Angular Correlation of Gamma Radiations from Oriented Nuclei*

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The angular correlation between the cascaded gamma rays following the decay of Co^{60} nuclei oriented at low temperatures has been determined. Comparison with the directional anisotropy of the individual gamma rays affords a test of the theory of Cox and Tolhoek largely independent of hyperfine interaction details and temperature determination. The theoretical predictions are confirmed.

INTRODUCTION

IN 1952 Cox and Tolhoek[1] published results of calculations predicting the manner in which the angular correlation between cascaded gamma rays should depend on spatial orientation of the parent nucleus. These first results were restricted to the special but physically interesting cases of two dipole or alternatively two quadrupole photon emissions, with maximum nuclear spin change for each transition; subsequent work[2–4] has expanded and generalized the treatment to cover other combinations of multipole emission and to account for the effects of preceding undetected radiation.[5] Steenberg[6] has developed expressions for the expected population distributions for some of the practicable orientation methods. The result, in the case of Gorter-Rose orientation used in the present experiments, is substantially more complicated than the Boltzmann distribution. Since it appears doubtful whether enough is known about the structure of the crystal employed to justify use of the more refined expressions for the population distribution, we utilize the Boltzmann distribution for making numerical estimates in the present paper.

Lemmer and Grace[7] have recently measured the angular correlation of the gamma rays in Ni^{60}, in a mixed Tutton salt, at 20°K and at 288°K. The correlation at 20°K was found to be identical with that observed at room temperature, establishing the absence at this temperature of any significant perturbation of the intermediate state in the gamma cascade, and giving confidence that none would be encountered at lower temperatures.

Currently available techniques for producing nuclear orientation through hyperfine interaction[8–10] in a paramagnetic salt cooled by adiabatic demagnetization yield appreciable degrees of polarization or alignment,[11,12] but introduce a number of obstacles to the measurement and interpretation of angular correlation distributions. The general purpose of the present experiment was to determine, first whether such measurements are feasible; and if so whether the results confirm the theoretical predictions; and second, whether the results will yield new information concerning either nuclear or crystalline structure.

We have determined the angular correlation between the cascaded gamma rays in Ni^{60} following beta decay of Co^{60} nuclei polarized at low temperatures in cerium magnesium nitrate. As will be seen, the results are in good agreement with the predicted distributions, and confirm the theory upon which they are based. Calculations and experimental procedures, together with the results, are given in the present paper. A discussion of the sensitivity of the results to the details of the hyperfine interaction (at best, imperfectly known for the salt in question), will be given in a forthcoming article.

I. THEORY

Angular Correlation and Distribution

For a gamma-ray cascade following decay of nuclei partially oriented with respect to some axis in space, the angular correlation function depends not only on the angle θ between the emission directions of the two photons, but also on the angles θ_1,θ_2 of each with respect to the orientation axis. For the case of present interest, two quadrupole radiations, each with maximum nuclear spin change, the expression for the angular correlation (apart from a normalizing factor) is[4]

$$W(\theta_1,\theta_2;\theta)=\sum_{k=0}^{4} N_{2k}f_{2k}G_{2k}(\theta_1,\theta_2;\theta). \qquad (1)$$

The coefficients N_{2k} depend on the spin I_i of the initial state in the gamma-ray cascade. In the present notation

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[1] J. A. M. Cox and H. A. Tolhoek, Physica **18**, 359 (1952).
[2] S. R. deGroot, Physica **18**, 1201 (1952).
[3] J. A. M. Cox, Physica **18**, 1262 (1952).
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[5] J. A. M. Cox and H. A. Tolhoek, Physica **19**, 673 (1953).
[6] N. R. Steenberg, Proc. Phys. Soc. (London) **A66**, 399 (1953).
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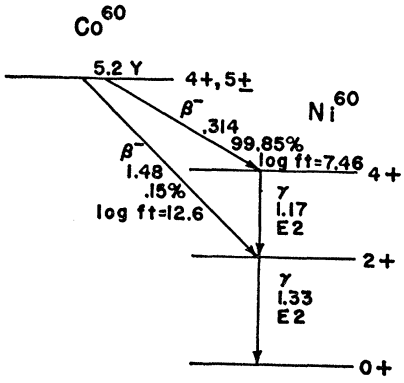

FIG. 1. Relevant part of the cobalt-60 decay scheme.

the general form given by Cox and Tolhoek[4,13] becomes

$$N_{2k}(I)=2^k I^{2k}(2I-2k)!/(2I)!, \quad k=0,1,2,\cdots.$$

The functions f_{2k} describe the state of orientation of the initial state; they depend upon the even moments of the population distribution over the nuclear magnetic sublevels, suitably weighted by expressions containing the nuclear spin. A general formula for f_k is given in reference 13 (p. 107), as well as explicit expressions for $f_1\cdots f_4$ in terms of the population moments. The functions f_6 and f_8 appear in reference 4 (page 1184). The products $N_{2k}f_{2k}$ characterize the orientation of the initial state for subsequent gamma emission. The factors G_{2k} contain the dependence of the angular correlation on the three angles which describe the direction of the two photons with respect to each other and to the axis of orientation. The explicit expressions are given in reference 4 (p. 1183), in terms of cosines of the various angles.

For random orientation, the parameters f_k all vanish except f_0; Eq. (1) then reduces to the well-known correlation

$$W(\theta)=(\text{constant})G_0(\theta)=1+\tfrac{1}{8}\cos^2\theta+(1/24)\cos^4\theta. \quad (2)$$

Nuclear orientation will cause the radiation pattern of each gamma ray to be anisotropic.[5,13–15] For quadrupole radiation with a nuclear spin decrease of two units the angular distribution is[13,16]

$$F(\theta)=1-(15/7)N_2f_2P_2(\cos\theta)-5N_4f_4P_4(\cos\theta), \quad (3)$$

where the parameters N and f have the same significance as before, and P_2,P_4 are Legendre polynomials. The distribution (3) has been well verified by experiments[11,12,17,18] on oriented Co^{60}.

The results (1) and (3) for the angular correlation and distribution are derived on the assumption that the oriented system has axial symmetry, and that the pertinent angles are measured with respect to the axis. If there are two orientation mechanisms with non-parallel axes then the resulting patterns must in general be expressed as the sum of two distributions of the form (1) or (3), suitably weighted and projected along the directions of measurement.

The most satisfactory test of the predicted effect of orientation on angular correlation would be a comparison between measured coincidence rates and values of $W(\theta_1,\theta_2;\theta)$ computed for various known temperatures and applied magnetic fields, which in principal determine the orientation parameters f_k. Unfortunately the details of the hyperfine interaction between the cobalt nucleus and its environment are insufficiently well known to allow this approach. A somewhat less direct but still decisive test is available in a comparison between angular correlation and angular distribution of the individual gamma rays. Two approaches are possible here: (a) to find the orientation parameters f_2 and f_4 empirically from the radiation patterns, inserting the values in the expression for the angular correlation; and (b) to assume some physically reasonable population distribution from which both correlation and angular distribution can be computed. Method (a) has the advantage of by-passing the (unknown) hyperfine interaction, except for the assumption of axial symmetry. It is subject to possible error in neglecting the contribution of f_6 and f_8 to the angular correlation; however, at the moderate orientations observed these are expected to be small. Method (b) entails no approximations in the calculation of either the angular distribution or correlation, but in order to carry any conviction requires a careful study of how sensitive the computed relations are to the specific population distribution assumed. Here we have chosen the second approach, assuming for the parent nucleus a Boltzmann population distribution with equally spaced magnetic levels:

$$a_m=Ce^{-\beta m}. \quad (4)$$

This distribution would indeed hold for a system of free nuclei in a uniform magnetic field, with

$$\beta=g\mu_N B/kT, \quad (5)$$

in the usual notation. A major convenience of the exponential distribution is that the degree of orientation is determined by a single parameter; although in general we will not expect that it correctly describes the population distribution over the nuclear magnetic sub-levels in Gorter-Rose orientation,[15] we may still look upon it as an approximation, and a useful tool for examining orientation effects.[19]

Since it is not experimentally feasible to determine

[13] H. A. Tolhoek and J. A. M. Cox, Physica **19**, 101 (1953).
[14] J. A. Spiers, Nature **161**, 807 (1948).
[15] N. R. Steenberg, Proc. Phys. Soc. (London) **A65**, 791 (1952).
[16] H. A. Tolhoek and J. A. M. Cox, Physica **18**, 357 (1952).
[17] M. A. Grace and H. Halban, Physica **18**, 1227 (1952).
[18] Poppema, Beun, Steenland, and Gorter, Physica **18**, 1235 (1952).

[19] Under certain conditions both the Bleaney and Gorter-Rose methods result in a Boltzmann population distribution over the nuclear magnetic sublevels. For Bleaney alignment the requirement is $A\gg B$ in the hyperfine part of the Hamiltonian $H=AI_zS_z+B(I_xS_x+I_yS_y)$; for the Gorter-Rose method, the applied magnetic field must be strong enough so that the spacing of the ionic levels is large compared with that of the nuclear levels.

the angular correlation in detail at a number of different angles, we determine the coincidence rates at 90° and 180°, and use the ratio $W(\theta_1,\theta_2;180)/W(\theta_1,\theta_2;90)$, for which the effect in the present case is expected to be greatest. This ratio is plotted against the anisotropy in the individual gamma-ray distribution defined by

$$\epsilon=1-[F(0)/F(90)]. \quad (6)$$

The angular correlation axis defined by the two counters placed on opposite sides of the source may be fixed at any desired angle θ_1 with respect to the orientation axis. The choice is of some interest: For $\theta_1=0$ (referred to subsequently as "Geometry I") the predicted Ni^{60} correlation decreases with increasing orientation, and appears to be comparatively sensitive to the detailed orientation mechanism. For $\theta_1=90°$ (Geometry II), the expected correlation increases rapidly, and at moderate degrees of orientation is quite insensitive to the type of population distribution assumed. The expressions for the angular correlation function reduce in the two cases to:

Geometry I: $(\theta_1=0)$

$$W(0,90;90)=0.1525-0.208N_2f_2 -1.47N_4f_4-0.10N_6f_6+1.5N_8f_8, \quad (7)$$

$$W(0,180;180)=0.1780-0.554N_2f_2 -0.532N_4f_4+3.7N_6f_6+4.0N_8f_8. \quad (8)$$

Geometry II: $(\theta_1=90°)$

$$W(90,180;90)=W(0,90;90), \quad (9)$$

$$W(90,270;180)=0.1780+0.277N_2f_2 -0.200N_4f_4-1.2N_6f_6+1.18N_8f_8. \quad (10)$$

The angular distributions are always measured with respect to the orientation direction:

$$F(0)=1-2.14N_2f_2-5.00N_4f_4, \quad (11)$$

$$F(90)=1+1.07N_2f_2-1.88N_4f_4. \quad (12)$$

Effect of Preceding Radiation

Usually the first gamma-emitting state is not the radioactive parent, but is reached by a transition such as beta decay in which the emitted quantum goes undetected. Co^{60} is an example; the relevant part of the decay scheme is shown in Fig. 1. Since it is the comparatively long-lived parent nucleus that is subject to orientation, one must know what effect the beta transition has on the Nf products appearing in (1). This problem also has been treated by Cox and Tolhoek,[5] who give relations connecting the orientation parameters $f_k(I_0)$, $f_k(I_i)$ belonging to the parent and initial daughter state respectively, both for beta and for gamma transitions.[20] For the case of Co^{60}, numerical values of quantities proportional to N_2f_2 and N_4f_4 computed on the assumption that the parent nuclear substates are exponentially populated, are tabulated as a function of the parameter β by Cox, deGroot, and Hartogh.[21] Despite the comparatively high value of log ft (7.4), the allowed shape of the beta spectrum and absence of beta-gamma angular correlation[22–24] indicate that the beta decay of the Co^{60} ground state to the Ni^{60} second excited state is probably allowed. This, together with the low intensity of the beta transition to the first excited level[25] in Ni^{60}, restricts the Co^{60} ground-state spin to the values 4 or 5.

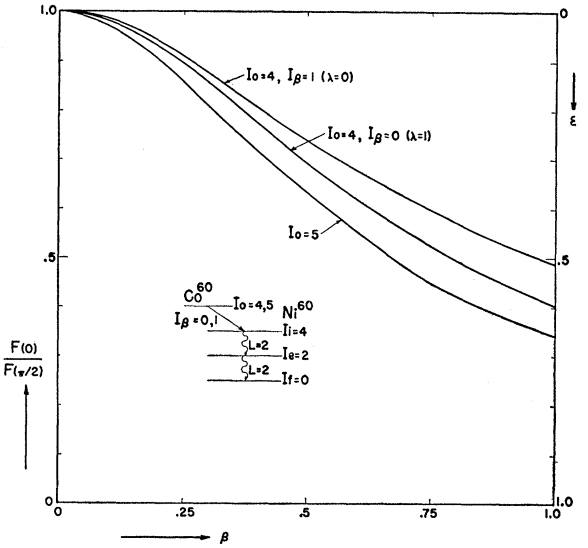

FIG. 2. Theoretical anisotropy of the gamma radiation pattern for quadrupole transitions with $\Delta I=2$. The ratio of emission probabilities parallel and perpendicular to the orientation axis are plotted upward at the left and the anisotropy $\epsilon=1-[F(0)/F(90)]$ is plotted downward on the right, both as functions of β. The curves correspond to three possible choices of Co^{60} spin and mode of allowed beta decay.

Curves of anisotropy *versus* β are shown in Fig. 2 for three choices of the Co^{60} spin and type of allowed beta decay: $I_0=5$, (Gamow-Teller only); $I_0=4$, (Fermi); and $I_0=4$, (Gamow-Teller). For the choice $I_0=4$, a mixture of Fermi and Gamow-Teller contributions produces a curve that lies between those for pure transitions. A similar plot, for angular correlation as a function of β is shown in Fig. 3. In principle a measurement of either anisotropy or correlation, combined with knowledge of the temperature and hyperfine interaction (which determine β) would permit determination of the mode of beta decay; but unfortunately in the present case neither the temperature nor (especially) the hyperfine structure are sufficiently well known

[20] A result of particular significance for the present work is that for any transition involving maximum change in nuclear spin consistent with the degree of forbiddenness, each N_kf_k product remains unaltered. As Cox and Tolhoek point out, the result in the case of Ni^{60} is that both gamma rays have the same directional distribution.

[21] Cox, deGroot, and Hartogh, Physica **19**, 1123 (1953).
[22] Deutsch, Elliott, and Roberts, Phys. Rev. **68**, 193 (1945).
[23] C. Y. Fan, Phys. Rev. **87**, 252 (1952).
[24] R. L. Garwin, Phys. Rev. **76**, 1876 (1949).
[25] G. L. Keister and F. H. Schmidt, Phys. Rev. **93**, 140 (1954). Their work strongly favors spin 4 for the Co^{60} ground state but does not appear to exclude 5 completely.

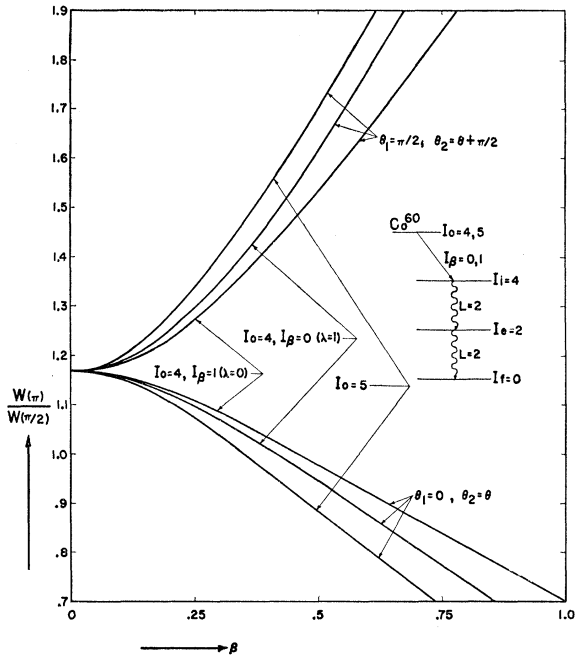

FIG. 3. Predicted dependence of angular correlation for the gamma-gamma cascade 4(2)2(2)0, on the parameter β. The upper curves correspond to geometry II, the lower curves to geometry I.

to distinguish among the three cases.[26] Although the unknown parameter β may be eliminated by plotting correlation against anisotropy, the resulting curves for the three modes of beta decay, throughout the range of feasible orientation, precisely coincide.

Observed Quantities and Corrections

We consider in this section the relation between the measured counting rates and the theoretical distributions. The coincidence counting rate, exclusive of accidental background, between counter 1 at θ_1 and counter 2 at $\theta_2=\theta_1+\theta$ is approximately

$$C_{12}=2\nu W(\theta_1,\theta_2;\theta)[\eta_1\Omega_1\eta_2\Omega_2], \tag{13}$$

where ν is the source decay rate via the gamma-ray cascade, η the counter efficiency, and Ω the solid angle, expressed as the fraction of a sphere, which the counter subtends at the source. Equation (13) holds provided (a) the efficiency-solid angle product $\eta\Omega$ for each counter is the same for both gamma rays; and (b) the correlation function $W(\theta_1,\theta_2;\theta)$ is symmetric in θ_1, θ_2. Condition (a) holds very well for photons as close in energy as those in Ni^{60}; and (b) is satisfied wherever, as in Ni^{60}, the two photon transitions have the same multipolarity and entail maximum nuclear spin change.

Provided the cascade is the only significant part of the decay scheme in which either gamma ray occurs (no important competition leading either to or from the intermediate level), the corresponding singles rates also contain the factor ν:

$$C_1=2\nu F(\theta_1)\eta_1\Omega_1, \quad C_2=2\nu F(\theta_2)\eta_2\Omega_2. \tag{14}$$

For random orientation ($\beta=0$), $F(\theta)=1$:

$$C_1^0=2\nu\eta_1\Omega_1, \quad C_2^0=2\nu\eta_2\Omega_2. \tag{15}$$

The following ratios eliminate the solid-angle efficiency factors of the individual counters, and at the same time yield the desired anisotropy and correlation function: In Geom. I, with counter 2 at 90° and counter 3 at 180° with respect to both counter 1 and the polarization direction,

$$\frac{C_3/C_3^0}{C_2/C_2^0}=\frac{F(180)}{F(90)}=1-\epsilon, \tag{16}$$

$$\frac{C_{13}/C_3^0}{C_{12}/C_2^0}=\frac{W(180)}{W(90)}=\kappa. \tag{17}$$

Account must be taken of the effect of counter solid angle and source extension, in relating the distributions measured with counters of finite size to the inferred rates at the counter centers. If reasonably small, both effects may be combined and expressed in terms of a set of attenuation coefficients J_i in the Legendre polynomial expansions for the angular distribution and correlation[27–30]:

$$F(\theta)=\Sigma_0{}^2A_{2k}J_{2k}P_{2k}(\cos\theta), \tag{18}$$

$$W(\theta_1,\theta_2;\theta)=\Sigma_0{}^2B_{2k}J_{2k}{}^2P_{2k}(\cos\theta). \tag{19}$$

The functions F and W are the measured quantities that appear in Eqs. (16) and (17), reducing to the theoretical forms (1) and (3) only for point sources and infinitesimal counters ($J_{2k}\to1$). For our geometry we estimate: $J_2=0.943$, $J_4=0.80$; using these values, the theoretical distributions (7)–(12), corrected for counter solid angle and source extension, become

$$W(0,90;90)=0.1533-0.226N_2f_2 -1.34N_4f_4-0.26N_6f_6+0.97N_8f_8, \tag{20}$$

$$W(90,180;90)W=(0,90;90),$$

$$W(0,180;180)=0.1755-0.544N_2f_2 -0.421N_4f_4+2.7N_6f_6+2.7N_8f_8, \tag{21}$$

$$W(90,270;180)=0.1755+0.267N_2f_2 -0.158N_4f_4-0.88N_6f_6+0.71N_8f_8, \tag{22}$$

$$F(0)=1-2.02N_2f_2-4.00N_4f_4, \tag{23}$$

$$F(90)=1+1.01N_2f_2-1.50N_4f_4. \tag{24}$$

The net effects on the ratios $W(180)/W(90)$ and $F(180)/F(90)$ are 1 to 2% and less than 1%, respectively.

[26] The case of Co^{58}, in which the disparity among the possibilities is much greater, has been studied by J. M. Daniels *et al.* [Phil. Mag. **43**, 1297 (1952)] who find an anisotropy corresponding to $\Delta I=0$ (G.T.).

[27] S. Frankel, Phys. Rev. **83**, 673 (1951).
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[29] Walter, Huber, and Zunti, Helv. Phys. Acta **23**, 697 (1950).
[30] A. M. Feingold and S. Frankel, Phys. Rev. **97**, 1025 (1955).

II. EXPERIMENTAL METHODS

Cobalt ions including a few microcuries of Co^{60} were introduced as divalent impurities in cerium magnesium nitrate, a procedure used by Ambler *et al.*[12] in the first successful attempt at Gorter-Rose orientation. Relevant thermal and magnetic properties of this salt at low temperatures have been studied by Cooke, Duffus, and Wolfe[31] and by Daniels and Robinson,[32] who have carried out a detailed calibration of cerium magnesium nitrate to determine the relation between the Kelvin and magnetic temperatures near the Curie point. This salt offers two important advantages for orientation work: it has a sufficiently low Curie temperature (~0.003°K) to permit appreciable orientation at moderate values of applied field, and, in addition, the electronic *g*-factor of the cerium is highly anisotropic, so that the orienting field, directed along the axis of minimum susceptibility, may be applied without significantly raising the crystal temperature.

The sample mounting is shown in Fig. 4. Eight single crystals with a total mass of about 2.5 grams, containing in all between 10 and 20 microcuries of Co^{60} were glued with Cutex lacquer to a mica sheet. The crystals were all mounted in the same spatial orientation, with axes parallel. The mica sheet was supported by a thin-walled Pyrex tube, which in turn was fastened to a guard salt composed of ten grams of iron ammonium alum. The entire assembly was attached with Araldite 101 to a glass pedestal of thin-walled tubing. The sample was brought to about 1°K in a field of 24 kilogauss, and adiabatically demagnetized to about 0.003°K.

The magnetic temperature of the specimen was determined from susceptibility measurements at 150 cps, carried out with a Hartshorn mutual inductance bridge.[33] Some contribution from the guard salt was detected, but was significant only in the calibration range between 4° and 1°, and, with sample absent, could be measured with sufficient accuracy to correct the calibration. The susceptibility at low temperatures could be obtained by using the calibration curve determined in zero field, then correcting analytically for the effect of the applied magnetic field.[34] In the present work the anisotropy in the radiation pattern of the individual gamma rays forms the most useful index of the state of the nuclear spin system, the temperature playing an essentially corroborative role.

The gamma rays are detected by means of scintillation counters, shown in place around the cryostat in Fig. 5. Each counter employs a 1-inch by 1-inch thallium activated sodium iodide crystal coupled by a 7-inch lightpipe of cast Lucite to a Type 6199 multiplier

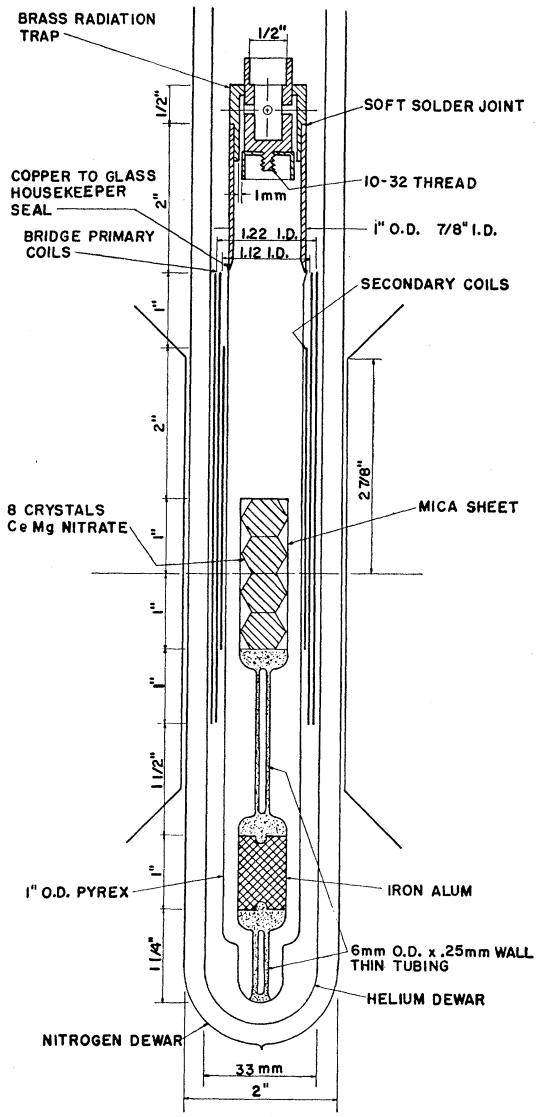

FIG. 4. Detail of the sample mounting.

phototube. The latter is protected from the orienting magnetic field by a mu-metal shield surrounded by an outer shield of $\frac{1}{4}$-inch soft ion. The orienting field is produced by two coaxial opposed pairs of Helmholtz coils (Fig. 5) with exciting currents chosen so that the dipole moments of the two pairs just cancel. This arrangement caused the fringing field to fall off rapidly enough to have no detectable effect on the photomultiplier tubes, even along the field axis, where the cylindrical magnetic shield is least effective. In the sketch three counters are shown mounted at 0°, 90° and 180° with respect to the polarizing field—the arrangement designated "Geometry I." Counters and Helmholtz coils are fastened to separate ring-shaped platforms which may be independently rotated about

[31] Cooke, Duffus, and Wolfe, Phil. Mag. 44, 623 (1953).
[32] J. M. Daniels and F. N. H. Robinson, Phil. Mag. 44, 630 (1953).
[33] D. DeKlerk and R. P. Hudson, J. Research Natl. Bur. Standards 53, 173 (1954).
[34] E. Ambler (private communication).

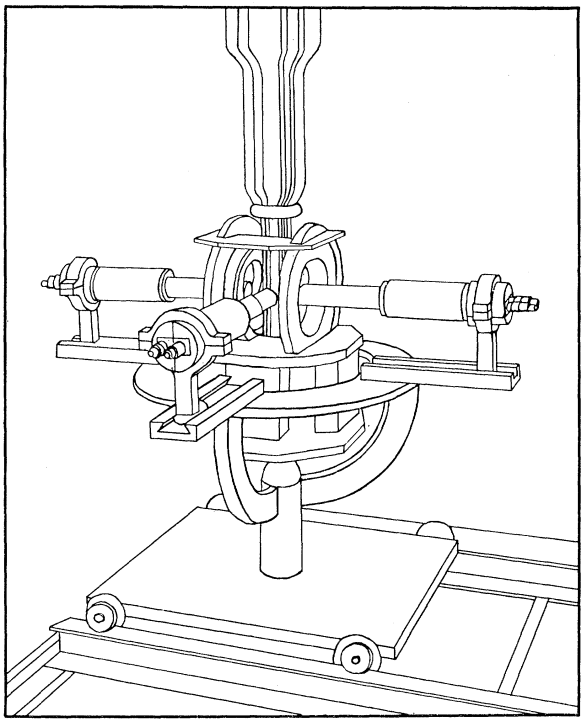

FIG. 5. Scintillation counters and Helmholtz coils in position around the cryostat.

the vertical axis; the combined assembly is mounted on a table which rolls on accurately placed rails with a preset stop at the cryostat. After demagnetization, the 24-kilogauss magnet (not shown) is rolled away, the counter table moved in place, the counters and Helmholtz coils rotated to the correct positions and the orienting field applied; this operation can be completed consistently within thirty-five seconds.

A block diagram of the circuitry for two counter channels is shown in Fig. 6. The linear amplifier and discriminator[35] in each channel select the photoelectric absorption lines of the appropriate gamma rays in the scintillation counter. Quanta scattered from the paramagnetic crystal or cryostat, being degraded in energy, are rejected, so that only unscattered photons are counted; this is of particular importance here, since the angular correlation is especially sensitive to scattering in the vicinity of the source.[27,29,36] Counter-to-counter scattering, which would otherwise result in spurious coincidence counts, is also eliminated. The fast amplifiers are designed to provide amplitude limited pulses of short rise time at the fast coincidence circuit. The coincidence resolving time of the system, including counters, is approximately 20 millimicroseconds. At typical counting rates—one to five coincidences per second and 1500 to 4000 single counts per second—the correction for accidental coincidence background amounts to between 4 and 6%, and in the ratio of 180° to 90° coincidence rates to less than 2%.

The warm-up time, from demagnetization to the point ($\sim$0.1°K) of vanishing radiation anisotropy, varied from ten to twenty minutes. Single counting rates were recorded at fifteen-second intervals, (with five seconds off for reading the scalers) and coincidences every twenty seconds. Several counting runs were required to accumulate a sufficient number of coincidence counts in any given interval of the warm-up period to be statistically significant. The coincidence rates in different runs were then correlated according to corresponding ranges of anisotropy (rather than temperature). Ten demagnetizations yielded approximately 3% precision in an anisotropy interval of 0.05.

In order to eliminate efficiency and solid-angle parameters of the individual counters, both the coincidence and single counting rates must be normalized by dividing by the single rates in the absence of orientation (see Eqs. (16), (17)). These rates were determined at the end of each warm-up period over three successive 100-second intervals. The normalized rates

$$F_i=C_i/C_i^0, \quad W_i=C_{1i}/C_i^0, \tag{25}$$

and the corresponding anisotropy and correlation values

$$\epsilon=1-(F_3/F_2), \quad \kappa=W_3/W_2, \text{ Geom. I}, \tag{26}$$

are quantities from which, as far as is possible, dependence upon instrumental factors peculiar to a given run have been eliminated, and the values of which from

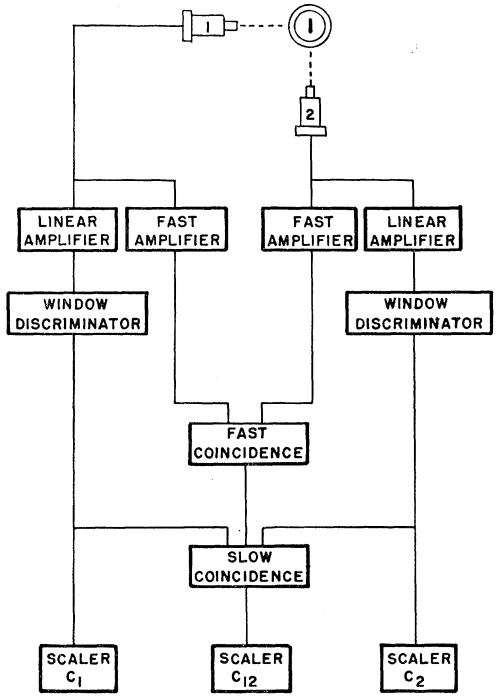

FIG. 6. Block diagram of two single counting channels and associated coincidence channel.

[35] P. S. Jastram (to be published).
[36] H. Frauenfelder, Ann. Revs. Nuclear Sci. 2, 149 (1953).

different runs may be compared and combined. This treatment offers no protection against drift in counter sensitivity during a given warm-up period. One may not normalize the coincidence rates by the low-temperature singles rates, for the crux of the experiment is to determine the relation between correlation and anisotropy. The best checks available against drift are intercomparison of the final three long singles counts at the end of each run, and requiring a reasonably consistent relation between magnetic temperature and single counting rate in each channel.

III. RESULTS

The CO^{60} angular correlation is shown expressed as a function of the anisotropy, for an applied orienting magnetic field of 200 gauss, in Fig. 7. The experimental points are compared with the relations (solid lines) that hold for an exponential population distribution of parent-nucleus magnetic sublevels, which were obtained using Eqs. (20) to (24). In Geom. I (lower curve) the angular correlation decreases with increasing nuclear polarization; $W(90)$ and $W(180)$ both decrease, but the latter more rapidly at low degrees of polarization. In Geom. II ($\theta_1=90°$), $W(90)$ is the same as in Geom. I, but $W(180)$ increases, with the result that the angular correlation rises rapidly with anisotropy, so that even with the moderate degree of orientation obtained the correlation increases from the 17% "room temperature" value to over 50%. Figure 8 shows similar results for Geom. II only, corresponding to an orienting field of 330 gauss. Except for the two high points in Fig. 8, the experimental results are in excellent agreement with the predicted values, and confirm the theory of angular correlation for oriented nuclei. A number of comments are still needed to support the generality of this conclusion.

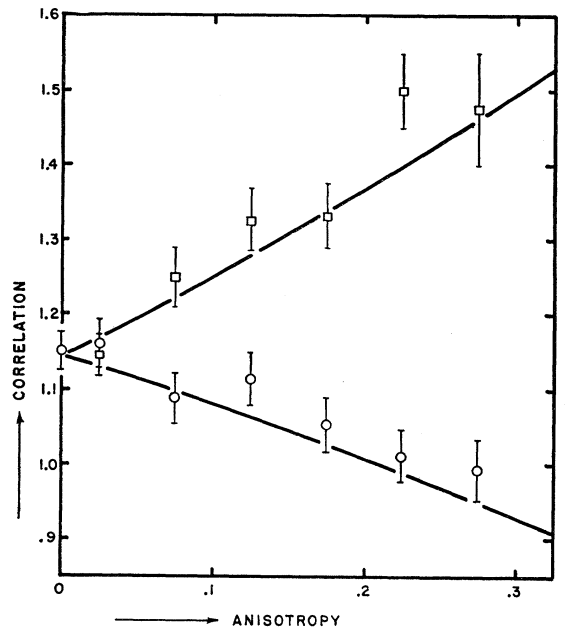

FIG. 7. Measured correlation as a function of anisotropy at corresponding temperatures in a polarizing field of 200 gauss. The upper points are for geometry II and the lower ones for geometry I. The curves for a Boltzmann distribution over equally spaced nuclear magnetic levels, corrected for finite geometry of source and detectors, are plotted from Figs. 2 and 3 at corresponding values of β.

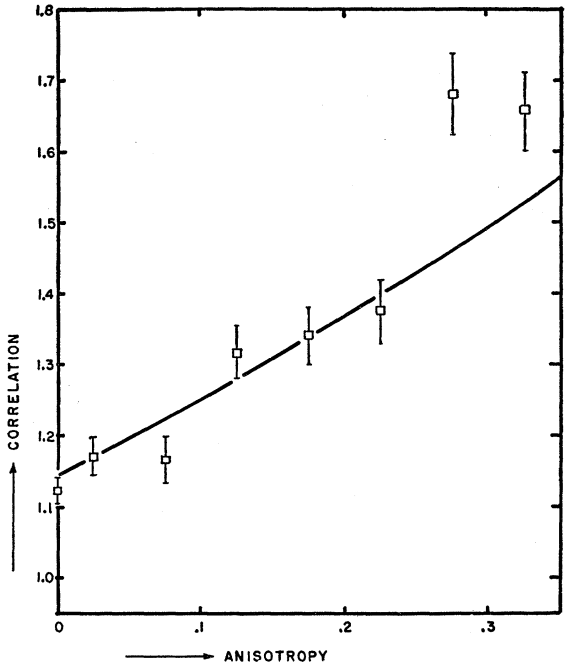

FIG. 8. Measured correlation *versus* anisotropy in a 330 gauss polarizing field for geometry II. The theoretical curve is identical with that given in Fig. 7.

IV. DISCUSSION

1. *Spin of* Co^{60} *and mode of beta decay.*—For the assumed exponential distribution over parent substates, all the possible choices of Co^{60} spin and allowed beta decay ($I_0=5$, Gamow-Teller; $I_0=4$, Fermi; $I_0=4$, Gamow-Teller) yield curves of correlation *vs* anisotropy that are identical within the thickness of the lines on the graph. That a more pronounced divergence may be expected at higher degrees of orientation is probably largely of academic interest: the corresponding curves of anisotropy and correlation *versus* orientation (Figs. 2 and 3) diverge much more rapidly, indicating that hope for determining the parent spin and beta-decay mode of Co^{60} and similar cases by orientation methods lies mainly in combining angular distribution measurements with improved knowledge of the nuclear environment.

2. *Hyperfine interaction.*—Some calculations based on more refined estimates of the parent ground-state population distributions in Gorter-Rose orientation[15] than the simple exponential distribution lead to the result that within the experimental range of orientation

the correlation-anisotropy relation in Geom. II is largely insensitive to specific assumptions about the hyperfine interaction, over a considerable range of reasonable choices. Accordingly, within the experimental statistics, comparison between theory and experiment in Geom. II suffers from no ambiguity resulting from the present lack of information on the hyperfine spectrum of the cobalt ion in cerium magnesium nitrate. In Geom. I, on the other hand, the theoretical curve is comparatively sensitive to the type of ionic structure assumed, and it is in fact somewhat puzzling that the exponential population distribution appears to give as good a fit with experiment as it does.

3. *Perturbation of the intermediate state.*—If the intermediate state in a cascade of two successive radiations has a lifetime comparable with the Larmor precession time of the nucleus in the field of the atomic shell, the angular correlation may be disturbed, usually in the direction of decreased anisotropy,[37,38] In terms of the hyperfine splitting $\Delta\nu$, the critical time is

$$t_c=(2\pi c\Delta\nu)^{-1}. \tag{27}$$

Cobalt ions (in other salts) are found to have hyperfine level separations of about 0.01 cm^{-1}, which, assuming a g-factor of the order of unity for the first excited state in Ni^{60}, leads to an estimated critical time of approximately 10^{-9} second. On this basis no detectable attenuation of the angular correlation is to be expected, for both room-temperature angular correlation measurements with a metallic source in a strong magnetic field[39] and direct lifetime measurements[40] place an upper limit of 10^{-11} second on the mean life of the first excited state in Ni^{60}. Moreover, in Geom. I one of the coincident gamma rays is always directed along the axis of orientation; the component of a perturbing magnetic field in this direction can cause no attenuation of the correlation.[41] The most pronounced effect is to be expected in Geom. II, where the 180° coincidences occur in the plane normal to the axis of the applied field[41]; however, it turns out that in the present case the attenuation is not expected to increase appreciably with orientation[42]; the curves corresponding to maximum attenuation cannot be distinguished, within our statistical precision, from the unperturbed correlations plotted in Figs. 7 and 8.

The results of the coincidence measurements of Bay *et al.* include an upper limit to the mean life of the 2.50-Mev level in Ni^{60}—also 10^{-11} second—ensuring negligible perturbation of that state by extranuclear fields.

A question on which the present experiments throw some light but cannot presume to settle is whether angular correlation measurements can be expected to yield information beyond that given by the technically simpler observation of angular distributions of individual gamma rays. That they should in principle give additional information about the radioactive parent and initial mode of decay follows from the more detailed dependence on the initial population distribution: in the case of two successive quadrupole radiations the angular correlation depends on the sixth and eighth moments (through f_6 and f_8), while the angular distributions contain nothing higher than the fourth moment. The contributions of the higher moments are too small, however, at moderate orientation, to be of any use. It is not by any means clear whether the insensitivity of the correlation—anisotropy relation to the mode of prior decay will persist for decay schemes that differ materially from that of Co^{60}; calculations of the dependence of correlation on orientation are needed for other commonly occurring special cases, notably the gamma cascade between successive states with spins 2,2,0.

As in certain cases the angular correlation may be substantially increased by practicable degrees of orientation, we may reasonably expect that details of the correlation where one of the gamma transitions comprises a mixture of two different multipoles will also be usefully enhanced.

Angular correlation measurements on oriented nuclei are found to be feasible in favorable cases, and the predicted dependence on polarization is confirmed. At present it remains an open question whether correlation measurements will in practice be of sufficient advantage over angular distribution observations to be worth the additional technical complexity.

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