MILLISECOND THALLIUM ISOMERS

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A technique of investigating short-lived (10^{-4} – 10 sec) isomers is described. The isomers Tl^{199m} and Tl^{201m} have been identified and a new isomer Tl^{200m} with a 34.1 ± 1.0 msec half-life and 205- and 520-keV γ lines has been discovered by irradiating gold with 22-MeV α particles and mercury with 11-MeV deuterons.

1. INTRODUCTION

ISOMERIC states having lifetimes from 10^{-5} sec to 1 sec (for E3, M3, and M2 transitions) have been observed in nuclei of different kinds. The identification of these transitions is often difficult[1] and unsatisfactory at the present time. It is therefore of considerable interest to discover and investigate millisecond isomers. The excitation of such isomeric states has been accomplished using 20-MeV[2-4] and 32-MeV[5] protons, 22-MeV,[6,7] 24-MeV,[8] and 26-MeV[9] γ rays, thermal neutrons,[10] and 14-MeV neutrons.[11] Similarly, 5.5-MeV protons and deuterons have been used for A < 100.[12,13] [1)] In the present work we investigated short-lived thallium isomers using 22-MeV α particles and 11-MeV deuterons accelerated in a cyclotron.

It is known that a 7^+ isomeric level of even-mass thallium isotopes exists. Deexcitation of this level by electron capture and M4 transitions characterizes the relatively long-lived isomers of Tl^{194}, Tl^{196}, and Tl^{198}[14], while E3 transitions occur in the short-lived isomers of Tl^{202} and Tl^{204}.[6,15] This level has been interpreted as a result of a configuration formed by the 81-st proton ($s_{1/2}$) and an odd neutron ($i_{13/2}$).[1,16] It was of interest to establish the existence of a Tl^{200} isomeric state and to determine its properties. It has been shown in [14] that direct excitation in an accelerator beam is a suitable procedure in searching for Tl^{200m}.

It has been proposed on the basis of experimental data regarding odd-mass thallium isomers[1] that the 42- and 5-msec activities observed in proton-irradiated mercury[2] belong to Tl^{199m} and Tl^{201m}, respectively. The isomeric transitions are interpreted as $h_{11/2} \rightarrow d_{5/2}$. A hypothesis has also been advanced regarding the upper limits of the Tl^{203m} and Tl^{205m} lifetimes. Subsequently, in [4] more precise data were recorded regarding the half-lives and γ-line energies of the proposed Tl^{199} and Tl^{201} isomers, but these were not identified by their mass numbers. Furthermore, it has not been reported that the γ spectrum includes an isomer having a shorter lifetime (1.8 msec) than the 0.36- and 0.33-MeV lines corresponding to $d_{5/2} \rightarrow d_{3/2}$ and $d_{3/2} \rightarrow s_{1/2}$ transitions of Tl^{201}.[15]

It was our aim to search for isomers of Tl^{200} and Tl^{203} and to identify the activities that are attributable to Tl^{199m} and Tl^{201m}.

2. EXPERIMENTAL PROCEDURE

Our procedure, like the techniques used previously, was based on the pulsed irradiation of targets, and the registration and analysis of radiation from the targets during the intervals between irradiation periods. It was necessary to provide for the monitoring of beam intensity, adequate registration speed, and synchronized operation of the different units.

The targets were irradiated with the external beam from a one-meter cyclotron. The α-particle or deuteron beam impinged on a tantalum diaphragm with an aperture of 10-mm diameter placed at a distance ~ 10 m from the cyclotron. The beam was adjusted by means of an arrangement whereby a quartz plate was placed in front of the diaphragm at 45° to the beam direction. The image on the quartz was observed with a PTU-3 television receiver. A lead shielding block with an opening for the beam was placed behind the diaphragm; the beam struck a target, 80 cm from the diaphragm, mounted in a vacuum at the end of a Faraday cylinder. The target thickness was several times greater than the

[1)] Investigations published prior to the end of 1960 are listed in the review article.[1]

deuteron or α-particle range, and its diameter was 30—40 mm. This arrangement excluded interactions between the charged-particle beam and extraneous materials located very close to the target.

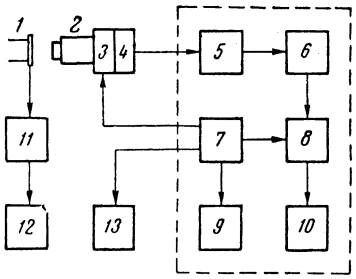

FIG. 1. Block diagram of apparatus. 1 – Faraday cylinder, 2 – scintillation counter, 3 – blocking circuit, 4 – cathode follower of scintillation counter, 5 – amplifier, 6 – pulse-height analyzer, 7 – control circuit (gating unit), 8 – time analyzer, 9 – cycle counter, 10 – registers, 11 – current integrator, 12 – scaler (PS-10000), 13 – modulator of rf cyclotron oscillator (located in the same room as the oscillator).

Figure 1 is a block diagram of the experimental arrangement. The ion beam was monitored by a current integrator with input from a Faraday cylinder. The sensitivity of the integrator was $(1.9 \pm 0.1) \times 10^{-10}$ Coulomb/pulse. Integrator pulses were counted by a PS-10000 scaler. The monitor background was about 30 pulses per hour. The maximum intensity of the ion beam was $\sim 9 \times 10^{12}$ deuteron/sec and $\sim 1 \times 10^{12}$ α particle/sec; both maxima were within the linear limits of the integrator.

The γ radiation from the target was detected with a scintillation counter having a NaI(Tl) crystal 40 mm in diameter and 40 mm high in conjunction with an FÉU-13 photomultiplier, which was of a type exhibiting high stability and high gain and which functions well for pulse loads up to $\sim 6 \times 10^4$ per second.[3] In order to obviate overloading of the photomultiplier diode system and the amplifier as a result of intense instantaneous γ-ray emission, a 40-V negative blocking potential was applied to the photomultiplier modulator during the target-irradiation periods. The blocking circuit 3 was a trigger operated by pulses from the control circuit 7.

Pulses from the cathode follower 4 were fed through a 15-meter RK-50 cable to the non-overloaded amplifier 5 and then to a 5-channel pulse-height analyzer 6 constructed on the basis of six Park threshold discriminators.[17] The resolving time of the analyzer was ~ 2 μsec. The threshold of the first discriminator could be set from 0 to 100 V, i.e., within the entire amplitude interval of the amplifier output signal. The channel width was variable from 1 to 20 V. The pulse-height spectrum was measured by shifting the discrimination threshold. In half-life measurements the pulse-height analyzer was operated with a single channel, from which pulses were fed to a 10-channel time-delay analyzer 8 consisting of a chain of ten triggers which at successive given intervals Δt switched the ten registers on and off. The ten time channels were divided into two sets of five each, separated by a delay; the channel width in each set was regulated separately.

The registers 10 comprised a fast scaler ($\tau \approx 1$ μsec) with a scaling factor of 256 and two OG-3 decatrons at the output. The capacity of the register was therefore 25600 pulses.

Blocks 5—10 in the figure are units of a single circuit with independently stabilized power supply. This circuit and the PS-10000 scaler were located in a room separated by a concrete shield from the room in which cyclotron beam emerged.

The control circuit 7 was based on a 10-kc generator operating continuously during the measurements. Pulses from this generator were fed to a dividing scaler, which operated successively, with given scaling factors, to determine the period of irradiation t_{ir}, of the first delay t_{d1}, of the first measurement t_{m1}, of the second delay t_{d2}, and of the second measurement t_{m2}. The repetition frequency of these measuring cycles varied from 10^4 to 3 cycles per minute. It was also possible to trigger single measuring cycles. The cycle counter 9 consisted of an OG-3 dekatron and SB-1M mechanical counter. The range of t_{ir} varied from 0.7 msec to 1.4 sec; its length was determined either by the duration of a single pulse, $t_{ir} = 0.7k$ msec (where $k = 1, 2, 3, \ldots, 10$), or by the duration of a series of pulses with $\Delta t = 0.7$ msec and a total of 10, 100, or 1000 pulses (with duty ratio $\frac{1}{2}$) in the series. The periods t_{d1}, t_{m1}, t_{d2}, and t_{m2} could be varied independently from 2×10^{-4} to 10 sec and assumed values given by integers from 2 to 10 multiplied by the factors 10^{-4}, 10^{-3}, 10^{-2}, 10^{-1}, and 1. The time instability did not exceed 1%.

The first delay t_{d1} was required to enable measurement following the decay of shorter-lived activities than the investigated one. The first measurement combines the sought effect and the long-lived background, while the second measurement gives the long-lived background. In the pulse-height analysis, pulses during the time t_{m1} go to the first set of five registers, while during the time t_{m2} they go to the second set of five. The difference between the numbers of

counts of the respective registers gives the magnitude of the effect. The second delay enabled enhancement of the ratio between the first and second measurements. In the time analysis the time t_{m1} is the width of each of the first five time-analyzer channels, while t_{m2} is the width of the channels in the second set of five.

The choice of the periods t_{ir}, t_{d1}, t_{m1}, t_{d2}, and t_{m2} and of the cycle repetition frequency was determined in each specific case by the half-life of the investigated isomer and the ratio between the intensities and time constants of the background and the investigated effect.

Energy calibration of the apparatus was performed using Ce^{144}, Cs^{137}, Hg^{203}, Na^{22}, and Zn^{65} samples. The resolution obtained with the Cs^{137} 661-keV line was 10.5%. The dependence of the crystal photoefficiency on γ-ray energy under the given experimental conditions was calculated using photofractions from [18]; the interaction efficiency for the given geometry was calculated from the conventional formula. Measurements of the γ spectra of several samples with known line-intensity ratios showed that the error in determining the relative intensity of γ lines using the derived relation did not exceed 10%. In order to take account of soft γ-ray attenuation the crystal packing closeness was determined by means of a Cd^{109} sample.

The technique was tested by measuring the characteristics of the well-investigated short-lived isomers Pb^{207m} (0.80 sec, $E_{\gamma 1}$ = 570 keV, $E_{\gamma 2}$ = 1065 keV)[15] and Na^{24m} (20 msec, E_γ = 470 keV).[12] The isomer Pb^{207m} was obtained by irradiating a lead target with deuterons to produce the reaction $Pb^{206}(d, p)Pb^{207m}$; Na^{24m} was obtained by irradiating NaCl to produce the reaction $Na^{23}(d, p)Na^{24m}$.

3. RESULTS AND DISCUSSION

Our search for Tl^{200m} and the identification of Tl^{199m} were performed by irradiating gold with 22.3-MeV α particles, since at this energy important yields are obtained practically in only two reactions: $Au^{197}(\alpha, n)Tl^{200}$ and $Au^{197}(\alpha, 2n)Tl^{199}$. A short-lived activity was observed; the γ spectrum is shown in Fig. 2, where the 720-, 520-, 365-, and 73-keV lines are clear, and there is an indication of a line at ~200 keV. We identified the 73-keV line as x-radiation accompanying internal K-shell conversion in thallium. From the numerous measurements, one result of which is shown in Fig. 3, we obtained the half-lives 28.9 ± 0.6, 34.1 ± 1.0, and 31.0 ± 1.4 msec corresponding to the 365- and 520-keV peaks and the x-ray line. It can therefore be assumed that when gold is irradiated with α particles the two isomers Tl^{200m} and Tl^{199m} are formed.

In order to test the foregoing hypothesis we measured the γ spectra obtained with 22.3-, 21.5-, and 20.6-MeV α particles. The energy was varied by means of aluminum foils. The results show (Fig. 4) that with decreasing energy E_α the 365-keV line intensity diminishes more rapidly than that of the 520-keV line. The existence of a ~200-keV line is also confirmed and its relation to the 520-keV line is found. From the following data (where B is the calculated yield and S is the area under the peak of the corresponding γ line) it appears that the area ratio of the 520- and 365-keV peaks varies with E_α like the ratio of yields from the reactions $Au^{197}(\alpha, n)Tl^{200m}$ and $Au^{197}(\alpha, 2n)Tl^{199m}$:

E_α, MeV:	20.6	21.5	22.3
$B(\alpha,n)/B(\alpha,2n)$:	0.75	0.41	0.28
S(520MeV)/S(365MeV):	0.55	0.25	0.16

In calculating these yields we used the procedure for calculating the cross sections for nuclear reactions that proceed via a compound nucleus.[19] The reaction thresholds were calculated from the binding energies,[20] taking into account the excitation energies of isomeric states (given below). Thus the activity with $T_{1/2}$ = 28.9 msec and E_γ = 365 MeV belongs to Tl^{199m} and the activity with

FIG. 2. γ spectrum of activity in gold irradiated with 22.3-KeV α particles; t_{ir}, T_{d1}, t_{m1}, t_{d2}, and t_{m2} are 14, 2, 100, 50, and 100 msec, respectively. N is the number of pulses in a channel per 1000 monitor pulses.

FIG. 3. Decay of 520-keV γ line obtained from α-particle-irradiated gold. Curve 1 – decay of total activity, curve 2 – decay after subtraction of constant long-lived background. $T_{1/2}$ = 34 msec; channel width ($t_{m1} = t_{m2}$) 30 msec; t_{ir}, t_{d1}, and t_{d2} are 14, 2, and 0.2 msec. N is the number of pulses in a channel.

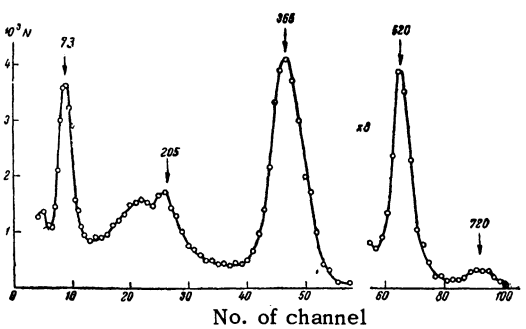

FIG. 2

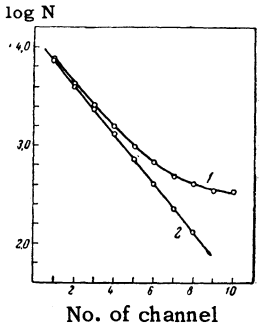

FIG. 3

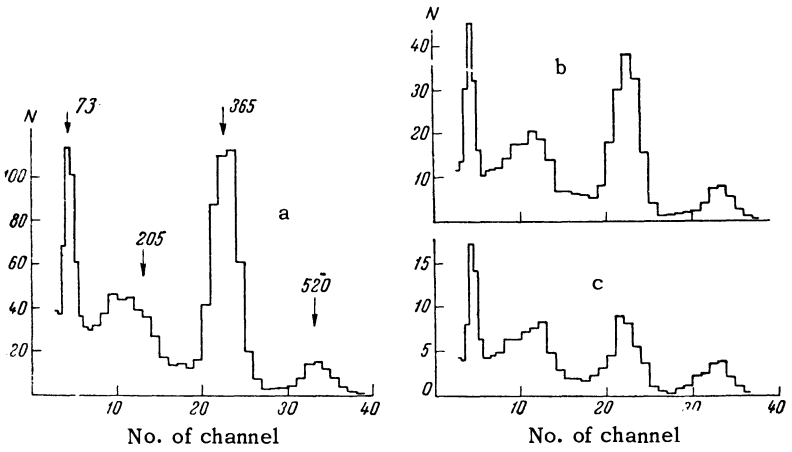

FIG. 4. γ spectra of activity resulting from the irradiation of gold with α particles having different energies: E_α = (a) 22.3 MeV, (b) 21.5 MeV, (c) 20.6 MeV. The periods t_{ir}, t_{d1}, t_{m1}, t_{d2}, and t_{m2} are 14, 2, 60, 60, and 60 msec, respectively. N is the number of pulses in a channel per unit monitor pulse. The scale of the ordinate axis was adjusted to maintain a constant area of the 520-keV peak.

$T_{1/2}$ = 34.1 msec and E_γ = 520 keV belongs to Tl^{200m}.

The isomer Tl^{199m}. Our measurements, $T_{1/2}$ = 28.9 ± 0.6 msec and $E_{\gamma 1}$ = 365 ± 5 keV agree with the results obtained from proton-irradiated mercury.[4] According to the decay scheme in [1], the 365-keV peak must be regarded as a superposition of three lines:

$$\sim 370 \text{ keV } (h_{11/2} \xrightarrow{E3} d_{5/2}), \quad 353 \text{ keV } (d_{5/2} \xrightarrow{M1+E2} d_{3/2}),$$
$$367 \text{ keV } (d_{3/2} \xrightarrow{M1+E2} s_{1/2}).$$

The line $E_{\gamma 2}$ = 720 ± 10 keV (Fig. 2) evidently represents a direct $d_{5/2} \rightarrow s_{1/2}$ transition.[15] The intensity ratio $N_{\gamma 1}/N_{\gamma 2}$ = 28 ± 5 yields the fraction 0.09 ± 0.02 of direct transitions.[2)] The ratio $N_{eK}/N_{\gamma 1}$ (where N_{eK} is the emission intensity of K-conversion electrons) was determined from the γ spectra for the three α-particle energies. The value 0.125 ± 0.020 is in good agreement with the result 0.11 calculated using tabular conversion coefficients[21] and data regarding mixed M1 + E2 transitions.[15]

The isomer Tl^{200m}. The sum of $E_{\gamma 1}$ = 205 ± 10 keV [3)] and $E_{\gamma 2}$ = 520 ± 10 keV corresponds to a smooth dependence of the energy difference between the 7^+ isomeric state and the 2^- ground state of even-mass thallium isotopes on the number of neutrons (Fig. 5). It can reasonably be assumed that Tl^{200} also has a 7^+ isomeric level. The relation between E_γ, $T_{1/2}$, and the multipolarity (according to Weisskopf[22]) is consistent with the following variants of the isomeric transition: E3 (205 keV), E3 (520 keV), and M3 (520 keV). The calculated values of $N_{\gamma 1}/N_{\gamma 2}$ and

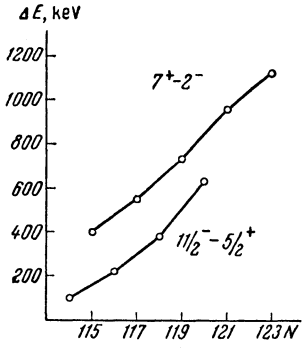

FIG. 5. Energy difference between the 7^+ and 2^- (ground) states of even-mass thallium isotopes, and between the $11/2^-$ and $5/2^+$ states of odd-mass thallium isotopes, as functions of the number of neutrons: $Tl^{196,199}$ from [14], $Tl^{202,204}$ from [6], $Tl^{195,197}$ from [15], $Tl^{199,200,201}$-present work.

$N_{eK}/(N_{\gamma 1} + N_{\gamma 2})$ are, respectively, 0.23 and 0.10, 0.78 and 0.10, and 0.22 and 1.15. The experimental values 0.24 ± 0.05 and 0.23 ± 0.06 are closest to the first variant. It can therefore be assumed that the Tl^{200m} decay scheme resembles the decay schemes of Tl^{202m} and Tl^{204m}:[6,15]

$$(7^+) \xrightarrow{205 \text{ keV}, E3} (4^-) \xrightarrow{520 \text{ keV}, E2} (2^-).$$

It should be noted that the hindrance factor of the 205-keV E3 transition is very close to those of the isomeric transitions of Tl^{202m} and Tl^{204m} (given in the accompanying table).

The isomers of Tl^{201} and Tl^{203} were sought by irradiating mercury with 11-MeV deuterons. Pure mercury is unsuitable as a target, and the use of mercuric oxide causes great difficulties because of the strong background of F^{17} annihilation radiation with $T_{1/2}$ = 66 sec. It is possible that the presence of β^+-activity background resulting from reactions with oxygen prevented the detection of Tl^{200m} when mercuric oxide was irradiated with protons.[3,4] We used a 1:2 mercury-bismuth alloy as a target. Deuteron-bombardment tests of targets made of tantalum, gold, and bismuth showed that bismuth gives the minimum short-lived background.

We observed four short-lived activities with the following values of $T_{1/2}$ and E_γ: 1) 34 msec, 0.52

[2)] It was shown that $N_{\gamma 1}/N_{\gamma 2}$ does not vary with increasing distance between the target and counter, i.e., it contains no appreciable contribution from the registration of coincidences.

[3)] The 325-keV line of Cr^{51} and the 511-keV line of Na^{22} were used as reference lines for our spectra.

E3 transitions in thallium

A	E_γ, keV	$T_{1/2}$(exp), sec	α (total)	$T_{1/2}$(theor), sec	F
200	205	$3.4 \cdot 10^{-2}$	3.35	$3.4 \cdot 10^{-2}$	4.4
202 [6]	460	$6.3 \cdot 10^{-4}$*	0.13	$1.1 \cdot 10^{-4}$	6.4
204 [6]	706	$6.2 \cdot 10^{-5}$	0.03	$5.5 \cdot 10^{-6}$	11.6
195 [15]	99	3.5	157	5.7	97
197 [15]	222	$5.5 \cdot 10^{-1}$	2.21	$2.0 \cdot 10^{-2}$	88
199	370	$2.9 \cdot 10^{-2}$	0.25	$5.2 \cdot 10^{-4}$	69
201 [4,9]	607**	$2.1 \cdot 10^{-3}$**	0.05	$1.6 \cdot 10^{-5}$	136

*Average of present work and [6].
**Average of present work and [4,9].

MeV; 2) 29 msec, 0.37 MeV; 3) 2.3 ± 0.2 msec, 345 ± 10 keV and 625 ± 20 keV; 4) 0.68 ± 0.04 msec, 475 ± 15 keV. The first two activities can be assigned to Tl^{200m} and Tl^{199m} (see above). The fourth activity can be associated with Tl^{202m}. Indeed, the half-life is in good agreement with previously known data, and the 475-keV peak can be regarded as the superposition of 460- and 490-keV lines.[6,15] According to [1] the activity with $T_{1/2}$ = 2.3 msec must be attributed to Tl^{201m}. This is confirmed by the presence of a 345-keV line, which coincides with the unseparated 361- and 330-keV Tl^{201} doublet.[15]

Our measurements of $T_{1/2}$ and E_γ for Tl^{201m} agree well with the properties of the activity observed when thallium was irradiated with 26-MeV γ rays ($T_{1/2}$ = 2.1 ± 0.2 msec; 335- and 597-keV γ rays in coincidence).[9] Since this activity was not observed with 22-MeV γ rays,[6] its production can be accounted for by the reaction $Tl^{203}(\gamma, 2n)Tl^{201m}$.

The energy of the isomeric transition and the lifetime of Tl^{201m} correspond to E3 multipolarity. Consequently, we can assign $11/2^-$ to the isomeric level as in the case of other odd-mass thallium isomers. The transition energy 625 keV is consistent with the idea of a smoothly increasing energy difference between the $11/2^-$ and $5/2^+$ levels as the number of neutrons increases (Fig. 5).[1]

We searched unsuccessfully for Tl^{203m} in the energy region 0.7 — 1.3 MeV for half-lives ranging from 10 to 0.1 msec. Either the lifetime is shorter than 10^{-4} sec, which would take us outside the limits of our technique, or isomerism similar to that in $Tl^{195-201}$ does not exist in this case.

In conclusion we consider the relation between the experimental and Weisskopf's theoretical half-lives of thallium isomers in E3 transitions.

The accompanying table gives theoretical half-lives calculated from the formula $T_{1/2}(\text{theor}) = 1.98\, A^{-2}E_\gamma^{-7} \times 10^{-2}$ ($r_0 = 1.2 \times 10^{-13}$ cm, S = 1, E_γ in MeV),[22] the total conversion coefficients calculated from the formula $\alpha(\text{total}) = \alpha_K + 1.3\alpha_L$[16] and tables in [21], and the hindrance factor

$$F = T_{1/2\ \text{exp}}\,(1 + \alpha_{\text{exp}})/T_{1/2\,\text{theor}}.$$

The table shows that the magnitudes of the hindrance factors are clearly divisible into two groups, for neutron and proton transitions (in accordance with the single-particle interpretation of isomeric transitions).[1,16] Within each group the values of the hindrance factors differ by less than a factor of three. For even-mass isomers the values are about one order of magnitude smaller than for odd-mass isomers. From the strictly single-particle model we would expect an inverse relation between the probabilities of neutron and proton transitions.[23]

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