Gamma-Gamma Angular Correlation in Pd^{106}

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The angular correlation of the 624- and 513-kev γ rays and that of the 1045- and 513-kev γ rays of Pd^{106} have been measured with a coincidence scintillation spectrometer using NaI detectors. For a dilute ruthenium chloride aqueous solution source, the correlation function of the 624–513-kev cascade is $W(\theta)=1+(0.3456\pm0.0079)P_2(\cos\theta)+(1.109\pm0.012)P_4(\cos\theta)$; that of the 1045–513-kev cascade is $W(\theta)=1+(0.0921\pm0.0027)P_2(\cos\theta)+(0.0350\pm0.0092)P_4(\cos\theta)$. In each of the above functions, the finite angular resolution of the equipment has been taken into account. The measurement on the 1045–513-kev cascade establishes the spin of the 1550-kev level as two and gives the intensity of the electric quadrupole radiation as 4.24±0.14 percent relative to the intensity of the magnetic dipole radiation in the 1045-kev transition.

I. INTRODUCTION

In a recent paper[1] Kraushaar and Goldhaber have reported a rather large discrepancy between the theoretically expected angular correlation of the 624–513-kev γ-γ cascade in Pd^{106} and the experimentally observed one. Previous measurements at Oak Ridge National Laboratory had indicated a much smaller discrepancy.[2] Since these measurements were made, the apparatus at Oak Ridge has been improved,[3] and it was decided to re-investigate the correlation with the new apparatus and to measure the angular correlation of the 1045–513-kev cascade. In addition the question of the angular resolution of the apparatus has been re-examined.

II. ANGULAR RESOLUTION CORRECTION

Finite solid angle corrections for directional angular correlation measurements have been treated in considerable detail recently by Walter,[4] Frankel,[5] Church and Kraushaar,[6] Rose,[7] and Lawson and Frauenfelder.[8] In our measurements we use the angular resolution corrections[7] which have been computed as a function of the geometry and the cross section of the NaI detector for γ radiation. Recently, Steffen[9] and Lawson and Frauenfelder[8] have indicated that the computed corrections[7] are useful as an approximation and that the corrections can only be achieved by using experimentally determined angular resolution curves. The latter statement is true for the arrangement and operation of the detectors which most other workers use in their angular correlation apparatus.

In view of this it seems appropriate to show that the computed corrections of Rose[7] are valid provided the detectors are operated under the conditions for which the calculations were made. These conditions are that the absorption of γ radiation in the NaI crystal is proportional to $(1-e^{-\tau x(\beta)})$ and that there is no radiation loss between the source and the NaI crystal. This latter condition has considerable significance in the finite angular resolution corrections.

In any practical geometrical arrangement of the source and detectors for an angular correlation apparatus, there is always degraded radiation, in addition to the undegraded radiation from the source, incident on the detectors. To shield against the degraded radiation resulting from the scattering of the primary radiation from the surroundings of the detectors, most workers have used lateral Pb shields around the NaI crystals and in some cases Pb shields on the front faces of the crystals. Unfortunately, this type of shield does not shield against degraded radiation scattered from the surroundings beyond the back face of the crystal with cylindrical symmetry. In addition, since the solid angle subtended at the source by the lateral Pb shield is usually larger than the solid angle subtended by the NaI crystal, the primary radiation scattered from the Pb shielding provides another source of degraded radiation incident on the detector. The Pb shielding in close to the detector thus increases the effective solid angle of the detector.

A differential pulse-height spectrum of 512-kev γ radiation incident on a NaI scintillation spectrometer has been measured to illustrate the points mentioned above. The solid curve of Fig. 1 is the pulse spectrum of undegraded radiation from a source of Sr^{85} located at 7 cm from the front face and on the axis of a NaI crystal 1.5 in. in diameter and 1.0 in. long. The lateral sides and front face of the crystal are covered with 30 mg/cm² of MgO and 0.010 in. of Al. The dashed curve at the lower pulse heights and the solid curve for pulse heights greater than 550 is the pulse spectrum of both the undegraded radiation and the degraded radiation resulting from scattering by the surroundings. The difference between these two curves represents the pulse spectrum of degraded radiation from the surroundings. The intensity of the degraded pulse spectrum relative to the undegraded pulse spectrum is 12 percent. The curve represented by the solid points is

[1] J. J. Kraushaar and M. Goldhaber, Phys. Rev. **89**, 1081 (1953).
[2] Arfken, Klema, and McGowan, Phys. Rev. **86**, 413 (1952).
[3] E. D. Klema and F. K. McGowan, Phys. Rev. **91**, 616 (1953).
[4] Walter, Huber, and Zünti, Helv. Phys. Acta **23**, 697 (1950).
[5] S. Frankel, Phys. Rev. **83**, 673 (1951).
[6] E. L. Church and J. J. Kraushaar, Phys. Rev. **88**, 419 (1952).
[7] M. E. Rose, Phys. Rev. **91**, 610 (1953).
[8] J. S. Lawson, Jr. and H. Frauenfelder, Phys. Rev. **91**, 649 (1953).
[9] Rolf M. Steffen, Phys. Rev. **90**, 321 (1953).

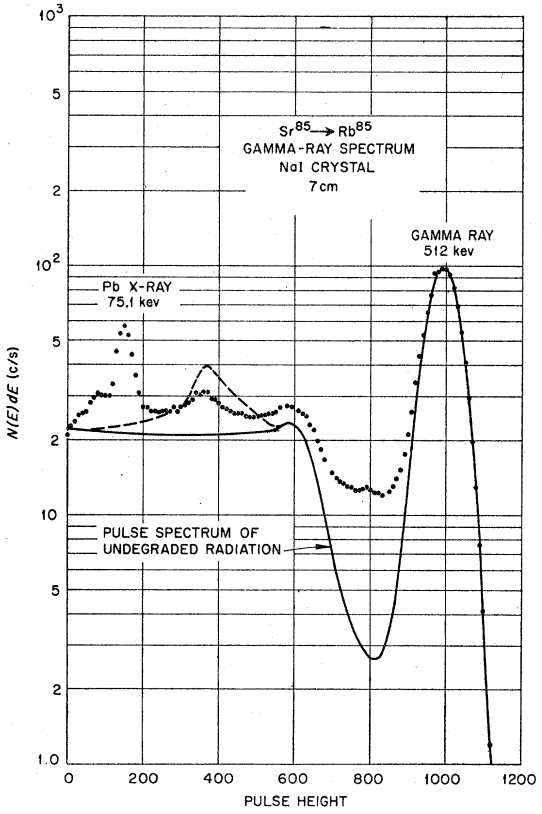

FIG. 1. Differential pulse spectrum of the Sr^{85} gamma radiation incident on a NaI scintillation spectrometer for the various experimental conditions discussed in the text.

the pulse spectrum of the γ radiation incident on the detector with 0.6 cm of Pb around the lateral sides and front face of the NaI crystal. This amount of Pb is less than that used by most workers, but is enough to attenuate the degraded radiation from the surroundings by a factor of 10^3. In this case the intensity of the degraded pulse spectrum relative to the undegraded pulse spectrum is 27 percent. In addition, the degraded spectrum is spread over a greater portion of the undegraded pulse spectrum.

In either arrangement it is desirable to operate the scintillation detector as a spectrometer rather than as a pulse counter, selecting only the full-energy peak of the pulse spectrum to insure that only undegraded radiation is counted. This method of operation immediately assures one that the solid angle subtended by the detector at the source, aside from the effect of the very small coherent scattering cross section of γ radiation by matter, is that subtended by the NaI phosphor.

In our equipment the windows of the differential pulse-height analyzers of the coincidence scintillation spectrometer in the angular-correlation apparatus are always set to include only the full-energy peak of the pulse spectrum of the γ ray in question. The full energy of the γ ray is dissipated in the phosphor by photoelectric absorption and also by multiple processes, i.e., Compton recoil electrons and the eventual absorption of the Compton-scattered γ ray. The ratio of the integral of the pulse spectrum in the full-energy peak to the integral of the total pulse spectrum of undegraded radiation is, of course, a function of the γ-ray energy incident on the NaI phosphor. Also, the peak to total ratio (ratio of the two integrals mentioned above) is expected to vary slightly over the NaI phosphor. This variation was neglected in the calculation of the corrections for finite angular resolution[7] for two reasons. The variation of the peak to total ratio over the crystal is not known, and preliminary estimates indicated that this effect on the finite angular resolution was very small.

After the computed angular resolution corrections were completed, the angular resolution was measured experimentally at $E_\gamma = 1.114$ Mev using a source of Zn^{65}. At this energy the contribution of the multiple processes to the intensity of the full-energy peak of the pulse spectrum is largest and is comparable to the photoelectric absorption contribution. If the measured and computed angular resolution corrections agree at this energy, then one is perfectly justified to use the computed corrections at all other energies.

The angular resolution curve for a collimated beam of 1.114-Mev γ rays is shown in Fig. 2. The radiation from a 2-mc source of Zn^{65} was collimated by a 0.3-cm diameter hole through a 4-in. Pb brick. To obtain the angular resolution curve in Fig. 2 one must, of course,

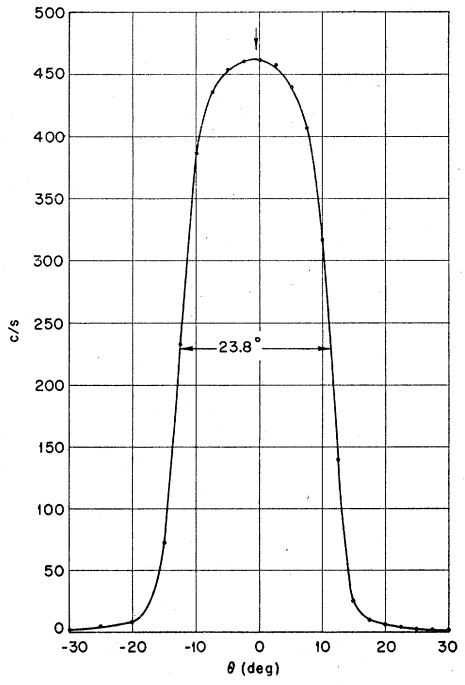

FIG. 2. Angular resolution curve for 1.114-Mev γ rays collimated onto a NaI crystal 1.5 inches in diameter by 1 inch long located at 7 cm from the axis of rotation.

measure the angular resolution with the collimator and with the collimator replaced by a solid Pb brick.

The integrals $J_l=\int_0^{\theta_{max}} P_l(\cos\theta)\epsilon(\theta)\sin\theta d\theta$ were evaluated by numerical integration for $l=0$, 2, and 4 using the observed angular resolution curve of Fig. 2 for $\epsilon(\theta)$. The results in the form J_2/J_0 and J_4/J_0 were 0.2 and 0.7 percent larger than the computed values.[7] The deviations are in the direction expected because the computed corrections are not weighted over the NaI phosphor to account for the variation of the peak to total ratio over the phosphor. Since the discrepancies are considerably smaller than the fractional standard deviations of the measured angular correlation coefficients due to statistical fluctuations, the use of the computed angular-resolution correction factors is certainly justified. To obtain the correction factors from measured angular-resolution curves, one is limited to about eight sources with a single monoenergetic γ ray having energies distributed between 279 and 1114 kev. Of these eight, only four may be obtained with sufficiently high specific activity to provide a source of well-collimated γ rays.

III. EXPERIMENTAL METHOD AND RESULTS

In the present experiments the data were obtained as discussed in the previous paper[3] and analyzed as discussed in the paper of Rose.[7] However, in the case of Pd^{106}, the energy resolution of the equipment has a very marked effect on the observed results.[2] The β^- shield for these measurements consisted of $\frac{1}{16}$ in. of Cu on the lateral sides of the crystal and $\frac{1}{2}$ in. of fluorothene on the front face. The γ-ray spectrum of the ruthenium chloride source is shown in Fig. 3. The bump on the high-energy side of the full-energy peak of the 1045-kev γ ray at about 960 pulse-height units is due mostly to sum pulses of a 513- and a 624-kev γ ray. Each point on the curve out to a pulse height of 1000 units contains 4096 counts, and the points for greater pulse heights contain 1024 counts each.

In the first set of experiments on the main cascade of Pd^{106}, the window of the differential analyzer of the fixed detector was set between the limits represented by A in Fig. 3 and the window of the moveable detector was set at B in the measurement of the correlation due to the composite 624–513, 870–513, 870–1045, 1045–513, and 870–1550 kev cascades and a source of coincidence counts due to inner and external bremsstrahlung correlated primarily with either the 513- or the 624-kev γ rays. Preliminary measurements indicate that the 870-kev γ ray is not in coincidence with the 1045. Alburger's[10] decay scheme predicts that they should be in cascade; thus the interference of the cascades other than the main one may not be as indicated above. A total of 6×10^5 coincidence counts were measured at 19 angular positions with this arrangement, and the angular correlation obtained was

[10] David E. Alburger, Phys. Rev. 88, 339 (1952).

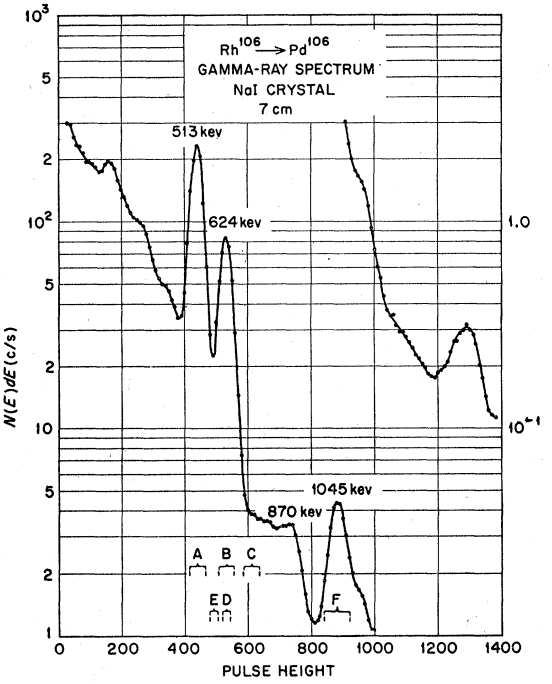

FIG. 3. Gammay-ray spectrum of the ruthenium chloride source. The right-hand section of the curve above pulse height 910 is to be referred to the scale at the right of the figure. The limits represented by A, B, C, etc. show the positions of the windows of the differential pulse-height analyzers for the various experiments described in the text.

$W(\theta)=1+0.3173\ P_2(\cos\theta)+1.058\ P_4(\cos\theta)$. The movable detector window was then set at C, and 2.5×10^4 counts were obtained at this setting, which represents the correlation due to the 1045–513-kev cascade and the bremsstrahlung correlated with the 512-kev γ ray. The position of the window at C is in the valley between the Compton distribution and the full-energy peak of the 870-kev γ ray; thus, virtually none of the cascades involving the 870-kev γ ray contribute to this measurement. The second correlation function was then subtracted from the first to give the correlation function for the 624–513-kev cascade as measured by this experiment. The function obtained is

$$W(\theta)=1+(0.3272\pm0.0047)P_2(\cos\theta)+(1.103\pm0.0025)P_4(\cos\theta).$$

Clearly, this is not quite the correct function because the correlation due to the higher energy cascades in which the 870-kev γ ray enters has not been subtracted off properly.

In order to get a better measurement of the correlation function of the main cascade, the following set of experiments was performed. The fixed analyzer was left at A and the movable one was set at D as shown in Fig. 3 in the measurement of the composite correlation. The random coincidences were not measured in this experiment, but were calculated from the known resolving time of the apparatus and the singles rates of

the two detectors. The resolving time is $(0.113\pm0.001)\times10^{-6}$ sec and remains constant to within 2 percent over long periods of time. The correlation function obtained was $W(\theta)=1+0.3310\,P_2\cos(\theta)+1.061\,P_4\cos(\theta)$. The effect of the higher-energy cascades was obtained by measuring the correlation with the window of the moveable detector set at E (both D and E are on the flat part of the Compton distribution of the 870-kev γ ray). However, the setting at E overemphasizes slightly the effect of the inner and external bremsstrahlung because of the rapid increase of the photon spectrum with decreasing energy. Also, in this case the correction for the changes in gain of the moveable detector and of the fractional acceptance of the window of its differential analyzer as discussed in the previous paper[3] does not apply since the product of the channel rates is not a good measure of the product of the individual efficiencies involved in the composite correlation. In this experiment, the windows remained constant to 1 percent with respect both to pulse height and their widths, and these data were not corrected. The correlation function obtained under these conditions in two runs with the window at D and a total of 2.8×10^5 coincidence counts and two runs with the window at E and a total of 1.2×10^5 counts is

$$W(\theta)=1+(0.3456\pm0.0079)P_2(\cos\theta)+(1.109\pm0.012)P_4(\cos\theta).$$

In the above function the standard deviations of the coefficients of P_2 and P_4 have not been calculated as discussed in the previous paper.[3] At present the standard deviation of each term in the expansion of the correlation function is calculated in the least-squares fit of the data of each run. The standard deviation of the mean of the runs is then calculated from the deviations of the individual runs. This can be done easily because the various runs of the experiment have the same number of degrees of freedom statistically and their individual variances can be combined directly.

The correlation function of the 1045–513-kev cascade was measured with one window at A and the other at F as shown in Fig. 3. Again in this case, since the counting rates were low, the correction for the random coincidence rate was calculated. The correlation obtained for four runs with a total coincidence count of 10^5 is

$$W(\theta)=1+(0.0921\pm0.0027)P_2(\cos\theta)+(0.0350\pm0.0092)P_4(\cos\theta).$$

IV. DISCUSSION

The discrepancies between the theoretical and experimental coefficients of P_2 and P_4 for the spin sequence 0–2–0 in the cleaner measurement of the present series on the main cascade are about 3 percent and are 1.5 and 2.8 times the standard deviation of the measurements. It is believed that these discrepancies are caused by interfering radiation in the windows of the differential analyzers even for the narrowest windows it was practical to use. Pound and Abragam[11] have recently attributed the considerably larger discrepancies (22.1 and 13.6 percent) found by Kraushaar and Goldhaber[1] to the interaction of the electric quadrupole moment of the intermediate state of the nucleus with the gradient of the electric field due to the electrons. The present results indicate that the attenuations of the cofficients of P_2 and P_4 by this interaction, if any, are exceedingly small. The discrepancy between the results obtained at Oak Ridge and those obtained at Brookhaven is probably to be explained in the use of differential pulse-height selection at Oak Ridge. This type of operation permits one to reduce the effect of the interfering cascades and also to measure their effect directly. This is shown by the fact that the discrepancies between the correlations uncorrected for the interfering cascades and theory were 12.5 and 7.9 percent for the coefficient of $P_2(\cos\theta)$ and 8.0 and 7.7 percent for the coefficient of $P_4(\cos\theta)$ in the two sets of experiments described in the foregoing. We have tried several different sources, both liquid and solid, and have obtained the same results with all of them. We have also used the equipment with only integral pulse-height selection, and in this case we can get correlation function similar to those obtained at Brookhaven.

The measured correlation function of the 1045–513-kev cascade allows the unambiguous assignment of spin 2 to the 1550-kev level in Pd^{106} in agreement with our previous measurements.[2] From the value of the measured coefficient of P_2 and its standard deviation, one finds that the 1045-kev transition consists of 4.24 ± 0.14-percent electric quadrupole radiation relative to magnetic dipole radiation. The value for the mixing ratio obtained from the measurement of the coefficient of P_4 is not so precise and differs from the above by about twice its standard deviation.

[11] R. V. Pound and A. Abragam, Phys. Rev. **90**, 993 (1953).