Gamma-Gamma Directional Correlations in Co^{59}, Xe^{131}, and Hg^{198} †,*

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The directional correlations of low intensity gamma-gamma cascades have been measured using NaI scintillation counters in a coincidence arrangement of 1.5×10^{-8} second resolving time. The correlation in the 3 percent intensity 191–1089-kev cascade in Co^{59} is compatible with a 3/2–5/2–7/2 spin assignment. The coincidences from the 284–80-kev cascade in Xe^{131} are isotropic within an error of 2 percent, supporting the assignment of spin $\frac{1}{2}$ to the 80-kev level. The large correlation observed in the ~1 percent intensity 680-411-kev cascade of Hg^{198} characterizes the spins of the levels involved as 2, 2, 0 and the 680-kev transition as a mixture of 60 percent $E2$ and 40 percent $M1$.

I. INTRODUCTION

THE measurement of gamma-gamma directional correlations has proven a valuable means of determining the spins of low-lying nuclear energy levels and the radiation character of transitions between the states. Previous experiments of this type have dealt with cascades which were present in a high percentage of the nuclear disintegrations. The present investigations involve cascades which represent only a small percentage of all the gamma-rays emitted from a given isotope. One has, therefore, to discriminate against the large number of gamma-rays not connected with the cascade. This discrimination, which can be accomplished by means of either lead absorption or pulse-height selection, reduces the efficiency for the detection of the cascade and in conjunction with the original low intensity results in a small coincidence rate. However, by shortening the resolving time of the coincidence circuit, one can increase the coincidence rate resulting from the cascade without at the same time increasing the chance coincidence rate. A practical lower limit for the resolving time is determined by the decay time of the scintillation crystal used for a detector.

There were two types of crystals available for these experiments: NaI crystals with a decay time of 2.5×10^{-7} second[1] and organic crystals with decay times up to 100 times shorter. NaI was chosen because its large photoelectric cross section can be employed as a basis for energy discrimination of the gamma-rays, while its comparatively large density results in a better gamma-detection efficiency than is obtained in organic crystals of comparable size. The use of NaI detectors with a coincidence arrangement of 1.5×10^{-8} second resolving time is roughly equivalent to an organic crystal arrangement with 10^{-9} second resolving time, in so far as gamma-gamma cascades are concerned.

The isotopes Co^{59}, Xe^{131}, and Hg^{198} were chosen for study because they each have only one gamma-gamma cascade. The cascades are all of low intensity, occurring in only 3 percent of the disintegrations in Co^{59}, 6 percent in Xe^{131}, and about 1 percent in Hg^{198}. In each case the extra coincidences resulting from bremsstrahlung and backscattering were so far reduced as not to affect the directional correlation measurement. The coincidences resulting from annihilation radiation were not reduced in the Co^{59} experiment but were corrected

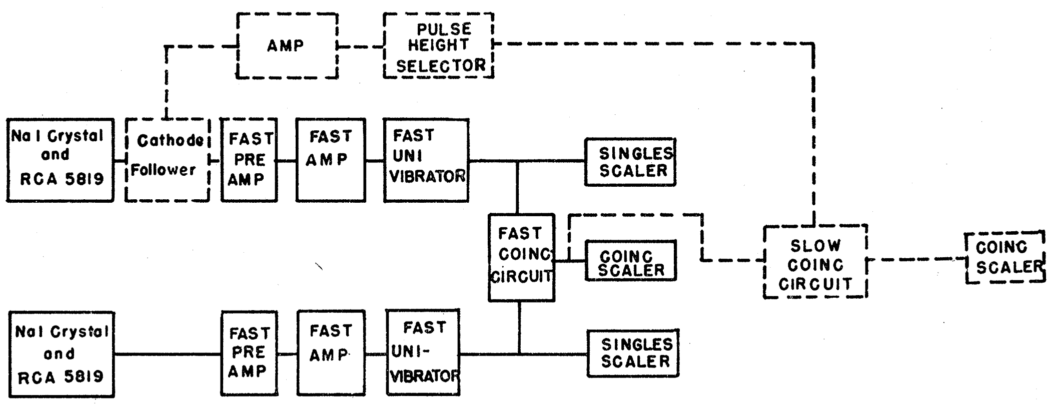

FIG. 1. Block diagram of electronic circuit.

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§ Now with the Bartol Research Foundation of the Franklin Institute, Swarthmore, Pennsylvania.
[1] R. Hofstadter and J. A. McIntyre, Nucleonics 7, 32 (1950).

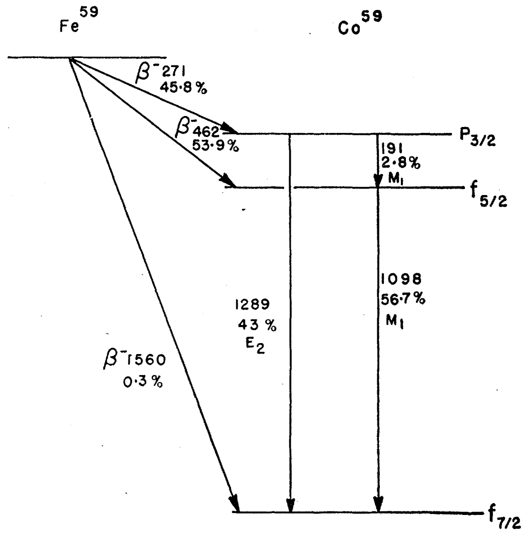

FIG. 2. Fe^{59} decay scheme.

for after a calculation. The limits of the equipment were reached in the measurements on the 0.2 percent cascade in Os^{186}, which are not reported here because of the large uncertainty produced by the above effects.

II. APPARATUS

The arrangement of the crystals and shielding was similar to that described by Deutsch.[2] The outputs of the RCA 5819 photomultiplier tubes fed amplifiers of 1.5×10^{-8} second rise time which were built after a design by W. C. Elmore. The amplifier output pulses of varying size were modified to constant voltage pulses by univibrators of 1.0×10^{-8} second rise time which led to either side of a fast coincidence circuit.[3] The arrangement of these units is shown in the block diagram of Fig. 1. The units represented by dashed lines were used only when pulse-height selection was needed, and, as shown, the pulse-height selector was installed in a side channel instead of after the main amplifier so that its comparative slowness and variable delay would not affect the fast coincidence procedure.

In using pulse-height selection on gamma-rays, it is often necessary to accept only the large pulses arising from photoelectric processes in the crystal. This discrimination against lower energies eliminates most of the pulses arising from Compton scattering. When the gamma-ray energy is above 500 kev the large percentage of rejected pulses in the Compton distribution results in a smaller coincidence rate. In many cases it is advantageous at these energies to replace the pulse-height selector by a lead absorber which greatly attenuates low energy radiation and yet does not appreciably affect the hard component.

One of the important uses of energy discrimination is to reduce those unwanted coincidences which are due mainly to the effects of bremsstrahlung, annihilation radiation, and backscattering. All beta-emitters produce bremsstrahlung, which is predominantly of low energy and can be effectively discriminated against. The isotopes chosen for investigation do not decay by positron emission, but annihilation radiation might occur owing to pair production in the source, absorber, or crystal if a gamma-ray of sufficiently high energy is present in the decay. Although the 511-kev annihilation radiation is sometimes difficult to discriminate against, it only affects measurements at 180° and may be eliminated by narrowing the acceptance angle and measuring at angles less than 180°. Backscattering, or Compton scattering in the 180° direction, is a problem peculiar to gamma-gamma directional correlation measurements and occurs to some extent in every such experiment. It affects the counting rates in positions near 180° where the lead cones do not protect the crystals.

The energy of the backscattered radiation is of the order of 200 kev, and the radiation can be discriminated against as long as the energy of the two cascade gamma-rays is somewhat larger. When, however, one of the cascade gamma-rays has almost the same energy as the backscattered radiation, energy discrimination no longer solves the problem. This situation occurs in Co^{59}, where the two cascade gamma-rays are of 191-kev and 1098-kev energy and the harder component is scattered in the backward direction with an energy of 207 kev. In this case a thin crystal, with only a copper absorber in front of it to reduce bremsstrahlung and

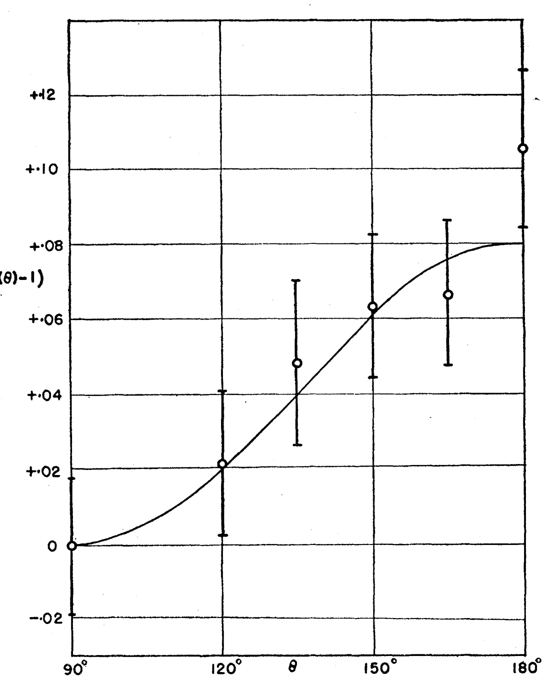

FIG. 3. Fe^{59} directional correlation. Data corrected for angular resolution.

[2] M. Deutsch, Repts. Progr. Phys. **14**, 196 (1951).
[3] K. P. Meyer *et al.*, Helv. Phys. Acta **23**, 121 (1950).

x-rays, is used on the side which detects mainly the 191-kev gamma-ray. The hard component alone is detected on the other side in a thick crystal with a lead absorber in front of it. The backscattered radiation originating in one crystal is thus prevented by the lead absorber from reaching the opposite crystal. If the Compton scattering occurs on the absorber surface away from the crystal so that the backscattered radiation can reach the other counter, then the Compton electron is absorbed in the lead. Thus coincidences resulting from the backscattering process are completely eliminated.

III. RESULTS AND DISCUSSION

Co^{59}

The decay scheme[4] of Fe^{59} is presented in Fig. 2. The spins were assigned on the basis of conversion experiments. The directional correlation measurement between the 191-kev and 1098-kev cascade gamma-rays was used as a confirmation.

After the backscattering had been eliminated by the method outlined in Sec. II, the 180° point was still somewhat too high because of the annihilation radiation following pair production by the 1289-kev gamma-ray. A calculation showed that the number of additional coincidences at 180° corresponded to the number expected from the pair creation process. Consequently, the 180° point was omitted in determining the best fit to the data, which is given by the correlation $W(\theta)=1+A_2\cos^2\theta$, with $A_2=+0.080\pm0.016$. These results are plotted in Fig. 3, corrected for angular resolution.

The most probable spin assignments to the 1289 kev, 1098 kev, and ground levels of Co^{59} are[4] 3/2–3/2–7/2, 3/2–5/2–7/2, 5/2–3/2–7/2, and 5/2–5/2–7/2. Of these, the measured correlation excludes only the 5/2–3/2–7/2 assignment, which is the most likely

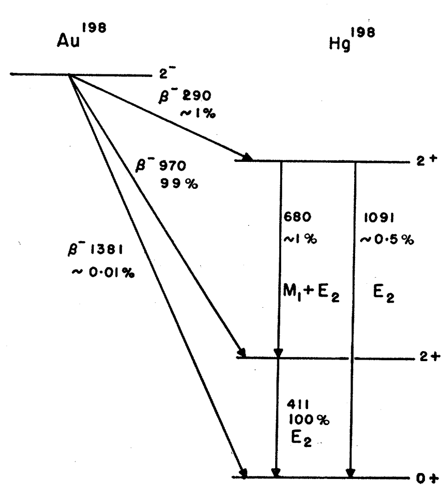

FIG. 4. Au^{198} decay scheme.

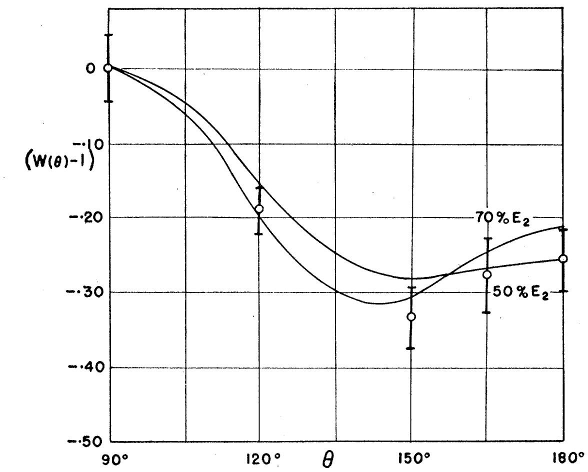

FIG. 5. Au^{198} directional correlation. Curves correspond to 2–2–0 spin assignment, zero phase, with anisotropy coefficients in $W(\theta)=1+A_2\cos^2\theta+A_4\cos^4\theta$ corrected for angular resolution.

alternative to the final 3/2–5/2–7/2 combination from the point of view of shell theory. The 5/2–3/2–7/2 assignment is excluded because all $M1-E2$ mixtures compatible with the measured internal conversion coefficients[4] of the 191-kev transition lead to anisotropies smaller than the lower experimental limit of $+0.06$.

Because of the possibility of $M1-E2$ mixtures, the directional correlation data are of no aid in determining which of the three remaining spin assignments is the correct one. The 3/2–5/2–7/2 assignment deduced from the conversion data is among the spin combinations compatible with the correlation data.

Xe^{131}

The radiations accompanying the decay of I^{131} have been studied by a large number of investigators. The recent paper by the Canadian group[5] gives the most complete account of this important isotope.

Although it had been shown in 1948 by coincidence experiments[6] that the 80-kev gamma-ray was not in coincidence with the 638-kev radiation, several investigators subsequently proposed a 638–80-kev cascade based mainly on uncertain energy considerations. In order to add further evidence for the nonexistance of the 638–80-kev cascade, expecially after the publication of coincidence experiments[7] supporting the opposite viewpoint, we reinvestigated the gamma-gamma coincidences in Xe^{131}. Coincidence absorption experiments showed that at least 90 percent of the gamma-gamma coincidences are due to the 284–80-kev cascade. By means of critical absorption it was shown that most of the remaining coincidences did not involve the 80-kev gamma-ray. The residual 10 percent coincidences

[4] F. R. Metzger, Phys. Rev. 88, 1360 (1952).

[5] R. E. Bell and R. L. Graham, Phys. Rev. 86, 212 (1952).

[6] F. R. Metzger and M. Deutsch, Phys. Rev. 74, 1460 (1948).

[7] Bell, Cassidy, and Kelley, Phys. Rev. 82, 103 (1951). Later retracted.

TABLE I. Au^{198} directional correlation data.

θ	$W(\theta)-1$
60°	-0.177 ± 0.048
90°	0.000 ± 0.045
120°	-0.193 ± 0.030
150°	-0.336 ± 0.040
165°	-0.280 ± 0.051
180°	-0.261 ± 0.041
180°[a]	-0.292 ± 0.057

[a] Additional lead absorber before crystal opposite pulse height selector.

consist partly of bremsstrahl-gamma coincidences and may also include cascades involving other very low intensity gamma-rays such as the 177-kev radiation reported by Cork.[8] The 638–80-kev cascade is therefore excluded.

The results of the directional correlation measurement between the 284-kev and 80-kev gamma-rays indicated a coincidence distribution which is isotropic within an error of 2 percent. This isotropy strongly supports the assignment of spin $\frac{1}{2}$ to the intermediate, 80-kev level, an assignment which was based on the magnetic dipole character of the 80-kev transition, the predictions of shell theory, and the observed trend of the $S_{\frac{1}{2}}$ and $D_{\frac{3}{2}}$ energy levels in this region of N and Z.[9]

Hg^{198}

The principal, 411-kev transition in Hg^{198} (Fig. 4) has been shown by accurate internal conversion measurements[10–12] to be a pure electric quadrupole transition. Since the ground-state spin of Hg^{198} is 0, the spin of the 411-kev level is 2. Recent investigations[10,13] have revealed a 1 percent intensity, 680-kev transition from a 1091-kev level to the 411-kev level, and a $\frac{1}{2}$ percent 1091-kev transition to the ground state. Internal conversion measurements[10] on the two weak gamma-rays show that the ground state and first two excited states have the same parity, that the 1091-kev transition is electric quadrupole, and that the 680-kev gamma-ray is magnetic dipole or an $M1-E2$ mixture. On this evidence Elliott and Wolfson[10] have proposed a 2–2–0 spin assignment to the 1091 kev, 411 kev, and ground levels of Hg^{198}.

The directional correlation measurement of the 1 percent intensity $680-411$-kev cascade is complicated by the presence of coincidences between the bremsstrahlung from the main 970-kev beta-spectrum and the subsequent 411-kev gamma-ray. It was found that these disturbing coincidences, which otherwise cause a large error, could be completely eliminated by means of pulse-height selection. A least square fit of $W(\theta)=1+A_2\cos^2\theta+A_4\cos^4\theta$ to the data in Table I yielded the values, corrected for angular resolution, of $A_2=-1.10$ and $A_4=+0.83$, with an error of approximately 10 percent in each coefficient.

The 1–2–0 and 3–2–0 spin assignments which require negative $\cos^4$ terms independent of the amount of $M1-E2$ mixture present are excluded by the experiment. The 0–2–0 combination is impossible in view of the presence of a cross-over transition; the 4–2–0 assignment is improbable by the same argument and, moreover, does not fit the measured correlation.

The 2–2–0 assignment fits the data for a multipole mixture of 50 percent to 70 percent $E2$ and 50 percent to 30 percent $M1$, zero phase (as defined by Ling and Falkoff[14]). The correlations obtained from these limiting mixtures are shown by the curves in Fig. 5. The measured value of 3×10^{-11} second for the half-life of the intermediate, 411-kev level[15,16] and the use of metallic gold for the source make it unlikely that the measurement was influenced by reorientation of the nucleus while in the intermediate state.[17] Even if such an effect were present, it would not change the signs of the coefficients in the correlation function, and the 2–2–0 combination would still be assigned.

Although the experimentally determined admixture of $E2$ is over a hundred times larger than the 0.2 percent expected from the Weisskopf formulation,[18] this case of Hg^{198} is no exception. In the 2–2–0 cascade of Te^{122} the 2–2 transition is 80 percent E_2,[19] and in the 2–2–0 cascade of Pt^{196} which differs from Hg^{198} by two protons, the 2–2 transition is approximately 95 percent E_2.[20]

The example of Hg^{198} shows the much more unambiguous character of directional correlations with even-even nuclei, in contrast with similar measurements on even-odd nuclei such as Co^{59}. This is due to the zero-spin ground state which requires gamma-transitions to it to consist of pure multipole radiation, thereby allowing only a few combinations for the spins of the excited states. In addition, the correlations for these different spin combinations vary greatly and are easily distinguishable.

[8] Cork, Rutledge, Stoddard, Branyan, and Childs, Phys. Rev. **81**, 482 (1951).
[9] R. D. Hill, Phys. Rev. **80**, 906 (1950).
[10] L. G. Elliott and J. L. Wolfson, Phys. Rev. **82**, 333 (1951).
[11] L. Simons, Phys. Rev. **86**, 570 (1952).
[12] P. Hubert, Compt. rend. **232**, 2201 (1951).
[13] P. E. Cavanagh, Phys. Rev. **82**, 791 (1951).
[14] D. S. Ling and D. L. Falkoff, Phys. Rev. **76**, 1639 (1949).
[15] L. R. Graham and R. E. Bell, Phys. Rev. **84**, 380 (1951).
[16] P. B. Moon, Proc. Phys. Soc. (London) **A64**, 76 (1951).
[17] G. Goertzel, Phys. Rev. **70**, 897 (1946).
[18] V. Weisskopf, Phys. Rev. **83**, 1073 (1951).
[19] M. Glaubman and F. R. Metzger, Phys. Rev. **87**, 203 (1952).
[20] R. M. Steffen, Phys. Rev. **89**, 903 (1953).