A NEW ISOMERIC STATE IN EUROPIUM

by

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ABSTRACT

Proton bombardment of europium produced a new activity of 96 ± 5 minutes half-life. The only 96 minute radiation seen is a 93 ± 3 kev gamma ray. This is classified as an E1 transition from measurement of the K conversion coefficient (α_K = 0.266 ± 0.04). The 93 kev gamma ray is considered to be in cascade with an isomeric transition; the isomeric transition is assumed to be highly converted and, hence, unobserved in the gamma-ray spectrum. The isomeric level is assigned to Eu^{152} by chemistry, excitation curves and bombardments of enriched isotopes.

ACKNOWLEDGEMENTS

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In particular, I would like to express my thanks to the research directors, Dr. R. E. Bell - Director of the Radiation Laboratory - and Dr. T. M. Kavanagh, for their interest and advice regarding various aspects of the work.

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SECTION I INTRODUCTION

Nuclei with an odd number of protons and neutrons usually have a higher ground state energy than the neighbouring isobars, and hence such nuclei are seldom formed by β-decay of neighbouring isobars. For this reason the energy levels of odd-odd nuclei often cannot be studied by the more conventional techniques of nuclear spectroscopy. However, the presence of a long-lived isomeric state does permit study of levels below the isomeric level. A further method of study of odd-odd nuclei is, of course, the direct detection of prompt gamma rays which follow nuclear reactions.

During measurements on (p,pn) cross sections of europium a new activity was observed. Gamma ray spectra showed a strong 93 kev gamma ray. The half-life of the gamma ray activity was 96 minutes. This work shows that the activity is associated with a new isomeric state in the odd-odd nucleus Eu^{152} .

SECTION II PREVIOUS WORK

Mottelson and Nilsson[1)] noted that there is a large increase in nuclear deformation between nuclei with neutron numbers 88 and 90. This has prompted many studies[2,3)] of the decay of europium 152 (N = 89). These earlier experiments have indicated that Eu^{152} has a 9.3 hour isomeric state, and that this state and the 13 year ground state both decay by electron capture to samarium and by negative beta emission to gadolinium. No evidence of a 96 minute isomer has been reported. In the previous experiments, however, the sources were usually prepared by pile neutron bombardment, and in most cases there was a delay of several hours between the bombardment and examination of the source. Furthermore, the present experiments indicated that the 96 minute state has a high spin, and hence pile neutrons probably would not penetrate the angular momentum barrier.

Recently Draper and Fleisher[4)] reported on soft gamma rays from Eu^{152} following resonance neutron capture. They observed a 95 $\pm$ 4 kev gamma transition. This is probably the same transition that has been observed in the present work.

SECTION III EXPERIMENTAL METHOD AND RESULTS

1. Introduction.

In the following subsections details are given of the methods used to find various properties of the new activity produced by proton bombardment of europium and samarium.

The properties studied are:-

1. The half-life.
2. The energy of the observed gamma ray.
3. The K conversion coefficient of this transition.
4. The form of the excitation function for production of the new activity by proton bombardment of
   a) europium and
   b) samarium.
5. The conversion electron spectrum.

2. Irradiation and Target Preparation.

Targets were prepared by placing 2 to 10 mg of the dried rare earth oxide in a 1/16" diameter aluminium tube, 0.0005" thick, sealed at one end. The powder was packed down and the other end of the tube sealed. The targets were mounted vertically, or horizontally, in the synchrocyclotron for the bombardments. The beam current for most of the studies was approximately 0.5 μA . Bombarding times were 5 to 15 minutes for gamma ray studies, and 3 hours for conversion electron studies.

3. Chemical Separations.

Often target impurities are the cause of the presence of unexpected gamma rays. A chemical separation, using an ion exchange column[5)] followed by fluoride precipitation, on a fraction of a bombarded target of natural europium showed the new activity was in a rare earth. A further chemical separation, using weak reduction methods[6)], showed the activity originated from the europium fraction. It is concluded that the new activity originates from europium.

The normal procedure used for source preparation, once the activity had been assigned to europium, was to dissolve the contents of the aluminium tube, after bombardment, in dilute hydrochloric acid. The solution was heated to dryness. More acid was added, and the process repeated. These operations are necessary so that the great activity produced by the products of proton bombardment of oxygen is removed. Further separations were required when natural europium was bombarded since considerable activity is produced from the reaction $Eu^{151}(p,3n)Gd^{149}$ (refer to figure 1). Europium was separated from gadolinium using nascent-hydrogen[6]. Sources for gamma ray studies were prepared by placing the contents of the test tubes - after the separations - in vials, or by placing the active material on scotch tape and evaporating to dryness.

4. Gamma Ray Spectra - Energy and Half-Life Measurements.

A sample of 5 mg of 95% Eu^{153} was bombarded at 36 Mev for 16 minutes. After the bombardment the target was opened, dissolved in HCl and a source prepared for study of the gamma ray spectrum.

The sources were placed 10 cm from a sodium iodide scintillation counter (crystal size 3" x 3"). The pulses from the counter were fed through a cathode follower to a 256 channel kicksorter.

Initial spectra showed the presence of a prominent low energy gamma ray. The energy of this radiation was measured by comparison with three others of known energy from the same source. They are the 121.8 kev gamma ray from decay of 9.3 hour Eu^{152m}, the 149.9 kev gamma ray from decay of 9 day Gd^{149}, and the K X-rays from

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RELEVANT TO THIS STUDY

Gd 149 9 d	Gd 150 3.10^5y	Gd 151 120 d	Gd 152 0·20%	Gd 153 225 d	Gd 154 2·15 %	Gd 155 14·7%	Gd 156 20·5%
Eu 148 54 d	Eu 149 120 d	Eu 150 14 h	Eu 151 47·8%	Eu 152 9·3h | 13y	Eu 153 52·2%	Eu 154 16 y	Eu 155 1·7 y
Sm 147 15·0%	Sm 148 11·2 %	Sm 149 13·8%	Sm 150 7·4 %	Sm 151 93 y	Sm 152 26·7%	Sm 153 47 h	Sm 154 22·7%

FIGURE I.

Sm, Eu and Gd (41.0 to 44.0 kev). A typical spectrum is shown in figure 2. The energy was measured to be 93 ± 3 kev.

The half-life of this gamma ray activity was measured by subtracting the Compton continuum from under the photopeak and following the decay for several half-lives. A typical decay curve is presented in figure 3. The half-life has been measured several times using sources produced from proton bombardment of natural europium, enriched Eu^{153} and natural samarium. These measurements assign a half-life of 96 ± 5 minutes to the activity.

5. Excitation Functions.

Excitation curves were measured to verify that the activity was in europium and to determine the mass number of the isotope involved. Excitation curves were obtained for proton bombardment of samarium and europium.

a) Europium.

Six targets of natural europium (47.8% Eu^{151} and 52.2% Eu^{153}) weighing from 3 to 7 mg, were bombarded at energies of 10 to 36 Mev intervals, each for 5 minutes. The proton beam was monitored by reading the beam current on the "frog" inside the cyclotron tank.

Sources for study of the gamma ray spectra were produced. The activity associated with the 93 kev gamma ray was evaluated at

FIGURE 2.

LOW ENERGY SPECTRUM USED FOR ENERGY DETERMINATION

[SPECTRUM TAKEN 287 MINUTES AFTER END OF BOMBARDMENT]

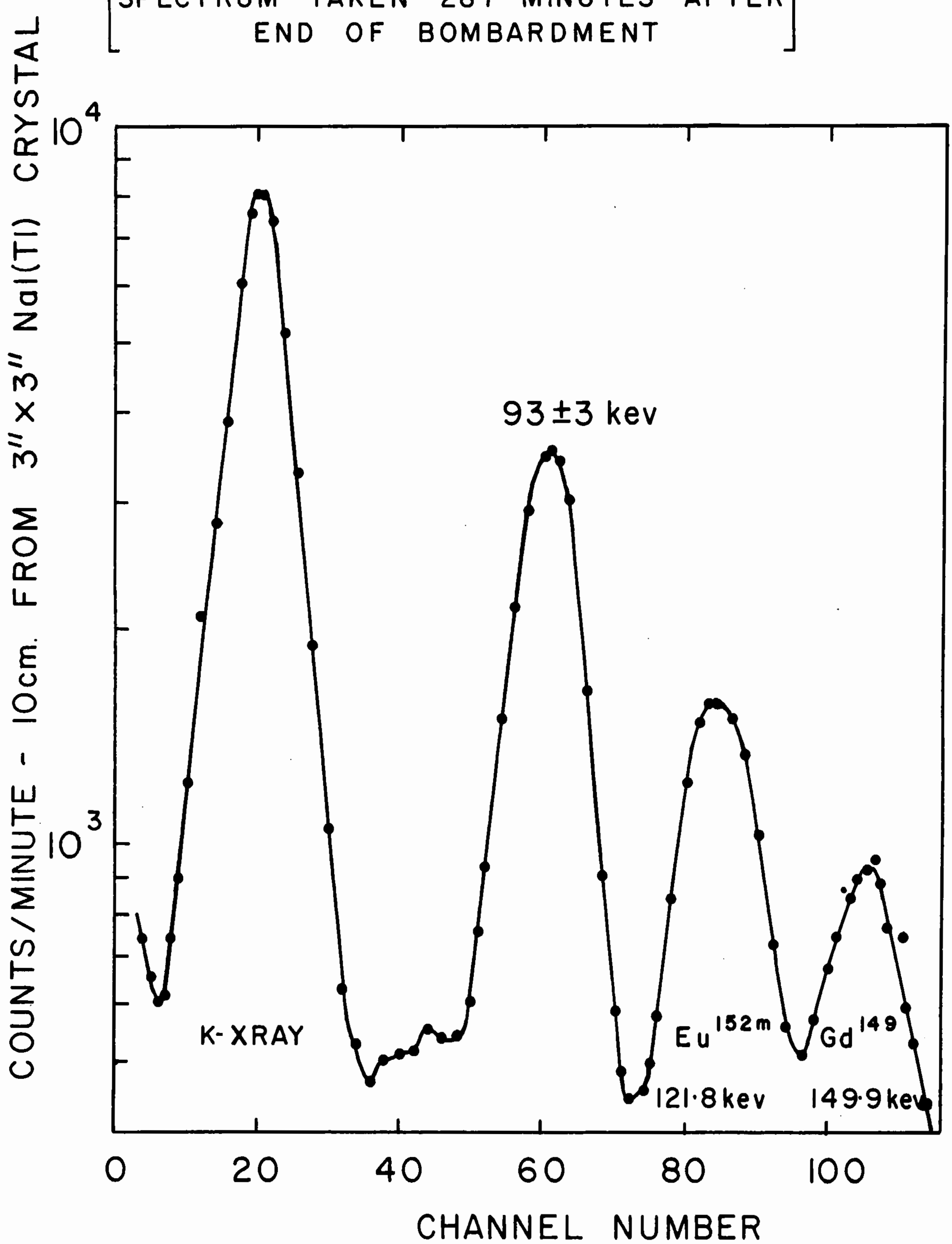
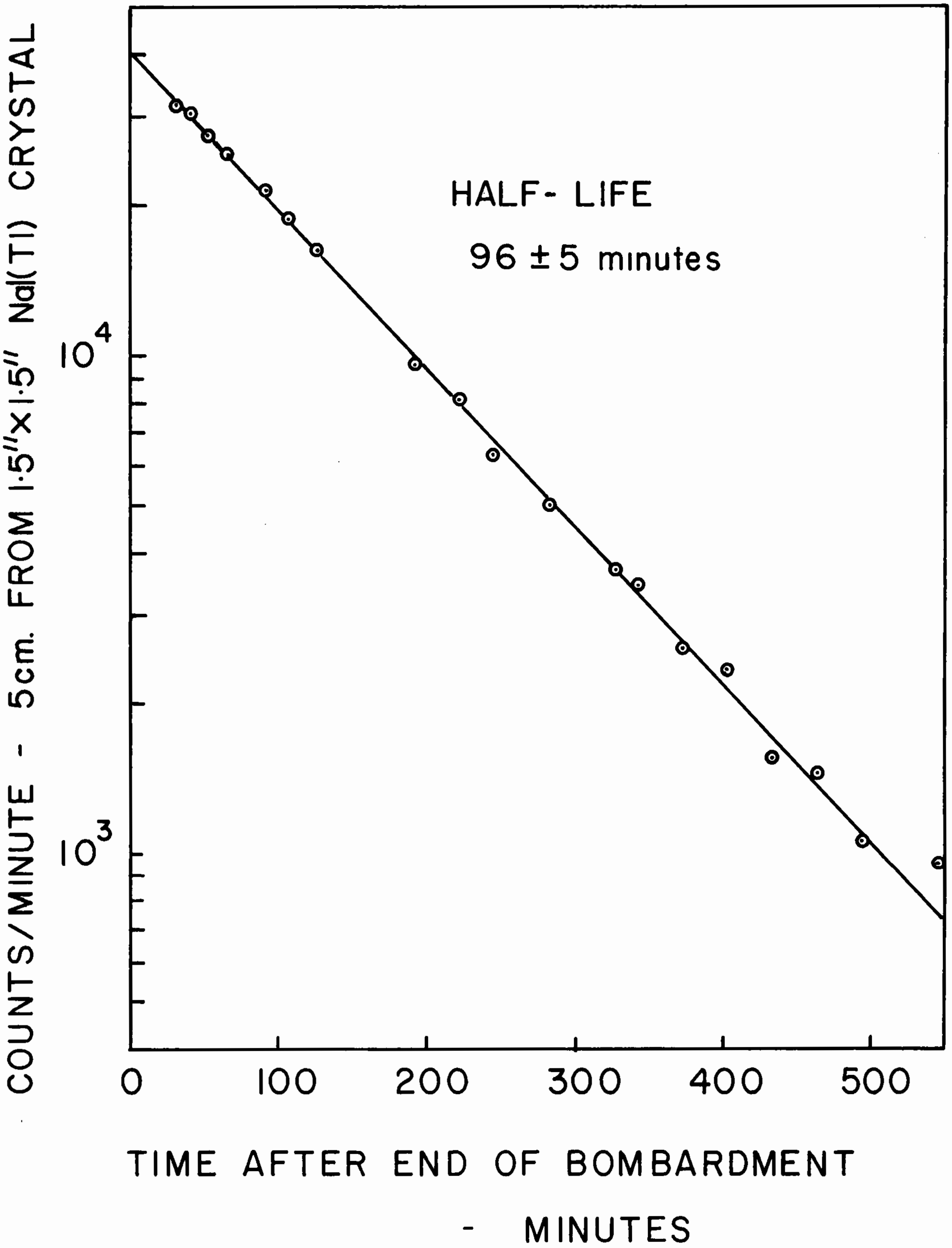


FIGURE 3.

TYPICAL DECAY CURVE

FOR 93 kev GAMMA RAY



80 minutes after the bombardment for each target. An excitation curve for the reaction $Eu^{151}(p,3n)Gd^{149}$ was also obtained from the source. This curve was based on measurements of the known 150 kev gamma ray from Gd^{149}, and served as a check on the experimental technique. Experimental results are shown in Table I and are represented graphically in figure 4.

TABLE I

Proton Energy Mev	Yield (Arbitrary Units) 93 kev	150 kev
10	0	0
15	0	0
19.8	94.5	85.8
25.2	209	420
29.6	449	684
35.5	689	474

The yield values have been normalized to constant target mass and to constant beam current as measured at a fixed orbit radius (i.e. that of the "frog"). The actual proton flux for the different bombardments decreases somewhat with increasing energy, but it is not expected that this will seriously influence the shapes of the excitation curves. The excitation curve for the reaction $Eu^{151}(p,3n)Gd^{149}$ has the shape expected for a (p,3n) reaction, and this serves as a rough check on the monitoring procedure.

EXCITATION FUNCTIONS FROM PROTON BOMBARDMENT OF NATURAL EUROPIUM

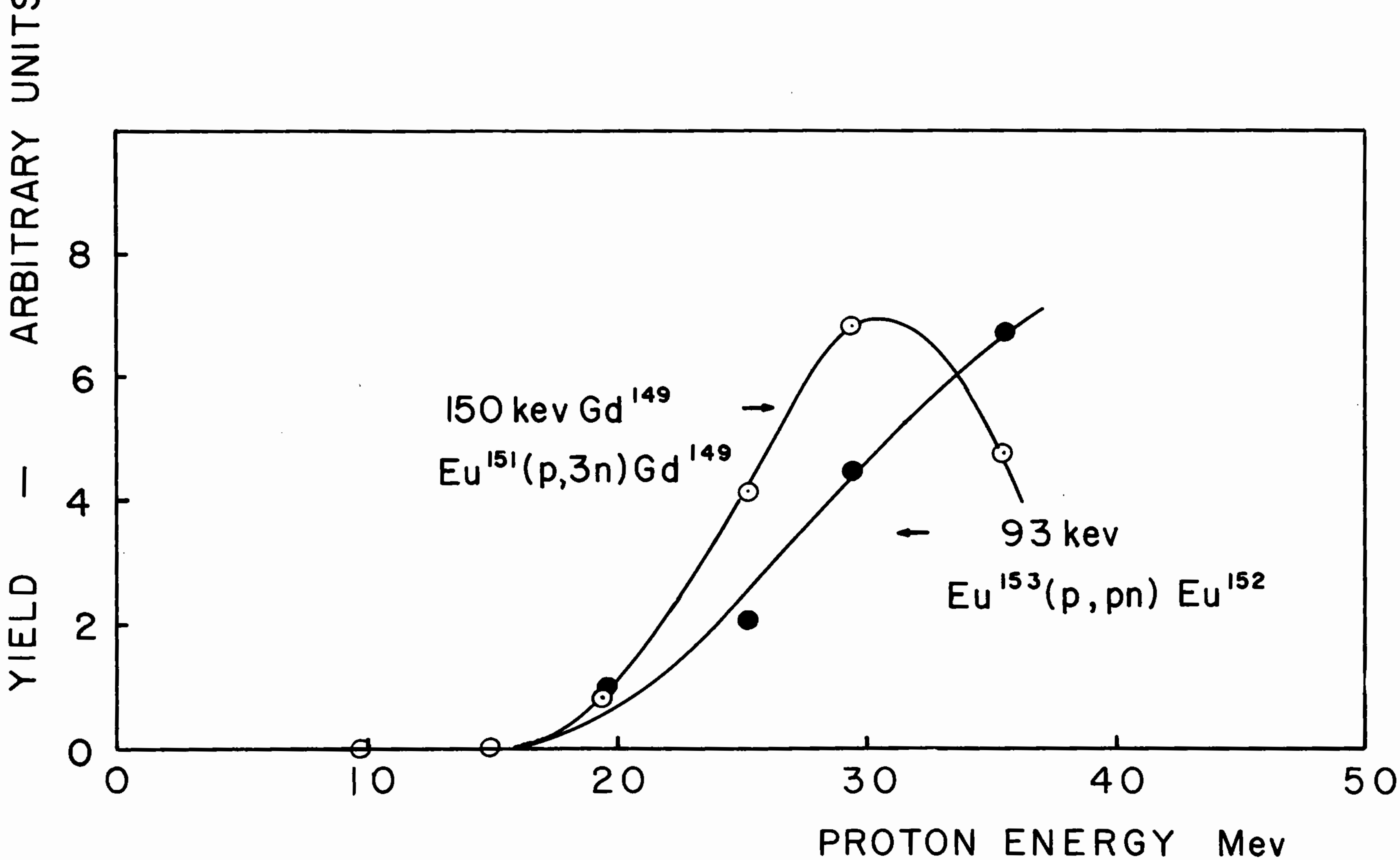


FIGURE 4.

The excitation curve for production of the 96 minute activity is a monotonically increasing function over the energy region 10 to 36 Mev. This is as expected since chemical analysis has indicated that the new activity is in europium - that is, the reaction involved here must be a (p,pxn) reaction. Comparison with (p,pxn) studies[7] indicate that the particular reaction involved is a (p,pn) reaction, and this establishes that the 96 minute activity is in either Eu^{152} or Eu^{150}.

In order to make a mass assignment, two bombardments were carried out at 30 Mev, using enriched targets (Eu^{153} - 95% enrichment, Eu^{151} - 91.9% enrichement). The 96 minute activity was produced in the bombardment of Eu^{153}, but not in the case of Eu^{151}. It is considered that the activity is due to a 96 minute isomeric state in Eu^{152}.

b) Samarium.

Six targets of natural samarium were prepared and bombarded under conditions similar to those described for europium. Three more targets were later prepared and bombarded at 22.4, 27.5, 32.6 Mev. These served as a check on the reproducibility of the method and enabled the form of the excitation function to be more clearly defined. As before, the yields were normalized to constant target mass and constant beam current as measured at a fixed radius. The activities were measured 160 minutes after each bombardment. The detector was a thin NaI crystal 0.080" thick and 1" diameter; the thin crystal was used to reduce the background due to Compton scattering of the high-energy gamma rays.

Samarium has seven stable isotopes, with Sm^{154} and Sm^{152} the most abundant (22.7% and 26.7% respectively). In view of the mass assignments made in the preceding subsection, it would be expected that the 96 minute isomer would be formed in the energy range 10 to 36 Mev by the reactions $Sm^{152}(p,n)Eu^{152m}$ and $Sm^{154}(p,3n)Eu^{152m}$. The sources used to obtain the excitation curve for the 96 minute state also yields an excitation curve for the well-known 9.3 hour isomer of Eu^{152} formed by the same reactions. The latter curve was based on a 122 kev gamma ray that was clearly resolved in these measurements from the 93 kev peak, and the curve was useful for comparison with that for the 96 minute state. Both curves are shown in figure 5 and the experimental values are summarized in Table II .

TABLE II

Proton Energy Mev	Yield (Arbitrary Units) 93 kev activity	122 kev activity
10	165	840
15	233	492
19.8	573	381
22.4	1600	735
25.2	2970	655
27.5	4060	1070
29.6	3930	792
32.6	1710	392
35.5	1180	477

EXCITATION FUNCTIONS FROM PROTON BOMBARDMENT OF NATURAL SAMARIUM

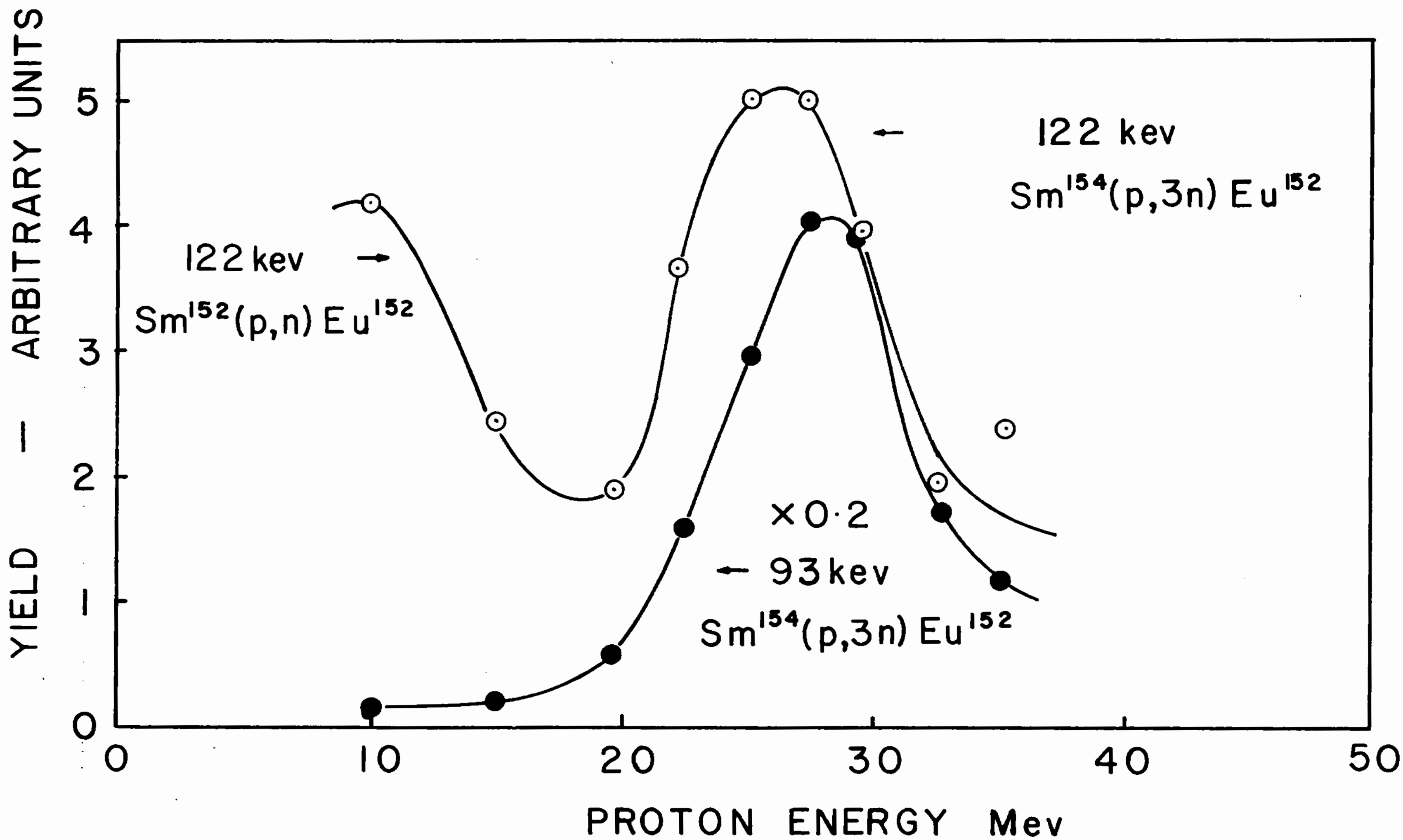


FIGURE 5.

The curve for the 9.3 hour state exhibits the usual peak for the (p,n) reaction at about 10 Mev and that for the (p,3n) at about 28 Mev. (This provides a very rough check on the monitoring procedure). It is interesting to note, however, that the curve for the new isomer does not exhibit the (p,n) peak, and the (p,3n) peak is 2 or 3 Mev higher in energy than for the 9.3 hour state. It is concluded that the 96 minute isomeric state has a much higher spin. The angular momentum barrier would prevent the formation of the high spin isomer by a (p,n) reaction, and the displacement of the (p,3n) curve to higher energy is in agreement with the assumption of large spin[8,9].

In a later section the isomeric state is tentatively assigned a spin of 7 –. The formation of the isomeric state by bombardment of samarium would require the incident proton to contribute 7 units of angular momentum. Classically this requires that the protons have energy in excess of 27 Mev. The results described here are consistent with the 7 - spin assignment.

It is to be noted that in the excitation curve for the 9.3 hour state the (p,n) peak is rather high with respect to the (p,3n) peak[10] even when allowance is made for the difference in abundances of Sm^{152}(26.8%) and Sm^{154}(22.7%). This is probably due to the procedure used for beam monitoring.

6. K Conversion Coefficient.

The K conversion coefficient of the 93 kev transition was measured to determine its multipolarity.

The source used for measurement of the transition energy was used for this study. The low energy spectrum was recorded for many days after the bombardment. The activities associated with the K X-rays and the 93 kev gamma ray were studied. The long-lived components in the K X-rays were removed resulting in a 96 minute component. Decay curves are shown in figure 6. Assuming all these K X-rays result from internal conversion of the 93 kev transition we may evaluate the corresponding K conversion coefficient.

The K conversion coefficient is defined by the following equation,

$$\alpha_K = \frac{N_{Ke}}{N_\gamma} = \frac{N_{KX}}{N_\gamma W_K} .$$

This may be rewritten $= \frac{N_{mK}}{N_{m\gamma}} \frac{E_\gamma}{E_K} \frac{e^{(\rho_K - \rho_\gamma)x}}{W_K} \frac{(1 + P_K)}{(1 + P_\gamma)}$

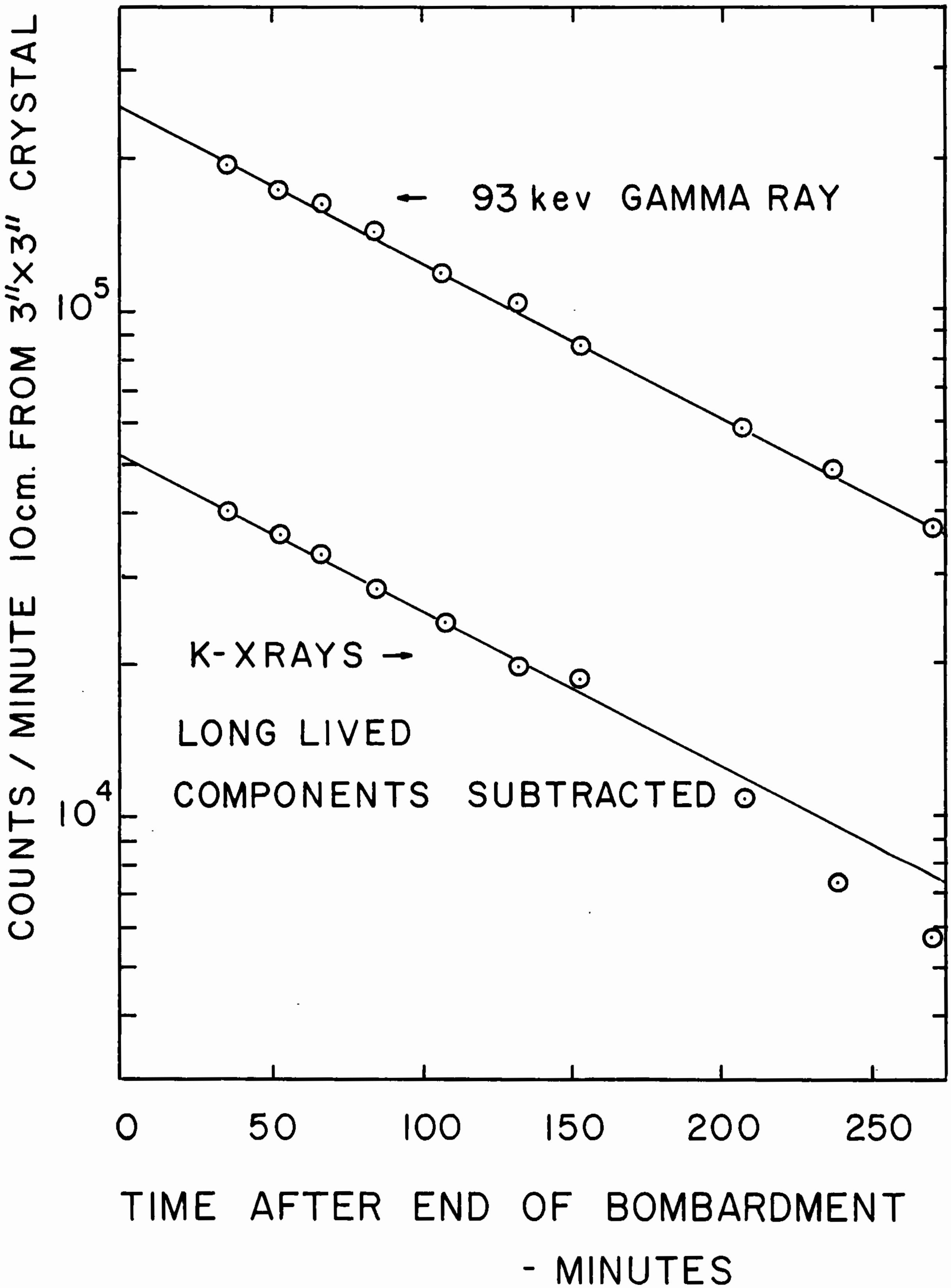
where

α_K K conversion coefficient

N_{Ke} Number of K conversion electrons/unit time associated with transition

N_γ Number of photons from same sample/unit time measured at same time as N_{Ke}

FIGURE 6.
DECAY CURVES OF 93kev GAMMA RAY
AND ASSOCIATED K-XRAYS



W_K K fluorescent yield

N_{mK} "Photopeak" activity for K X-rays associated with transition

$N_{m\gamma}$ "Photopeak" activity for gamma-ray measured at same time as N_{mK}

E_γ Crystal efficiency for photons

E_K Crystal efficiency for K X-rays

x Thickness of aluminium can

ρ_K, ρ_γ K X-rays and the gamma ray attenuation coefficients for aluminium

P_K, P_γ Ratio of escape peak to photopeak

TABLE III

Quantity	Value	Unit	Ref.
N_{mK}	40.4×10^3	counts/minute	-
$N_{m\gamma}$	196×10^3	counts/minute	-
E_γ	0.0320	-	11
E_K	0.0325	-	11
ρ_K	0.051	cm^2/gm	12
ρ_γ	0.0176	cm^2/gm	12
x	206×10^{-3}	gm/cm^2	-
W_K	0.917	-	13
P_K	0.272	-	13
P_γ	0.065	-	13

From these observations $\alpha_K = 0.266 \pm 0.04$.

From theoretical tables[14) the following values are obtained for the K conversion coefficients for a nucleus with Z = 63 and E1 transitions only.

TABLE IV

Energy kev	α_K
96	0.26
93	0.29
90	0.31

K conversion coefficients are at least a factor of 4.8 higher for all other multipolarities. By comparison of the experimental and theoretical values it is seen that the 93 kev transition may be assigned an E1 multipolarity. Also, as this is the lowest K conversion coefficient for a 93 kev transition in a nucleus with X = 63, this is strong evidence for believing that any other 96 minute activity is not highly converted in the K shell.

7. The Conversion Electron Spectrum

Measurement of the energy difference between the K and L conversion electrons determines the atomic number of a nucleus in which a transition originated. From the relative intensity of the L conversion lines often an assignment may be made as to the multipolarity of the transition. A study of the conversion electron spectrum is, therefore, a powerful method for study of nuclear structure.

The purpose of this investigation was to determine the atomic number of the nucleus in which the 93 kev transition is converted, and to search for conversion lines due to possible highly converted transitions not observed in the gamma ray spectrum.

A 180^{0} spectrograph was used for the study[15]. The magnetic field was calibrated using the standard thorium conversion lines[12]. The magnetic field was measured to 0.1%, the value being approximately 93 gauss. This permitted a possible study of conversion electrons in the range from 20 kev to over 160 kev.

About 8 mg of natural samarium oxide was bombarded at 27 Mev for 3 hours. Carrier free europium was prepared from this target using the ion exchange method. (Mr. G. S. Rayudu performed the separation). A source was prepared by pipetting the solution, containing the carrier free europium, in a 15 mil x 10 mil x 1/2" groove cut in an aluminium block and evaporating to dryness. This method of source preparation has not proved to be very effective. Further work on source preparation using electroplating methods[16] are being investigated. Further work had to be halted for cyclotron shut down.

One conversion line was observed on the last run on the spectrograph. The energy of the line was measured to be

$$54.85 \pm 0.1 \text{ kev.}$$

This observation is to be regarded as a preliminary measurement as the source preparation is not considered to be well developed. Discussion of the possible origin of this line is given in the next section.

SECTION IV PROPOSED DECAY SCHEME FOR Eu^{152}

The present study indicates there is an isomeric state in Eu^{152} of half-life 96 minutes. The 93 kev gamma ray observed to decay with this half-life is shown to be an El transition, and therefore unlikely to be responsible for the life-time. (The Weisskopf estimates indicate a mean-life of 2.5×10^{-13} sec for 93 kev El transition). There is probably a transition of high multipolarity preceding the 93 kev transition. The isomeric level would thus be a state of high spin, and this is in agreement with observations on the excitation functions produced by bombardment af natural samarium.

This proposed transition must be highly converted in shells other than the K shell. Measurements described earlier set an upper limit of 0.27 ± 0.04 for the K conversion coefficient of the 93 kev transition. This, according to Sliv and Band[14)], is the lowest K conversion coefficient that may be assigned to a transition of this energy. Thus, it is concluded there must be a very small K conversion intensity for any other 96 minute transition.

Studies of the conversion spectra indicated the presence of a 54.85 kev conversion line. This can not be assigned to the 93 kev transition, but it may be an L conversion line of the isomeric transition. We may then predict that the isomeric transition has an energy of 62.4 ± 0.5 kev.

We now investigate the various multipolarities that could be assigned to this level to be consistent with the present experimental observations. For this we require the conversion coefficients for a nucleus with Z = 63 and a photon energy of 62 kev[14].

TABLE V

	α_1	α_2	α_3	α_4	β_1	β_2	β_3	β_4
K	0.85	3.4	10.0	28	5.6	70	330	1,040
L_1	0.072	0.28	2.1	54	0.67	16.5	250	3,400
L_2	0.023	3.7	175	4,600	0.054	1.7	34	600
L_3	0.031	4.1	200	5,200	0.011	4.8	380	13,000
M	0.04	2.7	125	3,300	0.24	6	221	5,600
Total	1.01	14.2	512	13,200	6.6	99	1,220	23,600
$\alpha_K/\alpha T$	0.83	0.24	0.024	0.002	0.85	0.71	0.27	0.04

We may reject El, E2, Ml, M2 and M3 transitions due to the large $\alpha_K/\alpha T$ ratio. However, E3, E4, and M4 are consistent with experimental observation of a small $\alpha_k/\alpha T$ ratio and high total conversion coefficient.

Using the Weissope estimates of γ-ray emission it is found that the theoretical half-life is 0.14 sec for E3 transition.

Then $\log\left\{\frac{(T_{exp.})}{(T_{theo.})}\right\} = 4.6$.

Comparison with previous data[12] shows this value is similar to other values found for E3 transitions. We assign the isomeric transition an E3 multipolarity.

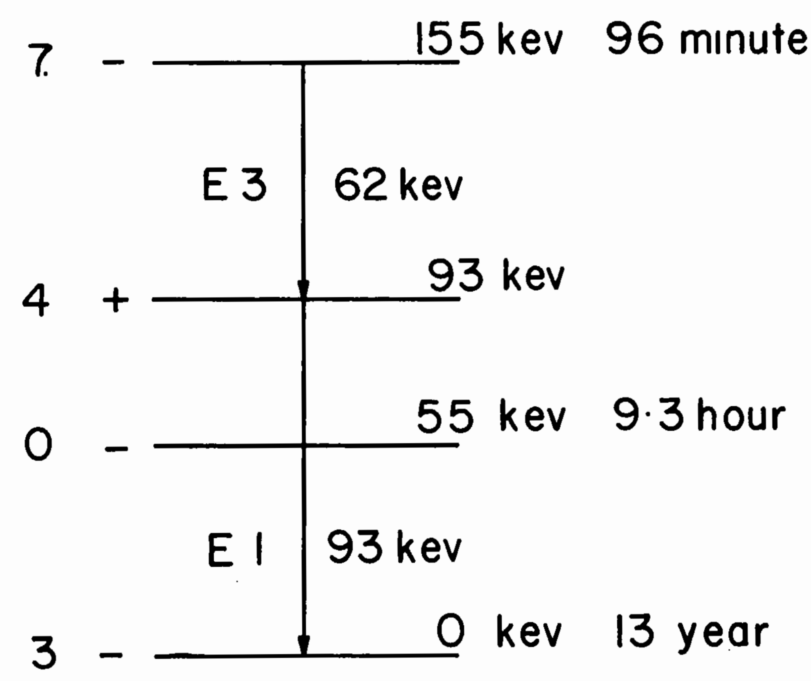
The 93 kev transition may be assigned to one of the isobars of mass number 152 in Sm, Eu or Gd. However, if we assume the prompt 95 ± 4 kev gamma ray seen from Eu^{152} following resonance neutron capture[4] is the same gamma ray observed from de-excitation of the proposed isomeric state in Eu^{152}, we may assign the 93 kev transition to Eu^{152}.

Under these assumptions we may propose a tentative decay scheme for the new isomeric state. (see figure 7).

The 96 minute activity is considered to feed the 13 year ground state for two reasons. If the 96 minute activity feeds the 9.3 hour level, the proposed isomeric state would be a 4- level, and it would decay by an E2 or M1 transition to the 3- ground state (that is, the lifetime would be very short, and the 93 kev transition would not be

FIGURE 7.

PROPOSED DECAY SCHEME



Eu^{152}

observed). Furthermore, if the 96 minute activity does feed the 9.3 hour level, the 9.3 hour activity would show a perceptible growth in the first few hours after bombardment. Rough preliminary measurements have indicated no such growth.

SECTION V CONCLUSION

A hitherto unobserved isomeric state of half-life 96 ± 5 minutes is assigned to the odd nucleus Eu^{152} .

A 93 ± 3 kev gamma ray is observed to follow the isomeric transition. The K conversion coefficient of the gamma ray shows that the 93 kev gamma ray is an E1 transition.

Conversion electron studies indicate that the 93 kev transition may be fed by a highly converted E3 transition of energy 62.4 ± 0.5 kev.

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