Radioactive decay of 47-min $^{154}Eu^m$

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A 47-min isomer has been identified in ^{154}Eu. Sources were prepared in (n,γ) and (d,p), $(d,2n)$ and (p,n), and (d,α) reactions on Eu, Sm, and Gd, respectively, and in each case the 47-min activity followed the Eu fraction in the separation of $EuSO_4$. Singles and coincidence γ-ray studies confirmed a number of the observed γ rays in a structure consistent with capture γ-ray studies. Owing to the high conversion coefficients for low energy γ rays a complete decay scheme has not been confirmed for $^{154}Eu^m$.

[RADIOACTIVITY $^{154}Eu^m$; measured $T_{1/2}$, E_γ, I_γ, $\gamma\gamma$ coin, deduced levels, J, π. Enriched targets. Ge(Li) and Si(Li) detectors, 170 eV at 5.9 keV.]

I. INTRODUCTION

Among the least studied and understood aspects of nuclear structure are the states of odd-odd nuclei, particularly those with high spin. As radioactive even-even nuclei have 0^+ spin and parity, their decay populates mostly low-spin states. In (d,p) studies on odd-Z nuclei, l values can be extracted but J values can span up to $2l+1$ possibilities. The best information comes from high resolution neutron-capture γ-ray and electron spectroscopy and from the decay of isomeric states. For many isomeric states their proximity to each other, their sizeable spin differences, and the relatively large Q_β common to odd-odd nuclei, combine to limit most odd-odd isomers to β decay.

Few areas of nuclidic mass show as much isomerism as the $N=89$, 91, and 93 isotones. The onset above $Z=61$ of the $h_{11/2}$-$d_{3/2}$ proton orbitals and the resulting nuclear deformations lead to odd-Z and odd-odd isomerism. Three isomers[1-5] are known for odd-odd $^{152}_{61}Pm_{91}$, $^{152}_{63}Eu_{89}$, $^{154}_{65}Tb_{89}$, $^{156}_{65}Tb_{91}$, and $^{158}_{65}Tb_{93}$. In this paper we report the presence and decay of a high-spin isomer in ^{154}Eu discovered[6] while searching for a low-spin β-decaying isomer in ^{154}Eu.

II. EXPERIMENTAL PROCEDURES

A. Isotopic identification

We show in Fig. 1 the low-lying levels of ^{154}Eu observed in[7-10] the $^{153}Eu(n,\gamma)^{154}Eu$ reaction and the $^{153}Eu(d,p)^{154}Eu$ reaction[11] and the levels of ^{154}Gd observed[12] in the β decay of $^{154}Eu^g$. The observation of two γ rays at 68.2 and 100.9 keV with a 47-min half life in neutron-irradiated enriched ^{153}Eu strongly indicated the presence of an isomer in ^{154}Eu, as these two γ rays were also observed in the $^{153}Eu(n,\gamma)^{154}Eu$ reaction.[7-9] Isotopic identification is confirmed by chemically separating $EuSO_4$ from Sm, Gd, and other rare earth elements following the irradiation of enriched ^{154}Sm with 7.5-MeV p beams, and enriched ^{154}Sm, ^{156}Gd, and ^{153}Eu with 14-MeV d beams. In each case, the above two γ rays are observed in the same ratio to each other and with a 47-min half-life in the Eu fraction.

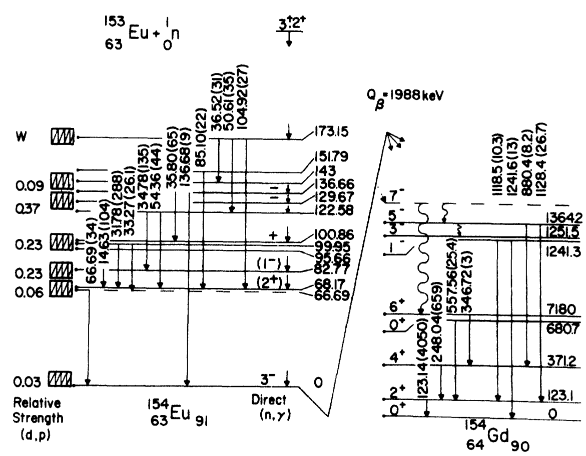


FIG. 1. Level scheme for ^{154}Eu from the capture γ-ray studies and the (d,p) studies, and the levels of ^{154}Gd that might participate in any β decay of $^{154}Eu^m$. The 27.5-, 28.8-, 68.2-, and 100.9-keV γ rays that deexcite the levels at 95.7, 129.7, 68.2, and 100.9 keV, respectively, in ^{154}Eu are not shown. The lined region associated with the observed (d,p) transitions reflects the ±5 keV uncertainty in the energy values. The intensities are given as γ rays per 10 000 decays of the $^{154}Eu^g$ and γ rays per 10 000 captures.

A least squares analysis of the decay of the two γ rays gives a half-life of 46.8 ± 0.6 min.

B. γ-ray spectroscopy

Subsequent studies of electron- and γ-ray spectra of $^{154}Eu^m$ have utilized primarily two types of sources, those obtained by the $^{154}Sm(d,2n)^{154}Eu$ reaction followed by the chemical isolation of $EuSO_4$, and those obtained by neutron irradiation of doubly enriched ^{153}Eu with thermal neutrons. The latter sources are made by passing 99% enriched ^{153}Eu through the isotope separater at the Lawrence Livermore Laboratory (LLL) to obtain sources ~99.99% enriched in ^{154}Eu. Such sources are required because of the ~3200 b cross section for the $^{151}Eu(n,\gamma)^{152}Eu^m$ reaction producing 9.3-h $^{152}Eu^m$. γ-ray spectra have been accumulated on several small very high resolution Ge(Li) detectors, one of which is shown in Fig. 2. The observed γ-ray energy and intensity values are tabulated in Table I along with energies[8,10,13] reported

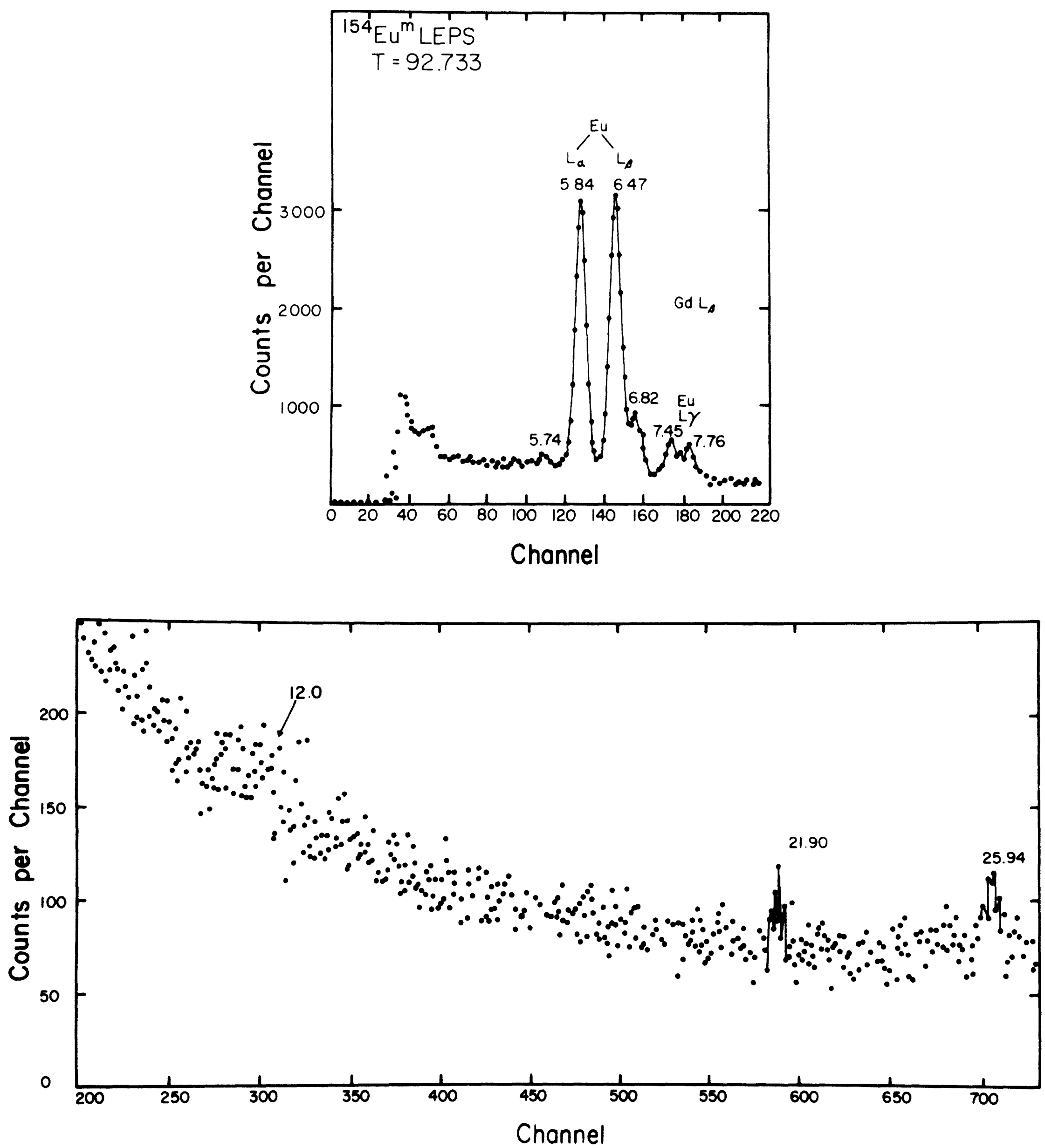
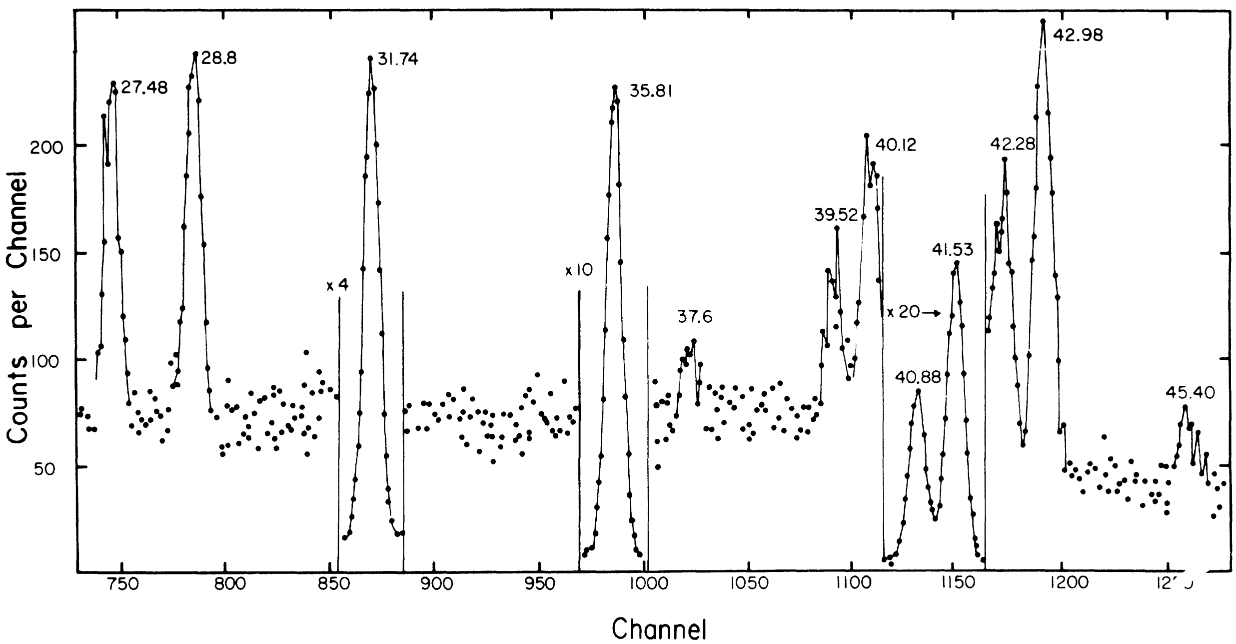


FIG. 2. γ-ray spectrum of $^{154}Eu^m$.



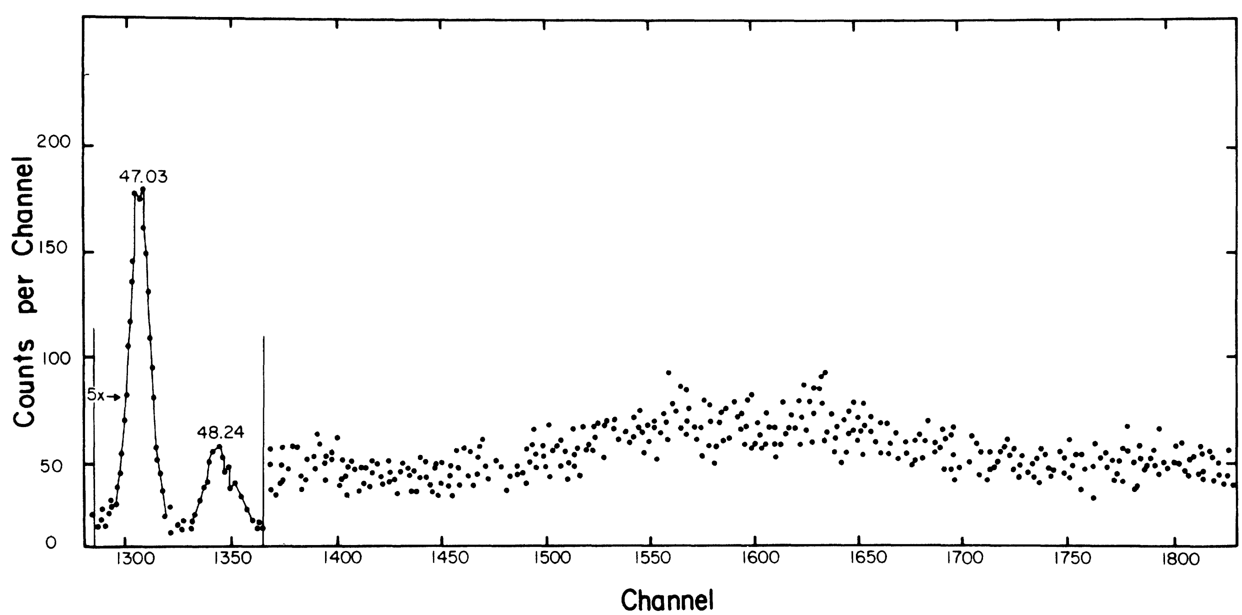


Fig. 2. *(Continued)*

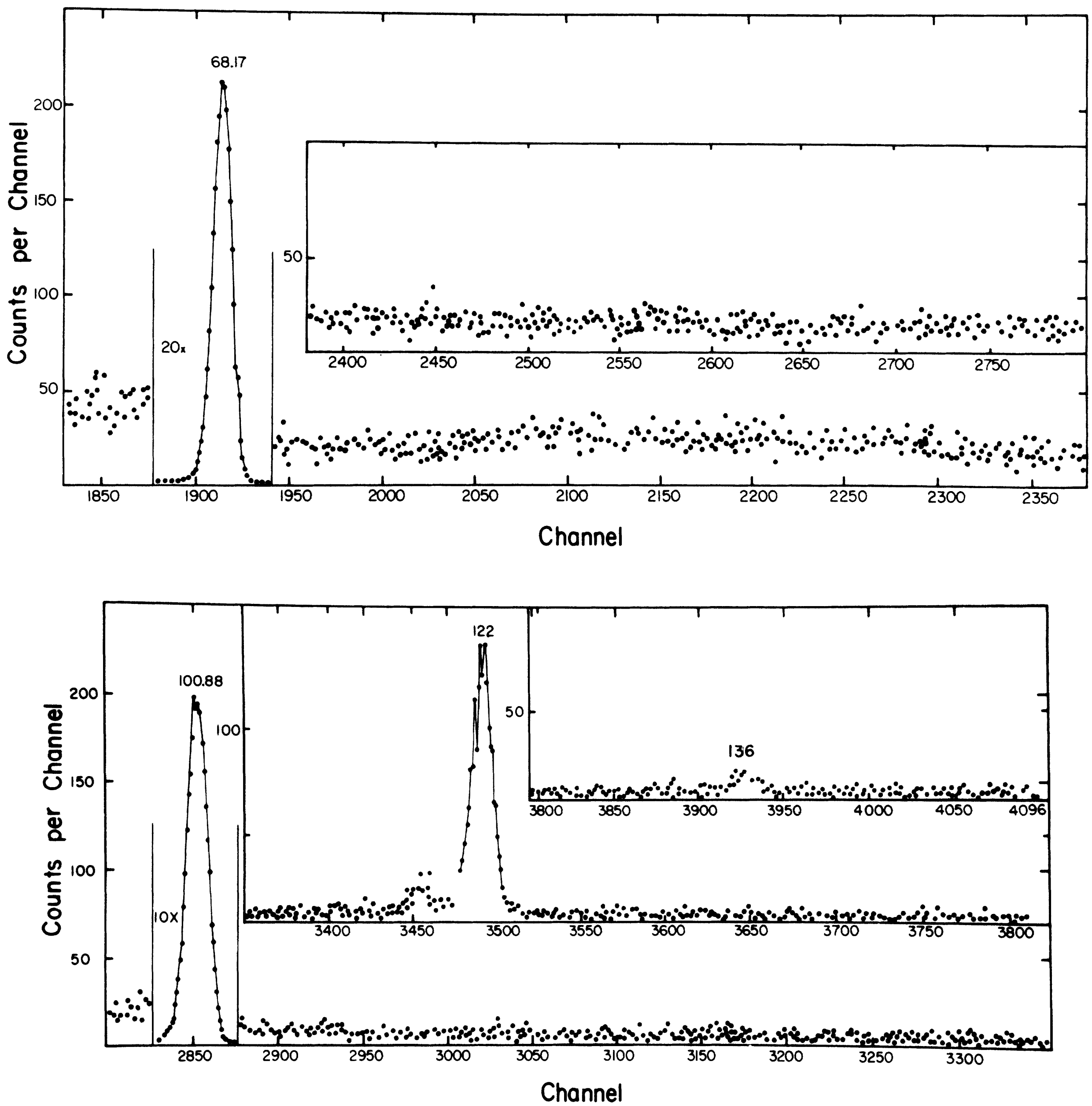


Fig. 2. *(Continued)*

by other investigators.

The possibility of β-decay branching from the isomeric state was investigated by searching for a 47-min component in the decay curve for the 123.14-keV 2^+ to 0^+ transition observed[4] in $^{154}Eu^{g}$ decay (see Fig. 1). A search was also made for the presence of the 6^+ to 4^+ 346.72-keV transition in the ^{154}Gd ground band that might be a part of any cascade from high-spin states fed in ^{154}Gd by any β decay from $^{154}Eu^{m}$. Limits of 0.3 and 0.03 relative intensity ($I_{68.2}=100$) can be set for a 47-min component in the 123.14- and 346.72-keV transitions, respectively.

Owing to the observed[9,10,13,14] lifetimes for the 68.2 and 100.9 keV γ rays of 2.3 μs and 55 ns, respectively, both prompt and delayed coincidence studies were made. A series of delayed coincidence measurements utilized a 55 cm^3 Ge(Li) detector sensitive only to γ rays with $E_\gamma > 50$ keV for a stop signal and a small high-resolution detector sensitive to γ rays with $E_\gamma > 5$ keV for start pulses. Coincidences were recorded only when the time

TABLE I. Energies and intensities of γ rays observed in ^{154}Eu.

This work E_γ [d]	This work I_γ	Zoller [a] E_γ	Orecher [b] E_γ	Stoffl et al. [c] E_γ	Stoffl et al. [c] Multipolarity
6.77					
7.70					
12.0(2)					
13.9					
21.9(2)		0.12(6)			
27.51(5)	1.75(20)	27.5(1)		27.469(11)	$M1+(\leq 2\%\ E2)$
28.78(2)	1.79(20)	28.9(1)	28.8171(10)	28.814(9)	$E1$
31.78(1)	11.4(7)	31.8(1)	31.7798(13)	31.776(14)	$M1+(\leq 0.5\%\ E2)$
35.802	2.2(8)			35.802(11)	$E1$
35.818(10)	26.1(15)	35.80(5)			
68.17(1)	100	68.20(5)	68.1717(18)	68.164(9)	$E1$
100.88(1)	76.6(25)	100.74(10)	100.859(3)	100.859(10)	$E1$
136.8(3)	0.3(1)			136.675(50)	

[a] Reference 13.
[b] Reference 8.
[c] Reference 10.
[d] The figures in parentheses indicate the uncertainties in the last digit(s) of the number they follow.

difference between the start and stop pulses was in the range of 150 to 2150 ns. Cascades through the 2.3-μs 68.2-keV level were thus emphasized.

Prompt coincidences utilized a time window of 200 ns centered at $t=0$ to include the events cascading through the 55-ns 100.9-keV level and other faster transitions. Two series were carried out, one using the detectors described above, and a second using two small high-resolution Ge(Li) detectors, the second being sensitive to γ rays with $E_\gamma > 20$ keV. The results of these experiments are tabulated in Table II, where we show the raw coincidence data corrected for contributions from the Compton background. Where two numbers are shown, the smaller represents the net counts obtained by correcting for random events. Few ran-

TABLE II. γ-γ coincidences in ^{154}Eu. The underlined zeros were set by the known lack of 69× 101 coincidences.

E_γ [a] \ Gate	100.9 Delayed [b]	100.9 Prompt Run 1 [b]	100.9 Prompt Run 2 [c]	68 Delayed	68 Prompt Run 1	68 Prompt Run 2	35.8 Prompt Run 2	31.8 Prompt Run 2	28.8 Prompt Run 2
100.9	86	2	27	72	5	63	256	1	18
(0.45)	$\underline{0}$		$\underline{0}$	$\underline{0}$		$\underline{0}$			
68.2	173	14	64	153	5	129	26	1	2
(1.0)	$\underline{0}$		$\underline{0}$	$\underline{0}$		$\underline{0}$			
35.8	509	350	852	219	6	82	0	68	
(0.33)	450		830	170		40			
31.8	17	0	16	95	1	69	28	0	
(0.13)	0		8	76		53			
28.8	43	26	69	0	3	9	3	10	
(0.02)	38		68			6			
27.5	6	0	2	20	0	13	2	14	
(0.02)	0		1	17		10			

[a] The number in parentheses below each γ-ray energy is its observed relative peak area, used to make random corrections.
[b] The numbers displayed are the peak areas in the small, thin window x-ray detector with the gate set in the 55 cm^3 detector (delayed and prompt, run 1).
[c] The numbers displayed are the peak areas in the small thin window x-ray detector and the gate was set in the spectrum of the larger thicker window x-ray detector (promt, run 2).

doms were observed in the prompt big-Ge(Li)–small-Ge(Li) experiments, owing to the relatively small time window and the high geometry. In the other two experiments the large time window in one and the two small detectors in the other led to relatively large random rates. Because of the shape and rise time of the low-energy pulses the random corrections useful for the 68.2- and 100.9-keV γ rays may not be applicable to the lower-energy transitions. This difficulty, coupled with the large statistical uncertainties associated with the small numbers of counts in a peak, hinders the clear identification of coincidence relationships.

C. Measurement of the isomer production cross section ratio

The production cross section ratio for the two isomers was measured by irradiating a fresh source of ~99.99% ^{153}Eu in the NBS reactor for four minutes and determining the initial activities of $^{154}Eu^m$ and $^{154}Eu^g$. The irradiation was carried out in the tube RT-4 at the NBS reactor. The tube terminates at the very edge of the core and the cadmium ratio at that position is 87 for Au. The 68.2- and 100.9-keV γ rays are known[10] to be $E1$ and were assumed to carry nearly all of the γ-ray feeding to the ground state (see Fig. 1). The 123-keV γ ray of $^{154}Eu^g$ was utilized along with the half-life value[15] of 8.5 ± 0.5 yr to determine the initial $^{154}Eu^g$ activity. The measured isomer ratio calculated from these data is $(3.1 \pm 0.3) \times 10^{-4}$. This value may be compared with the value[4] of 4.9×10^{-4} for $\sigma_{8^-}/(\sigma_{0^-} + \sigma_{3^-})$ in the $^{151}Eu(n,\gamma)^{152}Eu$ reaction. Comparison is made with the sum of the cross sections for the 0^- and 3^- states in ^{152}Eu, as the capture in ^{154}Eu to any similar 0^- state would be expected to decay to the 3^- ground state in ^{154}Eu.

D. Electron spectra

Several attempts were made to determine the energy of the 47-min isomeric transition (IT) using Si(Li) electron detectors. Because of the strong interference from x rays and ^{152}Eu β^- particles, no useable results were obtained, other than to limit the energy of the IT to values <50 keV.

III. LEVEL SCHEME

The proposed decay scheme shown in Fig. 3 for 47-min $^{154}Eu^m$ is much more complicated than that of the similar 8^- isomer in ^{152}Eu, as evidenced by the large number of γ rays and the widely varying intensities. Recent studies[16] of the decay of 8^- $^{152}Eu^{m2}$ have also revealed one additional weak γ ray at 77.23 keV whose presence requires at least one additional unobserved γ ray in that level scheme. The γ ray spectrum shown in Fig. 2 is seen to show none of the other γ rays shown in Fig. 1 as deexciting low-energy levels in ^{154}Eu. In particular, the 33.26-keV γ ray proposed by Stoffl *et al.*[10] to be $\sim\frac{1}{10}$ the intensity of the 31.2-keV γ ray deexciting the 99.9-keV level is seen to be absent.

We show in Fig. 3 a proposed level scheme for the decay of $^{154}Eu^m$ based on our data, the (d,p) data of Lanier[11] and the capture γ-ray work of Stoffl *et al.*[10] Owing to the complexity of the capture γ-ray spectrum, and the ±5 keV resolution for the (d,p) study, the association of the γ rays that we observe with γ rays and levels in those studies may not be unique. The seven γ rays on the left part of Fig. 3 are observed in these positions by Stoffl *et al.*[10] and our coincidence data are in reasonable agreement with such a scheme. Some of our data would indicate 27.5- by 31.8-keV coincidences and place the 27.5-keV γ-ray feeding into the 99.9-keV level. This would result in a level at 127.46-keV whose γ-ray feeding would come from the proposed 143.5-keV level. It would also upset the intensity balance into the 68.2-keV level and leave the possible 11.98-keV γ ray unplaced.

The two most serious difficulties in utilizing the observed data to construct a level scheme involve the overall intensity balance and the position of the 35.8-keV γ ray. As may be seen from the conversion coefficients[17] tabulated in Table III substantial

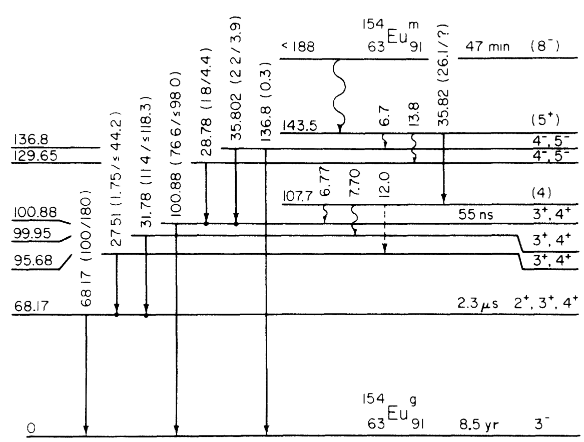


FIG. 3. Decay scheme of $^{154}Eu^m$. The solid lines represent firmly established γ rays, the dashed lines represent γ rays for which some evidence exists, and the wavy lines represent γ rays implied by the coincidence data and intensity balance. The values in parentheses represent γ-ray intensities and transition intensities calculated by using the conversion coefficients determined in Ref. 10. The new data described in the "note added in proof" would strongly indicate 2^+ and 4^+ spin and parity assignments for the 68.17- and 100.88-keV levels, respectively.

$E2$ admixtures in any of the observed low-energy transitions (or unobserved transitions) permit large quantities of intensity to flow to the 68.2- and 100.9-keV levels with little or no observed γ-ray emission.

Using the multipolarities indicated by Stoffl *et al.*[10] for the 27.5-, 28.8-, 31.8-, and 35.8-keV γ rays, it is clear that the total intensity of 278 feeding the ground state through the 100.9- and 68.2-keV γ rays cannot be accounted for.

As our placements for the 27.5-, 28.8-, and 31.8-keV γ rays are in accord with those of Stoffl *et al.*,[10] the placement of the 35.8-keV γ ray offers the only opportunity to account for the intensity into the ground state. Stoffl *et al.*[10] place an $E1$ (35.802 ± 0.011)-keV γ ray feeding the 100.9-keV level from a level at 136.675-keV. They also observe a 136.675-keV crossover transition to the ground state with an intensity 0.14 as large as the 35.802-keV γ ray. We also observe a weak γ-ray peak at 136.8 ± 0.3 keV, whose half-life is ~44 ± 4 min and its intensity is 0.3 ($I_{68.2} = 100$). If this γ ray is identified with the 136.675-keV transition observed in the capture γ-ray experiments, the assoicated intensity of the 35.802-keV γ ray would be only 2.2. This is far short of the total intensity of the peak at 35.818 ± 0.010 keV and far short of the intensity of the 35.8-keV peak in coincidence with the 100.0-keV γ ray. The 35.8-keV γ ray is ~10 times the intensity of the 28.8-keV peak in the spectrum in coincidence with the 100.9-keV γ ray. Thus, the 136.7-keV level cannot account for the total intensity of the 35.8-keV peak, its coincidence intensity, or the total intensity that must feed into the 100.9- and 68.2-keV levels.

TABLE III. Conversion coefficients for $^{154}Eu^{m}$ γ transitions (Ref. 17).

E_γ	$E1$	$E2$	$M1$
		α_K	
68	0.666	2.89	4.81
101	0.235	1.16	1.55
		α_L	
10	21.8	78 700	206
27.5	1.35	489	10.0
28.8	1.19	388	8.74
31.8	0.904	239	6.54
35.8	0.647	132	4.59
68.2	0.105	5.81	0.69
100.9	0.0035	0.94	0.22
		α_T	
10.0	26.7	96 400	250
27.5	1.64	601	12.2
28.8	1.45	478	10.6
31.8	1.10	294	8.0
35.8	0.78	163	5.6
68.2	0.80	10.5	5.7
100.9	0.28	2.4	1.8

The inclusion of a proposed level at 107.65 keV is suggested by the strong coincidences observed between the 35.8-keV peak and both the 100.9- and 68.2-keV γ rays. As it is clear that the intensity feeding into the 100.9-keV level from the 129.7- and 136.7-keV levels is inadequate, that the bulk of the intensity of the 35.8-keV peak must be placed so as to feed both the 100.9- and 68.2-keV levels, and that there is no highly converted transition between the 100.9- and the 68.2-keV levels, a level must exist that is fed by the 35.8-keV peak and decay to both the 100.9- and 68.2-keV levels. In the (d,p) reaction, a level is observed at 143 keV that is not observed in the capture γ-ray study. These characteristics suggest a high spin for the 143-keV level and permit us to suggest that it could participate in the deexcitation of a high-spin isomer. The presence of a weak γ ray at 11.98-keV permits us to propose a level at 107.65-keV that is ~35 keV below 143 keV. The proposed weak γ rays at 6.77 and 7.70 keV fall in the midst of the Gd, Eu, and Sm L x rays. Comparison of the γ-ray spectra and Eu fluorescence spectra do not rule out the existence of these γ rays. Possible transitions between the 143.5-keV level and the levels at 95.7, 99.9, and 100.9 keV also fall in the midst of the Gd, Sm, and Eu K x rays and cannot be ruled out.

The position of the isomer is also uncertain. As we observe no K x rays in excess of those arising from the $E1$ conversion of the 68.2- and 100.9-keV γ rays, we can conclude that the energy of the isomeric transition is less than ~40 keV above the 48.52-keV K edge in Eu. Below this energy, the decrease in γ-transition rate with decreasing energy is largely offset by the increase in conversion coefficient. Thus, the IT could be as small as 10 keV and as large as 88 keV. The analogous $E3$ transition in $^{152}Eu^{m2}$ was found by Takahashi *et al.*[2] to be highly hindered. Thus slight changes in the hindrance could easily offset any changes in the energy and conversion coefficient.

IV. SPIN AND PARITY ASSIGNMENTS

The ground state of ^{154}Eu is a 3^- state,[13] and the near equality of the isomer ratio for the (n,γ) reactions in ^{151}Eu and ^{153}Eu suggests an 8^- spin and parity assignment for the isomer. If the isomer is 8^- and the IT is $E3$, then the level fed by the isomer would be 5^+, the assignment suggested for the 143.5-keV level. Although both of these assignments appear to be quite speculative, a spin of <5 for the 143 keV level would permit an $E1$ or $M1$ transition to the ground state which was not observed ($I_{143}/I_{68} < 0.001$). As 7^- isomers are suggested for ^{154}Tb and ^{152}Pm, such an assignment could also be suggested for $^{154}Eu^{m}$. For such an assignment, direct $E3$ transitions to some of the possible 4^+ states near 100 keV would be anticipated. In fact, they would be almost necessitated by the long half-life of the isomer and the certainty that the isomer lies above 135 keV. The possibility of the 143-keV level being the isomer is limited by the need for the 35 keV to feed both the 100.8- and 68.2-keV levels. The possibility of $M2$ multipolarity for the isomer is limited by the long half-life of the isomer. The slow γ-ray transition rate for a low-energy $M2$ transition would be balanced by the increased conversion-electron rate. As the 68.2- and 100.9-keV transitions are $E1$ [9,10,14] (our observed K x-ray intensities are in the intensity that would be expected for the $E1$ conversion of these transitions), the levels at 68.2 and 100.9 keV must be 2^+, 3^+, or 4^+ levels. Stoffl *et al.*[10] report $E1$ multipolarity for a (28.814 ± 0.009)-keV γ ray that feeds the 100.9-keV level. If that γ ray is identified with the (28.78 ± 0.02)-keV γ ray that we see in coincidence with the 100.9-keV γ ray, the level at 129.6 keV would be negative parity with spin 4 or 5 to be fed by the 5^+ level above it. Stoffl *et al.*[10] report $M1+E2$ character for the 27.51- and 31.78-keV transitions. These are consistent with our intensity balances and indicate positive parity for the 95.7- and 99.9-keV levels. The parity of the proposed 107.65-keV level would be dependent upon the multipolarity of the 35.8-keV γ ray, and its spin is likely to be 4. If it were higher, we would expect strong feeding from the isomer (the possibility of weak feeding is not completely ruled out), and if it were lower, the 5^+ level would likely feed other levels with greater intensity.

V. DISCUSSION

The decay scheme of $^{154}Eu^{m}$ is characterized by the lack of any clear rotational character. Three or four of the observed transitions are $E1$ and represent p or n single particle transitions, and the two $M1+E2$ transitions both feed the same level so only one of them can be the lowest member of a rotational band. The presence of so many single-particle orbitals at these low energies is easy to account for by looking at the orbitals in $^{153}_{63}Eu$ and $^{153}Sm_{91}$. In ^{153}Sm, three odd-neutron Nilsson orbitals, $\frac{3}{2}^-[521]$, $\frac{11}{2}^-[505]$, and $\frac{3}{2}^+[651]$, are observed[18] below 100 keV, while $\frac{3}{2}^+[411]$, $\frac{5}{2}^-[532]$, and $\frac{5}{2}^+[413]$ orbitals are seen in ^{153}Eu below 103 keV. The $\nu\ \frac{11}{2}^-[505]$, $\Pi\ \frac{5}{2}^+[413]$ configuration suggested[2] for the 3^- and 8^- states in ^{152}Eu appear appropriate for ^{154}Eu also. The 35.8-keV transition which might be $M1+E2$ feeds out of the state at 143 keV which has considerable single-particle character.[11] The single-particle character of many of the levels is indicated by the long lifetimes and the lack of branching. The presence of only two ground state branches, both with long lifetimes, suggests that many of the excited states we observe have configurations in which neither the proton or neutron are the same as in the ground state.

Note added in proof: Recent studies of the conversion-electron spectrum of $^{154}Eu^{m}$ by Chu and Franz[19] have revealed a highly converted 32.61-keV $E2$ transition, set a limit of 0.6% for the $E2$ mixing of the 27.51-keV transition, and found $M1 + 0.9\%\ E2$ character for the 35.82-keV transition. The presence of the 32.61-keV $E2$ transition between the levels at 100.9 and 68.2 keV alleviates the intensity imbalance discussed above and would permit the 27.51-keV transition to feed the 99.95-keV level from a level at 127.46 keV. The new multipolarity for the 35.82-keV transition is in-accord with our discussion above and would indicate a transition intensity of ~218 in the place of the ? we show in Fig. 3, again alleviating the difficulties mentioned in the intensity balance. These new data would strongly indicate 2^+ and 4^+ spin and parity assignments for the levels at 68.2 and 100.9 keV, respectively.

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