The Application of Proportional Scintillation γ-Ray Counters to the Determination of the Decay Scheme of Au^{198}

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(Received November 14, 1950)

Besides the main γ-ray at 0.411 Mev, Au^{198} also emits two higher energy γ-rays of energy 0.67 and 1.09 Mev, present to the extent of only 1.4 and 0.4 percent, respectively. Using large clear blocks of naphthalene +1 percent anthracene as crude γ-ray separators, it has been established, by coincidence absorption measurements, that the high energy γ-rays are in coincidence with β-rays of maximum energy 295 kev. Moreover, it has been established that γ–γ-coincidences also occur, and that the 0.67-Mev γ-ray is in cascade with the 0.411-Mev γ-ray. The 1.09-Mev γ-ray is not in coincidence with either of the others. By comparing the γ–γ-coincidence rate and the β–γ-coincidence rate taken under appropriate conditions, it was possible to determine branching ratios for the 0.67- and 1.09-Mev γ-rays. These are (1.43 ± 0.10) percent, (0.33 ± 0.07) percent, respectively, these results being in excellent agreement with those derived from spectrometer measurements. The β–γ- and γ–γ-coincidence measurements establish that Hg^{198} has energy levels at 0.411 and 1.09 Mev.

INTRODUCTION

MANY workers have examined Au^{198} with rather conflicting results, especially in regard to the γ-rays emitted. A detailed study of the secondary electron spectrum has been made in this laboratory[1] using sources as large as 0.3 curie; and in addition to the main γ-ray of 0.411 Mev, evidence was found in the Compton and photoelectric spectra for γ-rays of energy 0.67 Mev and 1.09 Mev present to the extent of 1.4 and 0.4 percent, respectively, of the main branch. No evidence of any γ-rays of energy less than 0.411 Mev was found, in agreement with Siegbahn.[2] There is a possibility of the occurrence of a second mode of decay by K-capture to Pt^{198}, though this branch could not be greater than about 5 percent of the main branch by β-decay. There is consequently no *a priori* reason for assigning the higher energy γ-rays to the $\bar{\beta}$-decay branch, though the approximate equality of 0.41+0.67 and 1.09 Mev makes it rather tempting. The object of the present work was to establish if there were β-rays in cascade with the higher energy γ-rays and if so, of what maximum energy. It having been established that these results were consistent with a level scheme with energy levels at 0.41 and 1.09 Mev, it remained to show that the 0.67- and 0.41-Mev γ-rays are in cascade and to obtain some estimate of the branching ratio for the 0.67- and 1.09-Mev γ-rays.

γ-RAY COUNTERS

Since the three γ-rays are well spaced in energy, it was possible to use a proportional scintillation γ-ray counter to separate the higher energy γ-rays from the much more intense low energy γ-rays, in the same way as was done previously for the γ-rays of I^{131}.[3] A similar use of a scintillation counter as a crude γ-ray spectrometer had previously been reported by Bell,[4] but where he used pure anthracene, in this case large clear blocks of naphthalene plus 1 percent anthracene were used. These were grown from the melt by Mr. R. F. Jackson, Jr., of this laboratory, who has developed the technique to such a point that it is possible to obtain blocks of optical clarity and uniformity, completely free from any cracks or faults, several inches in diameter and up to two inches thick. These were cut to cylinders of one and three quarter inches in diameter and sealed directly onto an E.M.I. 5032 multiplier. This was followed by a head amplifier of small gain with an input time constant usually of 1 μsec, and a

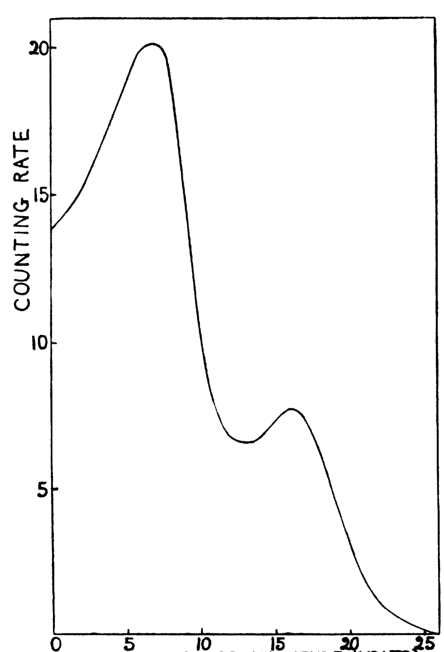

FIG. 1. Distribution of pulses from a well-collimated source of Na^{24} γ-rays showing the resolution of the 1.4- and 2.8-Mev γ-rays.

[1] Cavanagh, Turner, Booker, and Dunster, Proc. Phys. Soc. (London) **A64**, 13 (1951).
[2] K. Siegbahn and A. Hedgran, Phys. Rev. **75**, 523 (1949).
[3] P. E. Cavanagh, Nature **165**, 889 (1950).
[4] W. H. Jordan and P. R. Bell, Nucleonics **5**, No. 4, 30 (1949).

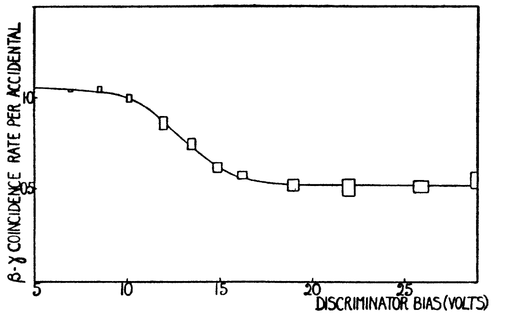

FIG. 2. Bias curve of the ratio of genuine β–γ-coincidences to accidental coincidences for Au^{198} with $\sim$8 mg/cm^2 Al β-counter window thickness.

1049A 2 Mc/sec pulse amplifier. The crystals were covered with a matte white reflector of magnesium oxide. The energy resolution obtained is illustrated in Fig. 1 with a pulse analyzer curve for the γ-rays of Na^{24}. The general form of the pulse distribution and the resolution is similar to that obtained by Bell.

$\beta-\gamma$-COINCIDENCE MEASUREMENTS

It was necessary to bias out completely the pulses from the 0.411-Mev γ-rays, and then to measure coincidences between the higher energy γ-rays and β-rays recorded by a short dead-time counter. Previously, in the case of I^{131}, it was possible to ascertain the point at which the lower energy γ-rays were biased out by means of absorption measurements in good geometry, as a function of the bias. In this case, however, the high energy γ-rays are relatively weaker by a factor of ten, and a correspondingly larger source was required for the absorption measurements. With a source of this magnitude, however, it was found that the higher energy γ-rays were completely obscured by "pile-up" pulses from the low energy γ-rays. Even when an input differentiation time constant of 0.1 μsec was used, the number of large pulses due to pile up was embarrassingly large. Moreover, it was realized that pile-up pulses would occur in the coincidence measurements, though to a lesser extent, and so it was necessary to devise a method to determine at what point all pulses and pile-up pulses from the 0.411-Mev γ-ray were eliminated, with the actual source and geometry to be used for the β–γ-coincidence absorption measurements. It was found this point could be determined by taking a bias curve of β–γ-coincidences.

If the higher energy γ-rays were not in coincidence with β-rays then the β–γ-rate per accidental coincidence would fall to zero at a value of bias such that no 0.41-Mev γ-rays were recorded. There would be precisely the same occurrence if the high energy γ-rays were in coincidence with soft β-rays which were completely absorbed by the β-counter window. Similarly, if the window were sufficiently thick to absorb the low energy β-group differentially with respect to the high energy β-group, say by a factor of two, then the β–γ-coincidence rate per accidental coincidence would fall to half-value as the bias was increased, and level out at that value of bias, such that the effect of the 0.411-Mev γ-rays was eliminated. Such a characteristic of the bias curve of β–γ-coincidences was in fact found.

The β-rays were recorded by means of a Geiger counter operated about 50 volts below the threshold and working into a 1049A pulse amplifier which has very good overload characteristics. The counter window, together with the air gap, was sufficient to reduce the intensity of 300-kev β-rays to about half-value, whereas it reduced that of the 960-kev β-rays by only a few percent. Both amplifiers were fed via discriminators to a coincidence unit of conventional design. Delays between the two channels were matched out using a variable delay unit, and the coincidence unit was used at such a value of resolving time that not only were all genuine coincidences recorded (to within 1 percent) but some margin was left for the small delays introduced by the variable setting of the two discriminators. This value was 0.3 μsec, and it was measured at intervals throughout the experiment and found to remain constant within the statistical accuracy of 1 percent.

For these measurements the source was in the form of an 0.2 mg/cm^2 layer evaporated onto 1 mg/cm^2 distrene sheet. The gold had previously been analyzed and found to contain not more than a few p.p.m. of any impurity. The source was activated in the larger Harwell pile, and a small amount of activity was found to be induced in the distrene. This was allowed to decay before coincidence measurements were started. The two counters were used in a close geometry with respect to the source, and counting rates in excess of 50,000/sec and 5000/sec for the β- and γ-counter, respectively, were recorded, together with the coincidence rate, with type 200 scalers.

The bias curve for β–γ-coincidences is shown in Fig. 2 in the form genuine rate/accidental rate, a small genuine rate arising from Compton scattering γ–γ and cosmic coincidences being subtracted. The experimental points are shown as boxes in which the height is the mean error and the width an estimate of the uncertainty of the overall gain of the γ-ray counting system. This gain usually fluctuated by 2 or 3 percent and sometimes drifted by more than this. The gain variations were followed by a recording ratemeter set at a low value of bias, and fed in parallel from the amplifier. It will be seen from the figure that the curve has substantially flattened out at a pulse height corresponding to 20 volts, and it was at about this value of bias that the β–γ-coincidence absorption measurements were made.

It was found possible to follow the absorption of coincidences to 30 mg/cm^2 aluminum and still retain reasonable accuracy. Beyond this point not only were the counts becoming excessively long, but, also, the genuine rate was rather small compared with the accidental rate and of the same order as the cosmic coincidence rate. It can be proved easily that to a high degree of approximation, the value of $G/N_\beta N_\gamma$ at zero absorber is the same as that at zero bias, and is, in fact, equal to the reciprocal of the source strength. G/N_γ is obtained by multiplying by the β-ray counting rate, corrected for paralysis loss, and extrapolated to zero absorber. This gives a zero point for the coincidence absorption curve, which is shown in Fig. 3. An absorption curve for the β-rays of Co^{60} of maximum energy 0.310 Mev was measured in the same geometry and under the same conditions of backscattering. The energies of the two β-rays were obviously very close, and the application of the Feather method of comparison gives the maximum energy of the β-rays in coincidence with the high energy γ-rays of Au^{198} as 0.295 Mev. This, together with the γ-ray energies of 0.41, 0.67, and 1.09 Mev, and the main β-ray of maximum energy 0.96 Mev, is compatible with a level scheme for Hg^{198} with energy levels at 0.41 and 1.09 Mev. γ-ray transitions occur between these levels and the ground state and also between the two levels. On this basis, one would expect coincidences between the 0.67- and 0.41-Mev γ-rays.

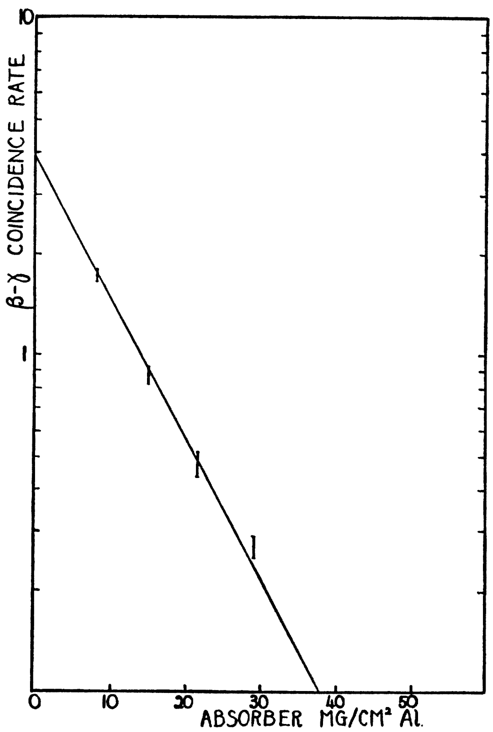

FIG. 3. Coincidence absorption curve of the low energy β-group in Au^{198}.

THE INVESTIGATION OF $\gamma-\gamma$-COINCIDENCES

Since the 0.67-Mev γ-ray is present only in a 1.4 percent branch, it was anticipated that it would only be possible to demonstrate in a qualitative manner that coincidences occur between this and the 0.41-Mev γ-ray. Two proportional scintillation counters were used in this work. One, which was normally used at high gain, so that the great majority of pulses arising from γ-rays were recorded, had an input time constant of 1 μsec; the other, with which a variable bias level was used, had an input time constant of 0.1 μsec to minimize "pile-up." In view of the expected small genuine coincidence rate, it was necessary to ensure that no spurious coincidences occurred owing to scattering processes. The two crystals were mounted facing each other with a lead absorber placed in between and a Na^{24} source to one side such that each counter had an unrestricted view of the source but was entirely obscured from the other by lead. The ratio of the genuine coincidence rate to the product of the two single channel rates was then measured as a function of the thickness of the lead. It fell by about 10 percent in the first 10 g/cm^2 and thereafter remained constant for increasing thickness of absorber. This geometry with a lead separator of 10 g/cm^2 was used for the measurements with Au^{198}. Again for these measurements a coincidence resolving time of 0.3 μsec was used.

Briefly, the method of the experiment was to hold one counter at high gain and low bias so as to record most of the γ-rays, while coincidences were measured as a function of bias on the second counter. At low bias on the latter, most of the low energy γ-rays were recorded, and there were a large number of accidental coincidences arising from 0.411-Mev γ-rays being recorded in both counters. The number of genuine coincidences between 0.411 and 0.67-Mev γ-rays recorded was relatively low. As the bias on the second counter was increased the number of 0.411-Mev γ-rays recorded fell off relative to that of the 0.67-Mev γ-ray; consequently, the number of accidental coincidences fell off relative to the genuine, or, to put it the other way, the genuine/accidental coincidence rate rises with increasing bias. But for the presence of the noncoincident 1.09-Mev γ-ray, this rise would continue until the 0.411-Mev γ-ray was completely biased out. As it is, the curve rises until the effect of the differential biasing out of the 0.67- and 1.09-Mev γ-rays becomes important, when it reaches a maximum, then it decreases and ideally falls to zero at that value of bias such that the 0.67-Mev γ-ray is completely eliminated. In the first experiment, it was possible to follow the curve barely to the maximum, the genuine coincidence rate becoming of the same order as the cosmic rate. At the same time, the single rate recorded by the second counter became comparable with the background rate. Fortunately, a bias curve of background coincidences showed that most were due to very large pulses, and it was possible to eliminate some 80 percent by a crude pulse analyzer technique. Using this it was possible to work at bias values well over the maximum of the curve. This is given in Fig. 4(a) and shows qualitatively very well the characteristics we have described.

A bias curve for the lower energy γ-ray alone, under the same conditions, was obtained by taking $\beta-\gamma$-

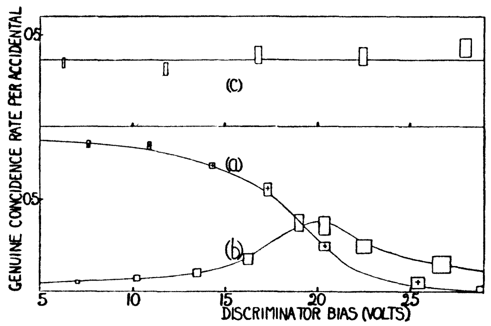

FIG. 4. (a) Bias curve of the ratio of genuine β–γ-coincidences to accidental coincidences for Au^{198}, with sufficient aluminum $\sim$80 mg/cm^2 over the β-counter window to eliminate the low energy β-group. (b) Bias curve of the ratio of genuine γ–γ-coincidences to accidental coincidences for Au^{198}, with the fixed channel set so as to count the great majority of pulses. (c) Similar to (b) except that the fixed channel is now set at a point corresponding to the maximum of curve (b), and the range of bias values is such as to change the single counting rate by $\sim$10.

coincidences between the variable γ-ray counter and a short dead-time β-counter, with a window sufficiently thick to cut off the low energy β-rays in cascade with the higher energy β-rays. Corrections were made for scatter and cosmic coincidences, and the curve obtained, plotted in the form genuine/accidental coincidence rate, is shown in the same figure. As expected, the curve falls off slowly at first, when the counting rate due to the 0.411-Mev γ-ray is much larger than that due to the higher energy γ-rays. As the bias increases, the genuine/accidental rate falls off more quickly, and thereafter approaches zero at that value of the bias such that the 0.411-Mev γ-ray is eliminated. The very gradual approach of the curve to the horizontal axis is almost certainly due to pile-up pulses. Qualitatively, therefore, these results are in agreement with a decay scheme in which the two lower energy γ-rays are in cascade, but not the high energy γ-ray. As further evidence of the coincidences having arisen from the 0.67-Mev and the 0.41-Mev γ-rays, the bias which had hitherto been varied was held constant at about the maximum of the γ–γ-coincidence curve. The bias on the other scintillation counter was then varied. This variation was such that, although the counting rate decreased by a factor of about 10 over all, the counting rate owing to the low energy γ-ray was always very much larger than that owing to the high energy γ-rays. Under these circumstances, if the coincidences are caused by 0.411-Mev γ-rays, the genuine/accidental rate should be approximately independent of bias value. That this is so is seen in Fig. 4(c).

Subsequently, it was found that more quantitative deductions could be made from these experiments.

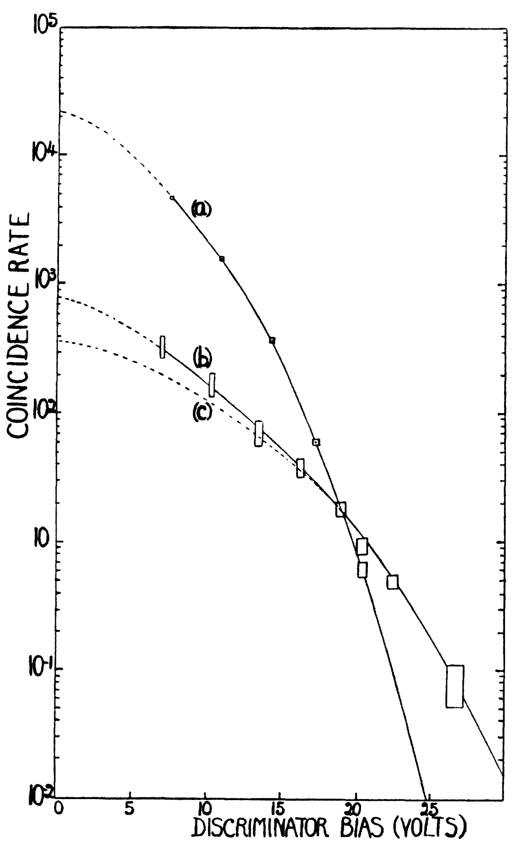

FIG. 5. (a) Bias curve of β–γ-coincidences corresponding to 4(a). (b) Bias curve of γ–γ-coincidences corresponding to 4(b). (c) Bias curve of 0.67-Mev γ-ray derived from (b).

DETERMINATION OF THE BRANCHING RATIOS OF THE 0.67- AND 1.09-MEV γ-RAYS

Let p be the branching ratio of the low energy β-ray, and let a fraction q of these β-rays be in cascade with the 1.09-Mev γ-rays, so that the 0.41- and 0.67-Mev γ-rays are in cascade to the extent of $p(1-q)$ per disintegration. Let the efficiencies of the 0.41-, 0.67-, and 1.09-Mev γ-rays in the fixed bias counter be E_1, E_2, and E_3, and for the variable bias counter, be ϵ_1, ϵ_2, ϵ_3, respectively. Then the counting rate in the fixed bias counter is

$$N_1 = N\{E_1(1-pq)+E_2p(1-q)+E_3pq\},$$

and in the variable bias counter

$$N_2 = N\{\epsilon_1(1-pq)+\epsilon_2p(1-q)+\epsilon_3pq\}.$$

The γ–γ-coincidence rate is

$$G_{\gamma\gamma} = Np(1-q)\{E_1\epsilon_2+E_2\epsilon_1\}.$$

Now $N_1 \doteqdot NE_1$, since the gain on this counter is high and p is small.

$$\therefore\quad G_{\gamma\gamma}/N_1 \doteqdot p(1-q)\{\epsilon_2+(E_2/E_1)\epsilon_1\} = 0.98p(1-q)\{\epsilon_2+(E_2/E_1)\epsilon_1\},$$

where the factor 0.98 arises on putting in approximate values for p, q, E_2/E_1, and E_3/E_1 in the expression for N_1.

In the β–γ-coincidence measurements, sufficient absorber was placed over the β-counter to exclude the low energy β-group.

$$\therefore \quad N_\beta = N(1-p)\epsilon_\beta,$$

and

$$G_{\beta\gamma} = N(1-p)\epsilon_\beta\epsilon_1, \quad G_{\beta\gamma}/N_\beta = \epsilon_1.$$

The curve for ϵ_1 [Fig. 5(a)] was extrapolated to low bias values by comparison with a bias curve of the single rate taken at high gain. A bias curve was measured for the counter normally held at fixed bias during the γ–γ-measurement and was extrapolated to zero bias. From this, on the basis that bias curves for γ-rays of different energies fall off in the same ratio at values of bias in the ratio of the energies, it could be determined that E_2/E_1 at the operating point was 1.20 times the value at zero bias.

$$\left(\frac{G_{\gamma\gamma}}{N_1}\right) \Big/ \left(\frac{G_{\beta\gamma}}{N_\beta}\right) = 0.98p(1-q)\left(\frac{\epsilon_2}{\epsilon_1}\right)_0 \left\{\frac{r_2}{r_1}+1.20\right\},$$

where $(\epsilon_2/\epsilon_1)_0$ is the ratio of the efficiencies for the two γ-rays, and r_2 and r_1 are the fraction of the values at zero bias to which the counting rates due to the two γ-rays are reduced at equal values of bias. The ratio $(\epsilon_2/\epsilon_1)_0$ is given by the ratio of the Compton cross sections, corrected for the finite absorption within the crystal and crystal holder. By experiments with several γ-ray energies ranging from 0.41 to 1.30 Mev, and using uncollimated sources, we have found, as an empirical fact, that within a few percent, the bias curves for γ-rays of different energies fall off in the same ratio at values of bias in the ratio of the energies, provided that there are no effects owing to "pile-up." On this basis, the bias curve for the 0.67-Mev γ-ray can be constructed from that due to the 0.41-Mev γ-ray, which is known experimentally, and in this way (r_2/r_1) may be determined. With this assumption it is possible to calculate a value for the branching ratio $p(1-q)$ for each of the experimental points. These are given in Table I. Over a range of values of bias corresponding to three decades in the bias curve of the 0.41-Mev γ-ray, the value of $p(1-q)$ derived in this way remains remarkably constant. Beyond this point the value falls off, and this may be ascribed to the effects of "pile-up" of the 0.41-Mev γ-rays. The constancy of the value of the branching ratio confirms that the assumption regarding the similarity of bias curves is substantially correct over a wide range of values of counting rate; moreover, it shows that it is the 0.67-Mev γ-ray which is in coincidence with the 0.41-Mev γ-ray. Taking an average over the first five values of bias, the value of the branching ratio for the 0.67-Mev γ-ray is 1.43 ± 0.10 percent.

By considering the ratio of genuine γ–γ-coincidences to accidental coincidences and making the same assumption concerning the similarity of the bias curves for the 0.67- and 1.09-Mev γ-rays as was previously made for the 0.41- and 0.67-Mev γ-rays, a value for the branching ratio of the 1.09-Mev γ-ray may also be determined.

TABLE I. Values of the branching ratio $p(1-q)$.

Bias value	7.0	10.3	13.5	16.3	19.0	20.4	22.5
Limits of values of $p(1-q)$, percent	1.76	1.86	1.66	1.62	1.55	1.15	1.08
	1.21	1.19	1.04	1.19	1.21	0.88	0.85

$$\frac{G_{\gamma\gamma}}{N_1N_2 2\tau} = \frac{1}{2\tau N} \cdot \frac{1.40\times10^{-2}(1+1.20r_1/r_2)}{[(0.83r_1/r_2)+1.40\times10^{-2}+pq(\epsilon_3/\epsilon_2)_0 r_3/r_2]},$$

and $1/(2\tau N)$ can be determined from the β–γ-measurements.

In this case, as would be expected, it is only possible to get values of reasonable accuracy from those points at values of the bias such that the counting rate due to the 0.411-Mev γ-ray is fairly small compared with that of the 0.67-Mev γ-ray. This limits us to values, obtained at 22.5 and 26.7 volts bias, of 0.34 percent and 0.20 percent, respectively, with an over-all statistical accuracy of about 15 percent and 40 percent. The value for the branching ratio of the 1.09-Mev γ-ray is therefore 0.33 ± 0.07 percent. These values for the branching ratios agree very well with those determined by the spectrometer, 1.4 and 0.4 percent, respectively, for the 0.67- and 1.09-Mev γ-rays.

CONCLUSIONS

It has been possible, using proportional scintillation counters, to establish a decay scheme where branching ratios of only about one percent are involved. Moreover it has been possible to measure these branching ratios with good accuracy, entirely by coincidence methods, and obtain results in good agreement with spectrometer measurements. It can now be taken as established that there are energy levels of the Hg^{198} nucleus at 0.411 and 1.09 Mev. Transitions occur between these states and the ground state, and also between the two states. The energy of the main β-branch to the 0.411-Mev level is 0.96 Mev, and that to the 1.09 Mev-level is 0.295 Mev. The branching ratio for the 0.295-Mev β-ray is (1.76 ± 0.12) percent. The branching ratios for the 0.67- and 1.09-Mev γ-rays separately are (1.43 ± 0.10) percent and (0.33 ± 0.07) percent, respectively. The accuracies quoted are statistical only. For the 0.67-Mev γ-ray this is believed to represent a fair estimate of the actual accuracy, but the value obtained for the 1.09-Mev γ-ray is probably less reliable.

This work was carried out at the Atomic Energy Research Establishment, Britain, and acknowledgment is made to the Director for permission to publish.