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ISOMERIC LEVELS IN THE LIGHT THALLIUM ISOTOPES

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R. M. Diamond and F. S. Stephens

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ABSTRACT

A study has been made of the metastable states produced in certain heavy-ion reactions by observing the gamma-ray and conversion-electron transitions occurring between the beam bursts of the HILAC at the Lawrence Radiation Laboratory. Targets and projectiles were chosen which produce thallium isotopes ranging from mass 200 to mass 191. New E3 isomers were found in Tl^{200}, Tl^{199}, and Tl^{193}. The isomer in Tl^{200} is apparently a 7+ level, in close analogy with other known even-mass thallium isotopes. Considerable evidence was accumulated that in the odd-mass thallium nuclei, including the previously studied Tl^{197} and Tl^{195}, the metastable state has spin and parity 9/2-. Such a state is not expected to occur at low energies in these nuclei, and its nature, therefore, presents an interesting problem.

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1. Introduction

This paper reports the beginning of a series of investigations of short-lived activities and of gamma-ray cascades occurring as the last step of a nuclear reaction. The work is being carried out at the Berkeley Heavy Ion Linear Accelerator (HILAC), and due to the pulsed nature of the machine, three time scales can be considered. If the de-excitation transitions are observed during the 2-3 millisecond beam bursts, all prompt transitions and those with periods up to as long as a few milliseconds can be observed. For half-lives of metastable or ground states in the milliseconds to minutes range, the transitions are best studied in the intervals between beam bursts (65 to 165 milliseconds, depending on the beam repetition rate used). Longer half-lives can be studied in the usual way with the machine turned off.

The technique of observing the target between the beam bursts has been used in the present work. By irradiating suitable targets with heavy ions from ${}_{2}He^{4}$ to ${}_{10}Ne^{20}$, excited thallium nuclei from mass number 200 to mass 191 have been produced according to the type reaction,

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

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After evaporation of the x neutrons, the residual thallium nucleus loses the last few MeV of its excitation by emission of gamma-rays. If a metastable state of millisecond to minutes half-life occurs in this cascade, those transitions following the decay of the isomeric level will be kept alive into the time interval between beam bursts, and can be observed without interference from all the more numerous prompt transitions occurring in the nuclear reaction. Of course, if the ground state itself is radioactive, the transitions in the decay of the thallium to the mercury daughter will also appear in the gamma-ray spectrum, but with the different half-life characteristic of the ground-state decay.

Because of the known existence of metastable states of the appropriate millisecond to seconds half-lives in some light thallium nuclei,[1,2,3] it was felt that a study of these nuclei would provide a good test of the experimental techniques. Because the levels in some of these nuclei have already been studied in the decay of the corresponding lead isotopes, many of the transitions observed in this work were previously known. The greater simplicity of the present method over the study of the decay of a mixture of lead isotopes has yielded some surprising results.

2. Experimental

The de-excitation spectra of the excited nuclei produced in the irradiation can be observed using either gamma-ray or conversion electron spectrometers. Because of the higher resolution possible with the latter, most of the work reported here has involved an electron spectrometer of the single wedge-gap type developed by Kofoed-Hansen, Lindhard, and Nielsen.[4] This spectrometer has been described previously.[5] The detector used is an anthracene crystal cemented to an RCA 6655A photomultiplier tube. After amplification, the pulses from the phototube are passed through a single-channel analyzer in order to reduce the background. The operation of the spectrometer is semi-automatic. After a certain (pre-set) amount of beam has irradiated the target, the number of counts that have passed through the single-channel analyzer is recorded in a 100-channel analyzer operating as a multiscaler, and the magnet current is increased by a small pre-determined quantity. The magnet current remains at this new value until the selected amount of beam has again struck the target and the number of counts at this new setting has been recorded in the next channel of the multiscaler; then the current is again raised by the small increment. This can be repeated until 98 points have been taken, and then the magnet must be re-cycled and the programmer reset in order to continue taking the spectrum.

The energies of the conversion electron lines observed were determined by interpolation between lines known from other measurements. The known and unknown lines were all measured in the same 98 channel magnet sweep so as to avoid errors due to hysteresis effects in the magnet.

The relative intensities of the conversion electron lines were obtained from the electron spectra by integrating the electron peaks, subtracting out background, dividing by the value of $H\rho$ for the line, and multiplying

by a correction factor $\partial(H\rho)/\partial I$ to account for the fact that the points in the spectra are taken for equal magnet current increments rather than equal increments in $H\rho$. The resulting relative electron intensities given in tables 1-5 are normalized to the strongest line of a particular spectrum. From these relative intensities, K/L conversion electron ratios were determined, and from the ratios, multipole assignments were made using the theoretical conversion coefficients of Sliv and Band.[6] Applying the theoretical K-electron conversion coefficients to the relative intensities of the K-lines allows calculation of the relative gamma-ray and total (conversion electron + gamma-ray) transition intensities.

Two- or three-point decay curves can be taken for the 98 channel spectrum by dividing the interval between beam pulses into two to three equal sections (timewise) and recording each section separately on an analyzer used as a multiscaler. For longer half-lives, a multi-channel analyzer can be used as a time analyzer for the decay of a single line in the spectrum. Thus, it is not difficult to obtain decay curves for the observed conversion lines.

The assignment of the transitions produced in a particular bombardment was done in various ways. The nature (projectile, target, and energy) of the irradiation allowed a very limited number of possibilities. Many of the transitions could be identified with previously known transitions either in the thallium nuclei themselves or in their decay products. When the irradiations were done at two slightly different projectile energies, the relative changes in the yields of the conversion electron lines showed which belonged together in the same decay scheme and indicated which of the (usually two) mass numbers possible was most probable. The elemental assignment of the lines, that is, whether they occurred in the original thallium nucleus, or in the mercury daughter or gold granddaughter, was made from the energy

difference between the K and L conversion electron lines. The half-lives of the electron lines also indicated which transitions were associated with a particular metastable state or radioactive ground state.

3. Results

3.1 Thallium-199m and 200m. These nuclides were produced by bombarding a thick Au^{197} target with 22 MeV He^4 ions. Essentially only the α,n and α,2n reactions occur at this energy so that there was no problem with interfering radiations. The beam energy was quite reproducible, but was not accurately measured. At the nominal energy of 22 MeV, where most of the data were taken, the ratio of Tl^{199m} to Tl^{200m} produced was about 4 to 1. This ratio could be lowered by lowering the beam energy; a decrease of about 1.8 MeV changed the above ratio to nearly 1 to 1.

An electron spectrum from such 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, is shown in fig. 1. The resolution of this spectrum is about 1.2% down to energies of about 250 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. At the end of a particular bombardment appreciable amount of 7.4 hr Tl^{199} and 26 hr Tl^{200} had accumulated, and a section of this spectrum, taken a few minutes after the beam was off, is shown in fig. 2. This spectrum is quite clean and contains mainly the lines of Tl^{199}, but also the 368-keV transition of Tl^{200}. Spectra taken later confirmed that the half-lives were consistent with the known values.

A gamma-ray spectrum taken with the counters gate off during the beam

pulse is shown in fig. 3. 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 later.

The energies, shell assignments, and intensities of the electron lines shown in fig. 1 are listed in tables 1 and 2. The energies are based on the value of 367.0 keV for the Tl^{199m} transition as measured by Andersson et al.[1] The assignment of the lines as those due to Tl^{200m} and Tl^{199m} is quite clear, and is based on the variation of the relative intensity of these lines with He^4 ion energy, the measured lifetimes associated with the lines, and the identification of the long-lived radioactivities produced in the bombardment.

The half-lives of the various electron lines were measured for the most part from three spectra which were taken with the counters gated on for a given time 14, 49 or 84 msec after each beam pulse. The beam pulses occurred 100 msec apart during this measurement. The transitions of 29, 367, 382, and 720 keV all showed a half-life of 27±4 msec; whereas those of 213 and 539 keV gave 37±4 msec. The 353-keV transition was too weak to measure without more care, and this was not considered to be necessary. The 353-, 367-, and 720-keV transitions were already known to be associated with Tl^{199} from studies of Pb^{199} decay.

The levels in Tl^{200} indicated by the present work are shown in fig. 6 as heavy lines. The lighter lines are previously known levels of Tl^{200}, and for simplicity the gamma-rays interconnecting these previously-known levels have not been included. The K/L ratio of the 213-keV transitions is 0.25±0.05. which strongly suggests an E3 transition, and the 37 msec half-life makes this assignment virtually certain. The theoretical K/L ratio[6] is 0.21. The K/L ratio of the 539-keV transition is 3.5±0.4, and for a pure E2 transition of

this energy Sliv gives 3.4. Thus, with a ground state spin[7] of 2-, the spin sequence, 7+(E3) 4-(E2) 2-, shown in fig. 6 is suggested, and this is in good agreement with the other even-mass thallium isotopes. As a final check we have computed in column 7 of table 1 the total transition intensities for the 213- and 539-keV transitions, and they agree very well, as the cascade arrangement of fig. 6 requires.

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.[8] The transitions in table 2 suggest a [illegible] level at 749 keV with a 27 msec half-life. We measure the K/L ratio of the 367-keV transition as 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 7 of table 2. The K/L ratio of the 382-keV transition is 0.88±0.05. The theoretical E3 K/L ratio[6] is 0.90, and taken together with the 27 msec half-life, this clearly indicates an E3 transition. Again the total transition intensity is listed in table 2. 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.

According to the level scheme of Tl^{199m} in fig. 7, 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 7 of table 2. The 29-kev transition (presumably M2) represents about a 7% branching of the 749-keV level.

The spin of the ground state of Tl^{199} is measured[9] 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 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. A more complete measurement of the distribution would probably have been worthwhile, but was not undertaken since the spin 3/2 intermediate state is rather well established. 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[10] (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.

3.2 Thallium-197m. In order to produce Tl^{197m}, the He^4 ion energy was increased to about 42 MeV, the full energy of the Hilac. Also, a relatively thin gold target (5 mg/cm^2) was used in order to reduce the amounts of the heavier thallium isotopes produced. Under these conditions, the α,3n and α,4n reactions occurred predominantly, producing Tl^{198} and Tl^{197}. However, it was not easy to make a reliable estimate of the relative amounts of these isotopes produced.

The electron spectrum taken under these bombarding conditions, and with the counters gated off during the beam pulse is shown in fig. 4. 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} on the basis that they have been seen previously in studies of Pb^{197m} decay, and also because we know from the long-lived lines observed that Tl^{197} is being produced. 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. 11).

The data on the Tl^{197m} electron lines are given in table 3. Here we have used as an energy standard the previously known energy of the 222.45-keV transition.[11] Our value for the K/L ratio for this transition is 0.28±0.05. The theoretical value for an E3 transition[6] 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. In the previous work from Pb^{197m} decay,[11] two transitions at about this energy were observed:

384.8 (predominantly M1) and 386.7 (predominantly E2). Our energy of 385.0±1.0 keV is in much better agreement with the lower energy transition; however, our K/L ratio agrees much better with the higher energy one. We can only say that possibly our energy is wrong by 1.7 keV, or the previously measured K/L ratios are wrong since the L lines of the two transitions overlapped in energy on their spectra.

The level scheme we deduce is quite simple and is shown as heavy lines in fig. 7. The cascade arrangement requires equal intensities for the two transitions, and the agreement here is shown in the last column of table 3 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 spin of the ground state of Tl^{197} has been measured[9] 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.

3.3 Thallium-195m and Thallium-193m. These nuclides can be produced by bombarding natural rhenium with C^{12} ions. The C^{12},4n reactions on Re^{187} and Re^{185} produce the above thallium isotopes, respectively, and such reactions can be rather clean if the C^{12} ion energy is slightly above the barrier energy. To separate Tl^{195m} from Tl^{193m} one can produce the latter only by bombarding Ta^{181} with O^{16} ions.

In order to identify unambiguously the various isotopes produced, we bombarded rhenium with C^{12} ions of 59 and 67 MeV energy and also tantalum with O^{16} ions of 74, 79 and 94 MeV energy. The resulting data were rather clear, and in good agreement with previous identifications of the light thallium isotopes.[12,13] We made no attempt to study the even-mass thallium isotopes, but could easily see lines belonging to mass numbers 196, 194, and 192. The half-life of the prominent 424-keV line of Tl^{192} and/or Tl^{192m} was measured, and gave a 9.5±1.5 min decay. This is in reasonable agreement with the previous value of 11.4±1.4 min;[13] however, we did not observe a prominent 109-keV transition associated with this activity as was previously reported. Perhaps we made predominantly the ground state of Tl^{192} rather than the 7+ isomer which was probably made previously. Possibly the lines of the 109-keV transition were just masked by other lines that were present in this energy region on our spectra. Electron lines from Tl^{195} decay were identified, and also the half-life of the very prominent 241-keV electron line in the decay of Tl^{193} was measured, giving 25±3 min. This is in excellent agreement with the previous value.[13] For almost all these isotopes, the decay of the mercury daughters was also observed. Thus the association with mass number of the short lived activities was quite clear.

Figure 5 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 2), and the data on these lines is collected in table 4. A single transition of energy 365 keV is assigned to Tl^{193m}, and the intensities of the lines of this transition are given in table 5. 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. 11). 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. 7. Our data do not show that the 99-keV transition is E3; however, it was so assigned previously[1,11] 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 7 of table 4, and we feel these are sufficiently similar to be consistent with the cascade arrangement of fig. 7. 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. 7. In this case the E3 transition is not observed, and our experimental limit on the energy is less than 25 keV. The 365-keV transition, which was shown to be converted in thallium and to belong to mass number 193, appears to correspond to the usual first excited state-to-ground state transition of the odd-mass thallium isotopes. The K/L ratio of 3.8±0.4 indicates 71±10% E2 and 29±10% M1.

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).

4. Discussion

In all the odd-mass thallium isotopes studied thus far, the ground state has spin 1/2, and states having spins 3/2 and 5/2 occur systematically as low-lying excitations (see fig. 7). In the early work on thallium isotopes, these states were assigned as the $s_{1/2}$, $d_{3/2}$, and $d_{5/2}$ shell model configurations, respectively,[2] Subsequent studies of these levels, while entirely consistent with the spins and parities implied by the above assignment, were not completely consistent with their explanations as pure shell model states. Thus, particularly in the 5/2 state, appreciable collective admixtures seem to be indicated.[14,15]

New, and considerably stronger, evidence that collective states play a part in the low-lying excited states of the light thallium isotopes comes from the present work. The $Tl^{199,197,195,193}$ results discussed in the previous section show that E3 transitions proceed to the above-mentioned 3/2+ levels, and therefore that they very likely come from 9/2- levels. There is no 9/2- level available in this region from a strict single-particle model approach except for the $h_{9/2}$ orbital from the next shell, which is expected at a considerably higher energy in these nuclei. Thus, the predominant component of the observed levels is most likely not a pure single-particle orbital. One

way of producing such a 9/2- level is to couple the $h_{11/2}$ proton hole explicitly to two unpaired neutrons or neutron holes. Another less detaied way is to couple the $h_{11/2}$ proton hole to a collective 2+ excitation (vibration) of the residual even-even core. These are not basically different models, but correspond to two different methods of analysis of the same type of (collective) state.

The first method is an extension of the well-known coupling of three (or five or seven) nucleons or holes of spin j to a total spin of j-1.[16-18] Such states occur as the ground states in various low Z nuclei, e.g., O^{19}, $Na^{23,25}$, Mg^{23}—$(d_{5/2})^3_{3/2}$; Ti^{47}, Mn^{55}—$(f_{7/2})^{-3}_{5/2}$. These were anomalous cases in the original independent particle model. They occur still more frequently as low-lying excited states, and there is even a family of E3 transitions occurring in nuclei with 43, 45, 47 neutrons or protons involving such a state, $(g_{9/2})^{3,5,7}_{7/2} \xleftrightarrow{E3} p_{1/2}$, somewhat similar to the 9/2- $\xleftrightarrow{E3}$ $d_{3/2}$ case under consideration in the thallium isotopes. Even the reduced transition probabilities are not too different in the two series; about $10^{-2.5}$ $B(E3)_{sp}$ in the thalliums and 10^{-2}-$10^{-3.5}$ $B(E3)_{sp}$ for the nuclei $Se^{77,79,81}$, Ge^{75}, $Kr^{79,81,83}$, $Ag^{107,109}$, $Rh^{103,105}$ (ref. 20). But the thallium 9/2- level would be the first example of a <u>single</u> 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.

The second approach is an extension of the treatment of coupling an odd particle to the collective excitation of an even-even core. A general expression for the shift in energy of the level of spin I of the multiplet formed by coupling the particle (p) of spin j to the core (c) of spin j_c has been given by de-Shalit,[14]

$$\Delta E(I) = \sum_k \langle j_c j I | T^{(k)}(c) \cdot T^{(k)}(p) | j_c j I \rangle$$

$$= \sum_k (-1)^{j_c+j+I} \langle j_c || T^{(k)}(c) || j_c \rangle \langle j || T^{(k)}(p) || j \rangle$$

$$\times \quad (-1)^{2j_c+2j} W(j_c j j_c j; Ik)$$

where $W(j_c j j_c j; Ik)$ is a Racah coefficient that contains all the spin dependence. For an $h_{11/2}$ particle and core excitation of spin two, the multiplet contains 7/2-, 9/2-, 11/2-, 13/2- and 15/2- levels, and k can take on the values 0,1,2,3,4. The monopole term does not contribute to a splitting of the multiplet, but the other four terms all do. Since the relative importance of the dipole, quadrupole, and the higher terms are not known without a detailed calculation using a specific nuclear force, the expression cannot be used a priori to calculate the level sequence, but it is of interest to note that with just the dipole term, and a smaller admixture of the quadrupole term, the 9/2- level can be made to lie lowest in the sequence of collective (vibrational) levels.

The important feature of the above discussion is that it is possible to find an explanation for the existence of a 9/2- level in these thallium isotopes, and even to expect that it might lie fairly low in energy. That it indeed lies lowest of all the odd parity levels is surprising to us, but seems quite clearly shown by the data of the previous section. We might hope that such a seemingly unexpected result can indicate something fairly explicit about the nuclear forces involved.

Although, the 9/2- level lies lowest, the 11/2- and/or 13/2- levels must not lie very much higher 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, consider the case of Pb^{197m}. Electron capture transitions from this level represent 80% of its decay.[1] With a half-life of 42 minutes, this yields a partial half-life for electron capture of 52 minutes. But then assuming a typical log ft value of 8.5 for such a first forbidden unique transition would require the very unlikely decay energy of ~55 MeV. 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. The discovery and identification of the latter levels in some thallium isotopes would certainly shed additional light on the nature of these states, and such an investigation is in progress.

In the case of the newly observed E3 transition in Tl^{199}, the energy and reduced transition probability fit very well into the pattern established by the mass 197 and 195 transitions, as shown in fig. 7 and table 6. These systematics also indicate why the E3 radiation in Tl^{193} was not seen directly (experimental limit < 25 keV) but only through the 2.1 minute decay of the 365-keV M1-E2 cascade transition to the ground state. From fig. 7, the estimated energy of the E3 transition in Tl^{193} would be less than 30 keV. A similar upper limit for the energy of the E3 transition can be obtained from its half-life by extrapolating a value for the reduced transition probability from the sequence for the corresponding decays in $Tl^{199,197,195}$, and assuming that there is negligible electron capture branching from the 9/2- level. Such a calculation shows that the energy is less that 25 keV, and probably below the L-electron edge at 13 keV.

The nature of the excited states in the even-mass thallium isotopes is

probably even more complicated than those of the odd-mass nuclei, and little can be said about them. The energy of the new E3 transition in Tl^{200} fits in well with those of the other even-mass isotopes, as shown in fig. 6, and the reasonably constant B(E3) values (table 6) indicate the similar nature of these 7+ → 4+ transitions. It is perhaps worth mentioning that these E3 transitions in $Tl^{200,202,204}$ have 10-20 times larger B(E3)'s than the 9/2- → 3/2+ transitions in $Tl^{195,197,199}$ or the 13/2+ → 7/2- transition in Pb^{205}.

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Table 1. Electron lines in Tl^{200m}

E_{e^-} [a]	Conv. Shell	E_γ	Ave. E_γ	Relative Intensity	Multi-polarity	Trans. Intensity
127.9	K	213.4		25		
199.0	L	213.3	213	100	E3	220
211.1	M+N	213.7		40		
453.7	K	539.2	539	3.4	E2	205
523.3	L	538.6		1.0		

[a] Energies measured relative to the 367.0-keV transition of Tl^{199m}, ref. 1.

Table 2. Electron lines in Tl^{199m}

E_e-	Conv. Shell	E_γ	Ave. E_γ	Relative Intensity	Multi-polarity	Trans. Intensity
25.5	M	28.9	29		(M2)	
28.3	N	28.9				
267.7	K	353	353	10	75% M1- 25% E2	69
281.5[a]	K	367.0[a]	367.0[a]	97	23% M1- 77% E2	1420±240[b]
352.0	L	366.7		28		
296.6	K	382.1	382.1	100	E3	1290
367.7	L	382.4		114		
379.2	M	382.2		34		
635	K	720	720	0.25	(E2)	27

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

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

Table 3. Electron lines in Tl^{197m}

E_{e^-}	Conv. Shell	E_γ	Ave. E_γ	Relative Intensity	Multi-polarity	Trans. Intensity
137.4	K	222.9		28		
208.0[a]	L	222.45[a]	222.45[a]	100	E3	230
219.2	M+N	221.9		34		
299.2	K	384.7	385.0	14.6	18% M1-82% E2	260±40[b]
370.8	L	385.6		4.2		

[a] Taken as standard from Andersson et al., ref. 11.

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

Table 4. Electron lines of Tl^{195m}

E_{e^-}	Conv. Shell	E_γ	Ave. E_γ	Relative Intensity	Multi-polarity	Trans. Intensity
85	L	99	99	100	E3	148
95	M+N	98		44		
297	K	383	383	7.8	26% M1-74% E2	120±20[a]
368	L	383		2.1		
380	M+N	383		0.66		

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

Table 5. Electron lines of Tl^{193m}

E_e -	Conv. Shell	E_γ	Ave. E_γ	Relative Intensity	Multipolarity
279	K	365		100	
350	L	365	365	26	29% M1 71% E2
362	M	365		9.6	

Table 6. E3 Transition probabilities

Mass No.	E_γ (keV)	Measured total E3 half-life (sec)	Total Conv. coefficient	Photon $T_{1/2}$ (sec)	B(E3) ($e^2 \times 10^{-72} cm^6$)	$\frac{B(E3)}{B(E3)_{sp}}$ [a]
199	382.1	2.9×10^{-2}	0.24	3.6×10^{-2}	2.8×10^{-5}	3.7×10^{-3}
197	222.45	5.3×10^{-1}	2.27	1.79	2.6×10^{-5}	3.5×10^{-3}
195	99.1	3.6	152.5	5.52×10^{2}	2.4×10^{-5}	3.2×10^{-3}
193	< 25	1.26×10^{2}				
204	703	6.2×10^{-5}	0.0362	6.42×10^{-5}	2.3×10^{-4}	2.9×10^{-2}
202	459.8	5×10^{-4}	0.125	5.6×10^{-4}	4.9×10^{-4}	6.3×10^{-2}
200	213.3	3.7×10^{-2}	2.78	1.4×10^{-1}	4.3×10^{-4}	5.6×10^{-2}

[a] $B(E3)_{sp}$ is taken to be: $\frac{e^2}{16\pi} R_0^6$, where $R_0 = 1.45\times A^{1/3}\times10^{-13}$ cm.

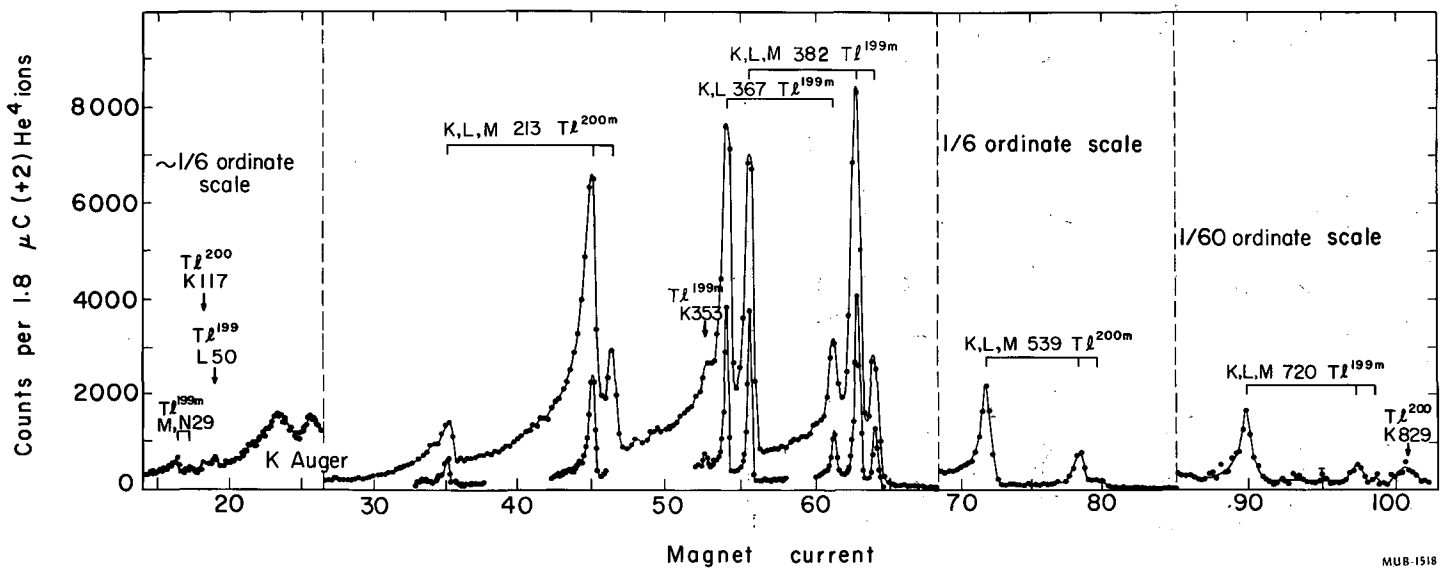

Fig. 1. Electron spectrum from Au^{197} irradiated by 22 MeV He^{4}. The counter was gated on only between the beam pulses. The upper curve was taken at 1.2% resolution, while the lower portions were taken at 0.6% resolution.

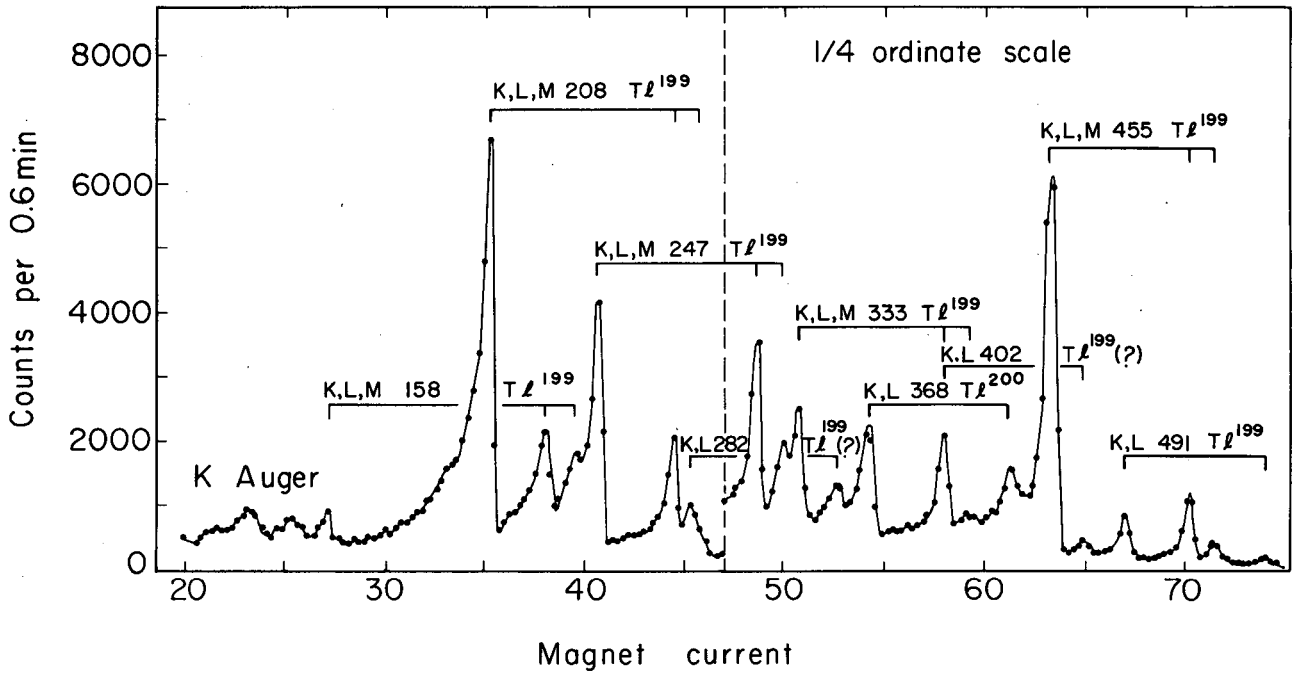

Fig. 2. Electron spectrum from Au^{197} after irradiation by 22 MeV He^{4}. Conversion lines are principally from the decay of $Tl^{199,200}$.

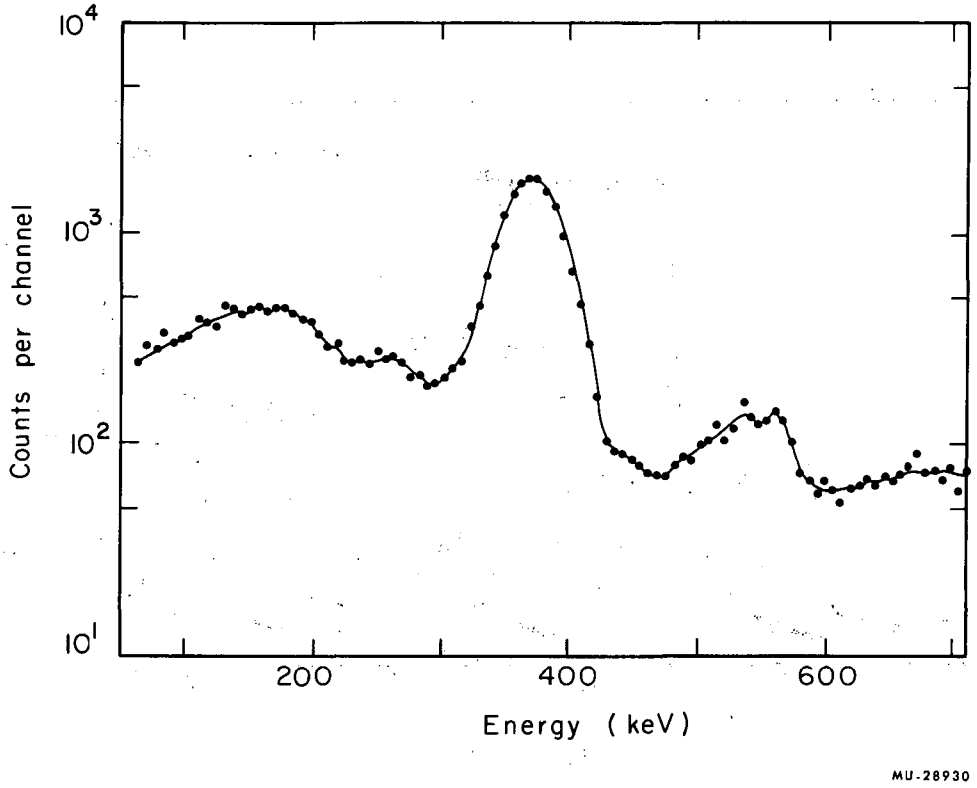

Fig. 3. Gamma-ray spectrum of Au^{197} irradiated by 22 MeV He^4. The counter was gated on only between the beam pulses.

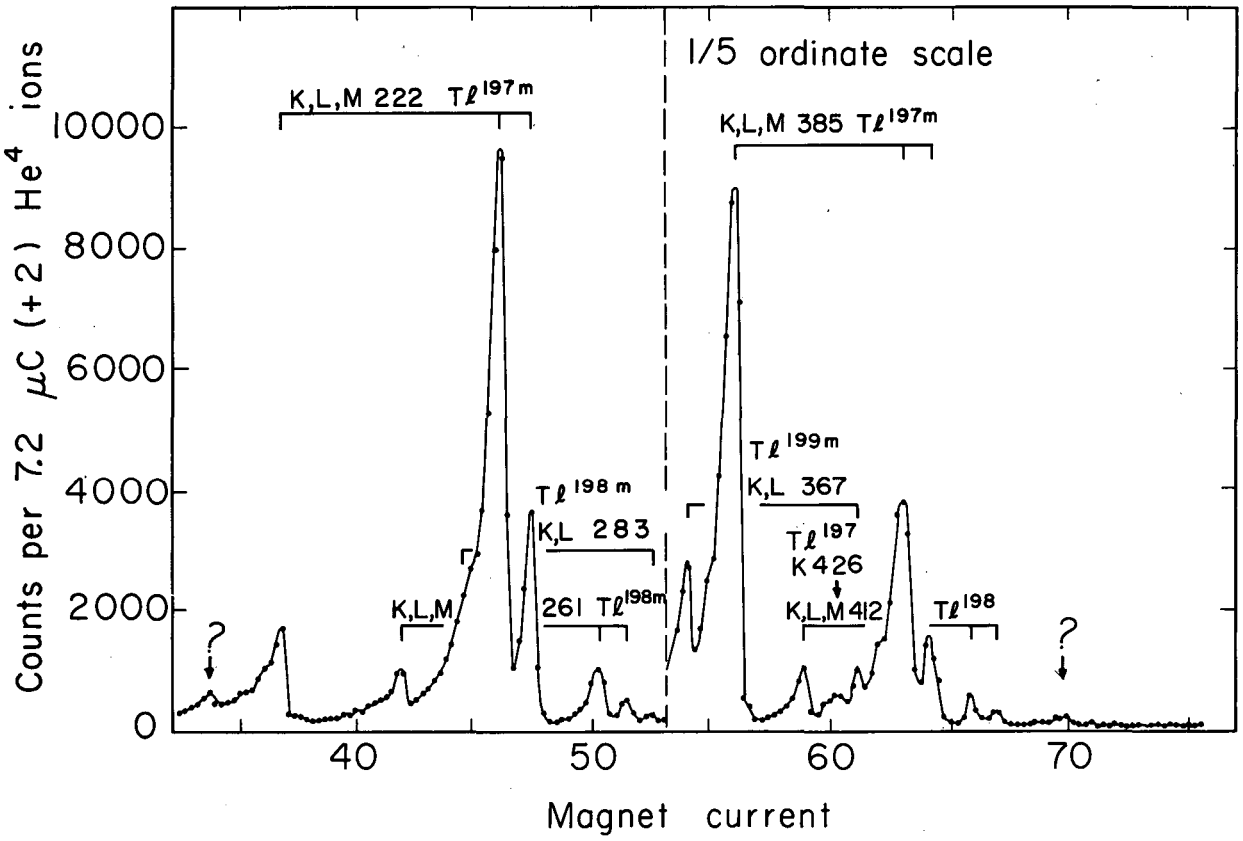

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Fig. 4. Electron spectrum from a thin Au^{197} target irradiated by 42 MeV He^4. The counter was gated on only between the beam pulses.

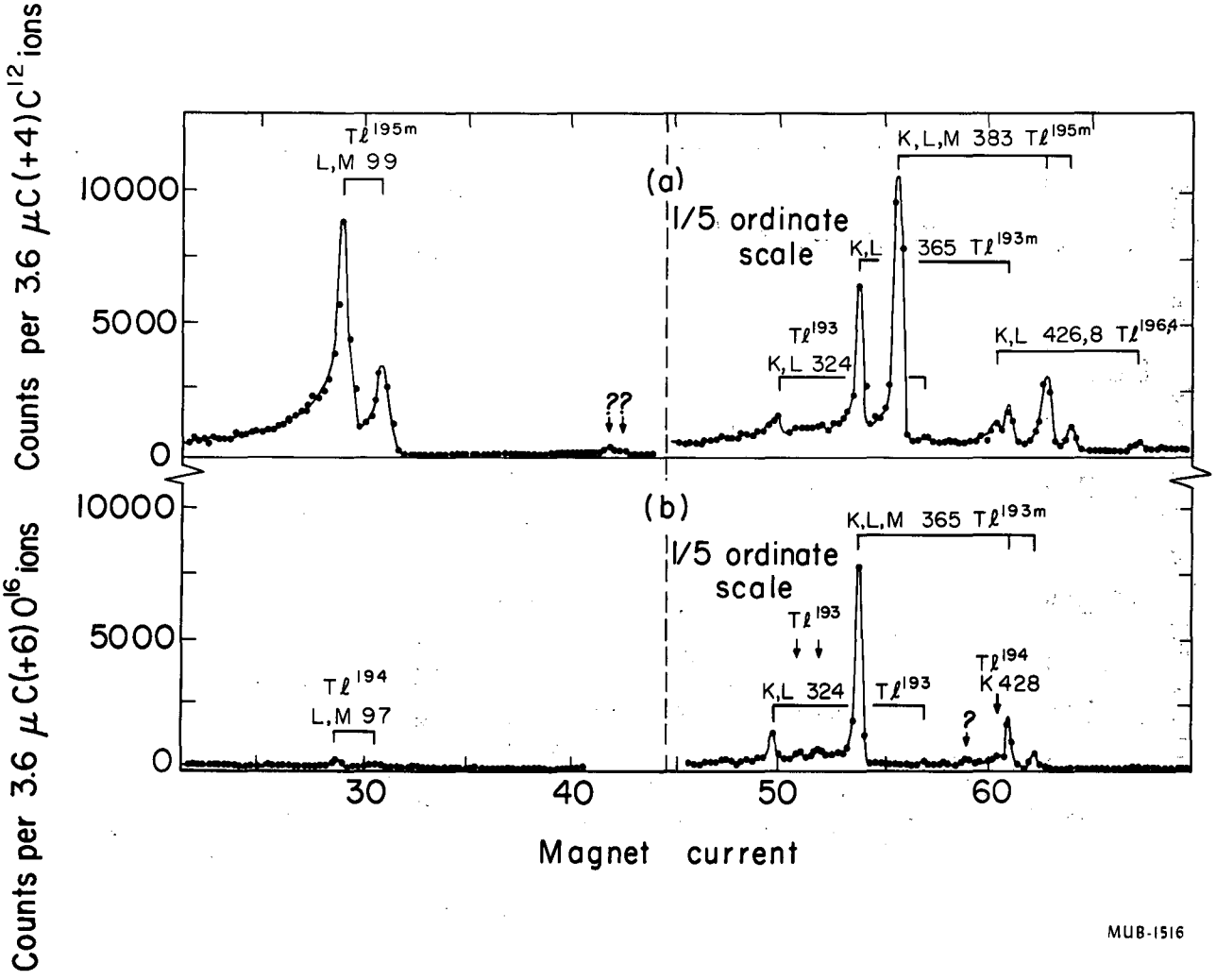

Fig. 5. Electron spectrum from (a) natural rhenium target irradiated by 67 MeV C^{12} and (b) Ta^{181} target irradiated by 79 MeV O^{16}. The counter was gated on only between the beam pulses.

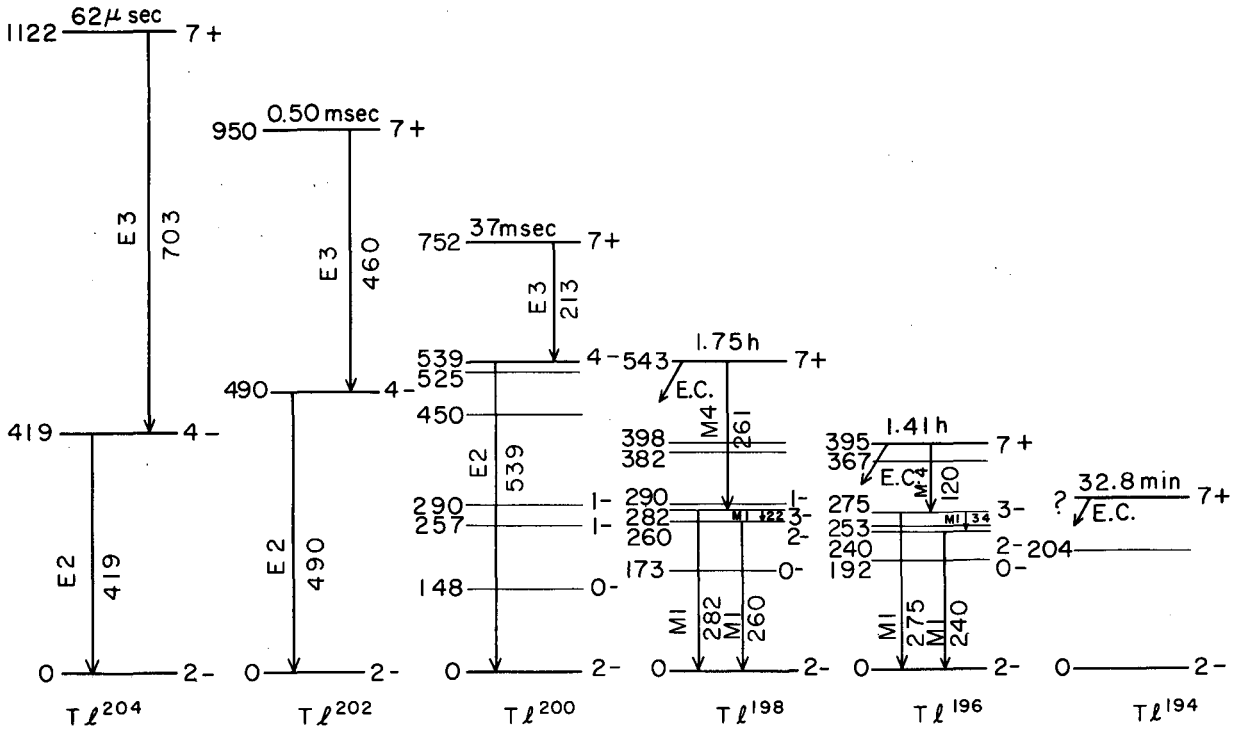

Fig. 6. Level schemes of the even-mass thallium isotopes. Transitions not relevant to the decay of the 7+ states have not been included.

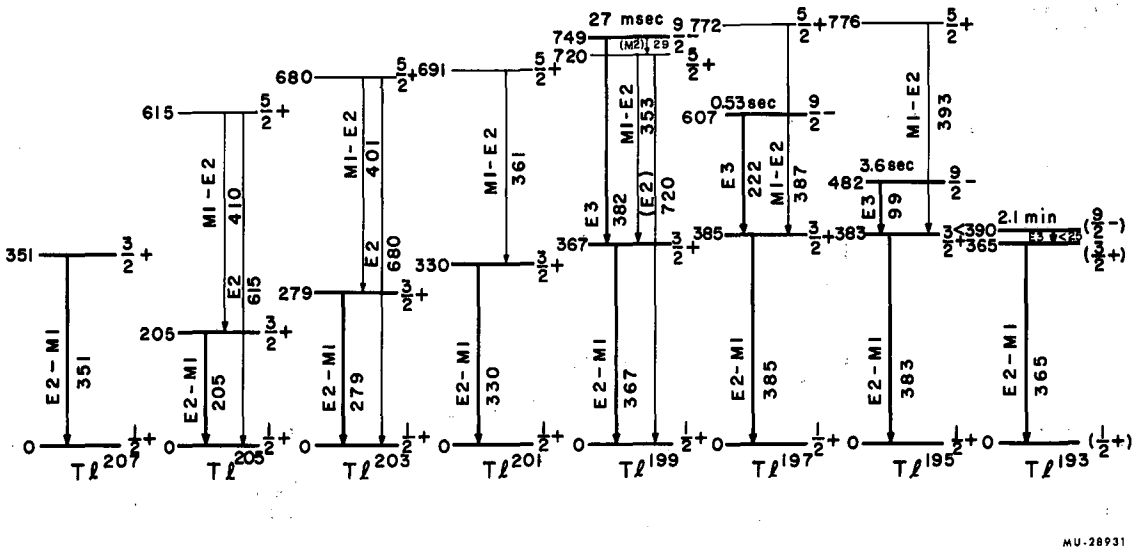

Fig. 7. Level schemes of the odd-mass thallium isotopes.

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