

## III. DISCUSSION

$\text{Ni}^{66}$  is an even-even nucleus and therefore a spin and parity of  $0^+$  can be assigned to its ground state. Using Moskowsky's nomograph<sup>9</sup> for  $ft$  values, the  $\log(ft)$  for the 0.20-Mev beta decay is 3.96, indicating an allowed transition. That this transition is allowed is also shown by the shape of the Kurie plot. As the Gamow-Teller selection rules forbid  $0 \rightarrow 0$  transitions and since the spectra indicate only a single beta and no gammas in the  $\text{Ni}^{66}$  decay, the  $\text{Cu}^{66}$  ground level must be  $1^+$ . This confirms its earlier assignment as well as the  $2^+$  assignment to the 1.03-Mev level in  $\text{Zn}^{66}$ .<sup>5,6</sup> The complete decay scheme for the isobaric triplet  $\text{Ni}^{66}$ - $\text{Cu}^{66}$ - $\text{Zn}^{66}$  is given in Fig. 3.

<sup>9</sup> S. A. Moskowsky, Phys. Rev. **82**, 35 (1951).

From the experimental mass decrements listed by Wapstra,<sup>10</sup> masses of  $65.946940 \pm 0.000200$  and  $65.949760 \pm 0.000200$  are found for  $\text{Zn}^{66}$  and  $\text{Cu}^{66}$ , respectively. If one uses an energy of  $0.20 \pm 0.03$  Mev for the difference between the ground states of  $\text{Ni}^{66}$  and  $\text{Cu}^{66}$ , a mass of  $65.949975 \pm 0.000203$  is calculated for  $\text{Ni}^{66}$ .

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<sup>10</sup> A. H. Wapstra, Physica **21**, 404 (1955).

## Gamma-Gamma Angular Correlation in the Decay of Chlorine-34†

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The directional angular correlation between the 1.16- and 2.10-Mev gamma rays which follow the decay of 32.4-minute  $\text{Cl}^{34}$  has been measured. Samples of  $\text{Cl}^{34}$  were produced in the cyclotron of the University of California, Los Angeles by the  $(p, pn)$  reaction and were produced in both solid and liquid form. Sodium iodide scintillation counters with subsequent pulse-height discrimination and standard coincidence techniques were used and the method was tested on the cascade of  $\text{Ni}^{60}$  giving satisfactory agreement with previous results. The results on  $\text{S}^{34}$ , although not unambiguous, are consistent with the assignment of spin 2 and even parity to both the first and second excited states of  $\text{S}^{34}$  with the 1.16-Mev gamma ray being 1.7% electric quadrupole and 98.3% magnetic dipole radiation.

## INTRODUCTION

CHLORINE-34 is a complex positron and gamma-ray emitter which has been the subject of numerous investigations.<sup>1-7</sup>

Figure 1 shows the probable decay scheme of  $\text{Cl}^{34}$ . The gamma-ray energies are in Mev and are due to Ruby and Richardson<sup>1</sup> and Ticho.<sup>2</sup> Arber and Stahelin<sup>4,6</sup> measured the lifetime of the ground state of  $\text{Cl}^{34}$  and calculated  $\alpha$ , the internal conversion coefficient of the 0.145-Mev gamma ray, from the results of Ruby and Richardson. The values of the half-life of the isomeric state of  $\text{Cl}^{34}$  and of the maximum energies of the positron groups were reported by Green and Richardson.<sup>7</sup>

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<sup>1</sup> L. Ruby and J. R. Richardson, Phys. Rev. **83**, 698 (1951). References to earlier works are given in this paper.

<sup>2</sup> H. K. Ticho, Phys. Rev. **84**, 847 (1951).

<sup>3</sup> David Green, thesis, University of California, Los Angeles (unpublished).

<sup>4</sup> W. Arber and P. Stahelin, Helv. Phys. Acta **26**, 433 (1953).

<sup>5</sup> W. Arber and P. Stahelin, Helv. Phys. Acta **26**, 584 (1953).

<sup>6</sup> P. Stahelin, Helv. Phys. Acta **26**, 691 (1953).

<sup>7</sup> D. Green and J. R. Richardson, Phys. Rev. **96**, 858 (1954).

The gamma-ray intensities were calculated from those of the positron groups and the relative intensities of the 3.22- and 1.16-Mev gamma rays measured by Green<sup>8</sup>; the intensity of the 0.145-Mev radiation includes the percentage which is internally converted.

Arguments based upon all the experimental evidence and upon the fact that  $\text{S}^{34}$  is an even-even nucleus lead to the choice of spin 2 and even (+) parity for both its excited states as most probable. This investigation of the directional angular correlation between the 1.16- and 2.10-Mev gamma rays was performed to confirm this spin assignment and to determine the ratio of the amplitudes of the electric quadrupole and magnetic dipole radiation in the 1.16-Mev transition. A preliminary and incomplete report of this work has been presented previously.<sup>8</sup>

## APPARATUS AND PROCEDURE

The gamma rays were detected by three scintillation counters, one of which served solely as a monitor. The electronic equipment was built following standard

<sup>8</sup> H. E. Handler and J. R. Richardson, Phys. Rev. **98**, 281(A) (1955).

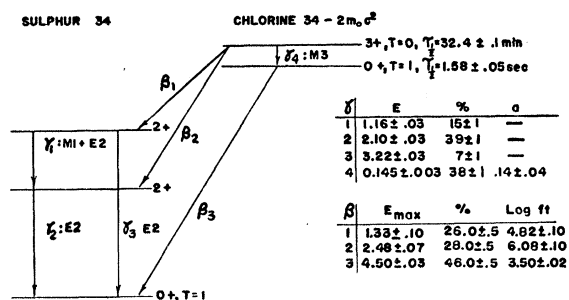


FIG. 1. Energy level diagrams of  $S^{34}$  and  $Cl^{34}$  together with a summary of the main gamma and beta transitions involved.

circuitry for the most part, and its performance was checked very frequently during the course of the work. As a test of the apparatus and procedure, the directional correlation of the well-known gamma rays which occur in the decay of  $Co^{60}$  was first measured; this test resulted in good agreement between the experimental correlation and that expected from theoretical considerations.

Figure 2 depicts the arrangement of the apparatus on the 36-inch correlation table. Counters 1 and 3 were permanently positioned on the table 90° apart, and Counter 2 could be fixed at any angular position from 45° to 225° with respect to Counter 1.

In the  $Co^{60}$  work, all three NaI(Tl) crystals were  $\frac{7}{8}$  inch in both diameter and thickness; in the  $Cl^{34}$  investigation, a crystal  $1\frac{3}{4}$  inches in diameter and 2 inches thick was substituted in the movable counter.

The  $Co^{60}$  source was prepared by dissolving a piece of a pile-irradiated cobalt wire in HCl and obtaining dry radioactive  $CoCl_2$  by evaporation. Adequate non-radio  $CoCl_2$  was mixed with the radioactive salt to provide sufficient bulk to fill the source volume of one of the source containers; care was taken to keep this

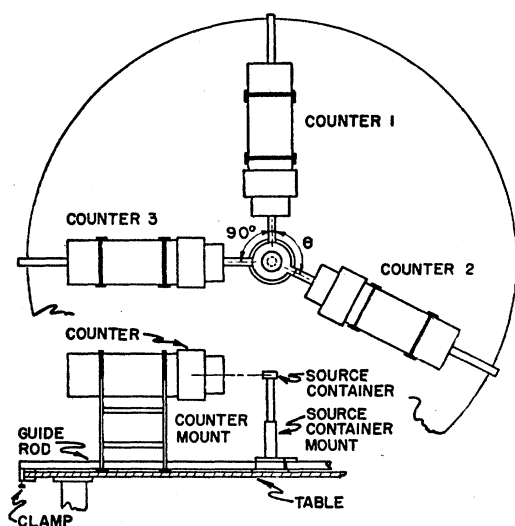


FIG. 2. The arrangement of counters on the correlation table. Counter 3 served as a fixed monitor counter.

source water-free. The approximate source strength was 0.05 millicurie.

Solid  $Cl^{34}$  samples were produced by bombarding pure, dry NaCl with the 20-Mev protons of the internal beam of the UCLA FM cyclotron; typical source strengths were about 0.1 millicurie at the beginning of the counting periods. Water solutions of radioactive NaCl were also used as sources, with an initial strength of about 0.03 millicurie. Care was taken to ensure that the most energetic beta particles were stopped in the source holders, and that the geometry for the absorption and scattering of the gamma rays in the source structure was the same in all cases.

Gross and accidental coincidences were counted simultaneously between counters 1 and 2; counter 3 served as a monitor.

The differential discriminator in each of the channels from counters 1 and 2 was set to accept pulse heights corresponding only to the full-energy peak of one of the gamma rays of the cascade under study. Coincidences arising from Compton scattering were thus reduced to a negligible number, and, in the  $Cl^{34}$  case, coincidences between the gamma rays and annihilation radiation were minimized. The large crystal which was installed in Channel 2 during the  $Cl^{34}$  work was used to enhance the full-energy peak of the 2.10-Mev radiation.

The minimum usable coincidence resolving times were about 0.3 microsecond for the  $Co^{60}$  work and about 0.4 microsecond during the  $Cl^{34}$  investigation. These were measured daily during the course of the counting runs and remained constant to within 0.5%.

Coincidence counting was performed with Counter 2 set at each of five angles from 90° to 180° relative to Counter 1. The  $Co^{60}$  data were accumulated in six two-hour runs at each angle; each run yielded about 8000 coincidences of which about 40% were accidentals. The  $Cl^{34}$  runs with the solid sources were done at the same five angles; each run required a freshly bombarded sample and lasted  $2\frac{1}{2}$  hours. A total of eighty-seven runs were made; typical counting periods yielded about 3000 gross coincidences and about 2000 accidentals. Twenty-eight two-hour runs at 90° and 180° only were made with the liquid solutions of radioactive NaCl; about 500 accidentals and 800 gross coincidences were obtained per run. The contribution of background to the coincidence rates were, in all cases, negligible.

Since the full-energy peak of the annihilation radiation in the  $Cl^{34}$  gamma-ray spectrum was very large, it extended somewhat above the lower energy limit of Channel 1, which was set to detect the 1.16-Mev radiation, and caused some coincidences to be registered between the 0.51-Mev quanta and the gamma rays of Channel 2. In order to determine the magnitude of this phenomenon, the over-all efficiency of Channel 1 for annihilation radiation was measured using a  $Na^{22}$  source and coincidence techniques. It was found that about 4% of the legitimate coincidences of the  $Cl^{34}$

work were due to this effect; corrections were made accordingly.

### RESULTS AND DISCUSSION

The angular-correlation function, expressed in terms of the quantities derivable from the experimental data obtained at angle " $\theta_i$ " during run " $j$ " is

$$w_j(\theta_i) = g \frac{N_3}{N_1 N_2} (C_G - C_A - C_B)_{ij}.$$

Here  $g$  is a constant which includes the appropriate branching ratios, parameters of the geometry, the counter efficiencies, the energy-selection limits, and the absorption properties of the source structure.  $N_1$ ,  $N_2$ , and  $N_3$  are the single-channel counts, corrected for scaling losses and background;  $C_G$  is the gross coincidence count.  $C_A$ , the number of accidentals, is the product of the number of counts registered in the "accidental coincidence channel" and the ratio of the

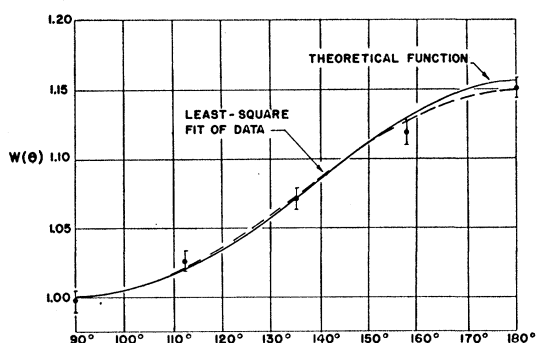


FIG. 3. The correlation function for the  $4 \rightarrow 2 \rightarrow 0$  cascade in  $\text{Ni}^{60}$ . The dashed curve represents a least square fit of the data and the solid curve is the theoretical function to be expected from this type of cascade.

resolving times of the two coincidence circuits.  $C_B$  is the number of "true" coincidences due to any phenomena other than the cascade under study; for the  $\text{Co}^{60}$  work,  $C_B$  was zero, and, for the  $\text{Cl}^{34}$  investigation,  $C_B = 2\rho\lambda N_2$ , where  $\rho$  is the over-all efficiency of Channel 1 for annihilation quanta and  $\lambda$ , calculated from the tables of Moszkowski,<sup>9</sup> is the fraction of the Channel 2 counts which were accompanied by positrons.

The  $w_j$  and their standard deviations, which were based solely on nuclear statistics, were calculated for all runs, and from these the weighted means for each angle,  $W(\theta_i)$ , and their standard deviations were computed. The internal consistency of the results at each angle was tested and justifies the use of these standard deviations. Table I lists the weighted means, arbitrarily normalized for each of the two cases. For the  $\text{S}^{34}$  cascade,  $W_L$  and  $W_S$  are the means for the liquid and solid samples, respectively; since these are equal at

TABLE I. Experimental weighted means and standard deviations for the angular correlation function.

$\theta$	$\text{Ni}^{60}$ $W(\theta_i)$	$W_S(\theta_i)$	$\text{S}^{34}$ $W_L(\theta_i)$	$W(\theta)$
$90^\circ$	$8094 \pm 66$	$1265 \pm 23$	$1278 \pm 39$	$1269 \pm 20$
$112\frac{1}{2}^\circ$	$8330 \pm 64$	$1396 \pm 28$	...	$1396 \pm 28$
$135^\circ$	$8690 \pm 67$	$1666 \pm 29$	...	$1666 \pm 29$
$157\frac{1}{2}^\circ$	$9073 \pm 68$	$1889 \pm 34$	...	$1889 \pm 34$
$180^\circ$	$9330 \pm 68$	$2035 \pm 39$	$1963 \pm 47$	$2006 \pm 30$

each of the two angles at which liquid samples were counted, they are combined to yield the grand means,  $W$ .

The coefficients,  $a_{2\nu}$ , in the expression

$$W(\theta) = \sum_{\nu=0}^{\infty} a_{2\nu} \cos^{2\nu}\theta$$

were fitted to each of the two sets of means by the method of least squares,<sup>10</sup> and the experimental anisotropy,  $R = [W(180^\circ) - W(90^\circ)]/W(90^\circ)$ , and its standard deviation were computed for each case. Table II includes these results and the theoretical coefficients and anisotropy for the  $4 \rightarrow 2 \rightarrow 0$  cascade, which is applicable to  $\text{Ni}^{60}$ . The latter quantities have been corrected for the finite solid angle of the detectors by the method described by Rose.<sup>10,11</sup> The normalization in all cases is such that  $W(90^\circ) = 1$ . The agreement between the least-square and theoretical coefficients and anisotropies for the  $\text{Ni}^{60}$  cascade is within statistical expectation and indicates that the experimental technique and method of analysis are reliable. The functions corresponding to the  $\text{Ni}^{60}$  coefficients are displayed graphically in Fig. 3.

The agreement between the means for the solid and liquid samples of  $\text{Cl}^{34}$  (see Table I) indicates that any attenuating effect due to electric interaction between the intermediate state of the  $\text{S}^{34}$  cascade with crystalline fields is smaller than the error of the measurements. Approximate calculations yield a life time of about  $10^{-12}$  second for a 2.10-Mev  $E2$  gamma ray; this is considerably shorter than the typical periods of precession induced by magnetic interactions (about  $10^{-8}$  second). Consequently, interpretation of the measured correlation of the  $\text{S}^{34}$  gamma rays in terms of theoretical angular correlation functions is meaningful.

Since the anisotropy is the best known of the least-squares quantities, the anisotropies were calculated

TABLE II. Angular correlation coefficients and anisotropies.

	$a_0$	$a_2$	$a_4$	$R$
$\text{Ni}^{60}$ : (least-squares)	1	$+0.138 \pm 0.041$	$+0.012 \pm 0.039$	$0.1502 \pm 0.0090$
(theoretical)	1	$+0.1219$	$+0.0342$	0.1561
$\text{S}^{34}$ : (least-squares)	1	$+0.66 \pm 0.10$	$-0.088 \pm 0.099$	$0.573 \pm 0.030$

<sup>10</sup> M. E. Rose, Phys. Rev. **91**, 610 (1953).

<sup>11</sup> E. D. Klement and F. K. McGowan, Phys. Rev. **92**, 1469 (1953).

<sup>9</sup> S. Moszkowski, Phys. Rev. **82**, 35 (1951).

TABLE III. Mixed-transition correlation parameters for  $R=0.573$ .

$J_1$	$J_2$	$\delta$	$a_2$	$a_4$
1	1	-2.05	+0.573	...
1	1	-0.488	+0.573	...
1	2	+6.89	+0.573	...
1	2	+0.464	+0.573	...
1	3	-16.6	+0.573	...
1	3	+0.738	+0.573	...
2	2	+0.133	+0.549	+0.0240
3	2	+0.401	+0.490	+0.0831
3	3	+1.42	+0.851	-0.278
3	3	-0.0426	+0.574	-0.00073
Experimental			+0.66±0.10	-0.088±0.099

from the theoretical functions<sup>12</sup> for pure multipole transitions for cascades with all combinations of spins 3, 2, and 1 for excited states and spin 0 for the ground state. After solid-angle corrections were made, these anisotropies were compared to the least-squares value. Since none of these was within two experimental standard deviations of  $R=0.573$ , it was concluded that a measurable mixture of magnetic and electric radiations occurs in the first transition in the  $S^{34}$  cascade.

For cascades in which such a mixture exists, the correlation function may be expressed

$$W(\theta) = W_{M, L} + \delta^2 W_{E, L+1} + 2\delta W_X = \sum_{\nu=0}^{\nu=\nu_{\max}} a_{2\nu} \cos^{2\nu}\theta,$$

where  $W_{M, L}$  and  $W_{E, L+1}$  are the functions, respectively, for pure magnetic  $2^L$ -pole and pure electric  $2^{L+1}$ -pole radiation in the otherwise mixed transition;  $W_X$  is an interference function; and  $\delta$  is the ratio of the reduced matrix elements of the appropriate electric and magnetic operators. The coefficients  $a_{2\nu}$  and the anisotropy are seen to be quadratic functions of  $\delta$ . These quadratic expressions were calculated for the same spin combinations as previously used and, after solid-angle corrections were made, those for the anisotropies were equated to the experimental value, 0.573. When solutions existed, two values of  $\delta$  were obtained and the corresponding  $a_{2\nu}$  calculated. Table III lists these results for all the cases for which no  $a_{2\nu}$  is more than three experimental standard deviations from the corresponding least-squares coefficient;  $a_0$  is again chosen to be unity for convenience. In Table III,  $J_1$  and  $J_2$  are the spins of the first and second excited states of  $S^{34}$ ; the experimental coefficients are included for comparison.

The gamma-gamma angular correlation alone thus leads to six possible sets of spin assignments for the excited states of  $S^{34}$ . In view of the other experimental

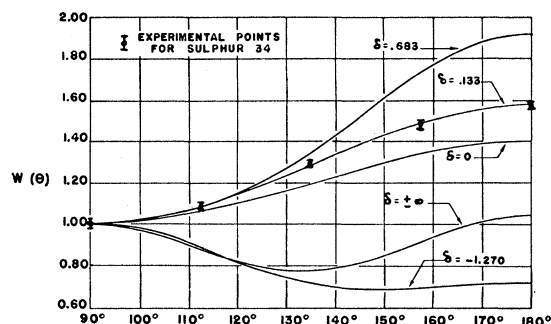


FIG. 4. The correlation function for the cascade in  $S^{34}$ . Assuming the cascade is  $2 \rightarrow 2 \rightarrow 0$ , the curves represent different mixtures of electric quadrupole and magnet dipole in the  $2 \rightarrow 2$  transition.  $\delta=0$  corresponds to pure  $M1$  and  $\delta=\infty$  corresponds to pure  $E2$  radiation.

information (see Fig. 1), the choice of  $J_1=J_2=2$  is by far the most probable of these. This assignment corresponds to  $\delta=0.133 \pm 0.024$  and leads to an intensity ratio of  $0.0177 \pm 0.0064$  or 1.7% electric quadrupole and 98.3% magnetic dipole for the 1.16-Mev transition. Figure 4 shows the experimental points for the  $S^{34}$  cascade and the correlation function for a  $2 \rightarrow 2 \rightarrow 0$  cascade for several values of  $\delta$  including that which best fits the data. The curves for  $\delta=0$  and  $\delta=\infty$  are those for pure  $M1$  and pure  $E2$  radiation in the first transition, respectively, and the curves for  $\delta=0.683$  and  $\delta=-1.270$  are those with the maximum and minimum anisotropies. Solid angle corrections have been applied to all these curves, and the normalization is such that  $W(90^\circ)=1$ .

It is to be hoped that eventually enough cases of the  $2 \rightarrow 2 \rightarrow 0$  cascade will have been investigated so that a comparison of experimental values of  $\delta$  with the predictions of the shell model will be possible. The published data, however, are not yet adequate for this purpose.

Since the completion of this work, Bleuler and Morinaga<sup>13</sup> have reported a weak positron transition to the third excited state of  $S^{34}$ . The effect of the gamma rays which originate from this state upon the results of the angular correlation measurement of this work would be negligible.

#### ACKNOWLEDGMENTS

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<sup>12</sup> L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953). This is a review of the theory of angular correlations of nuclear radiations and contains a complete bibliography of theoretical papers as well as many references to experimental work.

<sup>13</sup> E. Bleuler and H. Morinaga, Phys. Rev. **99**, 658(A) (1955).