

## Gamma-Gamma Angular Correlation in Ni<sup>60</sup>

E. D. KLEMA AND F. K. MCGOWAN  
Oak Ridge National Laboratory, Oak Ridge, Tennessee

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The angular correlation of the gamma rays emitted in cascade from the excited states of Ni<sup>60</sup> has been measured at increments of 10° from 90° to 270° with automatic apparatus using sodium iodide detectors and 5819 photomultipliers. For a dilute CoCl<sub>2</sub> source, the correlation obtained is  $W(\theta) = 1 + (0.131 \pm 0.017) \cos^2\theta + (0.024 \pm 0.017) \cos^4\theta$ . The anisotropy,  $(\alpha_2 + \alpha_4)/\alpha_0 = R$ , where  $W(\theta) = \alpha_0 + \alpha_2 \cos^2\theta + \alpha_4 \cos^4\theta$ , is  $0.1557 \pm 0.0010$ . The expected value of  $R$ , taking the finite angular resolution of the apparatus into account, is  $0.1517$ . A change in the anisotropy to a value of  $0.1453 \pm 0.0023$  due to a chemical change in the source was observed.

### I. INTRODUCTION

THE angular correlation of the gamma-rays emitted in cascade from the excited states of Ni<sup>60</sup> is known from experimental and theoretical considerations.<sup>1</sup> The present investigation was undertaken to find out how accurately the apparatus in use at Oak Ridge can measure the coefficients in the correlation function. This information is of considerable interest at present because of the finding of mixed radiation in parity-unfavored transitions.<sup>2</sup> The precision of the determination of the coefficients determines the limits which can be assigned to the mixing ratios. The Ni<sup>60</sup> correlation has been measured by several investigators<sup>3</sup> since the pioneer work by Brady and Deutsch. Recently Aeppli *et al.*<sup>4</sup> have measured the anisotropy precisely; and their value,  $0.148 \pm 0.002$ , is considerably lower than the expected theoretical value of 0.167.

### II. EXPERIMENTAL APPARATUS

The apparatus used in the present experiments is a modification of that described previously.<sup>5</sup> The detectors are sodium iodide crystals 1½ in. in diameter and 1 in. long, and the source is at the intersection of the axes of the cylindrical crystals at a distance of 7 centimeters from the front face of each crystal. The fractional acceptance of the windows of the differential analyzers included only the full-energy peaks of the two gamma rays in the cascade. The electronic equipment is the same as that described previously, but the apparatus has been equipped with a motor to change the angle between the two detectors. In the present experiment, data were taken automatically at 10° increments from 90° to 270°. The apparatus takes the data in sequence from 90° to 270° and then from 270° back to

90°, so that the point at 180° has only about half as much data as the other angles in the least squares fit of the data.

### III. EXPERIMENTAL METHOD AND ANALYSIS OF DATA

At any given angle  $\theta_i$  between the two detectors, the experimental method used was as follows: 4096 coincidence counts between the detectors were counted. The receipt of the 4096th count furnished a signal which started a sequence in which the time required for the collection of the counts was printed on a paper tape by a printing timer and the total number of singles counts of the fixed detector and of the moveable detector were also stamped on paper tapes.

The data for making the correction due to the random coincidences were obtained in a similar manner with the pulses from one detector delayed by 0.5 microsecond. In the present experiments the random rate was about 20 percent of the gross coincidence rate.

The above pieces of information are sufficient to enable one to combine the data for the various passes over a given angle properly, correcting for changes in the gain of the two detectors and of the fractional acceptance of the windows of the differential analyzers. Let us call the total counts for the  $j$ th pass over the angle  $\theta_i$ ,  $M_j$  and  $F_j$  for the moveable and fixed detectors, respectively; the number of coincidence counts observed  $C_j$ , and the time required for the collection of the  $C_j$  counts  $t_j$ . If the disintegration rate of the source is a constant  $N_0$ , then the total coincidence rate of the true coincidences plus the random coincidences is given by

$$\mu_j = N_0 e_F e_M W(\theta_i) + N_0 e_F N_0 e_M 2\tau,$$

where  $e_M$  and  $e_F$  contain the solid angles of the detectors, the detector efficiencies, and the fractional acceptance of the windows of the differential analyzers,

$$W(\theta_i) = \alpha_0 + \alpha_2 \cos^2\theta_i + \alpha_4 \cos^4\theta_i,$$

and  $\tau$  is the resolving time of the coincidence circuit.

The singles counting rates in the fixed and moveable channels are  $N_{0F} = N_0 e_F$  and  $N_{0M} = N_0 e_M$ , respectively. Thus the times  $t_j$  vary inversely with the product of

<sup>1</sup> E. L. Brady and M. Deutsch, Phys. Rev. **78**, 558 (1950).

<sup>2</sup> Aeppli, Frauenfelder, and Walter, Helv. Phys. Acta **24**, 335 (1951); E. D. Klema and F. K. McGowan, Phys. Rev. **87**, 524 (1952); R. M. Steffen and W. Zobel, Phys. Rev. **88**, 170 (1952); R. M. Steffen, Phys. Rev. **89**, 665 (1953).

<sup>3</sup> J. R. Beyster and M. L. Wiedenbeck, Phys. Rev. **79**, 411 (1950); R. M. Steffen, Phys. Rev. **80**, 115 (1950); H. E. Petch and M. E. Johns, Phys. Rev. **80**, 478 (1950); B. L. Robinson and L. Madansky, Phys. Rev. **84**, 604 (1951).

<sup>4</sup> Aeppli, Frauenfelder, Heer, and Rüttschi, Phys. Rev. **87**, 379 (1952).

<sup>5</sup> McGowan, Klema, and Bell, Phys. Rev. **85**, 152 (1952).

the channel rates,  $M_j F_j / t_j^2$ . One may then correct each  $t_j$  to an arbitrary fixed product of the channel rates  $N_{0F} N_{0M}$  as follows:

$$\text{Corrected } t_j \equiv t_{j0} = \frac{t_j (M_j F_j / t_j^2)}{N_{0F} N_{0M}} = \frac{1}{N_{0F} N_{0M}} \left( \frac{M_j F_j}{t_j} \right).$$

If  $n$  passes are made over the angle  $\theta_i$ , the corrected gross coincidence rate is given by

$$(\mu_{i0})_{\text{gross}} = n C_j / \sum_{j=1}^n t_{j0}.$$

Similarly, if  $m$  measurements are made of the random coincidence rate, collecting  $R_j$  coincidence counts each time, the corrected random rate is

$$(\mu_{i0})_{\text{random}} = N_{0F} N_{0M} m R_j / \sum_{j=1}^m \left( \frac{M_j \text{random} F_j \text{random}}{t_{j \text{random}}} \right).$$

We have finally for the corrected true coincidence rate at the angle  $\theta_i$ ,

$$\mu_{i0} = (\mu_{i0})_{\text{gross}} - (\mu_{i0})_{\text{random}}.$$

The variance of this rate,  $\sigma^2(\mu_{i0})$ , is given by

$$\sigma^2(\mu_{i0}) = (\mu_{i0})_{\text{gross}}^2 / n C_j + (\mu_{i0})_{\text{random}}^2 / m R_j.$$

The weighting factor,  $w(\mu_{i0})$ , to be used with  $\mu_{i0}$  in the least squares fitting of the data is then  $w(\mu_{i0}) = 1 / [\sigma^2(\mu_{i0})]$ .

#### IV. EXPERIMENTAL RESULTS

The data obtained in the various runs of this experiment are shown in Table I. The individual runs are based on total coincidence counts of from  $2 \times 10^5$  to  $4 \times 10^5$ . The first data obtained with a dilute solution source were those shown for August 27. This source consisted of 5 microliters of  $\text{CoCl}_2$  solution in a Lucite holder. After this run a continuous decrease in the anisotropy was noticed for a time, the values obtained being 0.148, 0.145, and 0.141. Then the anisotropy remained constant as is shown by the data in the table for the runs of September 4th through September 12th. It was hypothesized that this change in the anisotropy was caused by the fact that the liquid in the bottom of the Lucite source holder had disappeared, and that the chemical state of the source had changed (probably the cobalt changed from the divalent to the trivalent state). To test this hypothesis, 25 microliters of water were added to the source; and the data shown for the runs from October 3 through October 10 were obtained. Finally a dry source of  $\text{CoCl}_2$  was prepared, and the anisotropy found with this source for the runs of October 13 and October 15 agreed with that obtained with the dilute solution source.

The standard deviations of the  $R$ 's shown in Table I have been obtained from those defined in Eq. (30) of

TABLE I. Experimental values of the anisotropy,  $R = (\alpha_2 + \alpha_4) / \alpha_0$ , and the coefficients of the expansion  $W(\theta) = \alpha_0 + \alpha_2 \cos^2 \theta + \alpha_4 \cos^4 \theta$  obtained for the various runs. The  $\sigma(R)$  is the standard deviation of  $R$  in each case. The  $\epsilon^2$  is a figure of merit for each experiment; it is defined in the accompanying paper (see reference 6).

Date	$R$	$\sigma(R)$	$\epsilon^2$	$\alpha_2/\alpha_0$	$\alpha_4/\alpha_0$
August 27	0.1564	0.0072	0.898	0.201	-0.045
October 3	0.1554	0.0059	0.875	0.0936	0.62
October 6	0.1526	0.0082	1.088	0.156	-0.0032
October 8	0.1572	0.0091	1.564	0.125	0.032
October 10	0.1300	0.0071	1.124	0.0840	0.046
October 13	0.1533	0.0058	0.656	0.115	0.038
October 15	0.1592	0.0070	0.957	0.0972	0.062
September 4	0.1406	0.0078	0.691	0.109	0.032
September 8	0.1484	0.0034	0.234	0.113	0.035
September 10	0.1500	0.0058	0.679	0.132	0.018
September 12	0.1423	0.0047	0.688	0.106	0.036

the accompanying paper<sup>6</sup> as follows:

$$\begin{aligned} \text{variance} \left( \frac{\alpha_2 + \alpha_4}{\alpha_0} \right) &\equiv V \left( \frac{\alpha_2 + \alpha_4}{\alpha_0} \right) \cong \left( \frac{\alpha_2 + \alpha_4}{\alpha_0} \right)^2 \\ &\times \left[ \frac{V(\alpha_2) + V(\alpha_4) + 2 \text{Cov}(\alpha_2, \alpha_4)}{(\alpha_2 + \alpha_4)^2} + \frac{V(\alpha_0)}{\alpha_0^2} \right. \\ &\quad \left. - 2 \frac{\text{Cov}(\alpha_0, \alpha_2) + \text{Cov}(\alpha_2, \alpha_4)}{\alpha_0(\alpha_2 + \alpha_4)} \right], \end{aligned}$$

where

$$V(\alpha_\lambda) = \epsilon^2 \sigma^2(\alpha_\lambda),$$

and

$$\begin{aligned} \text{covariance}(\alpha_\lambda, \alpha_{\lambda'}) &\equiv \text{Cov}(\alpha_\lambda, \alpha_{\lambda'}) \\ &= \langle (\alpha_\lambda - \alpha_\lambda^0)(\alpha_{\lambda'} - \alpha_{\lambda'}^0) \rangle_N \\ &= [\sum_i v_i^2 / (m-1)] C^{-1}_{\lambda\lambda'}, \quad \lambda \neq \lambda'. \end{aligned}$$

On evidence obtained from an unpublished investigation<sup>7</sup> of the accuracy of the approximate formula for the variance of a ratio, the variance estimate from the above equation for  $V((\alpha_2 + \alpha_4) / \alpha_0)$  is probably within 2 percent of the true variance.

The  $\epsilon^2$  for each measurement is shown in column 4. Since the values of  $\epsilon$  cluster about unity, it has been concluded that the various runs have a single statistical distribution; and in each case of the final results of the experiment as shown in Table II, the standard deviation of the mean has been calculated from the mean square deviation of the individual runs from the average.

The means obtained have been calculated both without and with the run of October 10. If one applies a range criterion to see if the October 10 run can safely be omitted from the group of runs on the argument that it does not belong in the same statistical distribution which is represented by the other members,<sup>8</sup> one

<sup>6</sup> M. E. Rose, (preceding article) Phys. Rev. **91**, 609 (1953).

<sup>7</sup> A. W. Kimball, private communication.

<sup>8</sup> J. Moshman and G. J. Atta, Oak Ridge National Laboratory Report ORNL 1020 (unpublished).

TABLE II. Mean values of the quantities listed in Table I. The errors are standard deviations.

	Dilute solution and dried $\text{CoCl}_2$ sources	Chemically changed source
Without October 10 run	$\bar{R} = 0.1557 \pm 0.0010$	$\bar{R} = 0.1453 \pm 0.0023$
With October 10 run	$\bar{R} = 0.1520 \pm 0.0038$	
Without October 10 run	$W(\theta) = 1 + (0.131 \pm 0.017) \cos^2\theta + (0.024 \pm 0.017) \cos^4\theta$	$W(\theta) = 1 + (0.1149 \pm 0.0058) \cos^2\theta + (0.0304 \pm 0.0042) \cos^4\theta$
With October 10 run	$W(\theta) = 1 + (0.124 \pm 0.016) \cos^2\theta + (0.028 \pm 0.015) \cos^4\theta$	

finds that he can do so with less than a 1 percent probability of being in error.

In order to determine whether a real effect exists for the different source conditions, a "Student"  $t$ -test has been performed on the two sets of data. The test is used as a check on the consistency between the sample and the hypothesis that the two means in question are equal. If the computed  $t$  is large, as indicated by its level of significance, the hypothesis of equality of true means is rejected as incompatible with the differences in the observed means. When one considers the results without the October 10th run, he finds that if there were in fact no difference between the two sets of experiments, the chances of getting as large a difference of the means of the two sets of data as was observed are less than 1 out of 1000. The corresponding value with the October 10 run included is 12 chances in 100.

## V. DISCUSSION

The theoretical correlation function for two quadrupole transitions between levels of angular momenta 4, 2, and 0, when allowance is made for the finite angular resolution of the detectors,<sup>6</sup> is

$$W(\theta) = 1 + 0.1203 \cos^2\theta + 0.0314 \cos^4\theta,$$

which yields a value of 0.1517 for the anisotropy. This calculated anisotropy is unaffected by the 3 percent uncertainty in the absorption cross section of sodium iodide for the gamma-rays of the  $\text{Ni}^{60}$  cascade. The theoretical correlation function is modified to allow for finite angular resolution before comparing with the measured anisotropy because the anisotropy is measured more accurately than the individual coefficients.

We can summarize the results of the present experiments as follows: the measured effect on the anisotropy of the chemical state of the source seems to be a real one with some degree of certainty. This observed change

is surprising since the first measurement of the lifetime of the intermediate state in the cascade is  $< 10^{-10}$  second.<sup>9</sup> With this short lifetime one would expect that the correlation would be unperturbed by external fields. The explanation of the deviation of four times the standard error of our measurements from the expected value of the anisotropy is probably to be found in imperfection in the experimental apparatus, even though the  $\epsilon^2$  tests of the data show that the nonstatistical errors are small. However, it does seem that with a dilute source of  $\text{CoCl}_2$ , the anisotropy of the cascade is not attenuated by external effects.

Since the above experiments were completed, it was necessary to dismantle and reassemble the apparatus. The detectors have been changed to use a different light reflector around the sodium iodide crystals, and the coincidence circuit has been replaced by a new one of improved design. This apparatus has been checked by measuring the  $\text{Ni}^{60}$  cascade with a dilute solution source in a fluorothene holder. The results obtained for the anisotropy are  $0.1501 \pm 0.0054$  and  $0.1532 \pm 0.0027$ . The average of these measurements agrees with the expected anisotropy of 0.1517. If these results are combined with those of the previously described experiments, the discrepancy between the experimental and theoretical values is reduced to three times the standard deviation of the measurements.

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<sup>9</sup> Bay, Henri, and McLernon, Phys. Rev. **90**, 371 (1953).