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Short-Lived Isomers of Nuclei*

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A search for short-lived nuclear isomers of half-lives between 10^{-6} and 10^{-3} sec. has been performed using the method of delayed coincidences between two Geiger counters. A short-lived isomer is detected by the measurement of the time lapsed between β-decay and γ- (or internal conversion electron) emission. Out of 60 nuclei investigated, 4 short-lived isomeric states were found. These are: Ta^{181*} (22 μsec.), Re^{187*} (0.65 μsec.), Tm^{169*} (1 μsec.), and Tm^{171*} (2.5 μsec.).

THOUGH excited states of nuclei usually decay with a half-life too short to be directly measurable, the existence of numerous examples of metastable states with half-life long enough to be measured with the stop watch (*isomeric states*) is well established. It is therefore reasonable to assume that a number of metastable states of intermediate but directly measurable half-life (*short-lived isomers*) should exist; this paper summarizes a systematic search for the occurrence of nuclear excited states decaying with half-lives between 10^{-6} and 10^{-3} sec.

In order to detect such short-lived metastable states one has taken advantage of the fact that, when they are produced as a consequence of a β-disintegration, there is a time interval between the rays of the radioactive disintegration and the radiation emitted by the decay of the metastable state. The β- and γ-rays will not be, as usual, coincident, but will give two signals shifted in time, one corresponding to the formation of the metastable state, and the other to its decay.

Since the γ-rays of forbidden transitions are known to be strongly converted, it was thought that the decay of the metastable state could be detected efficiently through the observation of conversion electrons.

For the measurement of time intervals the method of delayed coincidences, which had proven successful in the measurement of the half-life of the mesotron, was chosen.

DESCRIPTION OF THE EQUIPMENT

For the detection of the radiations Geiger counters were used. The use of these instruments, though advisable because of their simplicity and reliability, involves two disadvantages. The first is their long dead-time, which prevents these counters from recording a ray following another after a time shorter than $\sim 10^{-4}$ sec.; and the second is the variation of the time lag between ionization and discharge which introduces an error in the measurement of the individual time intervals.

In order to circumvent the difficulty connected with the dead-time it is necessary to use two different counters, one for the detection of the immediate, the other for the detection of the delayed rays. The errors in the measurement of time intervals resulting from variations of counter lag times are estimated to be around 0.5 μsec. by some authors and are found to be much smaller by others. With the counters available at this laboratory they are of the order of 0.5 μsec. and cannot be reduced by improving the electronic circuits. For the moment we have limited our search to the measurement of time intervals of 1 μsec. or longer, and the shortest half-life detected is of this order of magnitude. However, Bittencourt and Goldhaber[1] have shown that, despite counter lag times of the same order as those observed by us, the method of delayed coincidences between Geiger counters can be refined and used for the measurement of half-lives as short as $5 \cdot 10^{-8}$ sec.

Three different counter sets were used during the experiments. Since the radiation from metastable states was expected to be in most cases strongly internally converted, two similar β-ray counters were used, one (counter 1) for the detection of the immediate radiation, the other (counter 2) for the detection of delayed conver-

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[1] P. T. Bittencourt and M. Goldhaber, Phys. Rev. 70, 780 (1946).

sion electrons. The first counter set (Fig. 1A) consists of two conventional mica window self-quenching counters, between which the source was introduced. Counter 2 of this set could be used for the detection of γ-rays by interposing between it and the source a convenient absorber. However, most of the effort was directed toward the detection of soft conversion electrons, and for this reason a second set of counters (Fig. 1B) was built in order to eliminate the absorption of the mica windows: it consisted of two counters contained in the same envelope, which is provided with a plug for the introduction of the source. This second set had the disadvantage of requiring evacuation and introduction of new gas in order to remove or change the source. To overcome this difficulty a third set (Fig. 1C) was prepared. In this set the counters are still contained in the same envelope, but this is provided with two arms, each containing a sliding magnetically operated source support. Each of these sets was provided with a small mica window W which was used to introduce canalized beta-rays through both counters in order to study their behavior when simultaneously traversed by a ray.

The block diagram of the coincidence circuit is shown in Fig. 2. The delay time T in the circuit of counter 1 is controlled by multivibrator 1, and can be varied from 1.5 μsec. to 975 μsec. in discontinuous steps, by changing the value of a condenser. The duration τ_1 and τ_2 of the pulses to be fed in coincidence is determined by multivibrators 2 and 3 and can be changed in a similar manner. Changes in resolving time $\tau=\frac{1}{2}(\tau_1+\tau_2)$ and in delay time could be effected by operating selector switches in the front panel of the instrument. Multivibrators 2 and 3 were always set to give pulses of nearly equal duration.

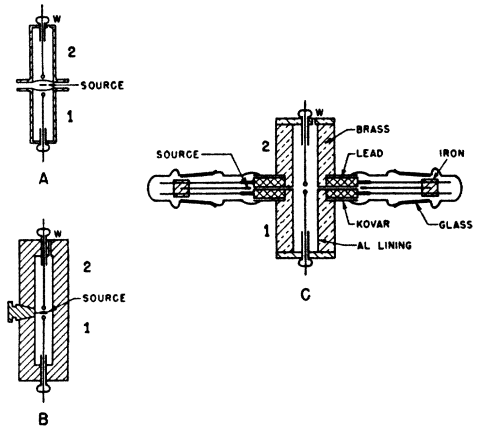

FIG. 1. (A) Two mica-window self-quenching counters; (B) Two self-quenching counters in the same envelope; (C) Similar to B but with magnetically controlled source supports.

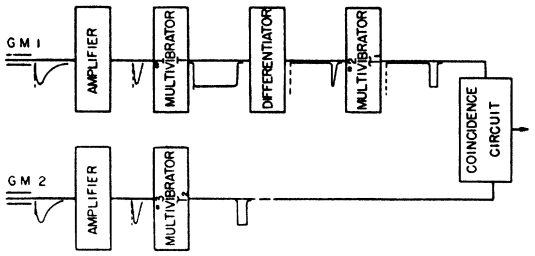

FIG. 2. Block diagram of coincidence circuit.

The length of the pulses from the multivibrators was measured on an oscilloscope (DuMont type 248) by comparison with the timing markers built in this instrument. The accuracy of the markers was checked with a calibrated wave generator. Finally, the value of τ was verified by counting delayed random coincidences.

DISCUSSION OF THE METHOD

Let us first consider the case of a source disintegrating with the emission of β-rays and neglect the corrections resulting from counter dead-time. Let N be the number of disintegrations per second of the source; if a fraction k of these disintegrations lead to a short-lived metastable state and if α is the conversion coefficient of the radiation from the metastable state, then there are $n_e=k\alpha/(1+\alpha)$ delayed electrons per disintegration, accompanied by the same number of atomic x-rays, and $n_\gamma=k/(1+\alpha)$ gamma-quanta per disintegration. One can assume that the efficiency of the counters, used without any absorber, is roughly equal to their useful solid angles ω_1 and ω_2 for electrons of all energies, while the efficiency for the detection of electromagnetic radiations is much smaller. Then, if α is large, the number of single counts per second in one of the counters (say counter 1) is

$$N_1=N\omega_1(1+n_e), \tag{1}$$

and the number of delayed coincidences per second is

$$D=2\tau N n_e\omega_1\omega_2\lambda\exp(-\lambda T), \tag{2}$$

TABLE I. Table of isotopes giving negative results in the range 10^{-6} to 10^{-3} sec.

Isotope investigated for short-lived isomeric state	Radio-active source	Half-life of the source	Mode of decay
$_{21}Sc^{45}$	$_{20}Ca^{45}$	180 d	β^-
$_{23}V^{51}$	$_{24}Cr^{51}$	26 d	K-capture
$_{25}Mn^{55}$	$_{24}Cr^{55}$	1.3 hr.	β^-
$_{26}Fe^{56}$	$_{25}Mn^{56}$	2.6 hr.	β^-
$_{27}Co^{59}$	$_{26}Fe^{59}$	46 d	β^-
$_{28}Ni^{60}$	$_{27}Co^{60}$	5.3 yr.	β^-
$_{29}Cu^{65}$	$_{28}Ni^{65}$	2.6 hr.	β^-
$_{28}Ni^{64}$ $_{30}Zn^{64}$	$_{29}Cu^{64}$	12.8 hr.	β^+, K, β^-
$_{32}Ge^{72}$	$_{31}Ga^{72}$	14 hr.	β^-
$_{31}Ga^{71}$	$_{32}Ge^{71}$	11 d	K-capture
$_{33}As^{75}$	$_{32}Ge^{75}$	1.5 hr.	β^-
$_{34}Se^{76}$	$_{33}As^{76}$	27 hr.	β^-
$_{33}As^{75}$	$_{34}Se^{75}$	115 d	K-capture
$_{36}Kr^{82}$	$_{35}Br^{82}$	34 hr.	β^-
$_{38}Sr^{86}$	$_{37}Rb^{86}$	19.5 hr.	β^-
$_{39}Y^{?}$	$_{38}Sr^{?}$	(fission product)	β^-
$_{39}Y^{89}$	$_{38}Sr^{89}$	55 d	β^-
$_{40}Zr^{90}$	$_{39}Y^{90}$	60 hr.	β^-
$_{41}Cb^{95}\rightarrow{}_{42}Mo^{95}$	$_{40}Zr^{95}$	65 d (fission prod.)	β^-
$_{41}Cb^{97?}$	$_{40}Zr^{97?}$	17 hr.	β^-
$_{45}Rh^{103}$	$_{44}Ru^{103}$	42 d	β^-
$_{45}Rh^{106}\rightarrow{}_{46}Pd^{106}$	$_{44}Ru^{106}$	1 yr. (fission prod.)	β^-
$_{45}Rh^{105}$	$_{44}Ru^{105}$	4.5 hr.	β^-
$_{46}Pd^{110}$	$_{47}Ag^{110}$	225 d	K-capture
$_{51}Sb^{?}$	$_{50}Sn^{?}$	26 hr.	β^-
$_{51}Sb^{?}$	$_{50}Sn^{?}$	long	β^-
$_{52}Te^{122}$	$_{51}Sb^{122}$	2.8 d	β^-
$_{52}Te^{124}$	$_{51}Sb^{124}$	60 d	β^-
$_{53}I^{127}$	$_{52}Te^{127}$	9.3 hr.	β^-
$_{54}Xe^{131}$	$_{53}I^{131}$	8 d	β^-
$_{55}Cs^{133}$	$_{54}Xe^{133}$	5 d	β^-
$_{55}Cs^{135}$	$_{54}Xe^{135}$	9 hr.	β^-
$_{56}Ba^{134}$	$_{55}Cs^{134}$	2.3 yr.	β^-
$_{57}La^{139}$	$_{56}Ba^{139}$	1.5 hr.	β^-
$_{58}Ce^{140}$	$_{57}La^{140}$	40 hr.	β^-
$_{59}Pr^{141}$	$_{58}Ce^{141}$	30 d	β^-
$_{59}Pr^{143}$	$_{58}Ce^{143}$	33 hr.	β^-
$_{59}Pr^{?}$	$_{58}Ce^{?}$	long (fission product)	β^-
$_{60}Nd^{142}$	$_{59}Pr^{142}$	19 hr.	β^-
$_{63}Eu^{153}$	$_{62}Sm^{153}$	46 hr.	β^-
$_{66}Dy^{160?}$	$_{65}Tb^{160?}$	73 d	β^-
$_{67}Ho^{165}$	$_{66}Dy^{165}$	2.5 hr.	β^-
$_{69}Tm^{169}$	$_{68}Er^{169}$	9.4 d	β^-
$_{71}Lu^{175}$	$_{70}Yb^{175}$	4 d	β^-
$_{71}Lu^{177}$	$_{70}Yb^{177}$	2.4 hr.	β^-
$_{74}W^{182}$	$_{73}Ta^{182}$	97 d	β^-
$_{75}Re^{185}$	$_{74}W^{185}$	74 d	β^-
$_{76}Os^{186}$	$_{75}Re^{186}$	92 hr.	β^-
$_{76}Os^{188}$	$_{75}Re^{188}$	18 hr.	β^-
$_{77}Ir^{191}$	$_{76}Os^{191}$	31 hr.	β^-
$_{77}Ir^{193}$	$_{76}Os^{193}$	17 d	β^-
$_{78}Pt^{192}$	$_{77}Ir^{192}$	60 d	β^-
$_{78}Pt^{194}$	$_{77}Ir^{194}$	19 hr.	β^-
$_{80}Hg^{198}$	$_{79}Au^{198}$	2.7 d	β^-
$_{82}Pb^{206}$	$_{81}Tl^{206}$	3.3 yr.	β^-
$_{84}Po^{210}$	$_{85}Bi^{210}$	5 d	β^-
$_{91}Pa^{231}$	$_{90}UY^{231}$	1 d	β^-

where λ is the disintegration constant of the metastable state, and where it has been assumed $\lambda\tau \ll 1$. From these equations it is possible to compute the number of delayed electrons per disintegration of the source from measurements of single counts and of delayed coincidences.

The number of random coincidences per second is expressed by

$$R = 2\tau N_1 N_2. \tag{3}$$

Within the limits of validity of Eqs. (1) and (2) one can evaluate the sensitivity of the method for the detection of delayed conversion electrons. Considering that the minimum detectible number of delayed coincidences is of the same order as the random coincidence rate, one obtains

$$n_{e\,\min} \approx \frac{N_1/\omega_1}{\lambda e^{-\lambda T} - N_1/\omega_1}, \tag{4}$$

where $n_{e\,\min}$ is the minimum number of delayed electrons which can be observed per disintegration.

In order to discuss the more general case, let us call *preceding radiation* that part of the radiation of the source which leads to the metastable state; *delayed radiation*, the radiation emitted by the metastable state itself; and *parallel radiation*, the radiation from the source which does not lead to the metastable state. The *immediate radiation* is then the sum of the preceding and parallel radiations. The fraction of source disintegrations through preceding and delayed rays is k, while the fraction through parallel rays is $1-k$. If ϵ_{1pr}, ϵ_{2pr} are the efficiencies (including geometric factors) of counters 1 and 2 for preceding rays as a whole, and if ϵ_{1d}, ϵ_{2d}, ϵ_{1par}, ϵ_{2par} are similarly defined, then

$$N_1 = N\{k\epsilon_{1pr} + k\epsilon_{1d} + (k-1)\epsilon_{1par}\}, \tag{5}$$

$$D = 2\tau N k \epsilon_{1pr}\epsilon_{2d}\lambda \exp(-\lambda T). \tag{6}$$

If the immediate, delayed, and parallel radiations are in turn complex, the efficiencies ϵ can be expressed as sums. For instance,

$$\epsilon_{2d} = n_{\mathrm{Kel}}\epsilon_{2\mathrm{Kel}} + n_{\mathrm{Lel}}\epsilon_{2\mathrm{Lel}} + \cdots + n_\gamma\epsilon_{2\gamma} + n_{KX}\epsilon_{2KX} + n_{LX}\epsilon_{2LX} + \cdots,$$

where n_{Kel} is the number of K conversion electrons per metastable state formed, $\cdots n_\gamma$ is the number of unconverted γ-rays, n_{KX} is the number of KX rays, $\cdots$, and the ϵ's are the efficiencies for these radiations. Equations (5) and (6) are, in general, too complicated to be of any use for the computation of k and α. However, they are important for the interpretation of the results,

in particular when the efficiency of the counters for different radiations is changed by the use of absorbers. For instance, they make clear that if absorbers are used in front of counter 1 and single counts are recorded, the measurement corresponds to the absorption of the total radiation from the source. By counting delayed coincidences with absorbers in front of counter 1, a measurement of the absorption of the preceding radiation is obtained; likewise, with absorbers in front of counter 2, a measurement of the absorption of the delayed radiation is obtained.

The sources used in the experiment were so weak that the corrections resulting from counter dead-time for pulses distributed at random (sometimes called coincidence correction) could be neglected. However, when a short-lived isomer is present there is a definite correlation between the time of emission of an immediate and of a delayed ray, and this requires special corrections for delay times shorter than counter dead-time. Both the expressions for single counts and for the number of delayed coincidences must be modified because the counters will miss the delayed ray after having detected a preceding one. Thus (1) and (2) become

$$N_1 = N\omega_1[1+(1-\omega_1)n_e], \qquad (1')$$

$$D = 2\tau(N\omega_1 - C)n_e\omega_2\lambda \exp(-\lambda T), \qquad (2')$$

where C is the number of simultaneous coincidences between the two counters. Similarly, (3) becomes

$$R = 2\tau(N_1 - C)(N_2 - C). \qquad (3')$$

NEGATIVE RESULTS

Sources of the 60 isotopes listed in the first column of Table I were tested for delayed coincidences with the windowless counter arrangement of Fig. 1B or 1C, and found not to give any appreciable number of delayed coincidences above the random background.

The sources were prepared either by neutron activation in the Oak Ridge Pile, or were obtained as products of uranium fission.[2] When necessary for the identification of the radioisotope, their decay was followed for a sufficient length of time. The active material was deposited on aluminum foil 1.5 mg/cm², with the thickness of the source never exceeding 1 mg/cm². The foil was introduced between the counters with the activity facing the counter used for the detection of the delayed radiation.

[2] We are indebted to members of the Chemistry Division of Oak Ridge National Laboratory for supplying us with many samples of separated fission products.

TABLE II. Experimental conditions.

Delay time T	Resolving time $\frac{1}{2}(\tau_1+\tau_2)$	Single counts $N_1 \approx N_2$	Expected random coincidences R
1.7 μsec.	0.75 μsec.	7500 c/m	1.4 c/m
13 μsec.	6.5 μsec.	4000 c/m	3.5 c/m
140 μsec.	70 μsec.	1000 c/m	2.3 c/m

For each isotope three different ranges of delays were investigated and sources of different strength were used for each range. Typical experimental conditions of these tests are shown in Table II.

The number of delayed random coincidences was obtained in each experiment by direct comparison with a source of nearly the same intensity prepared from an isotope known not to give any delayed coincidences. Initially, RaE was used for this purpose, but in later experiments any isotope previously tested with negative results (and in particular Tl^{206} and Ca^{40} which are convenient because of their long half-life) was used. Single counts and simultaneous coincidences were recorded for both sources and formula (3′) was used for the comparison of the random coincidences.

A negative result has to be interpreted as a failure to detect delayed conversion electrons emitted by a metastable state of the nucleus indicated in the first column of Table I.

The sensitivity of the method for the detection of delayed conversion electrons has been com-

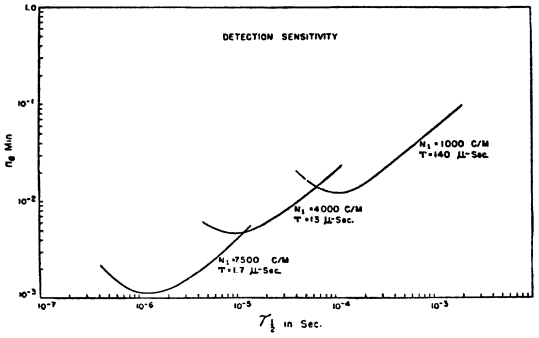

FIG. 3. Sensitivity of method for detection of delayed conversion electrons.

puted by means of (4) as a function of the half-life, assuming $\omega_1=0.5$ and using the figures of Table II. The results are plotted in Fig. 3; the curves of Fig. 3 indicate the maximum number of delayed electrons per β-disintegration which could have escaped observation.

The sensitivity for the detection of metastable states is obviously smaller than the sensitivity for the detection of delayed conversion electrons and will depend on the value of the conversion coefficient. Since this quantity increases with the third power of the atomic number and with decreasing energy, the method used is most sensitive for low energy metastable states in nuclei of high atomic number. For the detection of high energy metastable states in elements of low atomic number, it would be advantageous to measure also $\beta-\gamma$-delayed coincidences.

Independently from our work, and essentially at the same time, Hirzel, Stoll, and Wäffler[3] have used the method of delayed coincidences for the study of short-lived isomers. These authors report a 70-μsec. Pr^{141}* (daughter of Ce^{141}) and a 1200-μsec. Te^{124}* (daughter of Sb^{124}). Our attempts to verify their findings have failed, and we can state that there are less than 4×10^{-3} electron delayed with a half-life of 70 μsec. per disintegration of Ce^{141}, and less than 10^{-2} electron delayed with a half-life of 1200 μsec. per disintegration of Sb^{124}.

Several isomeric states of half-life considerably smaller than 1 μsec. have been reported by other authors. Our failure to detect these metastable states cannot be regarded as a disagreement since the sensitivity of our instrument decreases rapidly for half-lives smaller than 1 μsec.

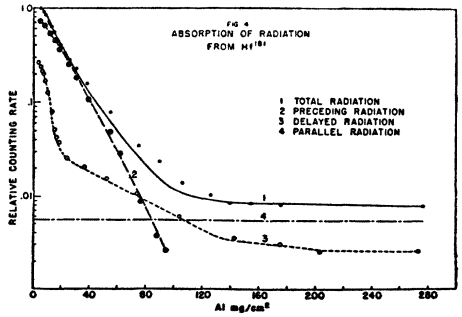

FIG. 4. Absorption of radiation from Hf^{181}.

[3] O. Hirzel, P. Stoll, and H. Wäffler, Helv. Phys. Acta 20, 241 (1947).

22 μsec. Ta^{181}*

It is known that isomeric states have been found in most nuclei of spin 9/2. Considering that the efficiency of the instrument described increases with atomic number, it seemed natural to start the experiments by looking for short isomeric states in heavy nuclei of spin 7/2.

In effect, the first short-lived isomer to be found[4] by the method of delayed coincidences was an excited state of Ta^{181} which is the heaviest nucleus of known spin 7/2. This state was detected using sources of Hf^{181}.

The decay curve of this metastable state corresponds to a half-life of 22 μsec. The decay curve was obtained by plotting delay times as the abscissae and the delayed coincidence counting rate, divided by the resolving time, as the ordinates.

From the number of single counts and delayed coincidences recorded it appears (using Eqs. (1′) and (2′) with $\omega_1=\omega_2=0.3$ as it is obtained by rough geometrical considerations) that there are around 0.5 delayed electrons per disintegration of Hf^{181}. Accordingly, 74 percent of the single counts are due to beta-disintegration electrons, and 26 percent to conversion electrons from Ta^{181}*.

Absorption curves of the radiations from Hf^{181} are plotted in Fig. 4. All these curves were obtained with the counter set of Fig. 1A whose high geometry, necessary for intensity reasons, is obviously poor for the purpose of energy determination. The full points indicate the absorption of the total radiation of this isotope, arbitrarily normalized to unity for no absorber, and were obtained by counting single counts with the absorber inserted between the source and the counter. The radiation of Hf^{181} appears to consist of soft radiations and of a hard γ-component, whose energy was found to be 0.5 Mev by absorption in Pb. Curve 2 is the absorption curve of the radiation preceding the metastable state and was measured by counting delayed coincidences (with constant T and τ) with absorbers between the source and counter 1; curve 2 is normalized to 0.74. Curve 3 is the absorption curve of the delayed radiation measured by counting delayed coincidences with absorbers in front of counter 2 and is normalized to 0.26.

[4] S. DeBenedetti and F. K. McGowan, Phys. Rev. 70, 569 (1946).

Curve 4 is obtained by extrapolating the absorption curve of the 0.5-Mev γ-rays.

Curve 1 is the result of the addition of curves 2, 3, and 4. Since this fits fairly closely the points obtained by absorbing the total radiation, we can consider that Hf^{181} emits a single beta-spectrum, whose absorption is given by curve 2. The maximum energy of the β-spectrum of Hf^{181} was estimated by applying Feather's method to curve 2, and the value $E_{max}=0.42$ Mev was obtained.[5] The γ-rays of 0.5 Mev, which are not found either in the preceding or in the delayed radiation, are emitted in parallel to the metastable state.

The radiation from the metastable state consists of K or L conversion electrons, x-rays, and possibly unconverted γ-rays. The maximum range in Al of the delayed electrons is 19 mg/cm^2. Their energy is therefore around 120 kev; if the maximum range measured corresponds to K conversion, the energy of the metastable state is about 185 kev, while, if mostly L electrons are present (as can be the case for a highly forbidden γ-transition), its energy can be estimated to 132 kev.

For the interpretation of these data one has to consider the other information available on the isotope considered. Cork, Shreffler, and Fowler[6] have studied the conversion lines from Hf^{181} by means of a photographic β-ray spectrograph. They report γ-rays of 0.478, 0.345, and 0.133 Mev, and suggest that the γ-rays of 0.345 and 0.133 Mev are in cascade, with the 0.478-Mev γ-radiation in parallel. If one assumes that the metastable state of 22 μsec. has an energy of 0.133 Mev, this scheme is in agreement with our observations. Our failure to detect the γ-rays of 0.345 Mev can be due to the low sensitivity of the counters for γ-rays. However, there is some evidence that the 0.345-Mev radiation is strongly converted. In effect, Cork *et al.* do not report any large difference in the intensities of the conversion lines of the 0.345- and 0.133-Mev gamma-radiations, and the 0.133-Mev radiation is known to be strongly converted. Furthermore, some careful absorption experiments of $\beta-\gamma$ immediate coincidences by Wiedenbeck and Chu[7] show

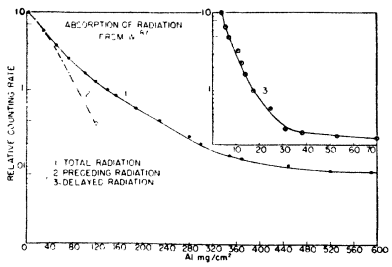

FIG. 5. Absorption of radiation from W^{187}.

the presence of a strong conversion line having an energy of 0.35 Mev. However, if the 0.345-Mev radiation is strongly converted, and therefore of high polarity, it would be difficult to explain why it should compete favorably with the γ-rays of 0.478 Mev.

Careful investigation of the continuous spectrum and of the conversion lines from Hf^{181} by means of a β-ray spectrometer is needed in order to elucidate these points and to confirm the present interpretation.

0.65 μsec. Re^{187*}

As previously reported,[8] sources of 24-hr. W^{187} give rise to delayed coincidences. From the decay curve the half-life of the metastable state (Re^{187*}) appears to be 0.65 μsec.

The absorption curves of the total (curve 1) and preceding (curve 2) radiations are shown in Fig. 5, indicating that the preceding radiation is softer than the total β-radiation from W^{187}. The delayed radiation (curve 3, Fig. 5) consists of electrons having a maximum energy of 0.13–0.15 Mev, accompanied by an electromagnetic radiation of less than 0.1 Mev. The mica window of counter set 1A absorbs a much larger percentage of the rays of Re^{187*} than of Ta^{181*}. This seems to show that the electrons from these two short-lived isomers, though of about the same maximum energy, have a different energy distribution, possibly due to a different ratio of K to L conversion. The β-spectrum of W^{187} is known to consist of two components of 1.31- and 0.62-Mev maximum energy and intensities 23 and 77 percent respectively, accompanied by a rather large number of conversion lines.[9] Though many authors have investigated this isotope, there is no general agreement as to the disintegration scheme.

[5] In a previous communication (see reference 4) this energy was erroneously reported as 0.8 Mev, because of an error in transcription superimposed on a different method of analyzing the curves.

[6] J. M. Cork, R. G. Shreffler, and C. M. Fowler, Phys. Rev. 72, 1209 (1947).

[7] M. L. Wiedenbeck and K. Y. Chu, Phys. Rev. 72, 1164 (1947).

[8] S. DeBenedetti and F. K. McGowan, Phys. Rev. 71, 380 (1947).

[9] P. W. Levy, personal communication of unpublished work.

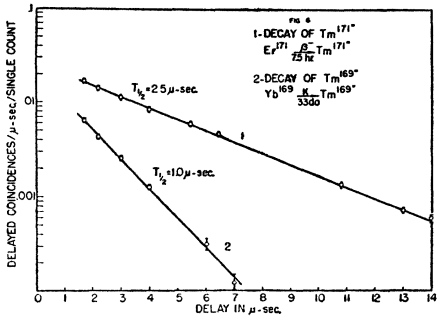

FIG. 6. Decay of Tm^{171*} and Tm^{169*}.

Analyzing the absorption curve of the preceding radiation by Feather's method, one obtains a maximum energy consistent with 0.62 Mev, indicating that the metastable state follows only the lower energy component of the β-spectrum. Some delayed coincidences, not numerous enough to permit the measurement of an absorption curve, were observed after absorbing all the preceding β-rays, indicating that some γ-rays are present in the preceding radiation.

A number of soft γ-rays which, within the accuracy of our absorption measurements, could be due to the decay of Re^{187*}, have been observed in the spectrum of W^{187}. However, one might feel inclined to associate this metastable state with a radiation of 0.130 Mev, since this is strongly internally converted.[10]

There are about 0.1 delayed electrons per disintegration of W^{187}, or around 0.13 delayed electrons per β-ray of the softer component of the continuous spectrum. However, the number of delayed electrons per metastable state could be higher, if there were other γ-rays in parallel.

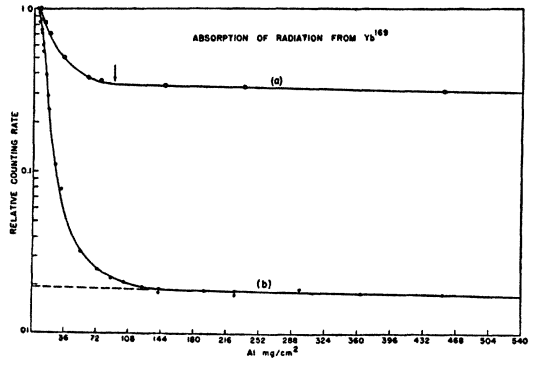

FIG. 7. Absorption of radiation from Yb^{169}.

[10] We are indebted to L. C. Miller and L. F. Curtiss for the communication of this result.

1 μsec. Tm^{169*}

Delayed coincidences corresponding to a half-life of 1 μsec. were observed with sources of 33-day Yb, which, according to Bothe,[11] decays by K capture to Tm^{169}. The assignment of the 33 days' activity to Yb was verified by the ion exchange technique of separating rare earths.[12] The fact that the decay process is K capture was confirmed by our absorption measurements; no positive electrons were found in a rough investigation.

Curve 2 of Fig. 6 is the decay curve of the metastable state of Tm^{169*}; the delayed radiation consists mostly of electrons having an energy of 0.12 Mev, and the excitation energy of the metastable state appears to be around 0.19 Mev (assuming that the observed electrons are K electrons) or around 0.13 Mev (if most delayed electrons are from L conversion).

Figure 7 shows the absorption of the total radiation of Yb^{169} (curve b) and that of the radiation preceding the 1-μsec. metastable state (curve a). It is evident from these curves that the metastable state is preceded by x-rays and, at least in certain cases, by conversion electrons of harder gamma-rays (around 0.4 Mev). The ratio of electronic to x-radiation is much larger in the total radiation than in the radiation preceding the metastable state, indicating that some electrons must be emitted in parallel to this state (it does not seem possible to account for the excess of electrons in the total radiation by attributing it to the delayed electrons alone). These "parallel" electrons are probably due to the conversion of some other γ-component.

2.5 μsec. Tm^{171*}

After the observations on Tm^{169*} we examined for delayed coincidences a number of Er activities in the hope that one of them would lead by β^--decay to the 1-μsec. metastable state. Evidence of delayed coincidences was found with a 7.5-hr. activity, but the half-life of the corresponding metastable state was 2.5 μsec. (curve 1, Fig. 6). In order to prove the difference of these two

[11] W. Bothe, Zeits. f. Naturf. 1, 173 (1946).

[12] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. 69, 2800 (1947). We are greatly indebted to Mr. Ketelle, of the Chemistry Division of Oak Ridge National Laboratory, for the preparation of all the separated rare earth sources used in these experiments.

half-lives (which were originally supposed to be due to the same isotope) they were compared with the arrangement of Fig. 1C. The values of 1 and 2.5 μsec. were confirmed in this way.

Some subsequent work by Ketelle and Peacock[13] showed that the 7.5-hr. Er activity cannot be assigned to the mass number 169, since it decays into a long-lived β-active daughter. Furthermore, these authors studied the spectrum of the 7.5-hr. Er and were led to assign this activity to mass 171 on the basis of its disintegration energy. This assignment seems to be confirmed by other thus far unpublished evidence.

From β-spectrographic work Ketelle and Peacock suggest a disintegration scheme for Er^{171}. The 2.5-μsec. metastable state is attributed to a 0.113-Mev transition, whose strongly converted L line appeared in the beta-spectrum.

An approximate evaluation of the number of delayed electrons per disintegration of Er^{171} from the number of delayed coincidences leads to the value of 0.4. Since only 71 percent of the beta-rays of Er^{171} lead to the isomeric state, the number of delayed electrons per disintegration of the metastable state is 0.56, and the total coefficient of internal conversion

$$\alpha = \frac{0.56}{1-0.56} = 1.3.$$

DISCUSSION OF RESULTS

Practically the only information available about the four short-lived isomers observed is their half-life and a rough estimate of their energy. The radiation emitted is strongly converted, but from the experimental results it is not known (with the exception of Tm^{171*}) whether the conversion coefficients are high enough to change the order of magnitude of the half-life caused by γ-emission alone. It is therefore impossible to draw any definite conclusions on the nature of the transitions involved. Nevertheless, comparing the half-lives and energies experimentally determined with the theoretical estimates[14] of half-lives for γ-emission, it appears that the observed values fall between the theoretical predictions for changes in angular momentum of 2 and 3 units. The inclusion of internal conversion would lower the theoretically expected half-lives, and as a consequence one can consider that the observed transitions are more probably to be associated to a change of angular momentum of 3 units, rather than 2.

Though it is clear that any statistical consideration on the frequency of occurrence of isomeric states with half-lives between 10^{-6} and 10^{-3} sec. is premature, one cannot escape the impression that changes in angular momentum of 3 units occur less frequently than changes of 4 or 5 units, since a large number of isomeric states with half-lives ranging from 1 sec. to 10^{7} sec. (corresponding to the larger spin changes) are known, and only 4 short-lived isomers were found in these experiments.

One must keep in mind, however, that the method used in the present investigation cannot be expected to detect all the isomeric states of the 60 nuclei studied, with half-lives in the range covered in the present survey. A first requirement which has to be satisfied for a positive result is that the metastable state be reached as a product of β-disintegration; furthermore, the radiation from the metastable state must be strongly converted; and, finally, the efficiency of detection is not constant over the whole range explored, but increases for shorter lives.

These considerations might be sufficient to explain why only four cases were observed, and why these were found in heavy elements, with small excitation energy and with half-lives in the microsecond region. It is conceivable that using different techniques for the production of nuclear excited states and for the detection of their radiation, one could discover many more short-lived isomers in the range considered. The possibility of exciting nuclei by irradiation with x-rays, by capture of slow neutrons, or by particles bombardment is immediately apparent. Also, the newly discovered technique of efficient gamma-counting with organic scintillation counters might provide a method of detecting short-lived isomers whose radiation is only slightly converted, and some work along these lines has already been started.[15]

[13] B. H. Ketelle and W. C. Peacock, Phys. Rev. **73**, 1269 (1948).

[14] See, for instance, H. A. Bethe, Rev. Mod. Phys. **9**, 226 (1937).

[15] S. DeBenedetti, F. K. McGowan, and J. E. Francis, Jr., Phys. Rev. **73**, 1404 (1948).