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METASTABLE STATES IN THE LIGHT THALLIUM ISOTOPES PRODUCED IN HEAVY-ION REACTIONS

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R. M. Diamond and F. S. Stephens

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This paper reports an investigation of relatively short-lived isomeric states produced in the odd-mass thallium isotopes. Suitable targets were irradiated at the HILAC with heavy ions ranging from $_2He^4$ to $_{10}Ne^{20}$ to produce excited thallium nuclei of mass number 200 to mass 191 according to the type reaction,

$$_{81-z}X^a + {}_zY^b \rightarrow {}_{81}Tl^{a+b-x} + xn$$

Those transitions following the decay of a metastable state of millisecond to minutes half-life could then be observed very cleanly in the intervals between the three millisecond long HILAC beam bursts (65 to 165 milliseconds, depending upon the repetition rate used), by either a gamma-ray or, most often in the present study, an electron spectrometer. By looking during the beam bursts, the more numerous prompt transitions could also be studied, and after the irradiation, the longer-lived transitions (including those of the daughter activities) could be observed with the HILAC turned off.

From the electron spectra, the energies and intensities of the various lines could be determined. Mass assignments of these lines could be made from the nature (projectile, target, and energy) of the irradiation and, in many cases, the lines could be identified with previously known transitions.[1] Irradiations at two slightly different projectile energies helped with the mass assignments and showed which transitions belonged to the same cascade. Measurement of the half-lives of the principal electron lines observed also showed which transitions were associated with a particular

metastable state, as well as determining the half-life of that state. From the experimental K/L electron ratios, multipole assignments were made using the theoretical conversion coefficients of Sliv and Band.[2] The elemental assignments of the lines, that is, whether they occurred in the original thallium nucleus or in the mercury daughter or gold granddaughter, were made from the energy difference between the K and L conversion electron lines.

Figures 1-4 illustrate the type of experimental results obtained. The first figure is an electron spectrum from a 22 MeV He^4 irradiation of gold taken with the counting equipment gated off for an 8 msec period, centered about and including the 3 msec beam pulse. The resolution of this spectrum is about 1.2% down to energies of about 25 keV, where the target thickness causes additional broadening. In regions of particular interest better resolution was desirable; short sections at about 0.6% resolution were taken and are inserted in Fig. 1 beneath the corresponding portions of the lower-resolution spectrum. Essentially only the α,n and α,2n reactions occur with 22 MeV He^4 ions on gold, and the assignment of the lines to either Tl^{200m} or Tl^{199m} was quite easily made based on the variation of their intensities with He^4 ion energy and their grouping by half-life. The transitions of 29, 367, 382, and 720 keV all showed a half-life of 27 ± 4 msec; those of 213 and 539 keV gave 37 ± 4 msec. The 353-keV transition was too weak to measure without more care, but it and the 367- and 720-keV transitions were already known to be associated with Tl^{199} from studies of Pb^{199} decay.[1]

Figure 2 is a gamma ray spectrum taken under similar conditions to those of Fig. 1. Here the resolution is too poor to indicate much; however, coincidences between the prominent 370-keV photons and others of comparable energy were quite intense. An angular distribution of these coincidences was measured and will be discussed.

Levels in Tl^{199} at 367 and 720 keV were previously known,[1] and a metastable state of 42 msec half-life has been observed.[3] The transitions studied in this work and with the properties summarized in Table 1 suggest the isomeric level has a 27 msec half-life and is at 749 keV. We measure the K/L ratio of the 382-keV transition as 0.88 ± 0.05. The theoretical E3 K/L ratio[2] is 0.90, and taken together with the 27 msec half-life, this clearly indicates an E3 transition. The K/L ratio of the 367-keV transition is 2.5 ± 0.3, and since this is almost undoubtedly an M1-E2 mixture (from previous work,[1] and also by analogy to all the other known odd-mass thallium isotopes), we then obtain for the mixing, 77 ± 8% E2 and 23 ± 8% M1. This mixture gives the total transition intensity listed in column 5 of Table 1. To estimate the intensity of the 353-keV transition, we have used the value 75% M1-25% E2 determined previously;[1] the intensity of the 720-keV transition was calculated on the basis that it is a pure E2 transition. Our ratio of the intensities of the 720- to the 353-keV transitions is roughly in agreement with the previous value.

The spin of the ground state of Tl^{199} is measured[4] to be 1/2, and since the 367-keV level is connected with this state by an M1-E2 transition, it must have spin 3/2. The parity of these levels is very likely positive as will be discussed later. The 720-keV level is probably the 5/2 state which occurs systematically at about this energy in the heavier thallium isotopes. These assignments were previously suggested by Andersson et al.[1] The 749-keV isomeric state, then, very likely has spin 9/2 since it is connected with a state of spin 3/2 by an E3 transition. The parity would be negative if that of the lower two states is positive, as suggested.

A 9/2- state is not expected at this energy in Tl^{199}, and therefore we have measured the angular distribution of the 367- and 382-keV photons in order to confirm this spin. Because the intermediate level in the sequence has a spin of only 3/2, the simple angular correlation expression, $W(\theta) = 1 + A_2P_2(\cos\theta)$, is applicable. The

constant, A_2, was determined by measuring the yield of the 367-382-keV gamma-ray group in coincidence with itself, using two NaI counters set alternately at 90° and at 180° to each other. Our value for A_2, corrected for the finite solid angle of the NaI crystals is $+0.20^{+0.04}_{-0.03}$. The theoretical value for the 9/2 (E3) 3/2 (M1-E2) 1/2 sequence, with $\delta = \sqrt{\frac{M1}{E2}}$ taken to be positive as is found for the heavier odd-mass thallium isotopes[5] (and corrected for a 5% contribution from the 353-367-keV cascade), is +0.23. The agreement here is satisfactory. Unfortunately, the sequence 11/2 (E3) 5/2 (M1, unobserved) 3/2 (M1-E2) 1/2, gives the same theoretical value of A_2. However, this sequence seems unlikely as the energy sum, 29 + 353 = 382, would require that the energy of the unobserved M1 transition be equal to or less than our experimental error; that is, less than about 2 keV.

These results lead to the level scheme for Tl^{199m} shown in Fig. 5. By this scheme the intensity of the 367-keV transition must be equal to the sum of the intensities of the 382- and 353-keV transitions. This is shown to be the case in column 5 of Table 1. The 29-keV transition (presumably M2) represents about a 7% branching of the 749-keV level.

In order to produce Tl^{197m}, the He^4 ion energy was increased to about 42 MeV. To reduce the amounts of the heavier thallium isotopes produced, a relatively thin gold target (5 mg/cm^2) was used.

An electron spectrum taken under these conditions, and with the counters gated off during the beam pulse is shown in Fig. 3. The lines belonging to the relatively long-lived Tl^{197}, Tl^{198} and Tl^{198m} were easily identified by rerunning a spectrum immediately after the beam was turned off. These long-lived lines are indicated in the figure, as is one of the lines previously identified in Tl^{199m}, which is quite weak here. The remaining lines are associated with two transitions of 222 and 385 keV.

These we attribute to Tl^{197m}, and they have been seen previously in studies of Pb^{197m} decay. The half-lives we determined for the two lines were the same, namely 0.53 ± 0.03 sec, and this value is in good agreement with the 0.55 sec half-life previously measured for Tl^{197m} (ref. 6).

The data on the Tl^{197m} electron lines are summarized in Table 1. Our value for the K/L ratio for the 222-keV transition is 0.28 ± 0.05. The theoretical value for an E3 transition[2] is 0.24, and the previously measured value is 0.28, so we are in agreement with the previous assignment[1] of this transition as E3. For the 385-keV transition, our K/L ratio is 3.4 ± 0.4, which assuming an M1-E2 mixture, gives 82 ± 10% E2 and 18 ± 10% M1. The spin of the ground state of Tl^{197} has been measured[4] to be 1/2. Again, the E2-M1 mixture of the 385-keV transition requires that the first excited state can only have spin 3/2, and the isomeric state then most likely has spin 9/2. In no case can the spin be higher than 9/2, unless one again invokes a low energy unobserved transition.

The level scheme we deduce is quite simple and is shown as heavy lines in Fig. 5. The cascade arrangement requires equal intensities for the two transitions, and the agreement here is shown in the last column of Table 1 to be good. The main point of disagreement between this and the previous work[1] is that we have the E3 transition terminating at the first excited state rather than at the (former) second excited state of 772 keV. If the 772-keV state were populated by the E3 transition, we would not have resolved the resulting 385- and 387-keV cascade transitions in this work; however, the intensity of the composite K line would then have been well over three times the intensity observed. Such a situation can be completely ruled out on the basis of the present data.

The C^{12},4n reactions on Re^{187} and Re^{185} produce Tl^{195m} and Tl^{193m}, respectively, and such reactions can be rather clean if the C^{12} ion energy is only slightly above the barrier energy. To separate Tl^{195m} from Tl^{193m} one can produce the latter alone by bombarding Ta^{181} with O^{16} ions of the appropriate energy.

Figure 4 shows the electron spectrum of rhenium bombarded with 67 MeV C^{12} ions, and directly beneath is the spectrum of tantalum bombarded with 79 MeV O^{16} ions. In both spectra the counters were gated off during the beam pulse. Transitions of 99 and 383 keV have been assigned to Tl^{195m} (see also ref. 1 and 7), and a single transition of energy 365 keV is assigned to Tl^{193m}. The data on these lines is summarized in Table 1. The half-life of the 383-keV transition was measured and gave 3.6 ± 0.4 sec, in good agreement with the previous value of 3.5 sec for Tl^{195m} (ref. 6). The half-life of the 365-keV transition was found to be 2.11 ± 0.15 min.

The level scheme proposed for Tl^{195m} is shown in Fig. 5. Our data do not show that the 99-keV transition is E3; however, it was so assigned previously[1,6] and we find no evidence to suggest otherwise. Our K/L ratio for the 383-keV transition is 3.8 ± 0.3, which indicates 74 ± 8% E2 and 26 ± 8% M1. The level scheme suggested is strictly analogous to the previous two odd-mass thallium isotopes, and differs from the earlier work again in that the 99-keV E3 transition terminates at the 383-keV 3/2+ level rather than at the 776-keV 5/2+ one. This is clearly shown in our spectrum by the absence of the K line of the 393-keV transition, which would have been over twice as intense as the K line of the 383-keV transition if both transitions were in the cascade. The total intensities of the 99- and 383-keV transitions are given in column 5 of Table 1, and we feel these are sufficiently similar to be consistent with the cascade arrangement of Fig. 5. Our use of thick targets produced sizeable tails on the lines, and for energies as low as 90 keV estimation of this tail can be rather uncertain.

The level scheme proposed for Tl^{193m} is also shown in Fig. 5. In this case the E3 transition is not observed, and our experimental limit on the energy is less

than 25 keV. From the pattern established by the mass 199, 197, and 195 isomers (Fig. 5), such an upper limit appears to be quite reasonable and suggests why the isomeric state is observed only through the decay of the following 365 keV transition. The latter appears to correspond to the usual first excited state-to-ground transition of the odd-mass thallium isotopes; the K/L ratio of 3.8 ± 0.4 observed indicates 71 ± 10% E2 and 29 ± 10% M1. The systematics shown in Fig. 5 also indicate that the 9/2- level may well become the first excited state in Tl^{191}, and so can only decay by a relatively slow M4 transition or by electron capture; no E3 transition or any short-lived transitions, in fact, were observed for Tl^{191}.

Thus in all four of the odd-mass nuclei studied, the E3 transitions must come from a 9/2- level, rather than a 11/2- level, unless the sequence 11/2 (E3) 5/2 (unobserved M1) 3/2 (M1-E2)1/2 occurs systematically. We can set an upper limit for the energy of the unobserved M1 transition of 2 keV in Tl^{199}, and of 25 keV in the other three cases. We feel that an 11/2 spin is virtually completely ruled out by the necessity of such unobserved low energy transitions in all four cases, and by the difficulties in accounting for 5/2+ levels, never directly observed, but occurring systematically in all these nuclei a few keV above the first excited state (of spin 3/2).

One of the main results of this work, then is the assignment of the isomeric levels in Tl^{199} and Tl^{193} as 9/2- states, as well as the corresponding ones in Tl^{197} and Tl^{195}, previously assigned as $h_{11/2}$ single particle states.[1,7] Since there is no 9/2- level available in this region from a strict single-particle model approach, the surprising existence of such low-lying levels is strong evidence that collective states play a part in the lower excited states of the light thallium isotopes. One way of producing such a 9/2- level is to couple the $h_{11/2}$ proton hole to a collective 2+ excitation (vibration) of the residual even-even core.[8] Another more detailed way is

to couple the $h_{11/2}$ proton hole explicitly to two unpaired neutrons or neutron holes. This would be an extension of the well-known coupling of three (or five or seven) nucleons of spin j to a total spin of j-1,[9-11] the cases of anomalous spin in the original independent particle model.[12] But the thallium 9/2- level would be the first example of a single nucleon of spin j (proton $h_{11/2}$) coupling to the other type of nucleon pair (or pairs) to give a total spin of j-1 lying lowest.

Although the 9/2- level lies lowest, the 11/2- and/or 13/2- level must not lie very much higher in energy in $Tl^{197,195}$. The half-lives for the electron capture decay of $Pb^{197m,195m}$ rule out the possibility of direct electron capture from these $i_{13/2}$ isomeric states in lead to the 9/2- states in thallium; such first forbidden unique decays would require impossibly large decay energies to yield the observed half-lives, for example, ~55 MeV for Pb^{197m}. Thus the electron capture decays of the 13/2+ lead isomers most likely proceed via 11/2- or 13/2- levels in the odd-mass thalliums.

FOOTNOTE AND REFERENCES

*This work was done under the auspices of the U. S. Atomic Energy Commission.

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Table I. Electron lines in odd-mass thallium isotopes.

Mass No.	Transition energy (keV)	Conv. electrons observed	Multipolarity	Transition intensity
199m	29	M,N	(M2)	
	353	K	75% M1- 25% E2	69
	367.0[a]	K,L	23% M1- 77% E2	1420 ± 240[c]
	382	K,L,M+N	E3	1290
	720	K	(E2)	27
197m	222.45[b]	K,L,M+N	E3	230
	385	K,L	18% M1- 82% E2	260 ± 40[c]
195m	99	L,M+N	E3	148
	383	K,L,M+N	26% M1- 74% E2	120 ± 20[c]
193m	365	K,L,M+N	29% M1- 71% E2	

[a]Used as an energy standard from Andersson et al., ref. 1.

[b]Used as an energy standard from Andersson et al., ref. 6.

[c]Error limit from the uncertainty in the M1-E2 ratio.

FIGURE CAPTIONS

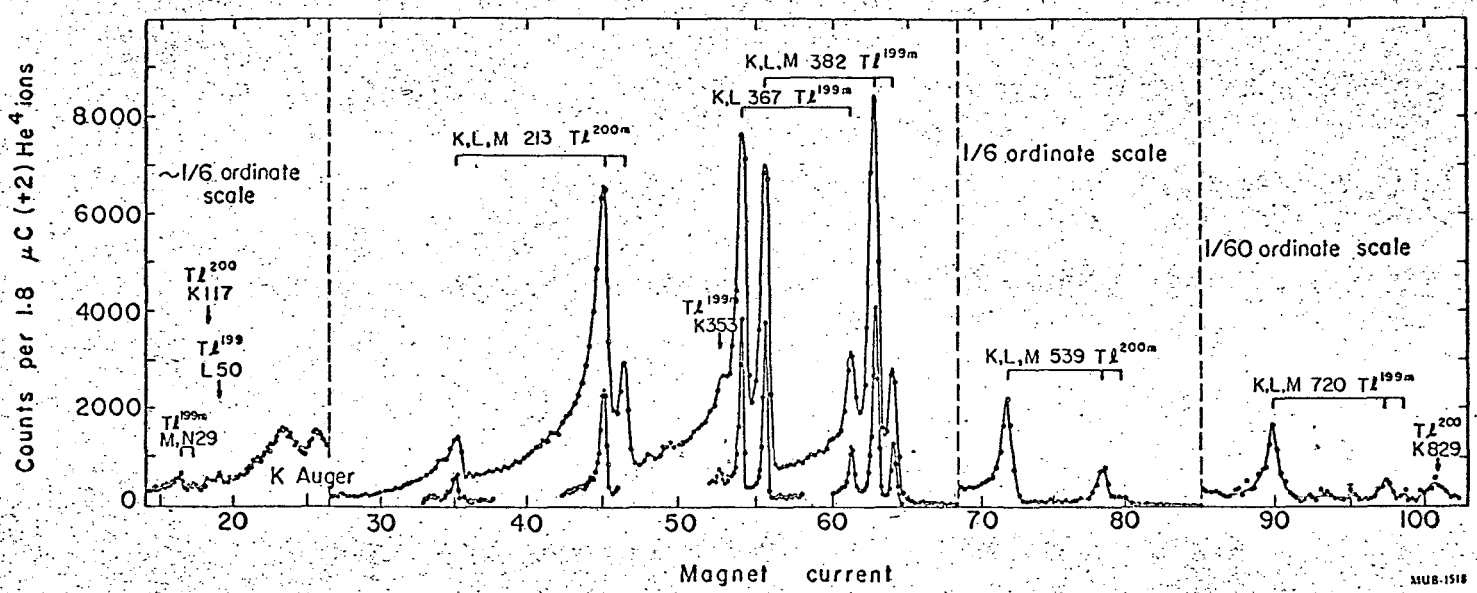
Figure 1. Electron spectrum from Au^{197} irradiated by 22 MeV He^4. The counter was gated on only between the beam pulses. The upper curve was taken at 1.2% resolution, while the lower portions were taken at 0.6% resolution.

Figure 2. Gamma-ray spectrum of Au^{197} irradiated by 22 MeV He^4. The counter was gated on only between the beam pulses.

Figure 3. Electron spectrum from a thin Au^{197} target irradiated by 42 MeV He^4. The counter was gated on only between the beam pulses.

Figure 4. Electron spectrum from (a) natural rhenium target irradiated by 67 MeV C^{12} and (b) Ta^{181} target irradiated by 79 MeV O^{16}. The counter was gated on only between the beam pulses.

Figure 5. Level schemes of the odd-mass thallium isotopes.


Counts per 1.8 μC (+2) He4 ions
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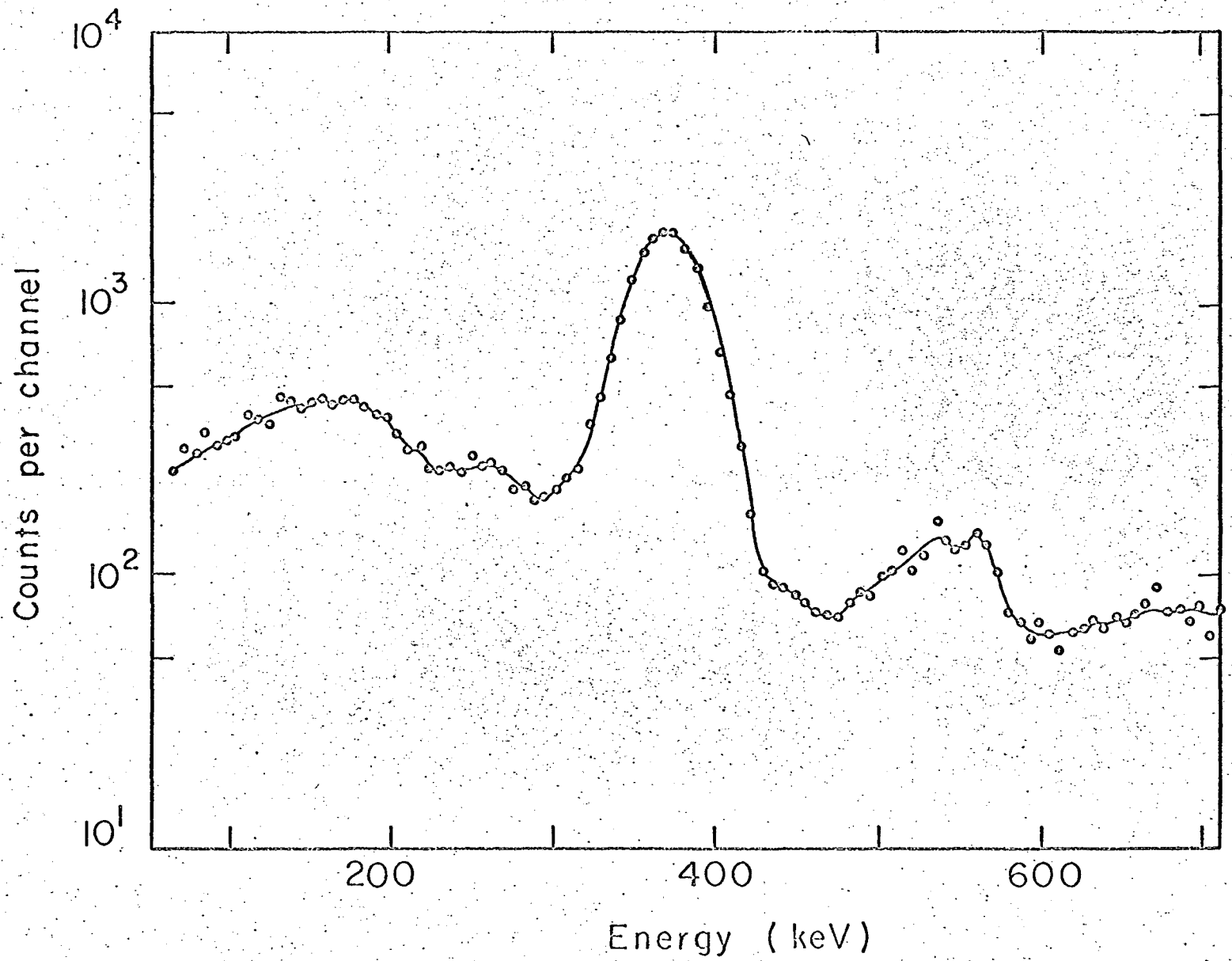
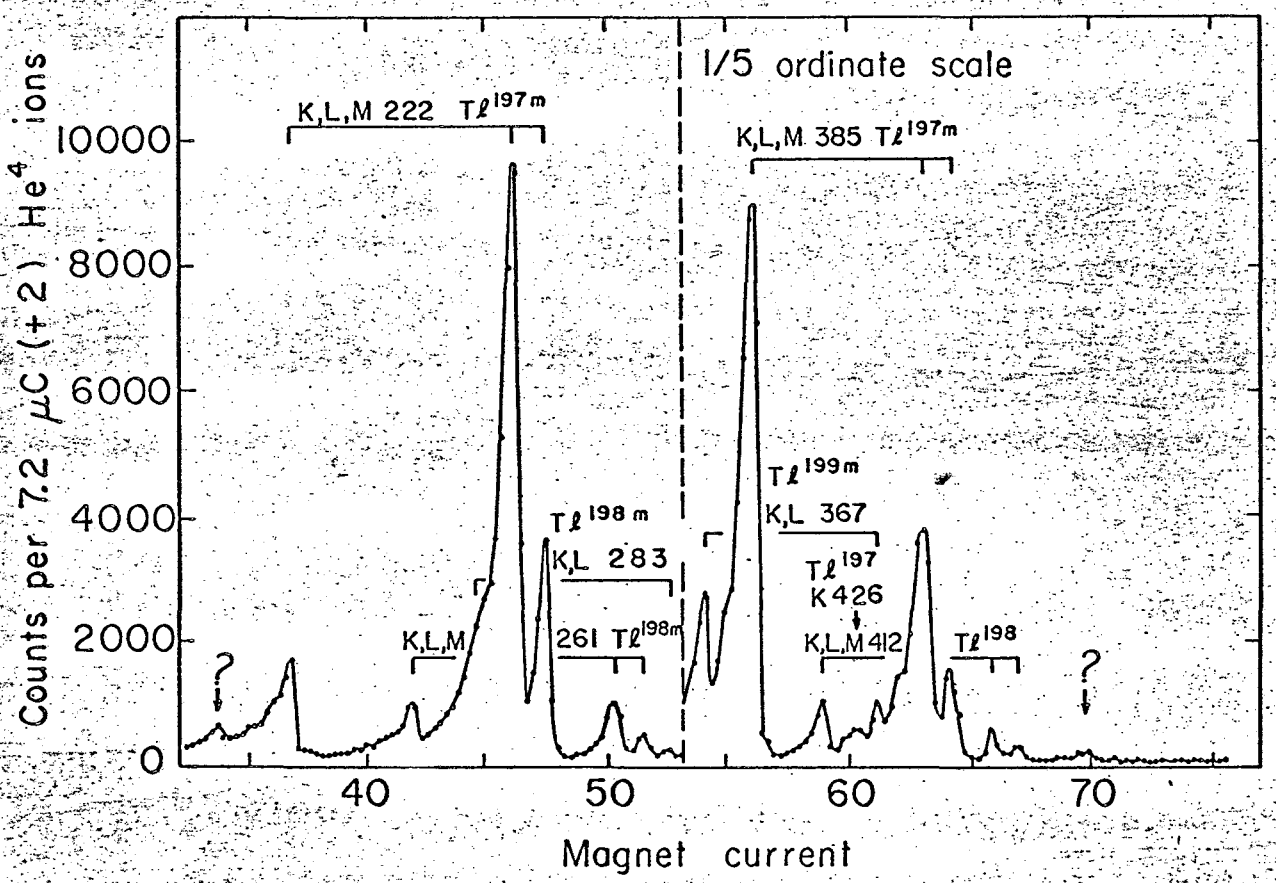
6000
4000
2000
0
~1/6 ordinate scale
Tl200 K117
Tl199 L50
Tl199m M,N29
K Auger
K,L,M 213 Tl200m
Tl199m K353
K,L 367 Tl199m
K,L,M 382 Tl199m
1/6 ordinate scale
K,L,M 539 Tl200m
1/60 ordinate scale
K,L,M 720 Tl199m
Tl200 K829
20
30
40
50
60
70
80
90
100
Magnet current
MUB-1518

Fig 1



MU-28930

Fig 2



MUB-1517

Fig 3

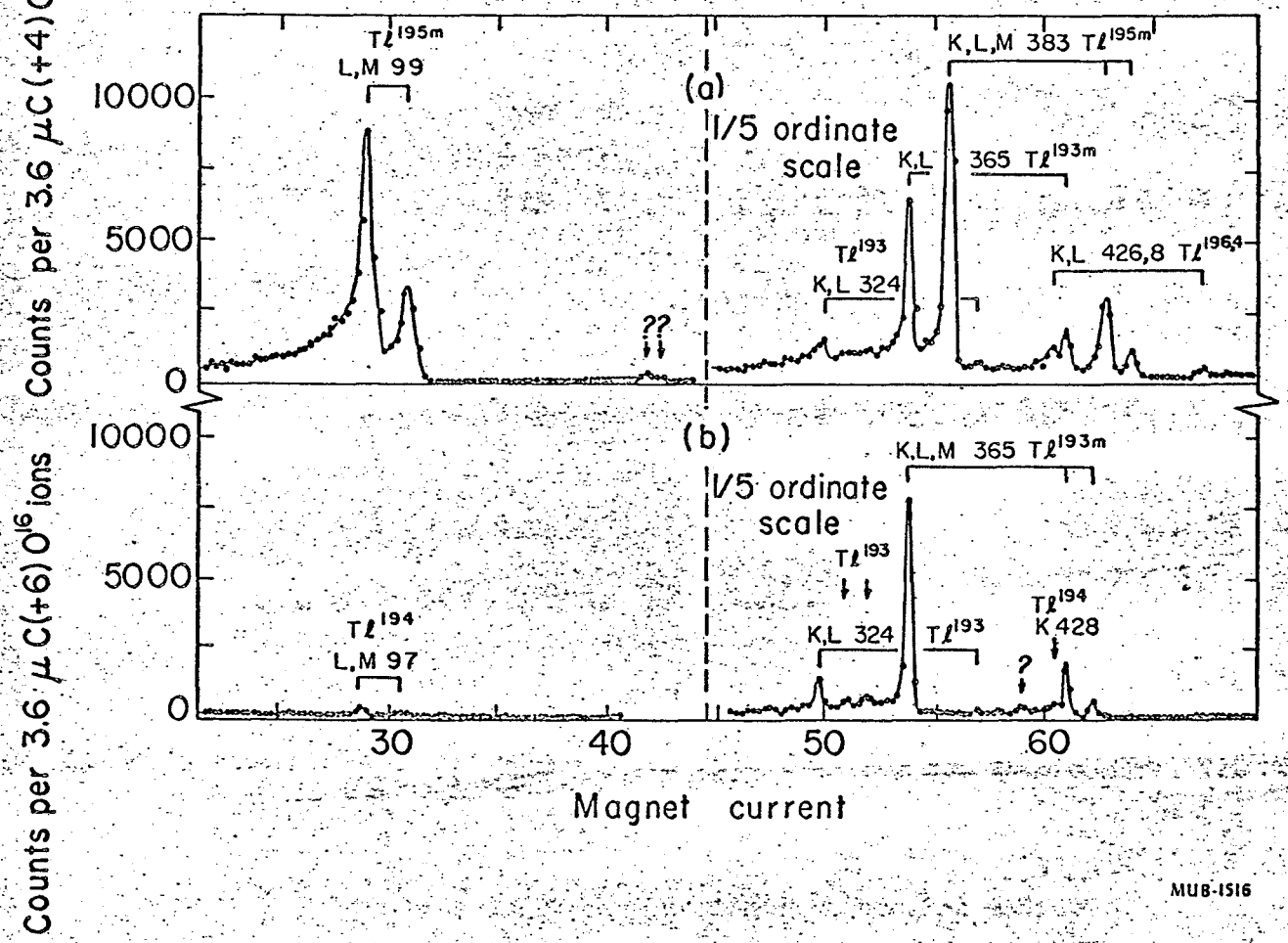
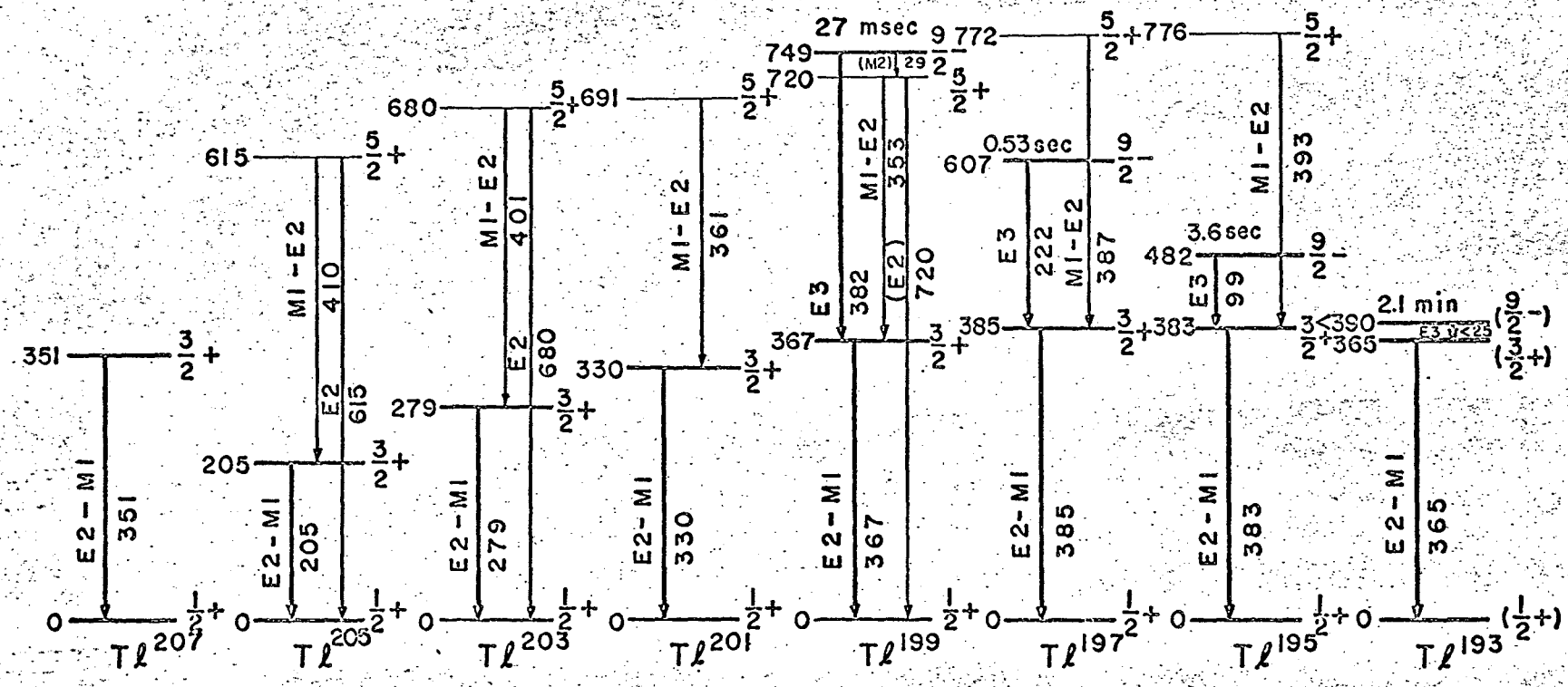

Counts per 3.6 μC(+4) C12 ions
Counts per 3.6 μC(+6) O16 ions
10000
5000
0
(a)
1/5 ordinate scale
Tℓ195m
L,M 99
??
K,L,M 383 Tℓ195m
K,L
365 Tℓ193m
Tℓ193
K,L 324
K,L 426,8 Tℓ196,4
(b)
1/5 ordinate scale
K,L,M 365 Tℓ193m
Tℓ193
K,L 324
Tℓ193
Tℓ194
K 428
?
Tℓ194
L,M 97
30
40
50
60
Magnet current
MUB-1516

Fig 4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749
720
772
776
680
691
615
607
0.53 sec
482
3.6 sec
2.1 min
390
365
351
367
385
383
279
330
205
0
(M2)
29
E3
382
(E2)
720
353
MI-E2
222
387
393
99
E3 V≤25
401
361
410
615
E2
680
E2-MI
351
205
279
330
367
385
383
365
Tℓ207
Tℓ205
Tℓ203
Tℓ201
Tℓ199
Tℓ197
Tℓ195
Tℓ193
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Fig 5

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