DM$$

where $M$ is the uniform magnetization per unit volume, then the temperature correction $\Delta$ is

$$\Delta = T^{\circ} - T^{*} = C\left(4\pi M - D\right).$$

In equation (7), $C$ is the Curie constant per unit volume. An approximate calculation based on the actual shape (flat hexagon) and size (about 1 cm across by 3 mm thick) of a single crystal gives $D \approx 2.0$ to $1.7$ with corresponding values of $\Delta \approx 1$ to $2$ millidegrees. Approximating the crystal by a flat disk or a thin oblate ellipsoid yields $D \approx 0.18$, and $\Delta \approx 3$ millidegrees.

For practical purposes the demagnetization correction may be estimated from $T_{\text{meas}}^*$ immediately after demagnetizing to zero field, assuming $T^{\circ}$ as given by Daniels and Robinson's work. This leads to a $\Delta$ of about 2 millidegrees.

---


8) Ibid., p.39
With these two corrections the observed $T_{\text{meas}}$ can be converted to $T = T^*$, giving a fair estimate of temperature in the region .006 to 1°K., say. Uncertainties in the local fields experienced by the ions cause additional complications in the interpretation of temperature measurements; see the discussions of De Klerk\(^9\) and Casimir.\(^10\)

If one follows the convention of using the Lorentz local field, the bridge data of a typical run is reduced as shown in Table V. A warmup curve is plotted in Fig. 63.


Frauenfelder\(^11\) and Deutsch\(^12\) have discussed appropriate corrections to be applied to data in a correlation experiment. These may be enumerated briefly as follows:

(a) Accidental coincidences will result from the finite resolving time $\tau_{\text{res}}$ of the coincidence circuit. This rate is

$$C_{12}^{\text{acc}} = 2 \tau_{\text{res}} C_1 C_2$$  \hspace{1cm} (8)

where $C_1$ and $C_2$ are the singles rates of the two counters. By placing a source of single gamma ray or non-coincident gammas in front of the counters, one measures this rate directly and computes $\tau_{\text{res}}$. In these experiments the

\(^10\)H. B. G. Casimir, op. cit., p. 56.
\(^12\)M. Deutsch, Repts, Prog. Phys. 14, 196 (1951).
fast coincidence resolving time was about 10 μsec, making the accidental correction of the order of a few per cent. This agrees well with the background observed by inserting 50 μsec. delay in the first fast channel. It was found to be rather low (about 1%) in the earliest experiments, and about 10% in later runs with a stronger source. The correction to the ratio C₁₃/C₁₂ was never more than 2%.

(b) "Prompt" singles and coincidence background due to contamination, nearby radioactive sources, or cosmic rays is observed with the sample removed, and was found to be completely negligible. Background due to other activities present in the source should be subtracted unless energy selection is made. Such a correction was not found to be necessary in these experiments.

(c) Counting losses may occur at high counting rates, particularly in scaling circuits. A check was made with a signal generator, and no consistent losses were noted up to 10 kcps in any of the scalers believed to be in good order. Observed rates were always very much less than this.

(d) Coincidences may arise from pair production and positron decay in the counters and source if the gamma energy exceeds 1 Mev, or from the decay of a positron-emitter.
<table>
<thead>
<tr>
<th>t (sec)</th>
<th>M</th>
<th>$T^{\circ}$</th>
<th>$T^{\circ}$</th>
<th>$T^{\circ}$</th>
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<td>.0017</td>
<td>.0037</td>
<td>(.0034)</td>
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</tr>
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</table>

\[
T_{\text{meas}}^* = \frac{1.60}{M+40.9}
\]
\[
T_{\text{meas}} = T_{\text{meas}}^* + 0.002
\]
\[
T^\oplus = \frac{0.00179}{\arctan\left(\frac{0.00179}{T_{\text{meas}}}\right)}
\]
Figure 63
Typical Warmup Curve
The resulting tendency to increase the correlation at 180° may be eliminated by energy selection.

(e) Scattering in the source, the cryostat, the detectors, and between detectors should be considered. The simplest way to eliminate this is to make rigorous energy selection by means of a discriminator circuit or by heavy lead shielding. As the former was done here, the scattering correction was assumed to be negligible.

(f) Finite geometry of the source and detectors was considered to be strictly a problem of geometry, and the theory was corrected accordingly (see Chapter II, Section 3).

(g) Asymmetry of the correlation due to source decentering is corrected to first order by taking the normalized coincidence rate \( \text{CR}_{12} \frac{C_{12}}{C_0} \), etc.

(h) Source decay need not be corrected for when the normalized rate is taken, since the activity is cancelled in the ratio.

Only correction (a) was considered to be important, although usually small. Now

\[
C_{12}^{\text{net}} = C_{12}^{\text{gross}} - 2 \gamma_{12} C_1 C_2
\]

so that

\[
\frac{C_{13}^{\text{net}}}{C_{12}^{\text{net}}} = \frac{C_{13}^{\text{gross}} - 2 \gamma_{13} C_1 C_3}{C_{12}^{\text{gross}} - 2 \gamma_{12} C_1 C_2}
\]

\[
\approx (\frac{C_{13}}{C_{12}})^{\text{gross}} \left[ 1 + 2 \gamma_{12} \frac{C_2}{C_{12}} \left( 1 - \frac{\gamma_{13} C_3}{\gamma_{12} C_2} \right) + \ldots \right]
\]
In case \( C_{13} \approx C_{12} = C \), a further simplification yields, using \( C_1C_2 \approx C_1^0C_2^0 \),
\[
\left( \frac{C_{13}}{C_{12}} \right)^{\text{net}} \approx \left( \frac{C_{13}}{C_{12}} \right)^{\text{max}} \left[ 1 + 2x \frac{C_1^0C_2^0}{C_{12}} \left( 1 - \frac{C_{13}C_3}{C_{13}C_2} \right) + \cdots \right] \quad (10')
\]

Now the correction may be estimated. For example, in early runs \( \frac{C_1^0C_2^0}{C_{12}} \approx 1.8 \times 10^6 \), \( \left( 1 - \frac{C_{13}C_3}{C_{13}C_2} \right) = .12 \), and
\[
2x \approx 2 \times 10^{-8} \text{ sec} \quad \text{so the bracketed correction factor in equation (13')} \text{ was about 1.01 for random orientation.}
\]
Note that the correction will be somewhat orientation dependent, decreasing with increasing orientation when \( \theta_1 = 0 \) and increasing when \( \theta_1 = \pi/2 \).

5. Typical Data.

Total counts recorded by observers at each of the scalers were subtracted to get singles and coincidence counts per timing interval. Singles readings correspond to an average over 15 seconds. Coincidences were integrated over 100 seconds in order to reduce scatter to the point where a definite trend could be seen.

Then the uncorrected ratios \( C_{13}/C_{12} \) and \( C_3/C_2 \) were computed to eliminate timing errors. These are plotted against time from demagnetization in Figure 64. The normalizing ratio \( C_3^0/C_2^0 \) was found from the tail of the curve, upon warm-up to bath temperature.

After being reduced as illustrated in Table VI,
individual values of CR_{13}/CR_{12} from all runs could be grouped according to the average value of N_3/N_2 to which they belong (with an uncertainty of about ±0.3 in the anisotropy resulting). This grouping was arbitrarily taken over intervals .05 units wide, all points in any one group being averaged together.

The observed angular distribution pattern anisotropy and the correlation are difficult to fit individually with any theoretical curve because of the presence and unknown contribution of the "Y*" ions. For the purposes of testing the theory of correlation, therefore, it was decided always to plot correlation versus anisotropy, effectively eliminating $\beta$ as a theoretical parameter and both H and T* as experimental parameters. This is to say that the measured radiation pattern defines an "equivalent" $\beta$ describing an ideal ensemble of nuclei which would give the same anisotropy and a corresponding directional correlation at a given temperature. The extent to which this procedure is valid when two orienting mechanisms are present will be discussed in the last chapter.
Figure 64

Plot of data from a typical run, Aug. 9, 1954 DEMAG 1.
TABLE VI

Reduction of the Data from a Typical Run (Aug. 9, 1954, Demag 1)

<table>
<thead>
<tr>
<th>Period (t)</th>
<th>Corrected $\frac{C_{13}}{C_{12}}$</th>
<th>Corrected $\frac{C_{13}}{C_{12}}$</th>
<th>Corrected $\frac{C_2}{C_3}$</th>
<th>Corrected $\frac{N_2}{N_3}$</th>
<th>Corrected $\frac{CR_{12}}{CR_{13}}$</th>
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<tbody>
<tr>
<td>0</td>
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<td></td>
<td></td>
<td></td>
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</tr>
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<td>40</td>
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<td>.79</td>
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<td>180</td>
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<td>.81</td>
<td>.83</td>
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<td>220</td>
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<td>.85</td>
<td>.81</td>
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</tr>
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<td>240</td>
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<td>260</td>
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<td>.87</td>
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<tr>
<td>280</td>
<td>1.07</td>
<td>1.08</td>
<td>.88</td>
<td>.84</td>
<td>1.07</td>
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<td>300</td>
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<td>.89</td>
<td>.85</td>
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<tr>
<td>380</td>
<td>1.05</td>
<td>1.06</td>
<td>.91</td>
<td>.87</td>
<td>1.01</td>
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<tr>
<td>400</td>
<td></td>
<td>.92</td>
<td>.88</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Period at t</td>
<td>$\frac{C_{13}}{C_{12}}$</td>
<td>Corrected $\frac{C_{13}}{C_{12}}$</td>
<td>$C_3$</td>
<td>$N_3$</td>
<td>$\text{CR}_{13}$</td>
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<tr>
<td>------------</td>
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<td>420</td>
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<td></td>
<td>.93</td>
<td>.89</td>
<td></td>
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<td>440</td>
<td></td>
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<td>.94</td>
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<td></td>
<td></td>
<td>.94</td>
<td>.90</td>
<td></td>
</tr>
<tr>
<td>480</td>
<td>1.06</td>
<td>1.07</td>
<td>.96</td>
<td>.91</td>
<td>1.02</td>
</tr>
<tr>
<td>500</td>
<td></td>
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<td>.94</td>
<td>.90</td>
<td></td>
</tr>
<tr>
<td>520</td>
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<td>.96</td>
<td>.91</td>
<td></td>
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<td>.95</td>
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<td></td>
</tr>
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<td>.90</td>
<td></td>
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<tr>
<td>580</td>
<td>.86</td>
<td>.87</td>
<td>.97</td>
<td>.92</td>
<td>.83</td>
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</tr>
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<td>620</td>
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<td>1.01</td>
<td>.96</td>
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<td>640</td>
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<td>1.23</td>
<td>1.23</td>
<td>1.025</td>
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<td>1.28</td>
<td>1.29</td>
<td>1.025</td>
<td>.98</td>
<td>1.23</td>
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<tr>
<td>765</td>
<td>1.11</td>
<td>1.12</td>
<td>1.04</td>
<td>.99</td>
<td>1.08</td>
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<td>880</td>
<td>1.26</td>
<td>1.27</td>
<td>1.05</td>
<td>1.0</td>
<td>1.21</td>
</tr>
<tr>
<td>H = 0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>985-1260</td>
<td>1.02</td>
<td>1.03</td>
<td>1.05</td>
<td>1.0</td>
<td>1.01</td>
</tr>
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</table>
CHAPTER V
RESULTS AND DISCUSSION

1. Angular Correlation at Very Low Temperatures and at Room Temperature.

The measured effect on the angular correlation of the Ni-60 gamma rays, produced by nuclear orientation for $\theta_1 = 0$ is shown in Fig. 65; the numerical values are tabulated in Table VII. Theoretical curves have been drawn in on the figure for comparison. It should be noted that the same theoretical curve applies to all beta-decay schemes, since for moderate degrees of orientation both distribution and correlation have similar dependence on orientation in all three cases (Chapter II). Hence the various beta-decay possibilities cannot be distinguished by these angular correlation measurements. For higher degrees of orientation it may be possible to make this distinction, provided sufficient statistical precision is attained.

Results have also been obtained on the arrangement $\theta_1 = \pi/2$. These are plotted in Fig. 66 and summarized in Table VIII. Table IX presents some preliminary data which illustrates quite well the striking difference between the two arrangements. Smaller anisotropies and faster warmup times, probably due to dehydration and
imperfection of the crystals, limited the amount of
data obtained from the latter runs.

The anisotropies observed are of the same magni-
tude as those found by the Oxford group for comparable
applied fields. No effect was noted in zero field at
temperatures in the range down to about .01°C.

As a test of the apparatus, the angular correla-
tion was measured for random orientation of the cobalt
nuclei in the crystals and also in a liquid solution
from which the crystals were grown. Table X summarizes
these findings.

2. Discussion of Results.

The theoretical curves with which the experimental
points are compared are obtained by use of Cox and
Tolhoek's formulas for angular correlation and radiation
anisotropy as a function of the degree of orientation.
The curves shown hold explicitly for a Boltzmann dis-
tribution over magnetic substates. As indicated in
Chapter IV, it is not to be expected that this is the
actual state of affairs; however, the comparatively good
agreement found may be interpreted to mean that the
departure of the actual population densities from the
exponential law is either (a) fairly small or (b) of
little consequence because summation over the initial
Table VII

Gamma-gamma Angular Correlation
of Oriented Co$^{60}$ Nuclei at Very
Low Temperatures, for $\theta_1=0$ and $H=200$ Gauss
(Aug. 4, 9, 1954)

<table>
<thead>
<tr>
<th>$N_3/N_2$</th>
<th>$CR_{13}/CR_{12}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.70 - .75</td>
<td>.993 ± .041</td>
</tr>
<tr>
<td>.75 - .80</td>
<td>1.012 ± .035</td>
</tr>
<tr>
<td>1.80 - .85</td>
<td>1.053 ± .036</td>
</tr>
<tr>
<td>.85 - .90</td>
<td>1.114 ± .035</td>
</tr>
<tr>
<td>.90 - .95</td>
<td>1.088 ± .034</td>
</tr>
<tr>
<td>.95 - 1.0</td>
<td>1.160 ± .033</td>
</tr>
<tr>
<td>1.0</td>
<td>1.151 ± .026</td>
</tr>
</tbody>
</table>
Figure 65
Angular Correlation as a Function of Anisotropy for $\theta$,=0 and $H=200$ Gauss
Figure 65

Angular correlation as a function of the anisotropy of the angular distribution.
Co\(^{60}\) in Ca\(_2\)Mg\(_3\)(NO\(_3\))\(_{12}\)·24H\(_2\)O

**Theoretical curve (Cox & Tolhoek)**

**Theory corrected for finite geometry.**
Figure 66.
Angular Correlation as a Function
of the Anisotropy for $\theta, = \pi/2$ and $H=200$ Gauss
Figure 66

Angular correlation as a function of anisotropy for $\theta = \pi/2$

Theoretical curve (Cox & Tolhoek)

Theory corrected for finite geometry.
Gamma-gamma Angular Correlation
in Nickel 60 at Very Low Temperatures
for $\theta_1 = \pi/2$ and $H=200$ Gauss
(Feb. 12, 1955)

<table>
<thead>
<tr>
<th>$N_2/N_3$</th>
<th>$\text{Gr}<em>{13}/\text{Gr}</em>{12}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.80 - .85</td>
<td>1.436 ± .048</td>
</tr>
<tr>
<td>.85 - .90</td>
<td>1.343 ± .046</td>
</tr>
<tr>
<td>.90 - .95</td>
<td>1.246 ± .056</td>
</tr>
<tr>
<td>.95 - 1.0</td>
<td>1.242 ± .022</td>
</tr>
<tr>
<td>1.0</td>
<td>1.180 ± .020</td>
</tr>
</tbody>
</table>
Table IX
Dependence of Angular Correlation on Position of the Counters; H=400 Gauss
(Jan. 11, 12, 1955)

<table>
<thead>
<tr>
<th>Anisotropy $\epsilon$</th>
<th>$\Theta_1 = 0$</th>
<th>$\Theta_1 = \pi/2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0$</td>
<td>$1.26 \pm .03$</td>
<td>$1.31 \pm .02$</td>
</tr>
<tr>
<td>$0 -.05$</td>
<td>$1.18 \pm .06$</td>
<td>$1.32 \pm .06$</td>
</tr>
<tr>
<td>$.05-.10$</td>
<td>$1.12 \pm .06$</td>
<td>$1.33 \pm .08$</td>
</tr>
<tr>
<td>$.10-.15$</td>
<td>$.97 \pm .10$</td>
<td>$1.62 \pm .09$</td>
</tr>
<tr>
<td>$.15-.20$</td>
<td>$.99 \pm .10$</td>
<td>$1.62 \pm .13$</td>
</tr>
</tbody>
</table>
magnetic substates washes out details of the specific statistical weights. To this extent the procedure appears justified of using an "equivalent $\beta$," defined (Chapter IV) as the parameter in the Boltzmann distribution $\exp (-\beta m)$ that gives the same anisotropy as the actual distribution of states. We recall that this parameter is related to the nuclear and environmental properties by the expression

$$\beta = \frac{g\mu \nu n B}{kT}$$

where $B$ is the average value of the component of the magnetic field at the nucleus in the direction of polarization.

Within the limits of statistical accuracy, the results show the expected dependence on temperature, and are quantitatively in agreement with the expression derived by Cox and Tolhoek for a 4-2-0, quadrupole-quadrupole cascade. The results lie about 2 to 3 percent above the theoretical curve 2-3% for $\Theta_1=0$, and almost 20% above for $\Theta_1=\pi/2$.

A rough calculation was made to determine whether this difference could result, in direction and magnitude, from breakdown of the assumption that none of the counters distinguishes between the two gamma-rays $\gamma_a(1.17 \text{ Mev})$ and $\gamma_b(1.33 \text{ Mev})$. If the scintillation counters are not equally sensitive to each gamma-ray and the
channel efficiencies are different, then the general equation (30) of Chapter II must be used, with the result that the efficiency factors can no longer be factored out of the correlation.

As an approximation, assume that the detection efficiency of the i-th counter for a particular gamma energy is

$$\varepsilon_i (\gamma) \propto C_i^0 \left[1 - \exp(-\mu_x x)\right],$$

where \(x\) is the thickness of the crystal, \(\mu_x\) is the linear absorption coefficient of sodium iodide for the given photon energy, and \(C_i^0\) is the counting rate at the end of a run. In our arrangement all the geometrical solid angles were equal, so that different \(C_i^0\), e.g. varying from 1200-1500 cps in the August runs, were due primarily to different discriminator settings. From the review article of Jordan\(^1\)), one may estimate values for the absorption coefficients \(\mu(1.33) \approx 0.015\) and \(\mu(1.17) \approx 0.024\) cm\(^{-1}\). These figures lead to a value of the efficiency-dependent factor in equation (30) near 1.00-1.01, so that individual experimental points would only be moved down 1% or less.

Magnetic hyperfine structure, besides making the nuclear orientation possible, might be expected to perturb the angular distributions and correlation if the

---

Table X
Angular Correlation at Room Temperature and 80°K; H=0.

Co$^{60}$ in Ce$_2$Mg$_3$(NO$_3$)$_{12}$ 24H$_2$O
(Combined results of Aug., 1954, Jan. and Feb., 1955 runs)

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T$</th>
<th>$C^{R_{13}}/C^{R_{12}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystals in Cryostat</td>
<td>80°K.</td>
<td>1.150 ± 0.005</td>
</tr>
<tr>
<td>Crystals in Cryostat</td>
<td>300°K.</td>
<td>1.173 ± 0.006</td>
</tr>
<tr>
<td>Crystals</td>
<td>300°K.</td>
<td>1.136 ± 0.005</td>
</tr>
<tr>
<td>Liquid Solution</td>
<td>300°K.</td>
<td>1.160 ± 0.007</td>
</tr>
</tbody>
</table>
excited states of nickel 60 were long-lived, particularly the intermediate state of the cascade (see Chapt. II, Sec. 5). Alder \(^2\) has given attenuation factors for the correlation in the presence of a paramagnetic electron shell. In the cobalt ions (and presumably in the nickel which results from the beta-decay) hfs fields of the order of \(10^5-10^6\) Gauss exist, corresponding to isotropic hfs splittings \(\Delta \nu\) near .01 cm\(^{-1}\); this leads to critical times \(\tau_c = (2\pi c \Delta \nu)^{-1} \approx 5 \times 10^{-10}\) seconds. If the intermediate state lifetime \(\tau_\theta\) is much less than \(\tau_0\) the directional correlation is unaffected.

The present results appear to be insufficiently good agreement with the unperturbed correlation to justify placing an upper limit to the mean life of the intermediate state of \(\tau_\theta \leq 10^{-11}\) sec. However, no calculation of the effect of static perturbation of the intermediate state on the angular correlation has yet been made for the case of initially oriented nuclei. It is in fact possible, though probably not likely, that reorientation of the intermediate state may account for the observed increased departure of angular correlation from the room temperature value with increased applied magnetic field. Steenberg\(^3\) has discussed the effect of

\(^3\)N. R. Steenberg, Phys. Rev. 95, 982 (1954).
Finally, an upper limit of $10^{-11}$ sec. on the lifetimes of both excited states of nickel 60 was estimated from delayed-coincidence measurements by Bay, Henri, and McLernon. 9)

It has been hitherto assumed that only one type of orientation mechanism must be considered. An effect that could reduce the agreement between experiment and any single-mechanism theory is the presence of "$Y$" type ions with hfs favoring Bleaney alignment in zero field. It is practically impossible to make a good estimate of this contribution to the distribution and correlation patterns, in view of the unexplained behavior of these ions in zero field; i.e. the measured anisotropy is zero down to .01°K. With two such patterns superimposed, one can no longer assume a single equivalent $\beta$-value; however it should still be possible to estimate what the effect would be if an equivalent $\beta$ could be assigned to each mechanism.

If for $\Theta_i = 0$ the "$Y$" contribution has a smaller anisotropy and an angular correlation correspondingly closer to the non-oriented value (as would seem reasonable) the resulting effect on the total angular correlation, for a given observed anisotropy, due to both $X$ and $Y$-

type ions is to reduce the departure of the angular correlation from the non-oriented value. The latter statement also holds for $\theta_1 = \pi/2$. See Fig. 67. Note that in combining the two contributions the radiation intensities must be added rather than the anisotropies.

Figure 67

Effect of a Small Admixture of Bleaney Alignment on the Relation Between Angular Correlation and Anisotropy

Estimates of a "Y" effect based on Trenam's resonance data, which would entail an anisotropy contribution of the order of 20%, lead to departures from the single mechanism relation which lie well outside statistical agreement with experimental results. A 5 to 10% contribution from the Bleaney ions would, on the other hand, be sufficient to bring the theory and experiment into almost exact agreement for the case $\theta_1 = 0$; however the
discrepancy between theory and experiment for $\theta_1 = \pi/2$ would be increased.

The room temperature results are in good accord with precise measurements recently reported by several workers, $^{10,11,12,13,14}$ which are summarized in Table XI. The expected angular correlation for the Swiss group was estimated from details of their equipment reported elsewhere. $^{15}$

That many workers observe the essentially full angular correlation at room temperature indicates that the nickel 60 correlation is very insensitive to intermediate state perturbations, presumably due to a very short $\tau_0$. Abragam and Pound $^{16}$ have given a comprehensive review of the perturbing mechanisms. Aeppli et al. could not change the correlation by the application of an 8000 gauss external field. First to report a search for a temperature-dependent effect, Lemmer and Grace could observe none down to 20°C.

TABLE XI

Summary of Angular Correlation Results for Randomly Oriented Nickel 60 Nuclei

Full theoretical value: \( W(\pi)/W(\pi/2) = 1.1667 \)

<table>
<thead>
<tr>
<th>Investigators</th>
<th>Measured Correlation</th>
<th>Corrected for Finite Geometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sapp, Jastram, Daunt</td>
<td>1.150 ± .005</td>
<td>---</td>
</tr>
<tr>
<td></td>
<td>1.161 ± .004</td>
<td>1.146</td>
</tr>
<tr>
<td></td>
<td>1.160 ± .007</td>
<td></td>
</tr>
<tr>
<td>Aeppler, Frauenfelder,</td>
<td>1.148 ± .003</td>
<td>---</td>
</tr>
<tr>
<td>Heer, Ruetschi</td>
<td></td>
<td>(1.153)</td>
</tr>
<tr>
<td>Klema and McGowan</td>
<td>1.1520 ± .0038</td>
<td>---</td>
</tr>
<tr>
<td></td>
<td>1.1453 ± .0023</td>
<td>1.1517</td>
</tr>
<tr>
<td>Lawson and Frauenfelder</td>
<td>1.159 ± .002</td>
<td>1.166</td>
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<tr>
<td></td>
<td>1.163 ± .003</td>
<td>1.168</td>
</tr>
<tr>
<td>Kloepper</td>
<td>---</td>
<td>1.170 ± .009</td>
</tr>
<tr>
<td>Lemmer and Grace</td>
<td>1.143 ± .002</td>
<td>---</td>
</tr>
<tr>
<td></td>
<td>1.142 ± .003</td>
<td>1.142</td>
</tr>
<tr>
<td></td>
<td>1.144 ± .005</td>
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</tr>
</tbody>
</table>
3. Conclusions.

An orientation-dependent effect upon the gamma-gamma directional correlation in nickel 60 has been observed, which may either increase or decrease the correlation. The magnitude of the change is in good agreement with the theoretical predictions for anisotropies in the radiation patterns up to 30%.

As the spins and multipole orders involved in the nickel 60 cascade are well established, this agreement confirms the theoretical predictions for oriented nuclei. A free-nucleus theory is suitable for the comparison when the correlation is studied as a function of anisotropy. It is not possible to distinguish between the possible modes of beta decay by such comparison, however. Small differences in detector efficiency for the gamma rays are not important in this comparison. The continued approximate agreement at low temperatures and with oriented nuclei supports the conclusion that the intermediate-state lifetime is less than $10^{-11}$ seconds.

The assumption of an equivalent $\beta$ associated with a Boltzmann population distribution appears to give a reasonably good comparison between theory and experiment, expressed as correlation versus anisotropy. This result is probably valid only for small values of $\beta$, such that
terms in \( F_6 \) and \( f_8 \) have negligible effect on the correlation.

Hence it is feasible to apply the method of coincidence measurements to more complicated decay schemes than that of cobalt 60 by use of the apparatus and techniques which have been described. In such cases the observation of directional correlation is expected to be more powerful in yielding new information as well as a check on the results of directional distribution measurements.
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AND RADIATIONS FROM ORIENTED NUCLEI
(IN Chronological Order)

   "Nuclear Paramagnetism."

   "Angular Distribution of Radiation Products."

   "A Suggestion for Aligning Certain Nuclei."

   "On the Production of Nuclear Polarization."

5. C. J. Gorter, D. de Klerk, O. J. Poppema, M. J.
   "Attempts to Align Nuclei."

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