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Isomeric Transitions in the Rare-Earth Elements*

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A search was made for isomeric transitions in the rare-earth elements with half-lives greater than 5 microseconds and less than ~30 minutes which arise in the decay of photoexcited nuclei. Four isomeric transitions were found and identified as follows: Tb^{158}, 50 kev$<E<$200 kev, 11.0±0.1 sec, $M3$ or $E3$; Ho^{163}, 305 kev, 0.8±0.1 sec, $E3$; $\mathrm{Er}^{165,167,\text{ or }169}$, 213 kev, 2.5±0.1 sec, $E3$; Lu^{174}, 133 kev, 75±2 μsec, $E2$. These results are compared with the predictions of the nuclear models which have been proposed by Nilsson and Gottfried.

I. INTRODUCTION

In a previous paper[1] a method was reported for finding isomeric transitions which arise in the decay of nuclei which have been excited by the x-radiation from the 75-Mev Iowa State College synchrotron. The purpose of this paper is to report on the results of the continuation of the search for isomeric states in the region of strongly deformed nuclei. The search of this region was undertaken because of the current interest in the nuclear models proposed by Nilsson[2,3] and Gottfried[4] which apply to nonspherical nuclei. These models have had considerable success in explaining the ground state spins and parities of the nuclei in the region $150<A<190$. It would be of further interest to test these models for their ability to predict spins and parities of low-lying, single particle excited states. La and all of the rare-earth elements, except Eu and Pm, were used in this investigation since these nuclei represent a considerable portion of the region for which Nilsson's and Gottfried's calculations are valid. A knowledge of the energy, lifetime, and conversion coefficient of the isomeric transition is sufficient to assign a spin difference between the ground state and the excited state.

II. EXPERIMENTAL PROCEDURE

The apparatus has been described in detail in a paper by Bureau and Hammer.[1] The targets used in the initial search were 1 to 1.5 grams of the rare-earth oxide pressed into a disk 1.2 inches in diameter. The disks were then mounted in a thin Lucite holder 0.025 inch thick and covered with 0.001 inch of aluminum foil.

The energy and short half-life measurements were made as previously described[1] by counting between beam bursts. However, for activities with half-lives greater than 10 milliseconds, the beam was turned on for a given number of cycles, then turned off for a given number of cycles. The phototube in this case was turned on about 5 milliseconds after the last beam burst and remained on for some desired time interval. The half-lives were measured using either a successively longer counting time interval or using an events per unit time counter which records the counts in successive one-second intervals.

III. RESULTS

Isomeric transitions were found for the following cases: Tb, Ho, Er, and Lu. These cases will be discussed separately. All of the data from the Lu was obtained using the original oxide target, but the data from the other three elements were obtained using pure metal targets.

A. Terbium

The spectrum of the activity obtained by bombarding a Tb target is shown in Fig. 1. The prominent peak is

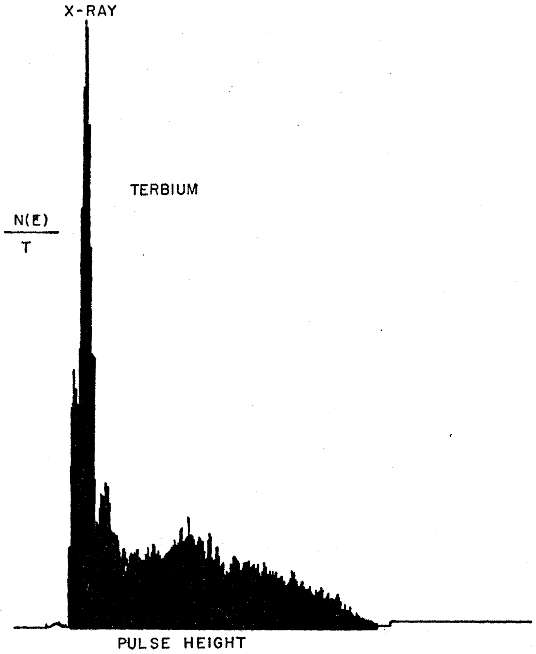

Fig. 1. Terbium activity. This is a spectrum that was plotted automatically by the pulse-height analyzer. The prominent peak is the Tb K x-ray. No other gamma rays could be detected above the background.

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* Contribution No. 517. Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

[1] A. J. Bureau and C. L. Hammer, Phys. Rev. **105**, 1006 (1957).

[2] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 16 (1955).

[3] B. R. Mottelson and S. G. Nilsson, Phys. Rev. **99**, 1615 (1955).

[4] Kurt Gottfried, Phys. Rev. **103**, 1017 (1956).

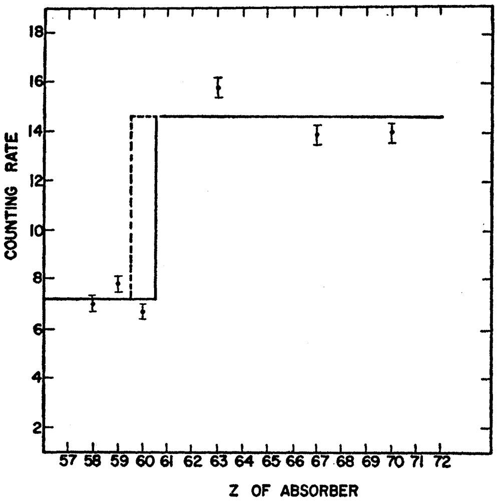

FIG. 2. Critical absorption of the Tb K x-ray. The dashed line is the expected transmission curve for the Gd x-ray and the solid line is the expected transmission curve for the Tb x-ray. The error flags represent statistical errors only.

the Tb x-ray while the smaller peak just above the x-ray is due to the background. No gamma rays could be detected above the background.

The half-life was measured as previously described (over a time interval of five half-lives) and was found to be 11.0 ± 0.1 sec.

The experimental data could result from two different decay schemes. One would be an 11-sec isomeric transition in Tb that is very highly converted, and the other would be an electron capture transition to the ground state of a Gd isotope. In the former case the Tb x-ray would be observed, while in the latter case the Gd x-ray would be observed. One can distinguish between the two by using critical absorbers. The results are shown in Fig. 2. The dashed line is the expected transmission curve for the Gd x-ray as a function of the Z of the absorber and the solid line is the expected transmission curve for the Tb x-ray. The results show that the x-ray is indeed the Tb x-ray and hence results from an isomeric transition and is not the Gd x-ray following electron capture. The possibility of this being a 45-kev transition and not the K x-ray can be ruled out, since for this case the gamma-ray would be almost entirely converted in the L shell and only the 6-kev L x-ray would be observed.

The threshold for the reaction was measured and the results are shown in Fig. 3. The abscissa represents the peak energy of the synchrotron beam and the ordinate represents the relative yield. The observed threshold of approximately 9.8 Mev corresponds to a (γ,n) reaction.

Since Tb is a single isotope of mass 159, the isomeric transition therefore occurs in Tb^{158}. This is an odd-odd isotope (65 protons and 93 neutrons) with a deformation of $\delta=0.31$,[5] if one assumes the deformation to be the same as that of Tb^{159}.[3] Nilsson's calculations[3] predict that the proton is in the $d_{\frac{5}{2}}(\Omega=\frac{3}{2}^+)$[6] state and that the neutron is in the $h_{9/2}(\Omega=\frac{3}{2}^-)$ state or possibly the $i_{13/2}(\Omega=\frac{5}{2}^+)$ state. One expects the ground state for an odd-odd nucleus to be a doublet with the spins given by

$$\Omega=|\Omega_p\pm\Omega_n|.$$

If the neutron is in the $\frac{3}{2}^+$ orbit, a ground state doublet with spins of 0^- and 3^- results. If the neutron is in the $\frac{5}{2}^+$ orbit, this would give rise to a 1^+, 4^+ doublet. Both of these cases predict an $M3$ transition between the doublet levels. Gottfried also predicts the same 0^-, 3^- doublet as Nilsson does. The observed half-life and large conversion coefficient are compatible with either a 50-kev to 200-kev $M3$ transition or a 50-kev to 100-kev $E3$ transition.

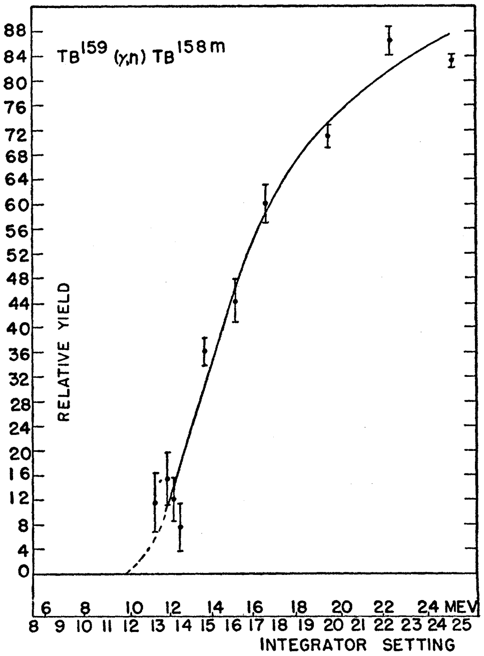

FIG. 3. Activation curve for Tb activity.

B. Holmium

The spectrum obtained by bombarding a Ho target is shown in Fig. 4. The background, taken with a Ta target, is shown as the lower curve. The energy of the gamma-ray is 305 kev as shown in Fig. 5 which is an energy calibration curve. There is some background not associated with the 305-kev activity that arises from the sample and could not be subtracted out.

The half-life was found to be 0.8 ± 0.1 sec. Figure 6 shows the counting rate as a function of the counting gate width. A least-squares fit to the curve

$$N(t)=N_0(1-e^{-\lambda t})+Ct$$

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[5] The notation of references 2 and 3 is used.

[6] Ω is the projection of j on the nuclear axis.

was made, where $N(t)$ is the number of counts recorded in a time interval t, λ is the decay constant, and C is a constant which is proportional to the amount of long-lived activity that arises from the target.

The threshold was measured to be 16.6 Mev as shown in Fig. 7. This corresponds to either a $(\gamma,2n)$ or a (γ,d) reaction. Since Ho is a single isotope of mass 165, the $(\gamma,2n)$ reaction would place the isomeric state in Ho^{163} and the (γ,d) reaction would place it in Dy^{163}. However, Dy^{164} is a stable isotope and if the 0.8-sec isomeric state were in Dy^{163}, we should have observed it from the $\mathrm{Dy}^{164}(\gamma,n)\mathrm{Dy}^{163}$ reaction. Since we did not, we assume that the reaction we observed was $\mathrm{Ho}^{165}(\gamma,2n)\mathrm{Ho}^{163m}$. Unfortunately the x-ray energy could not be determined since the 37-minute Ho^{164} activity gives rise to a Dy x-ray.

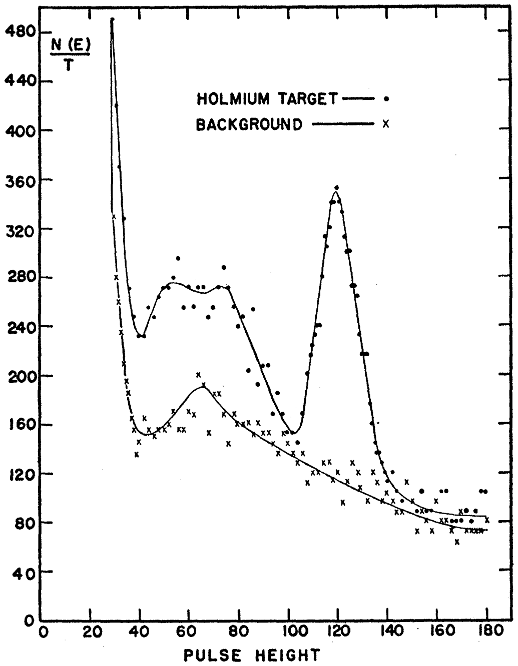

Fig. 4. Holmium activity. The lower curve is the background taken with a Ta target.

Ho^{163} is an odd-proton isotope with 96 neutrons and 67 protons, and thus the last unpaired proton will determine the spins of the single particle states. Assuming the deformation of Ho^{163} to be approximately equal to that of Ho^{165}, Nilsson calculates that the proton will be in a ground state $h_{11/2}(\Omega=\frac{7}{2}^-)$. At a slightly higher energy lies a $d_{\frac{3}{2}}(\Omega=\frac{1}{2}^+)$ state. A transition between these levels would correspond to $E3$ radiation. The observed energy and half-life are consistent with either $E3$ or $M3$ radiation. However, almost the entire x-ray peak can be attributed to the x-rays following the decay of 37-minute Ho^{164}, if the ratio of (γ,n) to $(\gamma,2n)$ cross sections is assumed to be ≥ 5. Since the theoretical conversion coefficients for $E3$ and $M3$ radiation for $E=305$ kev and $Z=67$ are 0.14 and 1.36,[7] respectively, the 305-kev transition must be $E3$ radiation.

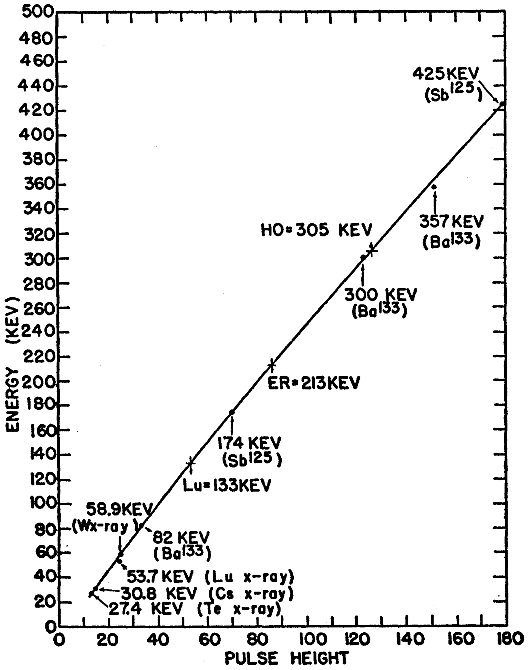

Fig. 5. Energy calibration curve.

Gottfried predicts a spin of $h_{11/2}(\Omega=\frac{7}{2}^-)$ for the ground state and spins of $f_{7/2}(\Omega=\frac{1}{2}^-)$ and $d_{\frac{3}{2}}(\Omega=\frac{1}{2}^+)$ for the first and second excited states, respectively. This scheme is consistent with an $M3$ but not an $E3$ transition since the $\frac{1}{2}^+$ state would depopulate by an $E1$ transition to the $\frac{1}{2}^-$ state. However, if the $\frac{1}{2}^-$ level did not cross below the $\frac{1}{2}^+$ level, then the $E3$ transition would be predicted and not the $M3$.

C. Erbium

The spectrum obtained by bombarding an Er target is shown in Fig. 8 with the background subtracted out.

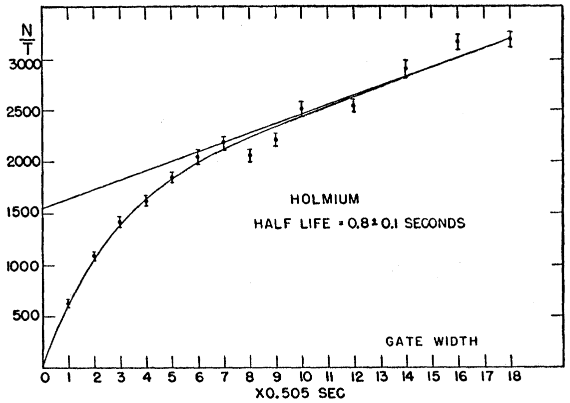

Fig. 6. Half-life of the 305-kev Ho activity.

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[7] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023, 1951 (unpublished).

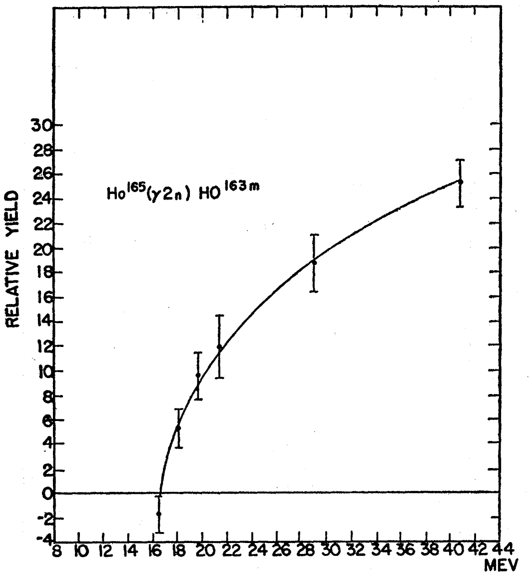

FIG. 7. Activation curve for the 305-kev gamma ray in Ho.

The gamma-ray has an energy of 213 kev as shown in Fig. 5 and the lower energy peak is the x-ray. The half-life was found to be 2.5±0.1 sec. Figure 9 shows the counting rate as a function of the counting time interval. A least-squares fit was made as in the case of Ho. An estimate could be made of the K-conversion coefficient by taking the ratio of the counts in the x-ray peak to the counts in the gamma-ray peak. This ratio was then corrected for the differences in efficiencies, peak-to-total ratios, fluorescence yield, and scattering[8] to find the desired coefficient. The measured conversion coefficient was $e_K/\gamma \sim 0.6$. The lifetime[9] and conversion coefficient[7] are consistent with only an $E3$ transition.

A 2.5-sec, 210-kev, $E3$ transition has previously been observed in Er following slow-neutron capture.[10,11] The five stable isotopes of Er are shown in Table I along with their percent abundances.

The isotopes that can be reached by (n,γ) and either (γ,γ') or (γ,n) reactions are 163, 165, 167, 168, and 169.

TABLE I. Stable isotopes of erbium.

Isotope	Percent abundance
162	0.1
164	1.6
166	33.4
167	22.9
168	27.1
170	14.9

The possibility of the isomer being in 163 can be ruled out because of the low abundance of the 164 isotope. Our targets contained only 15 mg of this isotope which is an insufficient amount for our purposes. The 168 isotope can be ruled out since it is an even-even nucleus and therefore has spins of 0^+ and 2^+ for the ground and first excited rotational states, respectively. An $E3$ transition to the ground state requires the excited state to have a spin of 3^-. However, since the 213-kev level is expected to be ∼100 kev above the 2^+ rotational state, the 213-kev level would decay by $E1$ radiation to the 2^+ level instead of by $E3$ radiation to the ground state. If the isomer were in the 167 isotope, this should also be reached by a (γ,d) reaction on Tm^{169}. Since this reaction may be only about one percent of the (γ,n) reaction in this region,[12] it would be too small for us to observe. Thus the isomer could be in either Er 165, 167, or 169.

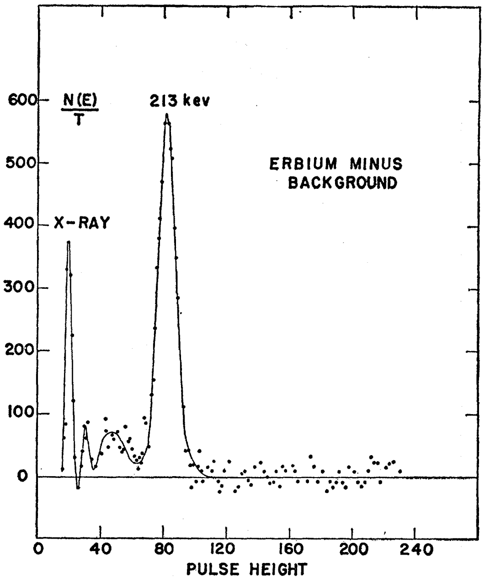

FIG. 8. Erbium activity. The background taken with a Ta target has been subtracted out.

For a deformation of $\delta \sim 0.28$, Nilsson predicts ground state orbits of $11/2^-$, $\frac{1}{2}^-$, and $\frac{7}{2}^+$ for the 97th, 99th, and 101st neutrons, respectively. The only level ordering consistent with the observed results is a $\frac{7}{2}^+$ orbit for the ground state and a $\frac{1}{2}^-$ orbit for the excited state. If the ground state had a spin of $\frac{1}{2}^-$, then the first excited rotational state would have a spin of $\frac{3}{2}^-$. Since the energy of the first excited rotational state is about 100 kev, a $\frac{7}{2}^+$ single particle excited state would decay by $M2$ radiation to the $\frac{3}{2}^-$ state rather than by $E3$ radiation to the ground state.

The ground state spin of Er^{167} has been measured as $\frac{7}{2}$.[13] Thus Nilsson's scheme can be made compatible

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[8] A. C. G. Mitchell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), pp. 224 ff.

[9] S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XIII.

[10] E. der Mateosian and M. Goldhaber, Phys. Rev. **76**, 187 (A) (1949).

[11] Campbell, Kahn, and Goodrich, Oak Ridge National Laboratory Report ORNL-1164, 1951 (unpublished).

[12] A. K. Mann and J. Halpern, Phys. Rev. **82**, 733 (1951).

[13] B. Bleaney and H. E. D. Scovil, Proc. Phys. Soc. (London) **A64**, 204 (1951).

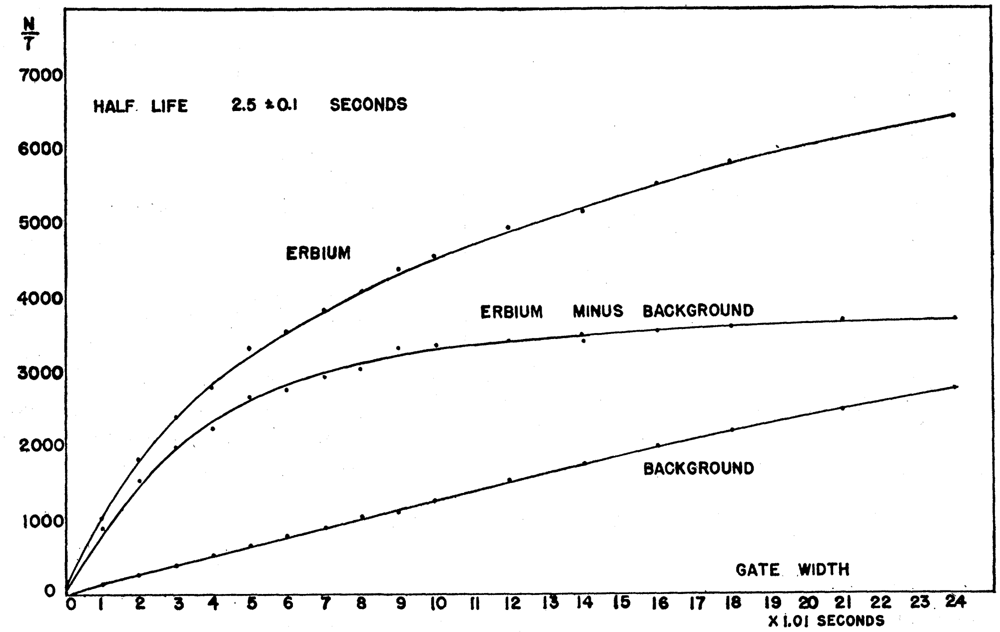

FIG. 9. Half-life of the 213-kev Er activity.

with this assignment by simply interchanging the $\frac{7}{2}^+$ and the $\frac{1}{2}^-$ levels; that is, by moving the point where the $\frac{1}{2}^-$ level crosses below the $\frac{7}{2}^+$ level out to a larger deformation. Depending on the exact crossing points of the three levels in question and on the exact deformation of the three isotopes, Nilsson's model can easily be made consistent with an $E3$ transition in either Er 165, 167, or 169.

Gottfried's model also can easily explain the isomeric transition, provided it is in either Er 165 or 167. If it is in Er^{169}, the assignment is not as good since in this case the $11/2^-$ orbit would have to be depressed several Mev.

If separated Er isotopes were used, the ambiguity in the assignment could easily be resolved.[14] Furthermore, since there is only one isomer present, the choice of the $\frac{7}{2}^+$ ground state and $\frac{1}{2}^-$ excited state can apply to only one of the isotopes.

D. Lutecium

The spectrum obtained by bombarding a Lu target is shown in Fig. 10. The background, taken with a La target, is shown as the lower curve. The gamma-ray has an energy of 133 kev and the x-ray was shown to be the Lu x-ray by using critical absorbers. The energy determinations of the gamma-ray and x-ray peaks are shown in Figs. 5 and 11, respectively. The half-life was found to be 75±2 microseconds by counting over five half-lives. An activation curve is shown in Fig. 12. The threshold for the reaction is about 8.0 Mev, which corresponds to a (γ,n) reaction.

Lu has only two stable isotopes, 175(97.5%) and 176(2.5%), and on the basis of these abundances we

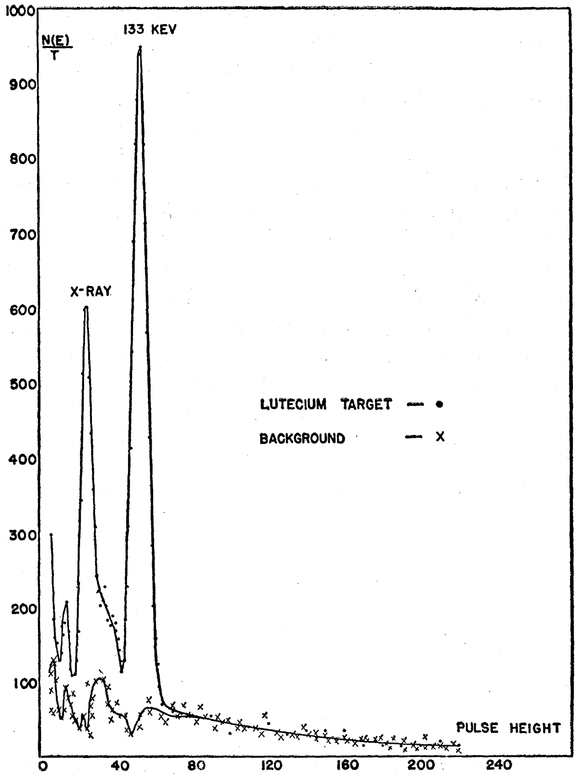

FIG. 10. Lutecium activity. The lower curve is the background taken with a La_2O_3 target.

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[14] J. W. Mihelich has communicated to us that he has observed a 208-kev $E3$ transition in Er^{167} following the electron capture decay of Tm^{167}. Since a half-life on the order of one second is expected for this $E3$ transition, there is little doubt that this is the same transition that we observe.

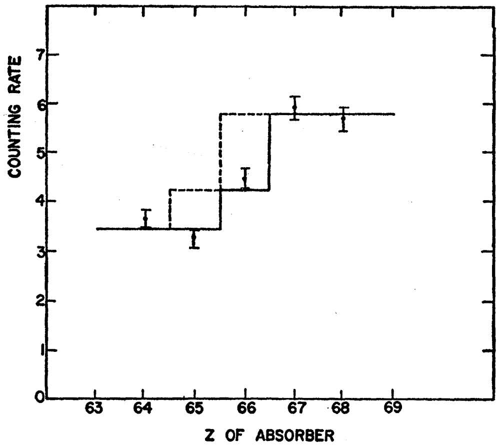

FIG. 11. Critical absorption of the Lu K x-ray. The dashed line is the expected transmission curve for the Yb x-ray and the solid line is the expected transmission curve for the Lu x-ray. The error flags represent statistical errors only.

assigned the isomeric state to Lu^{174}. By comparing the counts in the x-ray peak to the counts in the gamma-ray peak and making suitable corrections as was done for Er, we found the K-conversion coefficient to be $e_K/\gamma \sim 0.5$. The theoretical K-conversion coefficients for $Z=55$ and $E=133$ kev are shown in Table II.[15] For a Z of 71, these coefficients will be somewhat larger. The observed conversion coefficient is in agreement with that expected for either $M1$ or $E2$ radiation. The gamma-ray half-lives calculated on the basis of the single-particle model[8] for the $M1$ and $E2$ transitions are 10^{-11} sec and 10^{-7} sec, respectively. Thus the observed 75-microsecond isomer is most consistent with an $E2$ assignment.

Lu^{174} is an odd-odd isotope with 71 protons and 103 neutrons. For a deformation of $\delta=0.28$, the proton is in the $g_{7/2}(\Omega=\frac{7}{2}^+)$ state and the neutron is in the $h_{9/2}(\Omega=\frac{5}{2}^-)$ state. The ground state will then be a doublet with spins 1^-, 6^-. A transition between these levels would correspond to $M5$ radiation and would be very long-lived. However, below the $\frac{5}{2}^-$ level for the 103rd neutron is the $p_{\frac{3}{2}}(\Omega=\frac{1}{2}^-)$ level.[16] If a neutron is excited from the $\frac{1}{2}^-$ orbit to pair with the odd neutron in the $\frac{5}{2}^-$ orbit, the remaining odd neutron will now have a spin of $\frac{1}{2}^-$. If the coupling between the neutron and the proton is of the same type as for the ground state, the spin of the excited state will be

$$\Omega=\left|\tfrac{7}{2}^+\pm\tfrac{1}{2}^-\right|=3^-,\ 4^-.$$

Either a 3^- to 1^- or 4^- to 6^- transition will give rise to $E2$ radiation. If the first rotational state has an energy less than 130 kev, then the 4^- to 6^- transition is the only assignment that is allowed.

Gottfried's scheme does not predict the correct ground state assignment for Lu^{175} which is $g_{7/2}(\Omega=\frac{7}{2}^+)$. He predicts for the 103rd neutron a $g_{9/2}(\Omega=\frac{3}{2}^+)$ assignment. This neutron assignment together with the correct proton assignment gives for the ground state doublet spins of 2+, 5+. Below the $\frac{3}{2}^+$ level is a $g_{9/2}(\Omega=\frac{1}{2}^+)$ state. Thus if a neutron is excited from this level to pair with the odd neutron in the $\frac{3}{2}^+$ orbit, the spin of the excited state will be

$$\Omega=\left|\tfrac{7}{2}^+\pm\tfrac{1}{2}^+\right|=3^+,\ 4^+.$$

In this case either a 4^+ to 2^+ or 3^+ to 5^+ transition will give rise to the observed $E2$ radiation. Again if the first rotational level is less than 130 kev, only the 3^+ to 5^+ transition is allowed.

TABLE II. Theoretical K-conversion coefficients for $Z=55$, $E=133$ kev.

Type of transition	e_K/γ
$E1$	0.09
$M1$	0.38
$E2$	0.50
$E3$	2.5
$M2$	3.0

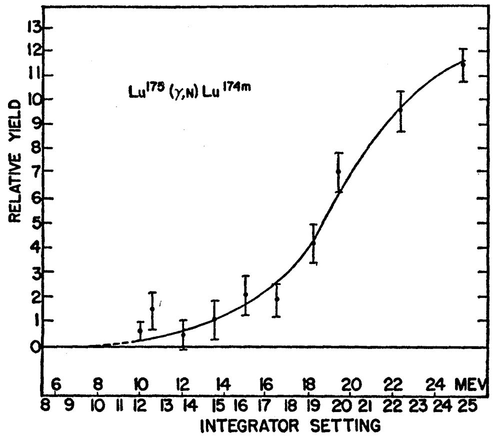

FIG. 12. Activation curve for the 133-kev gamma-ray in Lu.

IV. SUMMARY

In the search for isomeric transitions in the rare-earth region, isomeric states were found in Tb^{158}, Ho^{163}, $Er^{165,167,\text{ or }169}$, and Lu^{174}. The nuclear models for deformed nuclei proposed independently by Nilsson and Gottfried can be used to explain these transitions.

With only one small modification, the uncrossing of a single pair of levels, Nilsson's model successfully predicts all four of the isomeric transitions. Also with only slight changes, the Gottfried model predicts the isomers in Tb^{158}, Ho^{163}, and $Er^{165\text{ or }167}$. Gottfried's model does not predict the correct assignment for the odd proton in Lu. However, if the proper assignment for the proton is assumed, the Gottfried model can then also explain the $E2$ transition in Lu^{174}.

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[15] M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Appendix IV.

[16] This assumes that the $\frac{1}{2}^-$ level lies higher than the $\frac{7}{2}^-$ level at this deformation as was mentioned in the section on erbium.

The two models assign the same orbits to the states involved in the isomeric transitions in the Tb and Ho cases, but assign different orbits in the Er and Lu cases. In the Nilsson model, the isomeric state in Er arises from an $i_{13/2}(\Omega=\frac{7}{2}^{+})$ to a $p_{\frac{3}{2}}(\Omega=\frac{1}{2}^{-})$ transition, while in the Gottfried model it arises from an $i_{13/2}(\Omega=\frac{7}{2}^{+})$ to an $f_{7/2}(\Omega=\frac{1}{2}^{-})$ transition. For the Lu case, the $E2$ transition in the Nilsson model is between the neutron states $h_{9/2}(\Omega=\frac{5}{2}^{-})$ and $p_{\frac{3}{2}}(\Omega=\frac{1}{2}^{-})$. In Gottfried's model the transition is between the neutron states $g_{9/2}(\Omega=\frac{3}{2}^{+})$ and $g_{9/2}(\Omega=\frac{1}{2}^{+})$.

Both the Nilsson and Gottfried models predict a low-lying isomeric transition for the odd isotopes of Ho. In fact Nilsson's scheme assigns a $\frac{7}{2}^{-}$ orbit to the ground state and a $\frac{1}{2}^{+}$ orbit to all of the odd isotopes in Ho. Gottfried's scheme is not as clear-cut since there are three or four levels very close together in the region of 67 nucleons for a large deformation. However, for a ground state spin of $\frac{7}{2}$, as is the case for Ho^{165}, this scheme does predict an isomeric transition. These odd isotopes of Ho can be reached by (γ,p) reactions on even isotopes of Er, but using the Er target, we saw only a single isomeric transition which is known to be in one of the Er isotopes. However, since the (γ,p) cross section may be only one percent of the (γ,n) cross section for this Z[12] we would not expect to observe this reaction. Isomers in Ho^{165} can also be reached by a (γ,γ') reaction on stable Ho. However, this search should be carried out at energies below the neutron threshold in order to keep out the activity from Ho^{164}. The intensity of the Iowa State College synchrotron for energies below the neutron threshold is not sufficient for us to look for this predicted isomer.

It should be pointed out that there are some known isomeric transitions in the rare-earth region with half-lives greater than 5 microseconds and less than 30 minutes[17] that we did not observe. These are a 109-kev, 1.25-minute, $E3$ transition in Dy^{165}; and a 25-kev, 50-sec transition, a 200-kev, 6-sec transition, and a 455-kev, 0.15-sec transition, all in Yb isotopes.

The 1.25-minute isomeric state in Dy^{165} arises from the configuration of 99 neutrons, similar to that of Er^{167}, and hence an $E3$ transition is predicted. We could not produce this isomer since the highest stable isotope of Dy is 164.

The energies and lifetimes of the Yb isomers indicate that all of them may be either $E3$ or $M3$ transitions. An $E3$ transition is predicted for Yb^{169} since it has 99 neutrons. Thus, for example, the 200-kev, 6-sec transition could be assigned to Yb^{169}. It is doubtful if we could observe this since it would have to be reached by a (γ,n) reaction on Yb^{170} which is only 3% abundant or by a $(\gamma,2n)$ reaction on Yb^{171} which is 14% abundant.

The 25-kev, 50-sec transition could be assigned to Yb^{171} which has a ground state spin of $\frac{1}{2}^{-}$. An excited state corresponding to a hole excitation would have a spin of $\frac{7}{2}^{+}$. Since the first rotational state with a spin of $\frac{3}{2}^{-}$ is expected to have an energy of $\sim$100 kev, it would lie above the $\frac{7}{2}^{+}$ state and therefore the $\frac{7}{2}^{+}$ level could not decay by $M2$ radiation. The 25-kev state will be very highly converted in the L shell, and we would not expect to see such low-energy x-rays.

The 455-kev, 0.15-sec transition could be assigned to Yb^{177}. Either an $E3$ or $M3$ transition is compatible with either Nilsson's or Gottfried's scheme with only a slight reordering of the levels. Again since the highest stable isotope of Yb is 176, we could not reach an isomer in Yb^{177}.

V. ACKNOWLEDGMENTS

The authors wish to thank Dr. David Chase for many valuable discussions during the early phases of this problem, and they would also like to thank Mr. James Plimpton for performing the least-squares calculations.

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[17] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).