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March 22, 1954

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ABSTRACT

The electron spectrum of Tl^{198m} has been investigated in a 25-cm double focusing beta spectrometer with a resolution of approximately 0.4 percent. The electron lines have been assigned to three gamma rays with energies (in kev) 261.5, 284, and 48.4, respectively. The multipolarities assigned from conversion ratio and lifetime considerations are M4, M1 + E2, and E2, respectively. A tentative decay scheme is proposed which is consistent with all the available data.

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I. INTRODUCTION

A 1.9-hour isotope of thallium was first observed and assigned to Tl^{198} by Orth et al.[1] Recently this activity has been independently assigned by two different groups of investigators to the decay of an isomeric state. The previously unobserved ground state decays by electron capture to Hg^{198} with a half-life of 5.3 ± 0.5 hours.

Michel and Templeton[2] of this laboratory produced these activities by the Au^{197} (α, 3n) Tl^{198} reaction in the laboratory's 60-inch cyclotron. Mass separation was made on a time-of-flight isotope separator[3] and the 1.75-hour and 5.3-hour activities shown to be Tl^{198}. Bergström, Hill, and DePasquali[4] at the University of Illinois produced the same activities by bombarding mercury with 11.5-Mev deuterons. Among the many electron lines they observed were several approximately 1.9-hour lines assignable to two gamma rays converting in thallium with energies of 282.4 and 260.7 kev, and a third gamma ray of 48.7 kev whose assignment was not unambiguous. The authors suggested that all three gamma rays were in cascade from an isomeric state having the unusually high spin of 9 with odd parity. Because of the unusual decay scheme suggested, the ambiguity of the 48.7 kev gamma ray's assignment, and the uncertainties in the photographically determined intensities, further work on this isomer was thought to be desirable.

II. EXPERIMENTAL WORK

Means of production and chemical separation. --In this investigation 1-mil gold foil was bombarded in the laboratory's 60-inch cyclotron with 40-Mev helium ions to produce the Tl^{198m} by the (α, 3n) reaction. This choice of foil thickness and bombarding energy was effective in minimizing the production of Tl^{199}.

The thallium was chemically separated from the gold target by the following process: 1) the gold was dissolved in aqua regia leaving the gold and thallium in solution as Au^{+3} and Tl^{+3} ions, respectively; 2) sulfur dioxide gas was bubbled through the solution to reduce the Tl^{+3} to Tl^{+} and the gold to the metal; 3) the solution was centrifuged and supernatant containing Tl^{+} and traces of Au^{+3} carried through varying purification procedures including an ethyl ether extraction; 4) the solution containing thallium was then passed through a Dowex A-2 ion exchange column in 6 M HCl after first oxidizing the Tl^{+} to Tl^{+3} with persulfate. The Tl^{+3} and last traces of Au^{+3} stick at the top under these conditions. Successively more dilute hydrochloric acid solutions were used for washing out impurities, 5) the Tl^{+3} was finally stripped from the column with water saturated with sulfur dioxide gas, any Au^{+3} being reduced to the metal and remaining on the column. This carrier-free solution was used to make the spectrometer samples.

Sample preparation. --The sample which gave the most clearly resolved electron peaks was one in which the thallium was vaporized onto a palladium leaf from a tungsten filament. The palladium leaf had a thickness of 160 $\mu g/cm^2$. This vaporized sample had the dimensions 1 mm x 6 mm.

Beta spectrometer. --The double focusing beta spectrometer used was one previously described by O'Kelley.[5] The detector has been changed from an end window to a side window Geiger counter. The detector window consisted of three layers of a vinyl copolymer film supported by a grid of 0.001-in. tungsten wires spaced 0.008-in. apart on a copper ring. The window energy cut-off was about 4 kev.

Electron spectrum. --The experimental data are summarized in Table 1. The spectrum itself is shown in Figs. 1 and 2. The K Auger lines are not included since at the time they were observed >70 percent of them were being produced by 5.3-hour electron capture of the ground state. It might be of interest, however, to note that we obtained for the ratio KLL/KLY/KXY the values 1.0/0.53/0.063 (where X and Y denote M, N, etc. orbital electron shells). The L Auger lines were also observed, but no inferences can be made therefrom because of the large and uncertain window absorption correction necessary at such low energies. In Table 1 the gamma ray energies are also given. Except for the 48.4 kev gamma ray, the energy values of the Illinois group[4] are more accurate and will be used in the following discussion.

It should be mentioned that there was initially present a very small percentage of 7.4-hour Tl^{199}. The only Tl^{199} electron line which coincided with any of those from Tl^{198m} was the 50.0_{L_I}. This line was of insignificant abundance at the time the $48.4_{L_{II}}$ and $48.4_{L_{III}}$ lines of Tl^{198m} were observed, but several hours later it was a useful calibration point for determining the energy of the 48.4 kev gamma ray.

The ratios given in Table 1 depend considerably upon the half-life assumed for decay corrections. The limits of error do not

necessarily encompass those introduced by the unknown uncertainty in half-life. We used a half-life value of 1.75 hours which was measured by Michel and Templeton[2] on a mass-separated sample.

Table 1. Electron spectrum data of Tl^{198m}.

Line	Abundance (assuming $\Sigma 260\ e^- = 100$)	
48.4_{L_I}	<1.5	$L_{II}/L_{III} = 1.11 \pm 0.1$
$48.4_{L_{II}}$	37.3	
$48.4_{L_{III}}$	33.6	$L/M = 3.7_6 \pm 0.1$
$48.4_{\Sigma M}$	18.9	$L/N = 13.4 \pm 1.0$
$48.4_{\Sigma N}$	5.3	
$\Sigma\ 48.4\ e^-$	95.0	
260.7_K	44.3	$K/L = 1.0 \pm 0.1$
260_L	26.0	
$260_{L_{III}}$	17.8	
260_{M+N}	11.9	
$\Sigma\ 260\ e^-$	100	
282.4_K	19.1	$K/L = 9 \pm 1$
282.4_{L_I}	2.1	

III. DISCUSSION

The gamma rays. -- Bergström and co-workers[4] found that the 260.7 kev gamma ray was definitely the cascade initiator, being of the M3 or M4 type with a possible admixture of electric radiation. Our K/L ratio (1.0) is in excellent agreement with that for pure M4 radiation.[6-8] According to the curves of Tralli and Lowen,[9] where L_{III}/L_I is plotted as a function of Z^2/E, the isomeric transition would be of M3 type. (L_{III}/L_I = 0.80 for M3 and 1.4 for M4). These curves, however, are based on approximate calculations and were performed for Z = 35.

Mihelich[10] has summarized the experimental values of L_{III}/L_I for M4 transitions in the same region of atomic number as thallium. Figure 3 shows these values with the inclusion of our value of 0.68 ± 0.07 for the 260.7 kev gamma ray of Tl^{198m}. Since all of these transitions occur in the same region of atomic number, one may plot these values versus energy. It is apparent that our value would fit reasonably well with the other four on a smooth curve, supporting our M4 assignment for the 260.7 kev gamma ray.

The experimental mean life of the 260.7 kev gamma ray may be calculated using the theoretical K conversion coefficients for M4 radiation from the tables of Rose, et al.[11] (18), our $260.7_K/\Sigma 260$ ratio (0.44), and the half-life of the isomeric state (1.75 hours). The mean life thus calculated is 3.7×10^5 seconds. The theoretical value one obtains using the nomogram of Weisskopf's lifetime-energy-spin formula prepared by Montalbetti[12] is 5×10^5 seconds. This agreement may be fortuitous. The M4 assignment is therefore reasonable from

the comparisons we have been able to make with existing theories and empirical correlations.

Because of the high intensity of the Tl^{199} 50.0_{L_I} line, Bergström et al.[4] were unable to resolve the $48.4_{L_{III}}$ line. As a result they designated the $48.4_{L_{II}}$ line as 48.7_{L_I}. The two lines we observe can be only an L_{II}-L_{III} pair, and using the 50.0_{L_I} line of Tl^{199} as a standard, we arrive at an energy of 48.4 ± 0.2 kev for the gamma ray. The L_{II}/L_{III} ratio is in better agreement with E2 (1.6) than with E1 (~1.8) or M1 (~900) although only M1 can be ruled out on this basis.[13] The E1 assignment was eliminated by a scintillation spectrometer experiment which determined the total conversion coefficient of the gamma ray to be greater than 10. Therefore, the E2 assignment seems to be the only reasonable one.

The Illinois group has shown the 260.7 and 282.4 kev gamma rays to be converted in thallium. Coincidence measurements performed by Mr. Frank Stephens of this laboratory have shown the 282 kev gamma ray to be in coincidence with K X-rays. Thus it appears that the 282.4 and 260.7 kev gamma rays are in cascade. Assuming no electron capture from excited states of Tl^{198} the K conversion coefficient of the 282.4 kev gamma ray can be calculated from the intensity ratio $(282_K)/(260_{K+L+M...})$. The value (0.24) thus obtained may indicate a mixture of E2 (0.076) and M1 (0.52) radiation.[11] The high K/L ratio (9 ± 1) of this gamma ray supports an M1 assignment.[8]

If the 48.4 kev gamma ray is emitted in cascade with the other two and is assumed to be E2 radiation, the total intensity of its conversion electrons should equal the total intensity of those of the 260.7 kev gamma ray. As can be seen from Table 1 these intensities are equal within

experimental error. This does not exclude the possibility of an approximately 50 percent electron capture branching from the 1.75-hour isomeric state, with the 48-kev transition taking place in Hg^{198}. The L_{II}-L_{III} binding energy differences are too similar in mercury and thallium to allow an assignment on that basis. If, however, the electron capture were occurring, one would expect to observe gamma rays from levels in Hg^{198m} in high intensity with a 1.75-hour half-life. A scintillation spectrometer experiment gave no indication of such gamma rays. We can therefore conclude that the three gamma rays are very probably emitted in cascade as suggested by the Illinois group.[4]

Spin assignments. -- If one assumes that our assignments of multipolarity are correct and that the three gamma rays are in cascade, a tentative decay scheme can be constructed (see Fig. 4) in which the spin difference between the 1.75-hour Tl^{198m} and the 5.3-hour Tl^{198} can be as high as 7 with a parity change. The fact that no crossover radiation was observed supports this large spin difference. The positions of the 282.4 and the 48.4 kev transitions may be as shown in Fig. 4 or reversed. The spin alternatives in the left column in Fig. 4 are based upon the assumption of a 2- ground state for Tl^{198}.[4] The alternatives in the right column are based upon the assumption of a coupling between the spins of the 81st proton and the 117th neutron. The 260.7 kev M4 transition would then represent a transition of the 81st proton from an $h_{11/2}$ to a $d_{3/2}$ configuration, the 117th neutron remaining in an $f_{5/2}$ configuration; the 282.4 kev M1 + E2 transition may perhaps represent a transition of the same proton between the $d_{3/2}$ and $s_{1/2}$ configurations, the neutron again remaining in the $f_{5/2}$

configuration; the 48.4 kev E2 transition then represents a transition of the odd neutron from its $f_{5/2}$ to a $p_{1/2}$ configuration, the proton remaining in an $s_{1/2}$ configuration. This interpretation is, of course, extremely speculative.

Also shown in Fig. 4 is the electron capture decay from Pb^{198} which was first observed by Neumann and Perlman.[13] If the 25-minute Pb^{198} parent of Tl^{198m} is the 0+ ground state of Pb^{198}, then Tl^{198m} must have a low spin and Tl^{198} must have a high spin. The assignment of a high spin to Tl^{198} is in sharp disagreement with the interpretation of its electron capture decay given by Bergström and co-workers.[4] These workers' data indicate that the electron capture decay of Tl^{198} goes predominantly to the 2+ first excited state of Hg^{198}. Thus of the two isomeric states of Tl^{198} the 1.7-hour Tl^{198m} most probably has the higher spin. The fact that other even-even lead isotopes have high spin isomers suggests that Pb^{198} might also have one. For example, the isomeric state of Pb^{204} has been suggested to be 6+ [15] or 7-.[16] Very recently Molder and Wapstra[17] have found indication of a 9- isomeric state in Pb^{202}. Alburger[18] assigned the isomeric level in Pb^{206} as 7-. If Pb^{198} has a similar high spin isomer, direct electron capture decay from a high spin Pb^{198m} to a high spin Tl^{198m} would be possible. We therefore conclude that there is indeed another case of isomerism in Pb^{198}.

It is apparent that a study of the Tl^{198} electron capture decay is needed to make more definite spin assignments for Tl^{198} and Tl^{198m}. We can only state that the spin difference between the isomers may be as high as 7 and that Tl^{198m} most probably has the higher spin.

The authors wish to acknowledge the help and cooperation of Mr. G. B. Rossi and the crew of the 60-inch cyclotron in making the bombardments necessary for this investigation. We appreciate the assistance of Mr. Frank S. Stephens in making the coincidence measurement.

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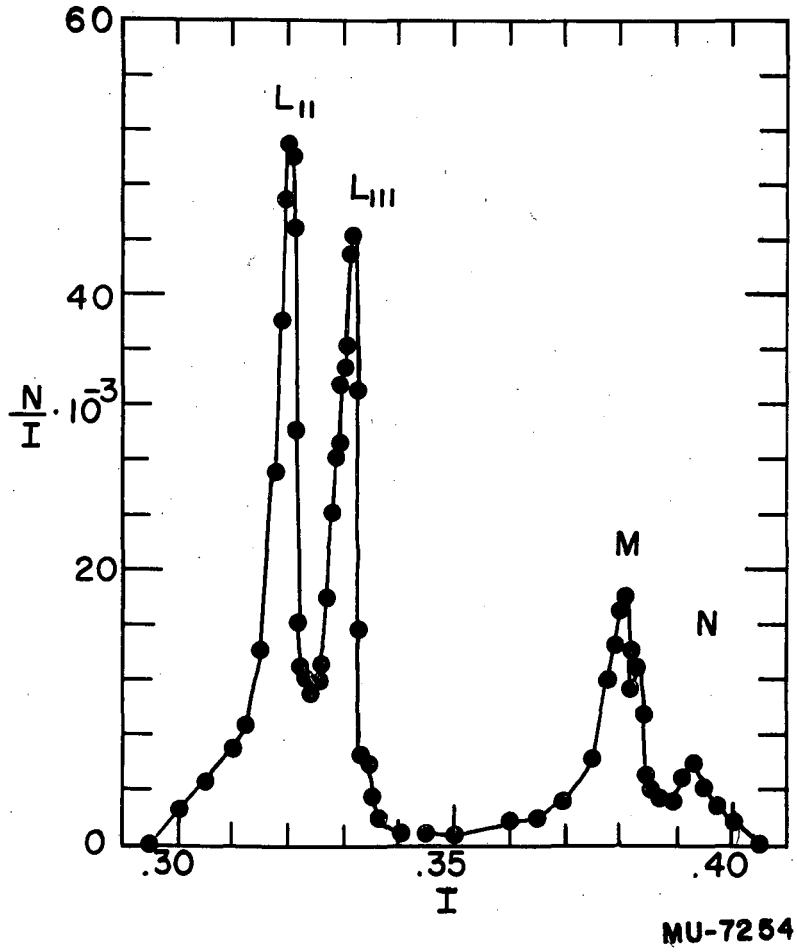

Fig. 1. Electron lines from the 48.4 kev gamma ray of Tl^{198m}.

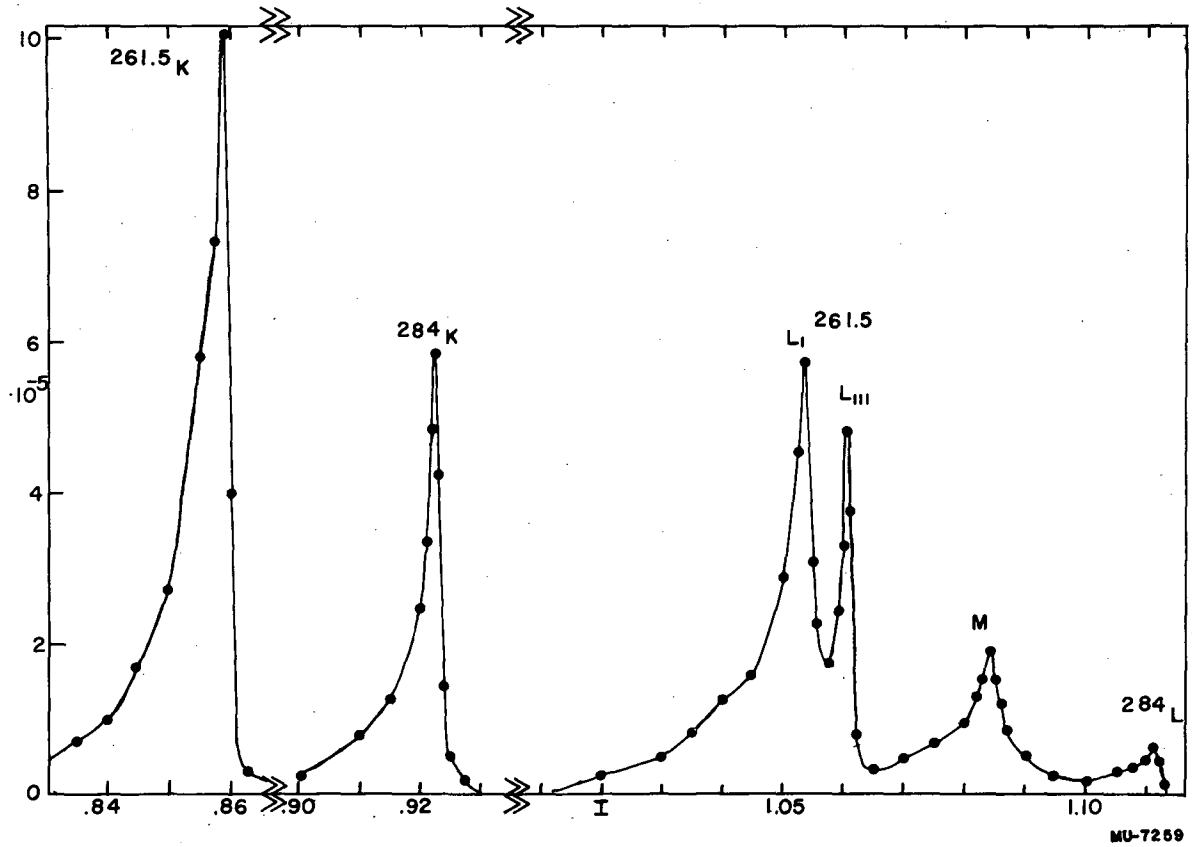

Fig. 2. Electron lines fromthe 261.5 kev and 284 kev gamma rays of Tl^{198m}.

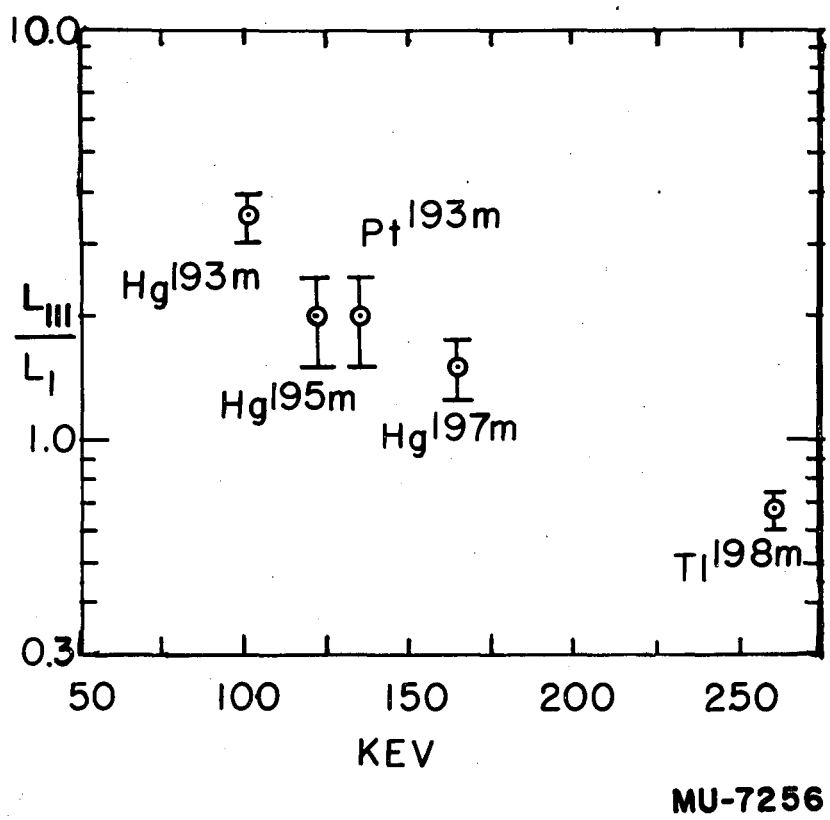

Fig. 3. Plot of experimental L_{III}/L_I ratios for some M4 transitions versus energy in kev.

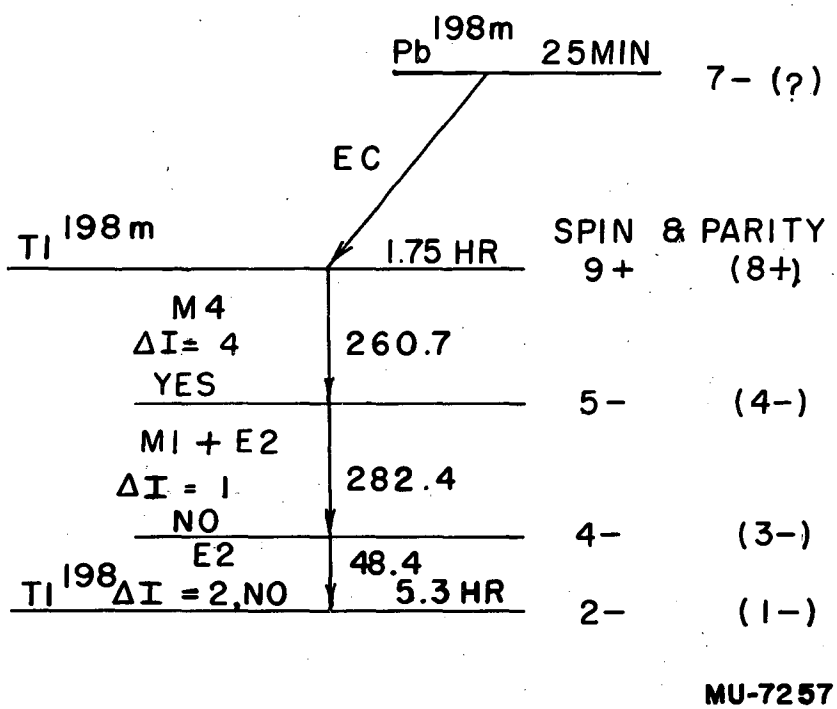

Fig. 4. Tentative decay scheme of Tl^{198m}.