SUM-COINCIDENCE MEASUREMENTS ON ^{154}Eu

By A. W. Parker*

[*Manuscript received February 24, 1961*]

Summary

By examining γ-rays from the radioisotope ^{154}Eu with a fast-slow sum-coincidence spectrometer any β-transition to a possible $1 \cdot 82$ MeV level in ^{154}Gd has been shown to have an abundance less than 1% of that of the β-transition to the neighbouring $1 \cdot 72$ MeV level. The presence of a $1 \cdot 60$ MeV γ-ray from the $1 \cdot 72$ MeV level is confirmed; the intensity of this γ-ray was found to be $6 \cdot 4$% of that of the $1 \cdot 28$ MeV γ-ray.

I. Introduction

The current knowledge of the energy levels of ^{154}Gd has been obtained from a study of the radioisotopes ^{154}Eu and ^{154}Tb. The results of these experiments have been tabulated by Way *et al.* (1959), and Figure 1 shows the levels populated by the decay of ^{154}Eu.

A level at $1 \cdot 82$ MeV was suggested by Cork *et al.* (1957) because, in an investigation of the decay of ^{154}Eu, they found a β-transition with an end point of 150 keV. They estimated the abundances of β-transitions to the $1 \cdot 82$ and. $1 \cdot 72$ MeV levels to be 12% and 28% of all β-transitions. However, Way *et al.* (1959) suggest that the 150 keV transition occurs in the decay of ^{155}Eu.

Cork *et al.* (1957) also suggested that the $1 \cdot 82$ MeV level de-excites by a 694 keV γ-transition. Toth and Rasmussen (1959) also detected a γ-ray of 692 keV in the decay of ^{154} Tb; and they recalculated the energy of an intense γ-transition, detected by Juliano and Stephens (1957), from fission-produced ^{154}Eu in which ^{152}Eu was absent. This they showed to be 692 keV. On the other hand, other workers (Gallagher and Thomas 1959; Nathan and Hultberg 1959; Sheline 1960; Marklund, Van Nooijen, and Grabowski 1960) have suggested that this γ-ray may result from the de-excitation of either a 692 keV level or a 815 keV level—via the 123 keV level—in ^{154}Gd.

Further evidence for the presence of a $1 \cdot 82$ MeV level is suggested by the measurements of Jha *et al.* (1959), who used a fast-slow sum-coincidence spectrometer to investigate the γ-rays of ^{154}Eu. However, their results were not conclusive because of poor statistics and because they did not completely consider the effects of triple coincidences.

No γ-ray from the $1 \cdot 29$ MeV level shown in Figure 1 to the ground state is known, and no 100 keV transition which might occur between the $1 \cdot 821$ and $1 \cdot 723$ MeV levels has been observed. Hence, by far the most probable mode

* Physics Department, University of Melbourne.

of de-excitation of a 1·821 MeV level will be via the 0·692–1·006–0·123 MeV cascade. The present paper describes sum-coincidence measurements aimed at detecting this triple cascade and thus obtaining information on the existence of a 1·821 MeV level.

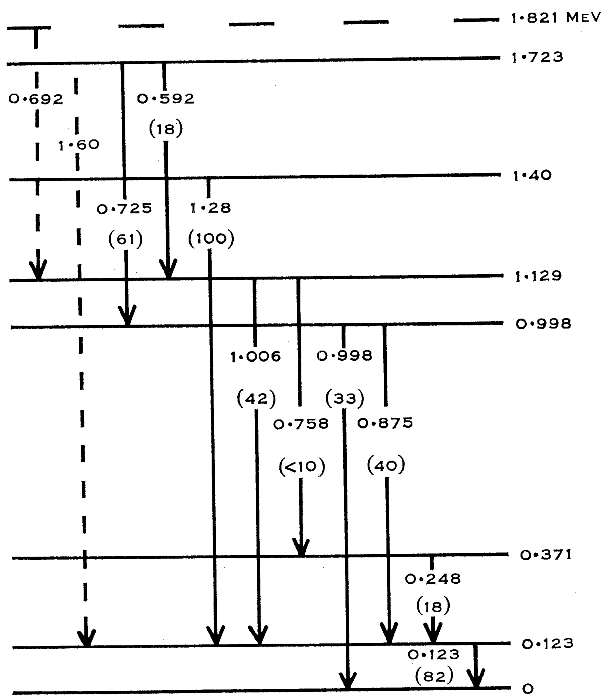

Fig. 1.—Energy levels of $^{154}_{64}$Gd as determined in the β-decay of $^{154}_{63}$Eu.

II. Method

The ^{154}Eu source used for these measurements was obtained from the Oak Ridge National Laboratory, and was prepared by the neutron capture of 99·4% enriched ^{153}Eu. γ-Rays from this source were detected by a fast-slow sum-coincidence spectrometer similar to that suggested by Hoogenboom (1958). Two 1·5 in. diameter by 1 in. thick Na(Tl) crystals were used, and β-rays were absorbed by the canning material which consisted of 0·7 mm of aluminium and 2·0 mm of magnesium oxide.

A block diagram of the apparatus is shown in Figure 2. Pulses from the two detectors are summed, and a narrow window includes only those pulses corresponding to the energy level of interest. If, for example, the level decays only by a simple double cascade, the coincidence spectrum will show two Gaussian peaks corresponding to the full energy peaks of the γ-rays. In this apparatus a fast-coincidence unit is used to reduce random coincidences, and so the coincidence spectrum does not show a full energy peak corresponding to both γ-rays being absorbed in one detector. Random-coincidence spectra were obtained by inserting a long delay at the input to one channel of the fast-coincidence unit.

Hoogenboom (1958) has shown that the half-width Γ_{S1} of a sum-coincidence peak is given by

$$\Gamma_{S1}=\Gamma_1(\Gamma_2^2+\Gamma_S^2)^{\frac{1}{2}}/(\Gamma_1^2+\Gamma_2^2+\Gamma_S^2)^{\frac{1}{2}},$$

where Γ_1 and Γ_2 are the half-widths of the two γ-ray peaks in the singles spectrum and Γ_S is the half-width of the sum channel. Further, the detection efficiency ε_{S1} of this peak is given by

$$\varepsilon_{S1}=2\{(\ln 2)/\pi\}^{\frac{1}{2}}\varepsilon_1\varepsilon_2\Gamma_S/(\Gamma_1^2+\Gamma_2^2+\Gamma_S^2)^{\frac{1}{2}},$$

where ε_1 and ε_2 are photo-peak efficiencies (including solid angle).*

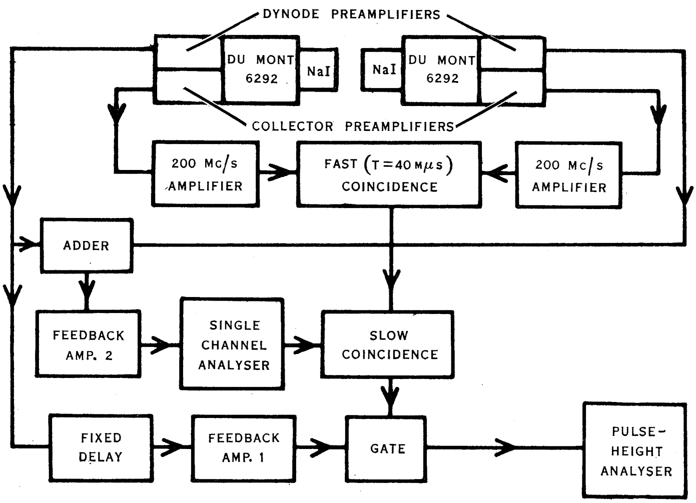

Fig. 2.—Block diagram of the fast-slow sum-coincidence spectrometer.

Triple cascades are detected by two γ-rays being absorbed in one detector whilst the third γ-ray is absorbed in the other, thus producing six peaks of energies E_1, E_2, E_3, E_1+E_3, E_2+E_3, and E_3+E_1 in the coincidence spectrum. Using assumptions similar to those of Hoogenboom (1958) it is easily shown that, while the sum peaks E_1+E_2 etc. are not strictly Gaussian, the half-width Γ_{S1} for the peak E_1 is given by

$$\Gamma_{S1}=\Gamma_1(\Gamma_2^2+\Gamma_3^2+\Gamma_S^2)^{\frac{1}{2}}/(\Gamma_1^2+\Gamma_2^2+\Gamma_3^2+\Gamma_S^2)^{\frac{1}{2}}.$$

However, the detection efficiencies of the peaks E_1 and E_2+E_3 are equal for matched detectors and

$$\varepsilon_{S1}=2\{(\ln 2)/\pi\}^{\frac{1}{2}}\varepsilon_1\varepsilon_2\varepsilon_3\Gamma_S/(\Gamma_1^2+\Gamma_2^2+\Gamma_3^2+\Gamma_S^2)^{\frac{1}{2}}.$$

In this experiment only the relative detection efficiencies for double and triple cascades are of interest and hence the factors involving half-widths may

* Values of these efficiencies were taken from Mott and Sutton (1958).

be neglected; but an additional factor is required when 123 keV transitions are involved because of internal conversion. Hence the double- and triple-coincidence rates C_D and C_T were estimated using the formulae

$$C_D = N_D \varepsilon_1 \varepsilon_2/(1+\alpha), \quad C_T = N_T \varepsilon_1 \varepsilon_2 \varepsilon_3/(1+\alpha), \tag{1}$$

where $\alpha = N_e/N_\gamma$ is the internal conversion coefficient of the 123 keV transition. Here N_D is the transition rate of the $1\cdot60$ MeV γ-ray leading directly to the 123 keV level and N_T is the transition rate leading to this level via a double cascade.

This sum-coincidence spectrometer was easily modified to display the summed output from the two detectors. The modification involved connecting amplifier 2 to the gate input and opening the gate with the fast-coincidence unit. Dead-time corrections to the kicksorter were avoided by inserting a scale-of-ten between the fast-coincidence unit and the gate. An energy calibration curve was obtained by removing one input to the adder so that pulses from one counter only were accepted when the scale-of-ten recorded a fast coincidence.

For a double cascade using this latter arrangement, the two photo-peaks sum to produce a peak E_1+E_2, and the coincidence rate is given by $2C_D$, where C_D is the rate calculated from equation (1); similarly, a triple cascade produces a peak $E_1+E_2+E_3$ and the coincidence rate is given by $6C_T$.

With either arrangement of this apparatus the ability to distinguish neighbouring energy levels is dependent on the energy resolution of the corresponding summed peaks. Thus reference to Figure 1 shows that any attempt to detect γ-rays from a possible $1\cdot821$ MeV level must allow for the overlapping of the summed peak associated with the $1\cdot723$ MeV level. This was done by using the first arrangement described above. In this case, the sum-coincidence spectrum displays the detailed composition of the sum peak presented to the window. A significant variation of this composition as the window height is increased above $1\cdot723$ MeV would then indicate the presence of the $1\cdot821$ MeV level. Since Figure 1 also indicates that the presence of a $1\cdot6$ MeV γ-ray from the $1\cdot723$ MeV level has not been definitely established, it is clear that the starting point of these measurements is the investigation of γ-rays associated with this level.

From this type of coincidence data it is also possible to calculate the relative abundance of β-transitions to these two levels. However, for this calculation the data were supplemented by measurements of the summed output of the detectors using the second arrangement, because this avoids uncertainties associated with the relatively wide window and its transmission characteristics.

III. Results

Figure 3 shows the sum-coincidence spectrum with the window height set at $1\cdot75$ MeV and the window width set at $0\cdot023$ MeV. The three peaks* shown correspond to the cascades $0\cdot725$–$0\cdot998$ and $1\cdot60$–$0\cdot123$ MeV. Using equation

* The fourth expected peak at 123 keV was removed by the bias applied to the linear gate. Only that portion of the spectrum above about $0\cdot3$ MeV is shown because the gate occasionally passed large non-coincident pulses at a reduced level, and hence the spectrum in the first five channels of the kicksorter was unreliable.

(1) with $\alpha=1\cdot5$, together with the relative intensities of Figure 1, the corresponding intensity of the $1\cdot60$ MeV γ-ray was found to be $6\cdot4\pm0\cdot5$, the error shown being the statistical error in the present experiment. The distance of the source from the crystals was $4\cdot5$ cm and, with this geometry, triple cascades can contribute less than 3% of the peak counts, while the true-to-random ratio for the cascades was found to be 12 and 18 respectively.

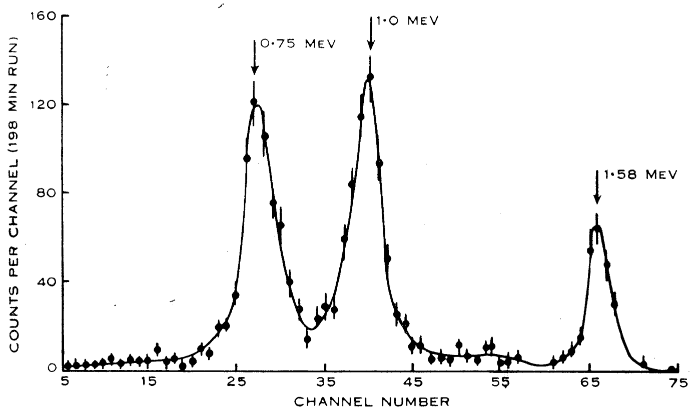

Fig. 3.—Sum-coincidence spectrum at $1\cdot75$ MeV with the source distance $4\cdot5$ cm.

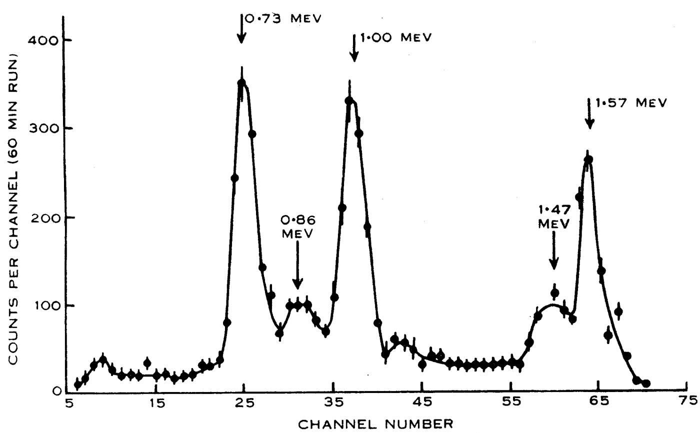

Fig. 4.—Sum-coincidence spectrum at $1\cdot725$ MeV with the source distance $0\cdot5$ cm.

Figure 4 shows the sum-coincidence spectrum with the window set at $1\cdot725$ MeV but with the crystals closer together and a weaker source placed $0\cdot5$ cm from each. With this geometry four cascades of $0\cdot725$–$0\cdot998$, $1\cdot60$–$0\cdot123$, $0\cdot725$–$0\cdot875$–$0\cdot123$, and $0\cdot592$–$1\cdot006$–$0\cdot123$ MeV should be detected. By use of the data from Figure 3 and the γ-ray intensities from Figure 1, the relative intensities of the peaks expected in this spectrum can be estimated. Table 1 shows these calculated intensities. These relative intensities are expected to be accurate to about 10%.

In the spectrum, the peaks at 0·725 and 0·715 MeV cannot be resolved. Hence it is expected that the peak at 1·60 MeV will have an intensity of 56/108=0·52 of that of the peak actually observed at 0·725 MeV. The experimental value of this ratio, calculated from the areas under the peaks, is

Table 1

EXPECTED INTENSITIES OF THE PEAKS IN THE SPECTRUM OF FIGURE 4

Energy (MeV)	0·123 1·60	0·592 1·129	0·715 1·006	0·725 0·998	0·848 0·875
Relative intensity	56	8	8	100	15

0·60±0·05, which is in reasonable agreement with this prediction. It is difficult to achieve any accuracy in the measurement of the intensities of the weaker peaks, but their intensities are certainly of the same order of magnitude as those predicted in Table 1, provided allowance is made for backscattering, which contributes to the peaks at 0·86 and 1·47 MeV.

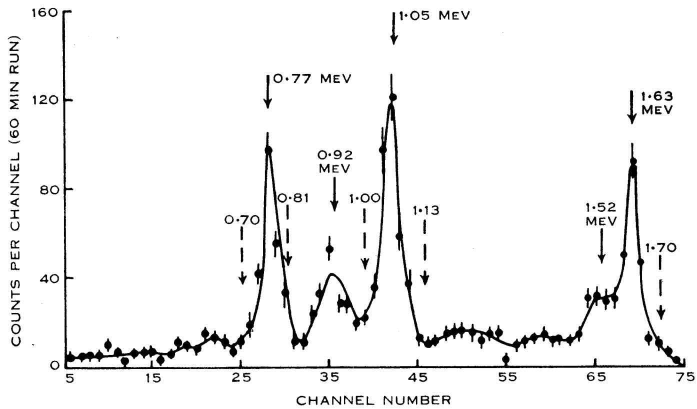

Fig. 5.—Sum-coincidence spectrum at 1·825 MeV with the source distance 0·5 cm.

Figure 5 shows the spectrum obtained with the window set at 1·825 MeV. This shift in the position of the window is less than the width of the sum peak at 1·723 MeV which is presented to the window. Hence it is expected that the peaks of the 1·723 MeV spectrum will still be present—although at lower intensities—but will be displaced in the direction of higher energies. This effect has been discussed by Hoogenboom (1959).

The 0·692–1·006–0·123 MeV cascade from the 1·821 MeV level, if present, should produce peaks at energies of 0·123, 0·69, 0·81, 1·01, 1·13, and 1·7 MeV and these peaks in the sum-coincidence spectrum should all have equal intensities. There is no evidence for any of these peaks in Figure 5.

The spectrum of summed pulses due to γ-rays from the $1 \cdot 723$ MeV level had a " tail " which would still be of appreciable intensity at a pulse height corresponding to $1 \cdot 825$ MeV, and might tend to mask any pulses in this region due to γ-rays from the $1 \cdot 821$ MeV level. With the window set somewhat above this pulse height, the reduction in the " tail " of the $1 \cdot 723$ MeV pulses should be considerably greater than that in the pulse-height distribution due to any genuine $1 \cdot 821$ MeV sum distribution. For this reason a spectrum was taken with the window set at $1 \cdot 88$ MeV, and the result is shown in Figure 6.

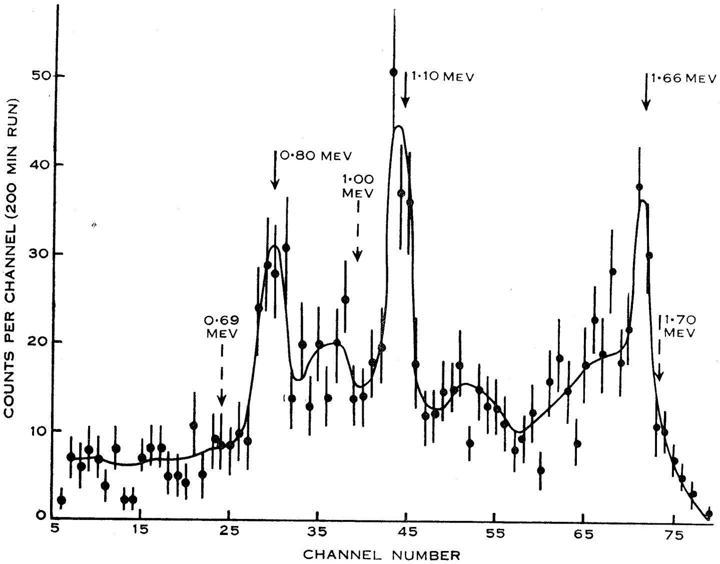

Fig. 6.—Sum-coincidence spectrum at $1 \cdot 88$ MeV with the source distance $0 \cdot 5$ cm.

Comparison of Figure 4 with Figures 5 and 6 strongly suggests that these latter are dominated by γ-rays from the $1 \cdot 723$ MeV level. This conclusion is reinforced by the following consideration. The ratio of the total area under the peaks at $0 \cdot 725$ and $1 \cdot 006$ MeV to that under the peak at $1 \cdot 62$ MeV in Figure 4 is $3 \cdot 2 \pm 0 \cdot 3$. The same ratio has the values $2 \cdot 9 \pm 0 \cdot 5$ and $2 \cdot 8 \pm 0 \cdot 5$ for the spectra of Figures 5 and 6 respectively.

An estimate of an upper limit for the contribution to the spectrum of Figure 6 from γ-rays associated with the $1 \cdot 821$ MeV level may be made as follows. No definite peaks are apparent at energies of $0 \cdot 69$, $0 \cdot 82$, $1 \cdot 0$, $1 \cdot 13$, and $1 \cdot 70$ MeV, and, since these peaks would not all be completely obscured by the displaced peaks associated with the $1 \cdot 723$ MeV level, a visual inspection of Figure 6 shows that it is reasonable to associate not more than 20 counts with each of these possible peaks. Also Figure 4 may be suitably normalized and superposed on Figure 6. Again 20 counts is an absolute upper limit to the intensity of any one of these peaks. Hence a total of 100 counts may be associated with the

1·821 MeV level, and this may be compared with the total number of counts, 1060, in Figure 6. It is therefore certain that not more than 10% of the pulses which sum to 1·88 MeV are associated with γ-rays from any 1·821 MeV level.

Figure 7 shows the spectrum of the summed output of the two detectors, the energy calibration spectrum, and the random-coincidence spectrum. These support the previous conclusion that any level at 1·821 MeV is very weakly populated. The ratio of counts at 1·723 and 1·821 MeV is 24 ; but the previous sum-coincidence measurements show that less than 10% of the pulses in the

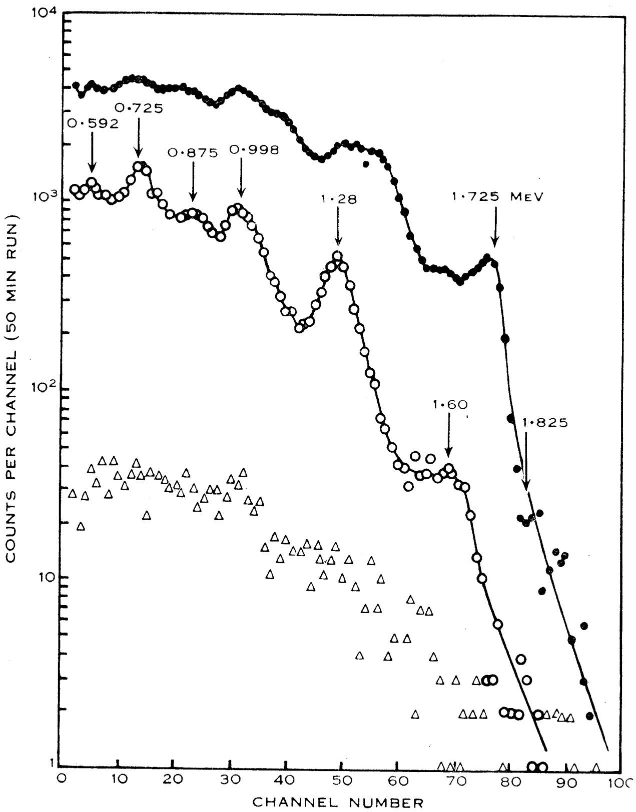

Fig. 7.—Summed spectrum (●), energy calibration spectrum (○) (15 min run), and random-coincidence spectrum (△) with the source distance 0·5 cm.

" tail " of the 1·723 MeV peak can come from γ-rays associated with the 1·821 MeV level. Thus the lower limit on R_C, the ratio of the counts associated with the 1·723 MeV level to those associated with the 1·821 MeV level is increased from 24 to 240. Now, if the internal conversion of γ-rays from the 1·821 MeV level is negligible, the ratio R of the abundances of the β-transitions to these levels is simply the ratio of the total γ-ray transition rates from the two levels.

The relation between R and R_C may then be obtained as follows. The 1·723 MeV level de-excites by three γ-rays of known relative transition rates, Since the total transition rate is the sum of these separate rates, these separate rates may be expressed as fractions of the total transition rate. Also, the branching ratios of γ-rays from lower levels are known, and hence the coincidence rate due to a definite cascade may be expressed in terms of the total transition rate by using equations (1) as explained in Section II. By summing the coincidence rates due to different cascades, the total coincidence rate is thus expressed in terms of the total transition rate. Similarly, the total coincidence rate due to γ-rays associated with the $1 \cdot 821$ MeV level may be related to the total transition rate from that level; in this case the calculation is simplified because it is assumed that this level de-excites only by the 692–1006–123 keV cascade. The ratio of these two total coincidence rates then leads to the result that $R = 0 \cdot 50 R_C$. Thus since $R_C > 240$, $R > 120$, i.e. any β-transition to a possible $1 \cdot 821$ MeV level has an abundance of less than 1% of that of the β-transition rate to the neighbouring $1 \cdot 723$ MeV level.

IV. Discussion

Measurements made by Juliano and Stephens (1957) and by Bhattacharjee, Raman, and Mitra (1958) using scintillation spectrometers indicated the possible presence of a $1 \cdot 60$ MeV γ-ray. The values of the intensity of this γ-ray found in these two experiments were $7 \cdot 4$ and 4 respectively, on the intensity scale shown in Figure 1. However, coincidence measurements made by them were inconclusive in that they failed to confirm the existence of this γ-ray.

The sum-coincidence measurements of Jha *et al.* (1959) showed a peak at $1 \cdot 6$ MeV, but the intensity of their peak is consistent with its being formed as a result of two triple cascades. Hence their results suggest the absence of a $1 \cdot 6$ MeV γ-ray.

The present experiment indicates the presence of this γ-ray, with an intensity, on the same scale mentioned above, of $6 \cdot 4 \pm 0 \cdot 5$. However, the accuracy of this result depends on the assumed value of the internal conversion coefficient of the 123 keV γ-ray, and also on the relative intensities of the other γ-rays as found by other workers. Taking these factors into consideration increases the possible range of error to 20%, so that the final result for the intensity of this γ-ray is $6 \cdot 4 \pm 1 \cdot 3$.

The present experiment also suggests that no level exists at $1 \cdot 821$ MeV in ^{154}Gd. If this is so, the query raised by Way *et al.* (1959) as to the interpretation of the angular correlation measurements of Hickman and Wiedenbeck (1958) no longer applies. Furthermore, this result removes one of the possibilities of fitting the 692 keV γ-transition into the level scheme of ^{154}Gd. Although this transition may come from the low-energy O^+ β-vibrational state predicted by Sheline (1960), the present experiment gives no direct information on the existence of low-energy states.

V. Acknowledgments

The author would like to thank Dr. J. A. McDonell for his interest and for many helpful discussions during the course of this work. The author also wishes to acknowledge the tenure of a Commonwealth Postgraduate Award.

VI. References

Bhattarcharjee, S. K., Raman, S., and Mitra, S. K. (1958).—*Proc. Indian Acad. Sci.* **47A**: 295.

Cork, J. M., Brice, M. K., Helmer, R. G., and Sarason, D. E. (1957).—*Phys. Rev.* **107**: 1621.

Gallagher, C. J., Jr., and Thomas, T. D. (1959).—*Nuclear Phys.* **14**: 18.

Hickman, G. D., and Wiedenbeck, M. L. (1958).—*Phys. Rev.* **111**: 539.

Hoogenboom, A. M. (1958).—*Nuclear Instruments* **3**: 57.

Hoogenboom, A. M. (1959).—In " Nuclear Electronics ", Vol. I. (International Atomic Energy Agency: Vienna.)

Jha, S., Devare, H. G., Rao, M. N., and Pramila, K. G. C. (1959).—*Proc. Indian Acad. Sci.* **50A**: 303.

Juliano, J. O., and Stephens, F. S., Jr. (1957).—*Phys. Rev.* **108**: 341.

Marklund, I., Van Nooijen, B., and Grabowski, Z. (1960).—*Nuclear Phys.* **46**: 533.

Mott, W. E., and Sutton, R. B. (1958).—In " Handbuch der Physik ", Vol. XLV. (Springer: Berlin.)

Nathan, O., and Hultberg, S. (1959).—*Nuclear Phys.* **10**: 118.

Sheline, R. K. (1960).—*Rev. Mod. Phys.* **32**: 1.

Toth, K. S., and Rasmussen, J. O. (1959).—*Phys. Rev.* **115**: 150.

Way, K., *et al.* (1959).—" Nuclear Data Sheets." (National Research Council: Washington, D.C.)