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AN EXPERIMENTAL INVESTIGATION OF THE BETA-GAMMA ANGULAR CORRELATION IN BETA DECAY

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BY
RICHARD L. GARWIN

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ABSTRACT

The beta-gamma angular correlation was investigated in the isotopes Na^{24}, Co^{60}, Ru^{103}, Ru^{105}, Cd^{115}, Ir^{192}, and Au^{198}. Measurements were made at ten angles between 45° to 180°. No departure from spherical symmetry was found in any case, within the statistical error (standard deviation 1% or 2% in most cases). Tests and arguments are set forth to establish that the lack of correlation exists in fact at the atom and probably at the nucleus itself. The discrepancy between the results of this experiment and the predictions of the beta ray theory is discussed.

INTRODUCTION

The present Beta-ray theory in all its forms predicts a correlation between the direction of emission of a beta ray and that of the gamma ray following the beta transition to an excited state. It was decided to undertake an experimental investigation of this angular correlation. This experiment may be expected[1,2] to yield information about the multipole order of the gamma ray and about the angular momentum of the emitted beta ray and the nuclear spins involved. It gives, in addition, information concerning the type of interaction operating in beta decay.

Similar experiments[3,4,5] have been carried out on the angular correlation between two cascade gamma rays, in which case the experimental results agree quantitatively with the theoretical predictions. The gamma-gamma experiment does not suffer from the necessity of utilizing thin sources, etc., which makes the beta-gamma investigation difficult.

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[1] D. L. Falkoff, Ph. D. Thesis, University of Michigan (1948).

[2] C. N. Yang, Phys. Rev. 74, 764 (1948).

[3] L. Brady and M. Deutsch, Phys. Rev. 72, 870 (1947).

[4] L. Brady and M. Deutsch, Phys. Rev. 74, 1541 (1943).

[5] M. Deutsch and F. Metzger, Phys. Rev. 74, 1542 (1943).

SUMMARY OF THEORETICAL PREDICTIONS

The qualitative results of the theory regarding beta-gamma angular correlation are simply summarized:[1,2] If the relative number of beta-gamma coincidences as a function of the angle θ between the directions of propagation of the beta and gamma rays is expressed as a power series in $\cos\theta$, the theory states that only even powers of $\cos\theta$ will appear. The highest power of $\cos^2\theta$ appearing is then the smallest of the three integers designating (a) the degree of forbiddenness of the beta decay, that is, 0 for permitted, 1 for first forbidden, etc.... (b) the spin of the intermediate state, (c) and the exponent of 2 in the multipole order of the gamma ray. (If the spin of the intermediate state is half-integral, one is to take J-1/2 rather than J.) It is to be noted that the theory does not distinguish between a magnetic dipole or, in general, multipoles. As a corollary of the general statement, the selection rules for obtaining a spherical angular correlation may be stated: If the beta transition is permitted, or if the intermediate state has spin 0 or 1/2, no angle-dependent term will appear in the correlation function. Since most beta transitions with which one is concerned are probably first forbidden, one expects in the majority of cases to obtain a correlation function of the form $1 + b\cos^2\theta$, where be may be identically 0. It should

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[1,2] See page 8.

be mentioned that weakly allowed beta transitions may still show a small correlation because of the non-permitted matrix elements.

All computations for the correlation have been made on the basis of zero nuclear charge. The effect of non-zero Z has been computed in first approximation and found not to change the qualitative results of the theory, nor to produce any large effect on the magnitude of the correlation.

Experimental

The general features of the experiment and apparatus are as follows: one needs a source of coincident betas and gammas, and two detectors movable in angle with respect to each other. In general, spurious angular correlations must be avoided as rigorously as possible. For this reason the beta counter is fixed relative to the source, while the gamma counter may rotate about the source in a plane, thus taking advantage of the relatively greater penetrating power of the gamma rays to maintain all possible effective symmetry. Since beta rays are easily scattered it is necessary, too, to use thin sources and to evacuate the path between source and beta counter in order not to lose correlation by scattering of the betas. Care must be taken to avoid the scattering of electrons or gamma rays from one counter to the other as this may introduce an effect much larger than that sought.

The experimental setup is indicated in Figure 1. The sources are deposited on organic films a few micrograms/cm^2 thick and the tank is evacuated to less than 1 mm Hg to reduce the beta scattering. The tank is made of black fiber 3 mm thick to obtain

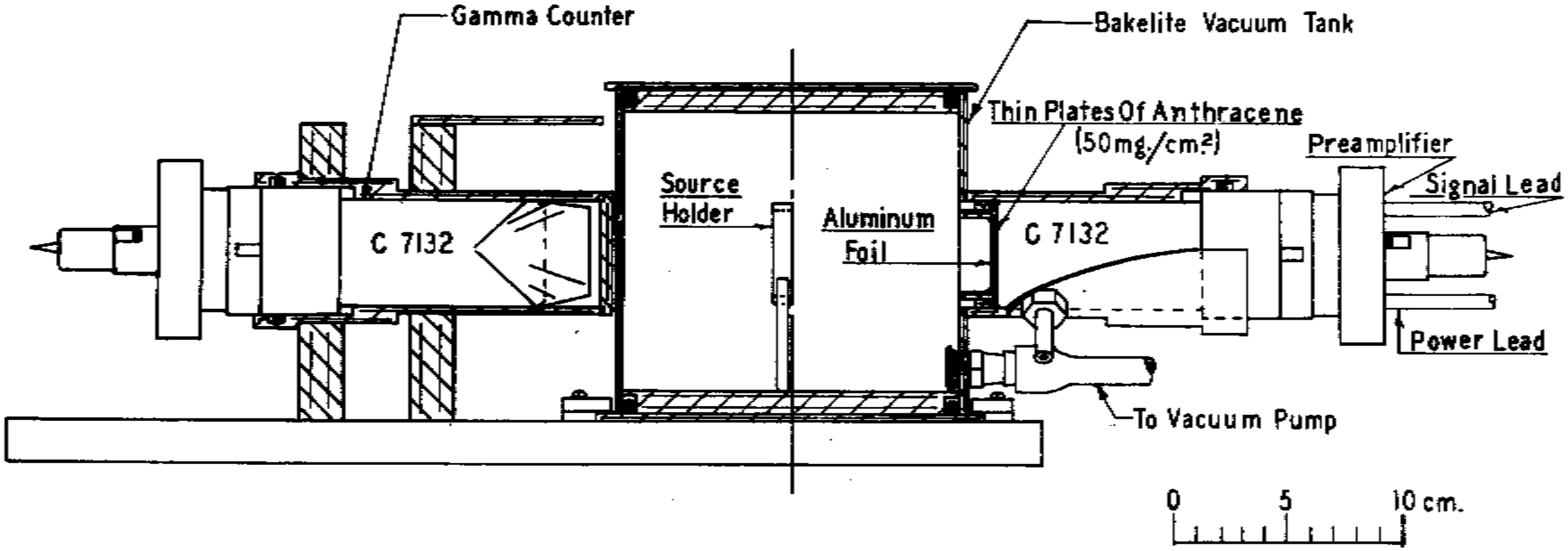

Fig. 1.--- Beta-gamma angular correlation setup.

a low Z material. Vacuum and light seals are made by the O-rings indicated. The tank was lined with paper further to reduce the back-scattering and to avoid possible contamination from broken sources. The connection to the rubber tube leading to the vacuum pump was packed with glass wool contained by soldered pierced disks, effectively removing the danger of breaking sources on letting air into the tank. The number of beta rays, originally not directed into the beta counter, which are scattered from the tank wall into the beta counter is determined by interposing between the source and the beta counter an obstacle near the source. The ratio of the resulting beta counting rate to that with the betas unblocked is a measure of the loss of correlation due to scattering. This was less than 5% for all substances studied. Such an effect independent of the angle between beta and gamma counters serves only to reduce the angle dependent term in the observed correlation function by 5%. This correction has been applied. The method adopted to avoid the double detection of gamma rays or electrons is to make the gamma counter totally insensitive to electrons by surrounding it with 2 g/cm^2 of low Z absorber, while the beta counter is rendered a poor gamma ray detector by making it very thin, as discussed below.

Detectors

Specifically, the gamma detector is an aluminum foil-backed, clear anthracene crystal 1 cm thick, mounted immediately before an RCA 5319 (C7132) photomultiplier, yielding an efficiency of 10 to 15 per cent for the usual gamma rays. It has been established that counting efficiency is within 10% of that corresponding to the Compton cross-section of the crystal. The beta counter consists of a mosaic of clear anthracene cleavings about 50 mg/cm^2 thick with sensitivity 100% for electrons, but only 1/2% for most gamma rays, thus reducing the contribution from double-detected gamma rays to less than 1% of the real beta-gamma effect. The error due to double-detection arises from the Compton scattering of gammas on detection and is readily seen to be of order $\frac{eff}{eff}$ where efficiencies refer to the beta counter. The residual 1% effect was compensated (together with 1% of gamma-gamma coincidences in some cases) by blocking all betas at the source and subtracting the resulting coincidence rate from that with the betas unblocked. No nuclear bremstrahlung gamma rays were detected with the sensitivity used.

The maximum source strength is determined only by the resolving time used. It is found by experience that a casual coincidence rate induced by finite resolving time is inconvenient if higher than about 10% of the real coincidence rate. A simple analysis then yields $N = \frac{1}{20\,\underline{t}}$ so that for the lowest easily attainable $\underline{t}$ (about 5×10^{-8} sec), the source strength N must be less than 10^6 disintegrations per second, i.e., about 25 microcuries, independent of counter efficiency or solid angle. If not all the electrons are accompanied by coincident gammas, N is still the number of electrons per second, but the coincidence counting rate is reduced by the branching.

The coincidence counting rate is then the product of the fractional solid angles of the two detectors, their efficiencies, and the number of disintegrations per second.

The solid angle subtended by the detectors at the source is limited by the complexity of the angular dependence investigated: for two detectors of equal solid angle, and for third order correlations (co6θ), each counter must subtend less than 0.12 steradian if a reduction of a factor 2 in the measured value of the coefficient of the cos6θ term is desired. The aperture of the beta ray counter was determined in three ways: geometrically, by comparison with a thin mica window counter, and internally by the ratio of beta-gamma coincidences to gamma ray counts. The solid angle subtended by the beta counter is 0.07 steradian, that by the gamma counter 0.13 steradian. Coulomb scattering in the source broadens the aperture effective for angular resolution in coincidence counting, but this effect may be made small compared to the geometrical aperture of the detectors. The source thickness has been chosen in all cases to broaden this aperture by less than 50% over the entire beta spectrum. In order to achieve this end, the sources are mounted on thin organic films less than 0.01 mg/cm^2 thick, and the allowable weight of active deposit calculated from the permissible small angle scattering. In the case of gold films, for instance, the scattering was completely negligible, as the metal deposit was less than 0.002 mg/cm^2 in weight. The geometrical size of the source (1.8 cm diameter) introduces only negligible aperture and second order effects.

Sources

All the sources were made by pile irradiation[6] of the natural isotope mixtures. In three cases where the equilibrium specific activity in the neutron flux available at the edge of the pile is sufficiently high to yield good statistical accuracy in a reasonable measuring time (five hours), some sources were prepared by irradiating the inactive material deposited on foils held on aluminum rings. The foils were then transferred to the plastic rings 3.8 cm diameter used in the apparatus. This scheme reduces the quantity of radioactive material to be handled and ensures uniformity density of active material, achieved by depositing the material by evaporation in vacuum from a crucible or from wire of the substance to be deposited. Gold sources 0.002 mg/cm^2 were readily formed by the evaporation of 1 gm of NaF from an alundum-coated tungsten crucible. In the case of Ir^{192} (63-day activity), the metal was deposited as the trichloride in the form of many discrete drops of water solution.

In the course of this investigation, a method was developed to prepare moderately large sources of uniform thickness. It was found that the insulin treatment was not sufficiently good, so that the following scheme was used: about one mg of detergent was added to each ml of solution to be deposited. A small drop (0.01 ml) was then deposited on the lacquer film from a pipette and the film warmed under an incandescent lamp for a moment until

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[6] I am grateful to the staff of the Argonne National Laboratory for their cooperation in performing the irradiation. The work on Ru^{103} and Ru^{105} was done at the Argonne Laboratory.

the drop had spread uniformly to a thin layer of the desired diameter.

The film and holder were then transferred to a bell jar evacuated by a mechanical pump and an infra-red lamp focussed on the film through the plate glass base plate. The solution freezes in a minute or so, trapping the dissolved matter since the freezing takes place over an interval of about a tenth of a second. Evaporation of water then takes place from the frozen solution. Sources may be prepared in this way in twenty minutes overall. It is necessary that there can be no constrictions in the line from the bell jar to the pump, a fixed glass capillary serving to allow atmospheric air to enter after the drying is complete. It was found that synthetic resin films (LC 600) were much less delicate chemically, although not as strong as Zapon or nylon. Aqua regia may be evaporated to dryness in vacuo on the LC 600 films.

In all cases, the sources were thin and uniform enough to make it impossible to attribute any lack of correlation to excessive source thickness. It was found necessary to preserve most prepared sources in a desiccator since the deliquescence of the active salts introduced non-uniformity of deposit by condensation of moisture from the humid atmosphere.

The particular isotopes studied were chosen for the relative simplicity of their decay schemes and their convenient half-lives.

Electronics

The electrical circuits are schematized in Figure 2. In order to obtain large gamma ray efficiency, a scintillation

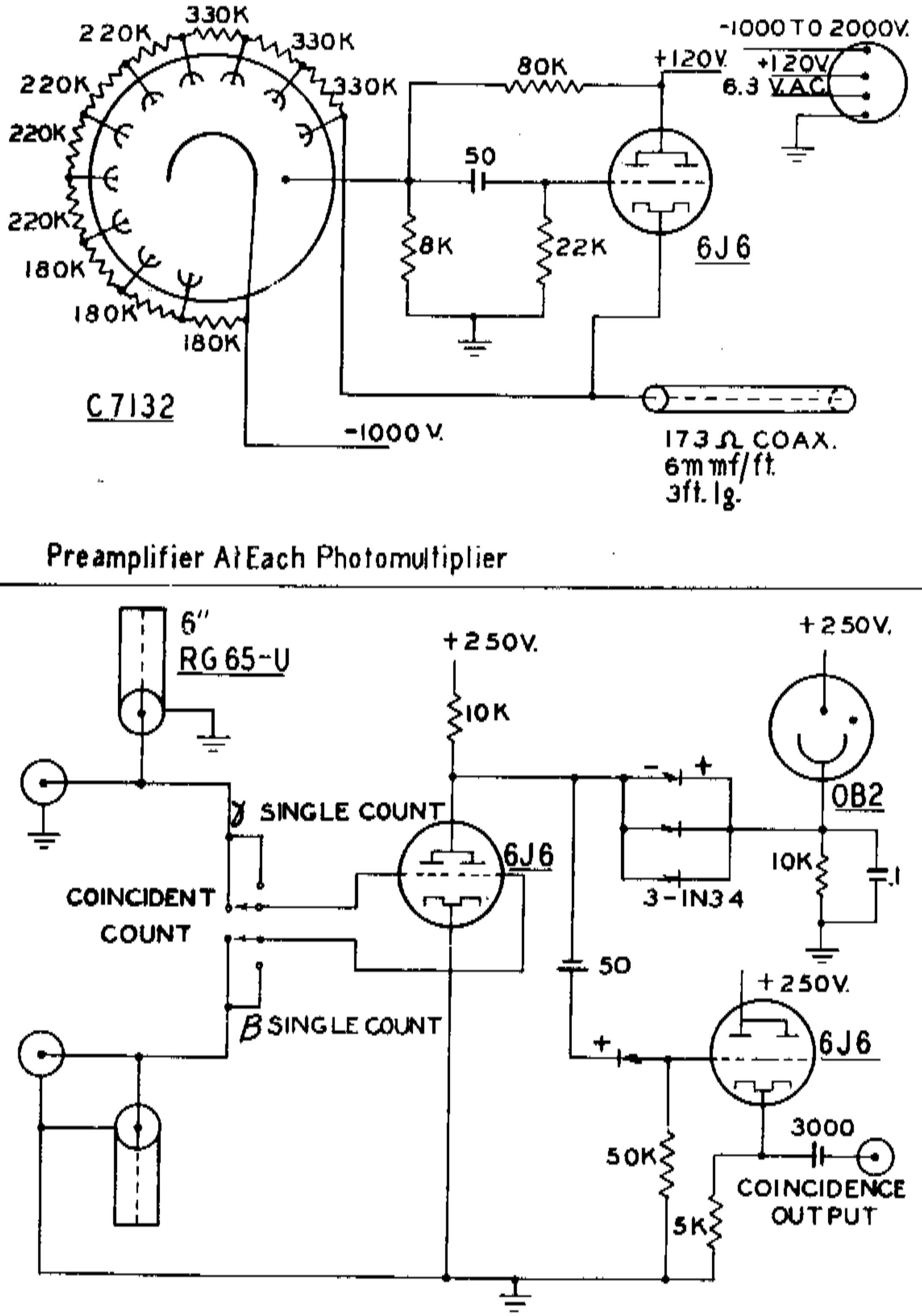

Fig. 2.--- Coincidence circuit.

counter was used, and to allow reasonable counting rates (200/min) fairly low coincidence times were necessary. It is always desirable to reduce the resolving time to a point where the uncertainty in the resolving time may be neglected; i.e., so that the casual rate is less than about 10% of the real rate. This was achieved in the present case with a resolving time of 0.04 microseconds determined by separate excitation of the beta and gamma counters with the betas and gammas of the activity studied. The coincidence circuit was constructed for simplicity and proved sufficiently stable. The coincidence circuit proper is noteworthy, since it is capable of resolving times as low as a few billionths of a second. This short resolving time is achieved with negative input pulses of amplitude 3 volts, particularly convenient for operation directly from the anode of photomultiplier tubes. Here the RCA photomultipliers are operated far above their normal operating range (1900 rather than 1250 volts) with some detriment to signal-to-noise ratio. At present, line-type amplifiers with bandwidths up to 150 or 200 mc/sec are available and will be used in further experiments.

The coincidence circuit requires about 15 ma at 230 v. These characteristics are obtained simply by clamping the plates of a Rossi pair above the potential to which the plates would rise were one tube cut off. Hence, with a low impedance clamp (3-IN34 in parallel), no pulse larger than 1 volt is transmitted to the pulse stretcher and scaler on driving one grid negative. With even 0.01 microsecond coincident pulses, it is readily seen that positive output pulses of amplitude 10 volts are obtained. The input pulse terminating networks here consist of shorted delay line used in a rather inefficient manner. It was determined that no real coincidences were lost and that a mean resolving time of 0.04 microseconds was available.

By using only 10 volts on the anode of the photomultipliers, the spurious multiplier pulses are reduced in amplitude for the following reasons: Since these noise pulses are sharp bursts of electrons (about 10^{-9} sec) and are integrated by the anode circuit, the peak current in a noise pulse must be ten times higher than that in an anthracene signal pulse to have the same integrated voltage amplitude. By operating at low anode voltage, space charge saturation is reached on the noise pulses and not on the signal, improving the action of the phototube.

Measurement

The schedule of measuring is as follows: Twenty angles at 15° intervals are used in order to have data for determining several coefficients of different powers of θ. A gamma single count is taken at each point to determine the shadow effect of the source holder and the increase in counting rate due to scattering from the beta counter housing. All coincidence counts are corrected by dividing by the corresponding gamma single count, thus compensating for source assymetries. Beta-gamma coincidence counts are then taken in rotation for one minute at each point, a total of five or six minutes being spent at each point in all. This method reduces the effect of long time drifts and sporadic disturbances. At least 500, and usually 1000, counts are taken at each point.

the results being corrected for decay of the source, single gamma count, and gamma-gamma coincidences where more than one gamma ray is involved. To these data are fitted, by least squares, curves of the theoretical form. In cases where the effect appeared very small, only the $\cos^2\theta$ term was fitted.

The statistical error obtainable in the amplitude of a $\cos^2\theta$ term by the least squares process is 140% of that which could be set were the same number of counts taken at 90° and 180° if the curve were known actually to be of the form $1 + \cos^2\theta$. The situation becomes much worse if the least squares process is extended to include the fitting of a $\cos^4\theta$ also, the statistical uncertainty in the angle dependent terms being 3.5% for 10^5 counts as compared with 0.9% for 10^5 counts if only the $\cos^2\theta$ term be fitted. For these reasons in several cases a long counting schedule was performed at 90° and 180° to obtain better statistical accuracy. The results are plotted in Figure 3; values of b from counts at 90° and 180° are in brackets.

Results and Conclusions

The general experimental results may be summarized briefly. In the seven cases studied and described in detail below, no angle dependent correlation has been observed outside the statistical error, which is for the most part 1% or 2%.

Au^{198} (0.96 MEV, 2.7d, $\log_{10} ft$ = 7.4; 0.41 MEV)[7] No correlation was found to within 1%. Here the spin of the final state

[7] Energies and lifetimes are taken from Seaborg and Perlman, Rev. Mod. Phys. 20, 535 (1948), unless otherwise stated. The product ft is used in the same sense as by Knopinski[10] where t is the half-life and f a function of maximum beta kinetic energy and nuclear charge.

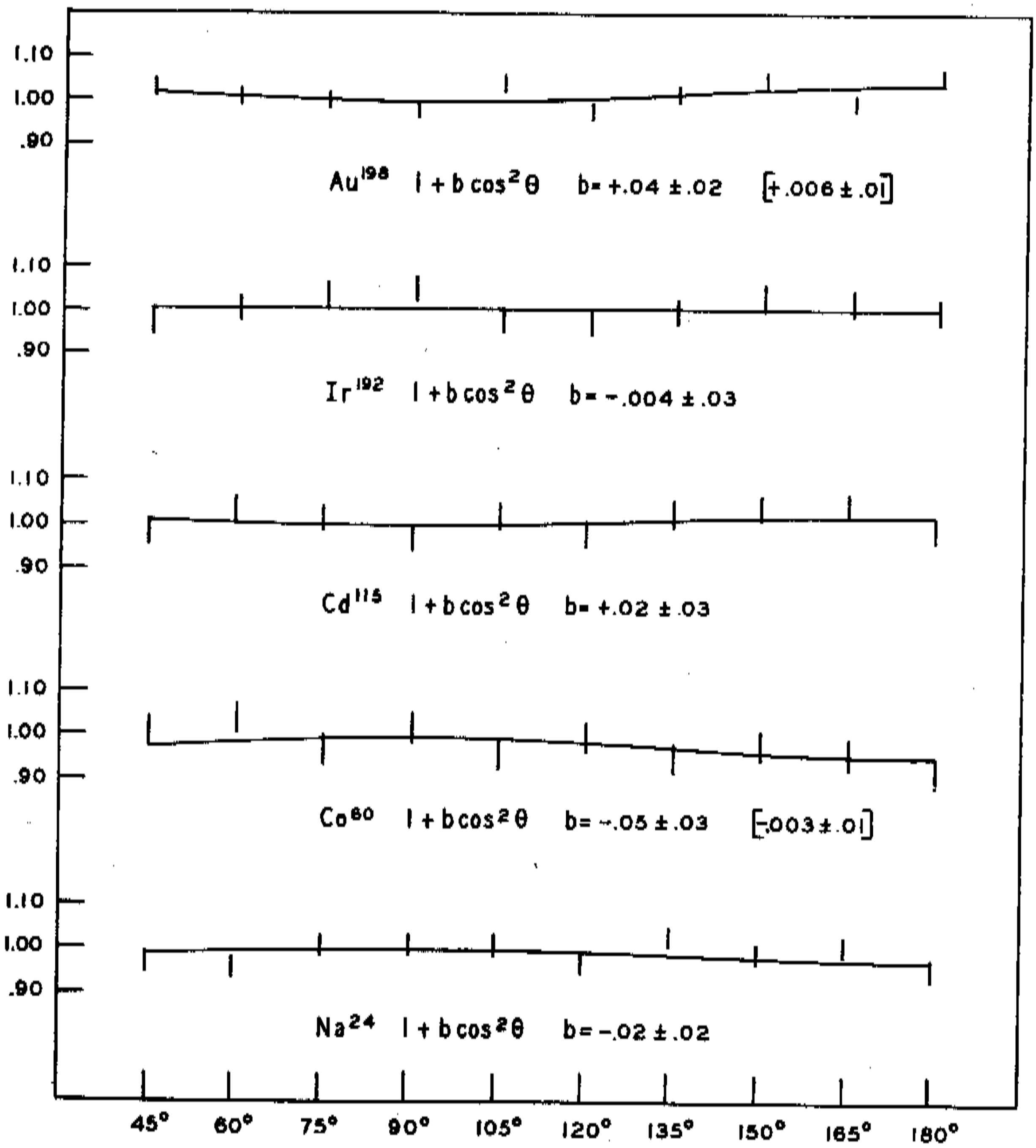

Fig. 3.--- Relative beta-gamma coincidence rate vs. angle.

(Hg^{198}, an even-even nucleus) is surely zero. The 0.41 MEV gamma has a lifetime of 0.023 microseconds[8], which probably implies electric octupole radiation. Such an assumption is consistent also with the measured conversion coefficients.[9] The spin of the intermediate state is thus 3. As pointed out in the introduction, the only other fundamental way of obtaining spherical angular correlation is to postulate an allowed beta transition. The high ft value for the beta leads Konopinski[10] to classify the beta as second forbidden, although a first forbidden assignment seems not excluded. Either of these choices would produce a measurable correlation on the basis of the theory. As an example, a first forbidden transition from spin 4 would yield b=0.2 on a non-relativistic scalar theory, without nuclear charge. The contradiction here is not as forceful as it might be because of the long gamma lifetime, a point which will be discussed in a further section, but the result adds statistical weight to the evidence here accumulated.

<u>Co^{60}</u> (0.3 MEV, 5.3y, log <u>ft</u> = 7.5; 1.1, 1.3 MEV) No correlation was observed to within the statistical error of 1%. Here gamma-gamma angular correlation measurements of the cascade gammas assign spins 0,2,4 to the ground and excited states of the Ni nucleus.[11] The high <u>ft</u> value for the beta leads to its classification according to Konopinski's criteria as second or possibly

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[8] W. J. MacIntyre, Phys. Rev. <u>76</u>, 312 (1949).

[9] D. Saxon and R. Heller, Phys. Rev. <u>76</u>, 909 (1949).

[10] E. J. Konopinski, Rev. Mod. Phys. <u>15</u>, 209 (1943).

[11] ~~E. Brady~~ and M. Deutsch, Phys. Rev. <u>74</u>, 1541 (1943).

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first forbidden. This, too, should then yield a measurable correlation. In this element and the following a small measured gamma-gamma effect was subtracted.

Na^{24} (1.4 MEV, 14.8 hr, log ft = 6.1; 1.4, 2.8 MEV) No correlation was observed to within the statistical error of 2%. Here the same gamma-gamma correlation experiments give spins 0, 2, 4 for the Mg^{24} nucleus and excited states. Konopinski lists the beta ray as first forbidden. A $\cos^2\Theta$ correlation should then be apparent, although none was apparent. Four other cases were studied, for which the spins are uncertain. In all cases no correlation was found.

Ru^{103} (0.3 MEV(95%) log ft = 6.4, 0.8 MEV, log ft = 8.9, 42d; 0.56 MEV) No correlation larger than 2% was observed. Here the ft value of the beta places it as first forbidden according to Konopinski's criterion. The spins are unknown, but it should be emphasized that according to the theory, spin 1/2 must be assumed for the intermediate state to yield no correlation.

Ru^{105} (1.4 MEV, 4.5 hr, log ft = 5.9; 0.7 MEV) Correlation less than 1%. This activity presented the only experimental difficulty encountered. The first run made on Ru^{105} showed a -9.0% $\cos^2\Theta$ term accompanied by a noticeable 5% dip in coincidence rate between 165 and 180°. Four additional runs have confirmed neither of these effects, yielding as the average of these last four runs -0.2% ± 1% for the $\cos^2\Theta$ amplitude. The first apparent correlation should probably be assigned to a malfunctioning of the equipment as all experiments designed to confirm it have failed. It was suspected that a small admixture of positron emitting impurity would produce the effect observed, but a test on Cu^{64} indicates the effect of positrons is to introduce high and sharp peak near 0°. This beta ray should also be first forbidden according to Konopinski. Thus to have no angle dependent term the theory forces spin 1/2 on the intermediate state.

<u>Cd^{115}</u> (0.6 MEV, log <u>ft</u> = 6.2, 1.1 MEV, log <u>ft</u> = 7.2, 2.3d; 0.6 MEV) Correlation less than 3%. The beta ray should be first forbidden from its <u>ft</u> value. The intermediate state must then have spin 1/2 to conform with the theory.

<u>Ir^{192}</u> (0.7 MEV, 70 hr, log <u>ft</u> = 7.9; 0.3, 0.5, 0.6 MEV) Correlation less than 3%. The high <u>ft</u> value leads to the classification of the beta ray as second forbidden, following Konopinski. The absence of correlation then implies spin 0 in the intermediate state.

In an attempt to clarify the contradiction, the decay schemes of Figure 4 were postulated, based naively on the results of this experiment. It was hoped that there would be sufficient rigidity in the existing spin data to bring a clear discrepancy between the demands of the beta ray theory and the results of this experiment. It was found, however, that the spin assignments are sufficiently flexible so that no real contradiction can be established in the cases studied, except for the one inherent in the experimental results; that in seven cases of forbidden beta decay no correlation was observed.[12] In three cases sufficient data are available (Au^{198}, Co^{60}, Na^{24}) to predict definitely the existence

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[12] In addition to the work reported here on the beta-gamma problem, M. L. Wiedenbeck and K. Y. Chu inform me (letter) that hey have found no correlation to 3% in Co^{60}, Na^{24}, Au^{198}, K^{42},

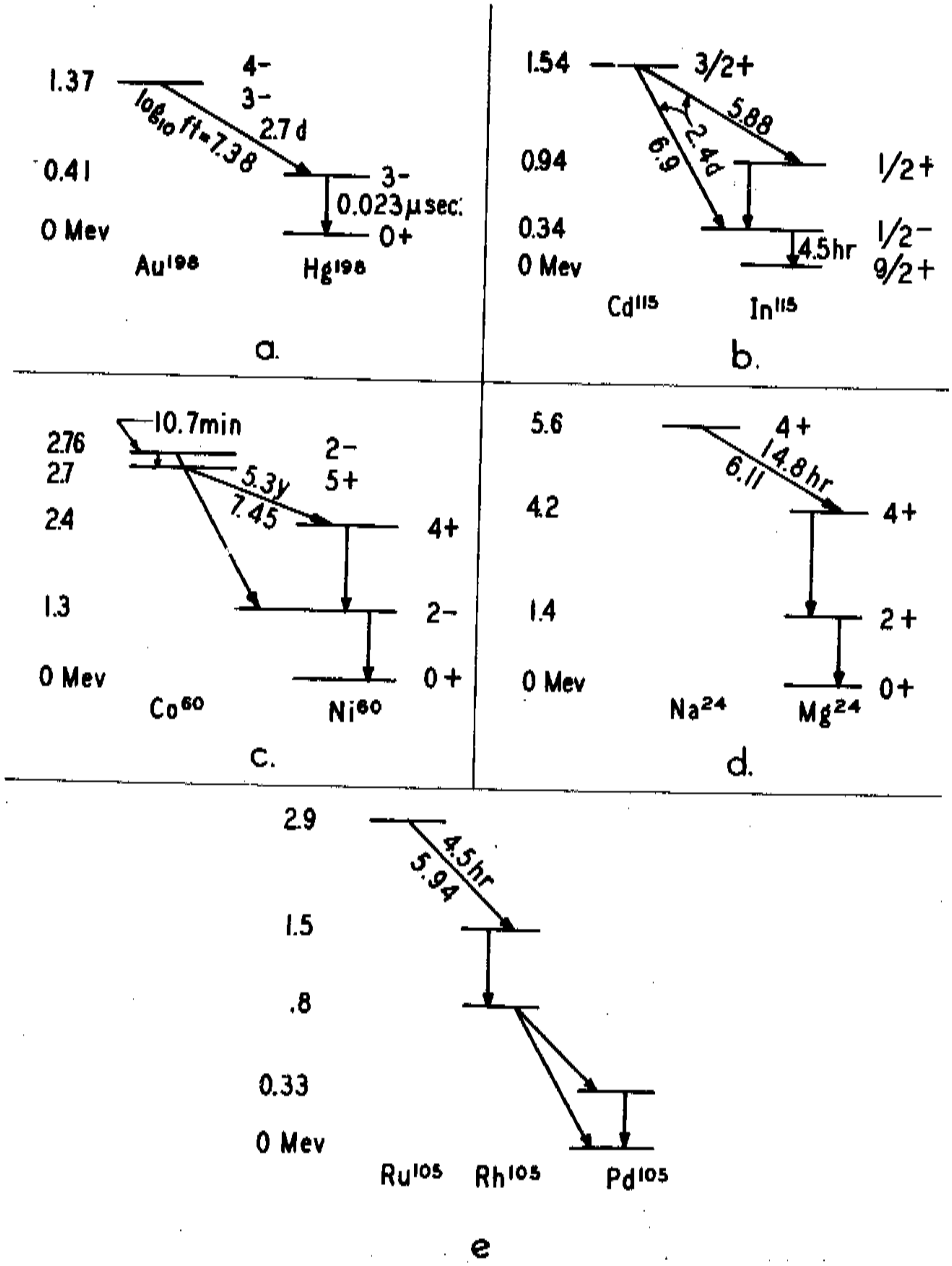

Fig. 4.--- Derived level schemes.

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of a correlation and to establish a clear discrepancy. It is extremely improbable that the lack of correlation in the other four cases should be due to chance in the selection of the spin of the intermediate state.

Discussion

Since neither the nuclear spins nor the degree of forbiddenness are known with absolute certainty in any single case, it is of some interest to calculate roughly the probability of the observed lack of correlation being due to the operation of one or the other of the selection rules stated in the theoretical summary. If one assigns to Na^{24} probability 1/2 for the beta being allowed, to Au and Co probabilities 1/3 each, and to cases in which the nuclear spin is unknown probability 1/2 for it to be zero, then the probability of these results being due to chance operation of the selection rules is of the order of 1%. It is thus necessary to look elsewhere for the solution.

A certain class of objections is answered by the fact that many gamma-gamma correlations have been observed. This problem has many elements in common with that of the beta-gamma angular correlation. For instance it cannot be supposed that the spin direction of the intermediate nucleus is lost be precession induced by the neighboring atoms of the material, or by the recoil since the recoil energies are entirely similar to those occurring in the gamma-gamma case. The gamma-gamma experiments show no sensitivity to the state of condensation of the source material.

Cs^{134}, and I^{137}. Also Na^{24} and Co^{60} by M. A. Grace, R. A. Allen, H. Halban, Nature <u>164</u>, 539 (1949).

The conclusion seems inescapable that the lack of correlation is present already at the atom in question. From the interactions of beta and gamma rays with electrons, it is known that the directions of propagation of the betas and gammas are not altered by passage through the electronic charge cloud. The only effects which prevent our supposing the lack of correlation to exist at the nucleus are concerned with the re-orientation of the spin of the intermediate state. It is estimated from the usual gyromagnetic ratios that a magnetic field at the nucleus of 20,000 gauss lasting for 0.01 microsecond would induce sufficient procession to diminish significantly the correlation, assuming that there is one from a bare nucleus. Since a magnetic field of this value is produced by a Bohr magneton at 0.8A°, it is not certain that the nuclear spin does not precess in the case of Au^{198}. Å X

The gamma-gamma correlations, however, are not affected by magnetic fields of the order of 10,000 gauss.[13] If one is to postulate a disorienting effect on the nuclear spin of the rearrangement of the electronic charge cloud, one must postulate that this be so fast that it does not interfere with the second gamma ray in a gamma-gamma experiment. Estimates of the rearrangement time than give about 10^{-15} sec. Enormous magnetic fields are needed to disturb the nuclear spin in so short a time. Coupling of the order evidenced in hyperfine structure thus seems unable to produce the observed lack of correlation.

If the lack of correlation is, as seems probable, present at the nucleus, it seems necessary to look for a substantial and

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[13] E. Brady and M. Deutsch, Phys. Rev. 74, 1541 (1943). X

(Repeat footnote # 4)

not obvious revision of the beta ray theory. The direction of this revision might be such as to make all observed beta transitions "permitted," or it might remove the presently predicted correlation between the direction of emission of "forbidden" electrons and the spin of the residual nucleus.

Acknowledgments

I should like to thank Professor E. Fermi and Dr. C. N. Yang for their very helpful discussions of this problem.