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POSSIBLE OBLATE SHAPE OF 9/2- ISOMER IN $^{199}\text{Tl}$

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Abstract: Low-lying 9/2- isomeric states have been previously observed in the neutron-deficient odd-mass thallium nuclei from mass 201 to 193. But there has as yet been no satisfactory explanation for the existence of such states, as the closest single-particle state is the $^9_9/2$ orbital some $4\text{ MeV}$ higher in energy. We have studied the transitions and states leading to this isomeric level in $^{199}\text{Tl}$ by in-beam spectroscopic techniques. Among the most prominent states observed above the isomeric level are an 11/2-, 13/2-, and possibly a 15/2- state. Very similar levels have been found in the other light odd-mass thallium nuclei. Combining a pairing-energy correction suggested by Blomqvist with the idea that the observed negative-parity levels form a rotational band based on the 9/2-[505] Nilsson state in a nucleus with oblate deformation seems to explain all the information presently available on these levels.

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1. Introduction

Low-lying $9/2^-$ isomeric states have been shown$^{1,2}$ to exist in the odd-mass thallium nuclei between mass number 193 and 201. The excitation energy of these isomers varies in a systematic way with mass number as can be seen in fig. 1. In order to throw more light on the nature of these and other states in the neutron-deficient thallium nuclei we have studied the gamma rays following production of the nuclides in (heavy-ion, xn) reactions. In addition, we have in some cases studied the $\beta$-decay of the odd-mass lead isotopes into the thalliums. This paper reports on $^{199}$Tl, and the results on the other nuclides will be presented in a later publication.

The (heavy-ion, xn) reaction has only rather recently been applied to studies of odd-mass final nuclei$^{3-5}$, though many studies have been made of doubly-even nuclei. In this work we have studied a final odd-mass nucleus near a closed shell. Such nuclei do not, in general, exhibit the rather simple systematic features, such as rotational bands, which occur in deformed nuclei and are a great help in interpretation. Therefore a much more detailed study is required to obtain useful information on these near-closed-shell nuclei than has often been done in the other cases.
2. Experimental Method

The helium beams were produced by the Berkeley Heavy-Ion Linear Accelerator, Hilac, and were magnetically analysed and focussed with a quadrupole lens onto the targets. The beam energies were measured by detecting ions scattered from a thin gold foil into a diffused-junction silicon detector, and comparing the pulse height with that obtained from the full energy beam of the Hilac (\( \sim 10.3 \text{ MeV/AMU} \)). The duty cycle ranged from 5 to 50%, and measurements were taken both during and between the beam pulses.

The details of the target arrangements for the gamma-ray measurements have been previously described\(^6,7\). Both thick and thin gold foils were used as targets. The gamma rays were detected in Ge(Li) counters, which had areas of 6 cm\(^2\) and thicknesses ranging from 0.8 to 1.3 cm. The resolution of these detectors at 1.33 MeV varied from about 5 keV in the earlier measurements to 2 keV in the later ones.

In the \( \gamma-\gamma \) coincidence measurements the Ge(Li) detectors were mounted opposite each other at 90° to the beam direction and about 2 cm from the target. A conventional fast-slow coincidence arrangement was used, and the whole detection and analysis system was interfaced to a PDP 7 computer. All the relevant information for each coincidence event was stored on IBM tape and was sorted afterwards. Without this arrangement, coincidence measurements of this type, where many gamma rays are involved, would be prohibitively expensive in time.

In order to obtain information on the multipolarities of transitions, the conversion-line spectra were also measured using a single-gap wedge spectrometer. The method of detection and the field stepping device have been described previously\(^1\).
Approximately 5 mg/cm$^2$ targets were used for the measurement of the conversion electrons. Gamma spectra and conversion-electron spectra were taken with the same targets at the same bombarding energies and at 90° to the beam direction. After the appropriate corrections had been made, relative conversion coefficients could be obtained. Absolute conversion coefficients could then be derived by normalising to that of a transition of known multipolarity.
3. Results

3.1. GAMMA AND ELECTRON SPECTRA.

The $^{199}\text{Tl}$ was produced by the $^{197}\text{Au}(^{4}\text{He},2n)^{199}\text{Tl}$ reaction. The yield of the observed gamma-ray lines, both during and between beam pulses, was studied with thin targets over a range of bombarding energies from 20 to 42 MeV. This was done both to ensure correct isotopic assignment and because the relative variation of transition intensity with bombarding energy in $^{4}\text{He},2n$ reactions can often give useful information about the spin of the state from which the transition arises.

An in-beam gamma-ray spectrum taken with a $6\, \text{cm}^2 \times 1.3\, \text{cm}$ Ge(Li) detector at $90^\circ$ to the beam direction is shown in fig. 2. The out-of-beam spectrum, shown in fig. 3, contains principally the lines from the decay of the 27 ms $^{199}\text{Tl}$ isomer. An in-beam conversion-electron spectrum, taken at the same energy and angle to the beam direction, is shown in fig. 4. The conversion coefficients at $90^\circ$ were obtained by normalising to the theoretical conversion coefficient (Sliv and Band\textsuperscript{8}) for the 382 keV E3 transition from the decay of $^{199}\text{mTl}$ (Ref. 1)). The $90^\circ$ conversion coefficients, $\alpha(90^\circ)$, may differ from the normal angle-integrated conversion coefficients, $\alpha$, when the gamma rays are emitted from aligned states and are therefore not isotropically distributed in space. The difference between the two does not usually exceed about 20%, even for very anisotropic angular distributions. However it does depend on the particular multipole mixture, being different, for example, for the same dipole to quadrupole admixtures of E1, M2 or M1, E2.
In table 1 are shown the energies, angle-integrated gamma intensities, 90° conversion data and angular distribution coefficients for the transitions which could be assigned to $^{199}$Tl. The isotopic assignments were made from excitation function data, which were in some cases confirmed by coincidence measurements; the degree of confidence in the assignment is indicated. The 90° conversion coefficients, rather than the angle-integrated ones, are listed, because values for the latter depend on definite multipolarity assignments which could not always be made.

3.2. EXCITATION FUNCTIONS

The excitation function for the $(^4\text{He},2n)$ reaction on gold is expected to have its maximum cross section at about 29 MeV, while the $(^4\text{He},n)$ and $(^4\text{He},3n)$ reactions should have their maxima at about 20 and 44 MeV, respectively. The probability of charged-particle emission is very small for such heavy nuclei not very far from the $\beta$-stability line, so that there is no problem in assigning the stronger gamma-ray lines to a given reaction. But ambiguities are bound to arise with the weaker lines.

The yields of various gamma rays, relative to that for the out-of-beam isomeric transition, as functions of bombarding energy are shown in fig. 5. For the $^{181}$Ta($^4\text{He},2n$)$^{183}$Re reaction$^3$, which leads to a rotational-type nucleus, it was shown that the shape of such a relative excitation function was strongly and in this case fairly uniquely related to the spin of the state from which the gamma ray arose. In the case of non-rotational nuclei, such as $^{199}$Tl, there is likely to be much less regularity in level structure than in a rotational nucleus. Thus, the relationship between the spin of a level and its relative excitation function may not be quite so strong. It
should, however, remain a useful tool in deciding between different spin assignments, and from fig. 5 one can see that there are indeed wide variations in the relative excitation functions.

3.3. COINCIDENCE MEASUREMENTS AND LEVEL POSITIONS

The coincidence relationships of most of the principal gamma rays were established by the measurements. The gamma rays found to be in coincidence with the various transitions in $^{199}$Tl are given in table 2.

The first three excited states had previously been established by Diamond and Stephens\(^1\) and by Andersson et al.\(^9\), fig. 1. Present coincidence data lends further support to this previous interpretation, which we shall take to be correct.

The 369 keV gamma ray is the strongest in the spectrum, apart from the 367 keV gamma from the first excited state. Also it has more gamma rays in coincidence with it than any other transition. The 332, 629 and 701 keV gamma rays are the next three strongest. The sum of the energies of the 369 and 332 keV lines are consistent with the energy of the 701 keV line, and all three are in coincidence with the 629 keV line. These coincidence results, taken together with the intensities, firmly establish states at 1117.8 keV, 1450.0 keV and 2079.5 keV. The 417 keV transition is in strong coincidence only with the 332 and 369 keV lines. Since it cannot come from the decay of the 749 keV state, it must precede the 332 keV transition and therefore arise from a state at 1866.6 keV. It is tempting from energy sums, to assign the 748.5 keV gamma ray as arising from this state also and leading to the 1118 keV state, though the coincidence data is not conclusive on this point. In
fact, the coincidence data, with its admittedly poor statistics in this case, might suggest that the 748.5 keV transition terminates in the 1450.0 keV level. The state at 1716.6 keV is also firmly established from the coincidence data on the 598.8 keV gamma ray. The 363.2 keV transition can be assigned to the decay of the 2097.8 keV state to the 1716.6 keV state, both from coincidence data and from energy sums. The states at 1984.8 keV and 1205.0 keV are assigned on the basis of coincidence data alone; there are no supporting energy sums. Rather more tentative are the proposed states at 1943.3 keV, assigned on the basis of coincidence data with poor statistics, and at 1394.2 keV. The latter assignment is based on the fact that the fairly strong 645 keV gamma ray hardly appears to be in coincidence with any other transition. This could be explained if it led directly to the ground state or to the 749 keV isomeric state or, rather less likely, if it led to another isomeric state which decayed by means of an unobserved transition of low energy. We have preferred to place the state at 1394 keV rather than at 645 keV for two reasons, neither of them very conclusive. There is no evidence from the systematics of the other thallium isotopes (see fig. 1) for any state, other than the 3/2^+ and 9/2^- states, lying below the 5/2^+ state. Furthermore, if the state were at 645 keV, its spin could hardly be more than 5/2. In this case one might have expected it to have a relative excitation function similar to that of the 353 keV gamma ray, which it does not appear to have. The evidence for all except the two last-mentioned states seems very firm.
4. Deduction of State Parameters

4.1. LIFETIME CONSIDERATIONS

The single-particle lifetime for an E3 transition of ~ 350 keV energy is of the order of a millisecond, so the fact that many of the gamma rays are seen to be in coincidence within a resolving time of 40 ns or less is sufficient to exclude them as being octupole or higher multipole transitions. For this reason we only consider dipole and quadrupole transitions in the following discussions.

4.2. INTERPRETATION OF ANGULAR DISTRIBUTION MEASUREMENTS

The gamma-emitting states formed in (heavy-ion,xn) reactions are normally strongly aligned because of the high orbital angular momentum brought into the compound system by the incoming projectile, this angular momentum being in the $m = 0$ magnetic substate \(^6,10\). The misalignment caused by the spins of the projectile and target and by the subsequent emission of neutrons and gamma rays is usually not great, and probably depends most strongly on the detailed way in which the state is fed in the last stages of the gamma-ray cascade. For example, if a significant number of the final gamma rays feeding a low spin state involve a non-stretched transition, the alignment may be appreciably less than it would be if predominantly stretched transitions were involved. The experimental data so far obtained suggest that the variation of alignment from case to case is not great, though not negligible either \(^6\).

If we have a state of spin $J_i$ decaying to a state of spin $J_f$ with a mixed transition of multipolarities $L_1$ and $L_2$, then the angular distribution of the gamma rays for complete alignment ($m = 1/2$) is given by
\[ W(\theta, \gamma) = 1 + A_2 P_2(\cos \theta) + A_4 P_4(\cos \theta) + \cdots \]  

(1)

where

\[ A_k = \frac{1}{1 + \delta^2} \left\{ f_k(J_f L_f L_i J_i) + 2\delta f_k(J_f L_f L_i L_i J_i) + \delta^2 f_k(J_f L_f L_f L_i L_i J_i) \right\} \]  

(2)

The coefficients \( f_k \) have been tabulated by Yamazaki\(^{11}\). The mixing ratio is given by

\[ \delta = \frac{\langle J_f L_f | J_i \rangle}{\langle J_f L_f | l_i \rangle} \]  

(3)

If the state \( J_i \) is not completely aligned, then the angular distribution is given by

\[ W(\theta, \gamma) = 1 + G_2(J_i) A_2 P_2(\cos \theta) + G_4(J_i) A_4 P_4(\cos \theta) + \cdots \]  

(4)

where the attenuation coefficients \( G_k \) depend only on \( J_i \) and the population of magnetic substates in the decaying state. The population distribution amongst the magnetic substates is not normally known, though it has been shown\(^6,10\) that a Gaussian distribution gives a rough approximation to it. If one assumes a Gaussian distribution, it is easy to show that the coefficients \( G_4, G_6, \text{etc.} \), decrease in magnitude much more quickly than does \( G_2 \) as the distribution is broadened. For example when \( G_2 \) has been reduced to 0.6, \( G_4 \) has been reduced to 0.2 and \( G_6 \) to 0.05. Thus in these reactions the coefficients of the Legendre polynomials of order 4 tend to be small and the still higher ones negligible.
Since the alignments for a particular state are not known precisely, we have adopted a number of procedures in order to interpret the experimental angular distributions. Some possibilities for the spin of a decaying state can be immediately rejected because the experimental values of the $P_2(\cos \theta)$ and $P_4(\cos \theta)$ coefficients are greater than those which would occur even if the state were completely aligned. Of course one must take account of sign as well as of magnitude. If two or more gamma rays come from one state then all of the angular distributions must be compatible with the same alignment, and hence $G_k$, for the state. This requirement may considerably reduce the number of possibilities and in some cases may allow the $G_k$ to be determined. A third and somewhat less certain procedure is to use the experimentally determined $G_k$ for other nuclei (mostly even-even) in order to interpret the angular distributions. Such experimental $G_2$ coefficients are plotted against the spin $J$ of the decaying state in fig. 6. For our purposes we have taken $G_2$ to have the values indicated by the solid line, with standard errors indicated by the broken lines. A similar procedure was used for $G_4$. Although it appears highly probable that the $G_k$ for the $^{197}$Au($^4$He,2n)$^{199}$Tl case should fit these curves, it is not certain, and therefore assignments made on this basis can only be regarded as having a good probability of being correct.

In order to simplify the discussion of the experimental angular distributions, examples of theoretical angular distributions for decay from completely aligned states as functions of $|\delta|(1 + |\delta|)^{-1}$ are shown in fig. 7. The quantity $\delta$ is the usual quadrupole to dipole amplitude mixing ratio. Although these curves refer to particular values of $J$, they do not vary very much with $J$ provided $J$ is not very small. Therefore for orientation purposes
one can, for example, take the 11/2 + 9/2 curve as being valid for any of the $J + J - 1$ cases which we discuss.

4.3. INTERPRETATION OF THE CONVERSION ELECTRON MEASUREMENTS

As indicated previously, the 90° conversion coefficients, $\alpha_k(90°)$, differ from the true (angle-integrated) conversion coefficients, $\alpha_k$, because the gamma rays and conversion electrons usually have different angular distributions.

Angular distributions for conversion electrons can be expressed in the same form as eq. (4). However the coefficients $A_k$ are now given by the expression

$$A_k = \frac{1}{1 + \delta_e^2} \left\{ b_{k1}(L_1,\pi)f_{k1}(J_{1L_1L_1}) + 2\delta b_{k2}(L_2,\pi)f_{k2}(J_{2L_2L_2}) \right\}. \tag{5}$$

The particle parameters $b_k$ depend both on the multipolarity of the transition and on its parity change $\Delta \pi$, i.e., on whether the multipoles are electric or magnetic. They also depend on the energy of the transition and the shell in which the conversion takes place. Values for these parameters have been given by Band et al.\textsuperscript{13}. The mixing parameter $\delta_e$ is given in terms of $\delta$ (eq. (3)) by

$$\delta_e = \delta \left( \frac{\alpha(L_2,\pi)}{\alpha(L_1,\pi)} \right)^{1/2}. \tag{6}$$
where the \( \alpha(L, \pi) \) are the conversion coefficients. From these equations it is possible to calculate the 90° conversion coefficient in terms of the normal conversion coefficient, provided that the transition multipolarities are assumed.

If one restricts oneself to dipole and quadrupole admixtures, one finds that the maximum deviation between the \( \alpha_k(90°) \) and the \( \alpha_k \) occurs for pure El transitions between states of the same spin. In this case \( \alpha_k(90°) \) can be about 50% higher than \( \alpha_k \); this large deviation occurs because \( b_2(\text{El}) \) is negative and larger than unity. For the other cases the \( b_2 \) are positive and the deviations smaller, the largest of about 30% being for mixed M1+E2, \( J \rightarrow J - 1 \), transitions with \( \delta \approx 1 \). In most cases the deviations are appreciably less than this. Since the \( \alpha_k \) for the different multipolarities differ by factors of about three, the \( \alpha_k(90°) \) are still a fairly good indication of multipolarity, even if no information on the gamma-ray angular distributions is available. If, as here, such information is available, then of course the appropriate corrections can be applied to the \( \alpha_k(90°) \). The K/L ratios for 90° differ little from the integrated K/L ratios and the corrections are less than the experimental errors in our case.

4.4. LEVEL SPINS

4.4.1. The 1118 and 1450 keV states. The conversion coefficient of the 369 keV transition, which arises from the decay of the 1118 keV state, indicates that it is mainly M1 or an El + 30% M2 mixture. Because of the necessity for resolving the 367 and 369 keV peaks in both gamma and electron spectra, the error on the \( \alpha_k \) is rather large. The K/L ratio of 6.8 ± 1.5
favours M1, but does not completely exclude the value of 4.3 expected for the El + M2 admixture.

Unfortunately the angular distribution measurements were also made with detectors having sufficient resolution to resolve accurately the 367 and 369 keV lines. It is easy to correct the measured angular distribution of the composite line for that part of the 367 keV radiation which arises from the decay of the isomeric state. The intensity of this radiation, which is isotropic owing to the long lifetime, can be deduced from the out-of-beam data and the in-beam intensity of the 382 keV line. However, as is seen in other data taken with good resolution at a fixed angle, there is also a contribution to the intensity of the 367 keV transition arising from direct feeding from higher states. The angular distribution of this component is not known but can be fairly safely estimated. From this and previous work\textsuperscript{1,9} the transition is known to be about 30\% M1 and 70\% E2. The transitions from the first excited states of the odd thallium isotopes vary smoothly in energy and in mixing ratios (see fig. 1). The mixing parameter is known to have positive sign in the heavier isotopes\textsuperscript{14} and it would be most surprising if it did not have this sign also in \textsuperscript{199}Tl. Knowing the sign and magnitude of \( \delta \), and with an estimate of \( G_2 \) from fig. 6, we can estimate the contribution to the angular distribution of the directly fed 367 keV radiation. After making rather liberal allowance for errors arising from non-statistical effects, we can conclude that the \( A_2 \) coefficient for the 369 keV transition has the value \(-0.7 \pm 0.2\).
The conversion coefficient data tell us that the spin of the 1118 keV state must be 7/2, 9/2 or 11/2. The sign and magnitude of the $A_2$ coefficient exclude the 9/2 possibility (the sign also excludes 5/2 or 13/2). It is not possible to decide between the 7/2 or the 11/2 spins using the experimental $G_2$ values, though the 7/2 possibility looks the least likely of the two; either parity is possible.

We consider next the excitation function measurement where, however, again the 367 and 369 keV peaks were not resolved. It is easy to subtract the 367 keV component arising from the isomer, but it is not clear how one should correct for the direct component. In order to get some idea of the excitation function for the 369 keV line, we used the value for the ratio of 367 (direct) to 369, obtained from another measurement with good resolution at 27.5 MeV bombarding energy, and then assumed arbitrarily that the excitation function for the 367 (direct) component had the same shape as that for the 353 keV transition, which also arises from a state of low spin. Both the "excitation function" deduced from this procedure, and the one for the composite peak with only the isomer contribution subtracted, are shown in fig. 5. The points for the "369 keV line" do suggest a slight rise with increasing bombarding energy, indicating that the spin of the 1118 keV state is greater than 9/2. However this evidence can only be considered as very weak, particularly as the points for the composite peak show the opposite tendency. The conclusion on the 1118 keV state, obtained from data on the 369 keV gamma ray, is that the state has spin 11/2 or 7/2, with the former being the most likely.
The 1450 keV state decays to the 1118 keV state by means of the 332 keV transition and to the 748 keV 9/2^- state by means of the 701 keV transition. Considering the 701 keV transition first, all spins between 5/2 and 13/2 would be allowed for the 1450 keV state if this transition were dipole or quadrupole. The value of \( \alpha_k(90^\circ) \) for the 701 keV gamma ray is consistent with it being pure E2, E2 plus a small admixture of M1, or E1 plus about 5% M2. The 13/2^+ and 5/2^- possibilities, which would require pure M2, can therefore be rejected. The 9/2^- assignment can also be rejected, because the observed sign of the angular distribution cannot be achieved for this spin with nearly pure E2 radiation.

The value of \( \alpha_k(90^\circ) \) for the 332 keV gamma ray is consistent with it being nearly pure M1, or being about 75% E1 plus 25% M2. The measured K/L ratio for this transition has the value 6.8 ± 1, which is consistent with the theoretical value of 5.7 for an M1 transition, but not with that of 4.2 for a 75% E1 + 25% M2 transition. This conclusion, deduced from \( \alpha_k(90^\circ) \), is still valid when the angular distribution correction is applied; for the E1+M2 admixture, \( \alpha_k \approx 1.2 \alpha_k(90^\circ) \). We conclude therefore that the 332 keV transition is mostly M1 and that the 1118 and 1450 keV states have the same parity.

The magnitude of the measured \( A_2 \) coefficient for the 332 keV gamma ray exceeds the theoretical value for \( J \rightarrow J \) transitions with complete alignment. We can therefore exclude 11/2 or 7/2, whichever is the spin of the 1118 keV level, as an assignment for the 1450 keV state.

In order to try to resolve the remaining ambiguities in the spin assignments of these states we appeal to the excitation function and intensity data. As can be seen from fig. 5 the intensities of the 332 and 701 keV gamma
rays, which originate from the 1450 keV state, rise rapidly relative to that of
the 382 keV gamma ray, from the 9/2^- isomeric state, as the bombarding energy
is increased. This implies strongly that the spin of the 1450 keV state is
higher than 9/2, particularly since much of the feeding of the isomeric state
comes from the 1450 keV state. This conclusion is supported by the fact that
the 1450 keV state is one of the most strongly populated states and yet is
700 keV higher in energy than the isomeric state. In (HI,xn) reactions it
would be most unusual if two states of the same spin were separated by this
large amount of energy, and the higher of the two were strongly populated.

From these considerations it seems certain that the 5/2 and 7/2 spins
can be ruled out and that 9/2^+ is extremely unlikely. We regard the 13/2^-
assignment as being almost certainly correct, though on this type of evidence
there must always be some small probability of error. If the 1450 keV state
is 13/2^- then the 1118 keV state must be 11/2^-. In the further discussion of
other levels we shall assume these assignments to be correct; if they are not,
then of course any conclusions based on them may also be incorrect.

4.4.2. The 1717 and 2080 keV States. The 2080 keV state decays to the
13/2^- level at 1450 keV by means of the very strong 629 keV transition and to
the 1717 keV level by means of the 363 keV transition. The 629 keV gamma ray
is E1, so that the 2080 keV level can have spins 15/2, 13/2, or 11/2 with
even parity. The 13/2^+ possibility is ruled out by the negative sign of the
A_2 coefficient, but the other two are consistent with the angular distribution
result. There is no information on the angular distribution of the 363 keV
gamma ray, since it is not well resolved from the stronger 367 and 369 keV
gamma rays, though its conversion coefficient indicates that it is E1.
The relative excitation function of the 629 keV gamma ray rises with bombarding energy at least as quickly as that of the 332 keV line (see fig. 5) which suggests that the spin of the 1450 keV state is at least $13/2$. Moreover the fact that this state is populated almost as strongly as the $11/2^-$ state, which is 962 keV below it in energy, makes the $11/2^+$ possibility unlikely. We therefore conclude that the 2080 keV state is almost certainly $15/2^+$, though $11/2^+$ cannot be rigorously ruled out.

The 1717 keV state decays by means of the 599 keV transition to the $12/2^-$ state at 1118 keV. The $\alpha_k(90^\circ)$ for this gamma ray indicates that it is either pure E2, E2 plus a small fraction of M1, or E1 plus about 13% M2. The negative sign of the $A_2$ coefficient excludes $15/2$ and $7/2$ as spins for the state. For complete alignment and $11/2 \rightarrow 11/2$ gamma ray, the negative value of $A_2$ of greatest magnitude is $-0.41$ and occurs for nearly pure quadrupole radiation. Since predominantly dipole radiation would give a positive sign for $A_2$, the E1 + M2 possibility is definitely excluded if the state is $11/2$. And although the observed value of $-0.48 \pm 0.08$ for $A_2$ is consistent with the value for complete alignment, it does not agree with the value of $-0.23 \pm 0.04$ deduced for the expected alignment. Thus $11/2$ is rejected on the basis of being inconsistent with the empirical $G_k$'s.

It is not possible to decide between a spin of $13/2$ or $9/2$ on the basis of the angular distributions and the $\alpha_k$ of the 599 keV transition, nor is it possible to decide between the two possible multipole mixtures. The relative excitation function, which has poor statistical accuracy, suggests rather weakly that the spin of the 1717 keV state is greater than $9/2$. However, the
13/2 assignment seems almost certain to be correct, since the 2080 keV state is almost surely 15/2, which rules out the 9/2 possibility. Since the 363 keV gamma ray appears to be E1, we assign the 1717 keV state as 13/2− and the 599 keV transition as mainly E2.

4.4.3. The 1867 keV state. This state decays to the 13/2− level at 1450 keV by means of the 417 keV transition. It also possibly decays to the 11/2− level at 1118 keV via the 749 keV transition. The latter decay is however deduced only on the basis of energy sums, since the 749 keV line is not strong enough to show conclusively in the coincidence spectrum. The sign of A2 and the value of αk(90°) for the 417 keV transition excludes 17/2, 13/2 and 9/2 for the spin. The value of αk(90°) indicates that the transition is almost pure M1, or E1 plus about 30% M2. Spins of 15/2− or 11/2−, with either parity, are consistent with the data on the 417 keV transition.

The value of αk(90°) for the 749 keV transition indicates that it is E2 plus about 6±20% M1, or E1 plus about 10% M2. With these admixtures the value for A2 of 0.35 ± 0.16 is not consistent with spin values of 11/2− or 15/2+. Thus, provided that we accept that the 417 and 749 keV gamma rays originate from the same state, the angular distribution and conversion coefficient data allow spins of 15/2− or 11/2− for this state. Of these two possibilities, the excitation function data somewhat favours 15/2 and so does the intensity data. This state is fairly strongly populated and is 750 keV above the 1118 keV 11/2− state. However the 2079 keV state, which is 200 keV higher in energy and also probably 15/2, is twice as strongly populated as the 1867 keV level, so that this
argument must be treated with caution. On balance it would seem that the most likely assignment for this level is $15/2^-$. The $11/2^+$ possibility cannot be entirely excluded even if the 748 keV transition does depopulate this state. Since this is not certain, $15/2^+$ and $11/2^-$ are also possible assignments.

4.4.4. The 1205 keV State. This state decays by means of the 838 keV gamma ray to the $3/2^+$ level at 367 keV. Since the gamma ray is observed to be anisotropic, spin $1/2$ is excluded but $3/2$, $5/2$ or $7/2$, with either parity, are all allowed. There is no information on the conversion coefficient of this transition. The excitation function is in accordance with the above possibilities for the spin. The fact that this level is significantly, though admittedly not strongly populated somewhat favours the $5/2$ or $7/2$ assignments in view of its being over 800 keV higher in energy than the 367 keV $3/2^+$ state.

4.4.5. The 1985 keV State. The 535 keV gamma ray, which depopulates this level, leads to the 1450 keV $13/2^-$ level. The value of $0.010 \pm 0.004$ for $\alpha_k(90^\circ)$ is consistent with $E1$ ($\alpha_k^{(\text{theor})} = 0.007$); but, though not in good agreement with $E2$ ($\alpha_k^{(\text{theor})} = 0.018$), we cannot entirely exclude this possibility. Since pure $M2$ is excluded, the $17/2^+$ and $9/2^+$ possibilities are not allowed. The sign of the $A_2$ coefficient eliminates $13/2^-$ for the spin. If account is taken of the expected alignment of the state, then $15/2$ and $11/2$, with either parity, can be rejected on the grounds that the quadrupole or dipole mixing required to give the observed value for $A_2$ would give too large a value for $\alpha_k(90^\circ)$. The angular distribution data is consistent with decay by pure $E2$ and $17/2^-$ for the spin of the state, but only in poor agreement with $9/2^-$ for
the spin. Both angular distribution and conversion coefficient data are in
good agreement with $13/2^+$ for the spin and decay by almost pure $E1$ radiation.
Therefore $13/2^+$ seems the most likely assignment for this state, with $17/2^-$
as a possible but unlikely one owing to poor agreement with the measured
conversion coefficient. The excitation function for the 535 keV line has a
statistical accuracy too poor to throw any further light on this. However, the
rather weak population of the 1985 keV level compared to those of the nearby
levels at 1867 and 2079 keV, which probably have spin $15/2$, supports the $13/2^+$
assignment. If it had spin $17/2$ one might expect it to have at least as much,
and probably more, population than these nearby levels.

4.4.6. The 1943 keV State. This state decays by means of the 825 keV
transition to the $11/2^-$ state at 1118 keV. There is no information on the
conversion coefficient, but the sign of the angular distribution excludes
$15/2$ or $7/2$ for the spin of the state. Spins of $9/2$, $11/2$ or $13/2$ with either
parity are allowed. The weak population of the level would somewhat favour
the $9/2$ or $11/2$ possibilities.

4.4.7. The 1394 keV State. As previously mentioned there is some
uncertainty about the placing of this level, but we are assuming here that
the 645 keV gamma ray with which it decays leads to the $9/2^-$ isomeric state.
The sign and magnitude of the $A_2$ coefficient excludes $13/2$, $9/2$ and $5/2$ for
the spin of the state. The value of $\alpha(90^\circ)$ is consistent with the transition
being about an equal admixture of $M1$ and $E2$, or else $E1$ with about 20% $M2$.
Taking into account the probable alignment of the state, both of these
admixtures are consistent with the angular distribution data. The spin of the
state can therefore be $7/2$ or $11/2$ with either parity. The excitation function slightly favours the $7/2$ assignment but the fairly strong population of the state favours $11/2$; it is therefore not possible to differentiate between the two possibilities.

4.4.8. Summary. The information on the states deduced in this section is summarized in table 3. The values of $A_2$ given in column four are for the (hypothetical) completely aligned state, e.g., the observed value divided by the $G_2$ read from fig. 6. The proposed decay scheme is shown in fig. 8.
5. Discussion

As has been pointed out\(^1\), the interpretation of the 9/2\(^-\) states at such low excitation energies presents a problem. The simple shell model would predict a low-lying 11/2\(^-\) hole state arising from the \(h_{11/2}\) orbit, but the 9/2\(^-\) state, which could arise from the \(h_{9/2}\) orbit, would be expected to have an excitation energy of about 4 MeV. No quantitative explanations of this phenomenon have been advanced so far, though it has been suggested that the 9/2\(^-\) state might arise from a coupling between the \(h_{11/2}\) hole and, either two unpaired neutrons, or a collective oscillation of the core\(^1\). A recent and promising suggestion\(^{15}\) is that the simple shell model considerably overestimates the energy required to excite the odd proton to the \(h_{9/2}\) state in thallium. The main reason is that in the ground states of the odd thallium nuclei very little energy can be obtained from pairing correlations, since the nearest unblocked level into which proton pairs can scatter is far above the Fermi surface. (This level will in fact be the \(h_{9/2}\) level across the closed shell of 82 protons.) However, when a proton is excited to the \(h_{9/2}\) orbit there will be an unblocked level close to the Fermi surface and the pairing energy will be correspondingly increased. The excitation energy to the \(h_{9/2}\) orbit will therefore be reduced over the single-particle spacing by an amount equal to the gain in pairing energy. It was estimated empirically\(^{15}\) (by comparing the proton binding energies in \(^{203}\)Bi and in \(^{201}\)Tl and then correcting for the Coulomb energy) that this type of effect might reduce the excitation energy of the \(h_{9/2}\) state in \(^{201}\)Tl from \(\sim 4.2\) MeV (the \(s_{1/2}-h_{9/2}\) gap) to about 1.6 MeV, and further suggested that interactions involving the neutron holes might lower this still further to the observed energy of 0.91 MeV. This
suggestion seems valid; however, a remaining difficulty is that similar calculations for the lighter thalliums yield similar excitation energies for their 9/2- states, while experimentally these levels fall monotonically in energy to less than 390 keV in $^{193}\text{Tl}$.

In addition, our measurements on $^{199}\text{Tl}$ show the existence, not only of the low-lying 9/2- state, but also of 11/2-, 13/2- and possibly 15/2- states above it. The measurements on the odd thallium nuclei between mass number 191 and 197, which will be published later, also show the systematic existence of 11/2- and 13/2- states above the 9/2- states. In each case the spacing between the 9/2- state and the other two states is strikingly similar, as shown in fig. 9. This highly systematic behaviour suggests strongly that these states are closely related to one another, possibly in some collective manner. It would seem most improbable, for example, if the 9/2- state arose from the $h_{9/2}$ level and the 11/2- state arose from the $h_{11/2}$ level, that the spacing between them would remain so constant while their excitation energies varied so much. The intense E2 crossover gamma rays between the 13/2 and 9/2 and between the 15/2 and 11/2 states are also notable and support a collective interpretation for these odd-parity states.

We wish to point out a possible explanation for these levels which at first sight looks implausible, but for which a reasonably strong case can be made, nevertheless. This is that the odd-parity states arise from a rotational band based on the 9/2- (505) Nilsson state. This state is derived from the $h_{9/2}$ shell-model orbit, and in order for it to be a low-lying level, the excited thallium nuclei must have oblate deformation.
We shall first consider the evidence which leads us to believe that the thallium nuclei may have oblate deformation in the (505) state. Kumar and Baranger\textsuperscript{16}, using a pairing-plus-quadrupole model, have calculated the potential energy of deformation, $V(\beta,\gamma)$ for a large range of doubly-even nuclei in the rare-earth region. They find that the neutron-deficient mercury nuclei ($Z = 80$) have their potential minima for negative values of the deformation parameter $\beta$ and for $\gamma = 0$. These nuclei would therefore have oblate deformation if they were indeed deformed. However, the zero-point energy of vibration is estimated to be larger than the difference between the potential energy of deformation for the oblate minimum and for the spherical shape, so that no permanent deformation occurs and these nuclei are expected to be soft vibrators rather than rotors. The recent calculations of Tsang and Nilsson\textsuperscript{17}, based on a method which synthesizes the liquid drop and Nilsson models, support this conclusion. The odd thallium nuclei are obtained by adding one proton to the doubly-even mercury core. If the proton were placed in the lowest available shell-model orbit to produce the ground state, then indeed we would expect to get a spherical nucleus, since we have a closed shell of protons with only one hole. However, if we put the proton in the $h_{9/2}$ orbit we have a different situation altogether. Now we still have two holes in the shell, as with the mercury nuclei. This, as previously mentioned, will give an increased pairing energy over the single-hole case and the core will tend to favour the oblate shape. In addition, however, the energy of (a component of) the $h_{9/2}$ state can be lowered considerably by deforming the nucleus to either the oblate ($K = 9/2$) or prolate ($K = 1/2$) shape. For a given change in deformation parameter, $\beta$, slightly more energy is gained by going to
oblate rather than prolate deformation. In Table 4 we have tried to estimate the deforming effect of the $h_{9/2}$ level in a simple-minded way. We show the deformation and depths of the oblate potential minima together with the zero-point energies for a number of mercury nuclei as calculated by Kumar and Baranger\(^16\). It is apparent that the zero-point energy exceeds the depth of the minimum in all cases. In addition we give the reduction in energy of the $9/2^{-}[505]$ state in an oblate nucleus compared with a spherical nucleus, as estimated from Nilsson's energy diagram\(^18\). This will increase the depth of the oblate potential minimum for the odd thalliums in this state, and it can be seen that the depth is now greater than the zero-point energy for all nuclei shown with mass number 199 and less. Furthermore, the trend of energies of this state clearly indicates that its excitation energy will be lower the more neutron-deficient the thallium nucleus. On average, the lowering is by about the observed amount. We would not expect any of the present calculations of deformation to be highly reliable in this clearly rather critical region near to the closed shell, nor would we expect the estimate of the deformation energy of the $9/2^{-}[505]$ to be very precise. Nevertheless, we feel these considerations show that oblate deformation for the neutron-deficient odd thallium nuclei in this state is not only possible, but also likely.

An objection to the rotational interpretation of these levels might be that their spacings do not appear to be characteristic of a $K = 9/2$ rotational band. However, deformed states arising from a shell-model state of high spin are well known to have large Coriolis matrix elements connecting them, and such mixing can produce irregular spacings in the rotational levels. In the present case, this irregularity can be thought of as arising principally from mixing
with the $K = \frac{1}{2}$ state. The decoupling parameter of this state is expected from Nilsson's wave functions to have a value of about $+4.7$. The effect of a decoupling parameter of this magnitude is to lower, from their regular positions, the sequence of rotational states with spins $\frac{5}{2}$, $\frac{9}{2}$, $\frac{13}{2}$, etc., and to raise those with spins $\frac{3}{2}$, $\frac{7}{2}$, $\frac{11}{2}$, etc. This band will Coriolis mix with the $\frac{3}{2}$- [532] band, which lies below it, transmitting to this band a lower $\frac{5}{2}$ ... sequence relative to the $\frac{3}{2}$ ... sequence. This effect will be passed on to all of the states arising from the $h_{\frac{9}{2}}$ level, and hence to our $\frac{9}{2}$- [505] state, by successive operations of the Coriolis operator, which mixes states with $\Delta K = \pm 1$. This is just the effect which we need to explain the departures from regular spacing in the odd thallium levels. It is, however, necessary to show that the effect can be as large as the one seen.

The energies of the levels in a rotational nucleus can be written as:

$$E_I = E_0 + \frac{\hbar^2}{2J} I(I+1)[1+B'I(I+1)+ \cdots] + (-)^{I+1/2} A_{2K} \frac{(I+1)!}{(I-K)!} \left[ 1 + \frac{B_{2K}}{A_{2K}} I(I+1)+ \cdots \right]$$

where the first series is the usual $I(I+1)$ expansion, and the second series gives rise to the irregular spacings mentioned above. For the $\frac{9}{2}$- band in $^{199}\text{Tl}$ this equation is a poor approximation but, if it is at all applicable, $A_9$ must be of order $+1 \times 10^{-6}$ keV in order to give the observed spacings. To see if this value is plausible, we can estimate that, due to mixing with a $K = \frac{1}{2}$ band, $A_9$ is given to lowest order by:
\[ A_9 = a \frac{\hbar^2}{2J} \left[ \frac{(K \pm 1|J_\pm|K)}{\bar{W}} \right]^8 \]

where \( a \) is the \( K = 1/2 \) band decoupling parameter, \( (K \pm 1|J_\pm|K) \) is the mean value of the operator \( J_\pm \) among the states involved, and \( \bar{W} \) is the mean excitation energy of the \( K = 1/2, 3/2, 5/2, \) and \( 7/2 \) components of the \( \hbar_{9/2} \) orbital relative to the \( 9/2 \) component. From the Nilsson wave functions\(^\text{18}\) at \( \beta = -0.1 \) we can estimate that \( a \sim 4.7, (K \pm 1|J_\pm|K) \sim 3.9, \) and \( \bar{W} \sim 1.2 \) MeV. With a value of 30 keV for \( \hbar^2/2J \) (see below), this leads to a predicted value for \( A_9 \) of \( +1.4 \times 10^{-6} \) keV. The agreement in order of magnitude of this value with the observed value of \( +1 \times 10^{-6} \) keV, shows that the level spacings are reasonable for this interpretation.

A further test which we can apply to this hypothesis is to compare the observed E2 branching in \( ^{199}\text{Tl} \) from the \( 13/2 \) level to the \( 11/2 \) and \( 9/2 \) levels with that predicted by the simple rotational model. This model cannot strictly be applied since the states are mixed; however, we would not expect large deviations due to the mixing. The expected value for \( I(332 \text{ keV})/I(701 \text{ keV}) \) is \( 0.154 \) and the experimental value of \( 0.11^{+0.2}_{-0.06} \) for this quantity is in satisfactory agreement with it. If one assumes that the 749 keV gamma ray does come from the probable \( 15/2^- \) state at 1866 keV, then the experimental value of \( I(417)/I(749) \) of \( 0.09^{+0.1}_{-0.06} \) is also in satisfactory agreement with the rotational model prediction of \( 0.164 \).

We can also compare the E2/M1 mixing ratio of the 332 keV, \( 13/2 \) to \( 11/2 \) transition with what we might expect from the rotational model. We first estimate the E2 transition probability. To do this a value for the
quantity $\hbar^2/2\gamma$ for the band is deduced by taking an average of the values calculated from the $13/2 - 11/2$ and $11/2 - 9/2$ spacings. The value of 29.5 keV obtained is typical for a poor rotor, which is as might be expected. With this value for $\hbar^2/2\gamma$ we can estimate the energy of the $2^+$ state of a corresponding doubly-even nucleus, whose transition probability we can then estimate from the statistics of Grodzins. From this we can now deduce the $E2$ transition probability for the 332 keV transition. The $M1$ transition probability can be estimated from Nilsson's wave functions. From this procedure we obtain a value of $5 \times 10^{-2}$ for the mixing ratio in satisfactory agreement with the experimental value of $(9^{-}_{-5}) \times 10^{-2}$.

We conclude that the pairing-energy correction suggested by Blomqvist, together with the hypothesis that the $9/2^-$ level in the odd-mass thalliums is the $9/2^-\{505\}$ state in a nucleus with oblate deformation, is in satisfactory agreement with the present data. In order to test this suggestion further, data on the lifetimes of the $11/2^-$ and higher members of the proposed band would be especially valuable.

Acknowledgments

We would like to thank Drs. Chin Fu Tsang and J. Blomqvist for helpful discussions.
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17) C. F. Tsang and S. G. Nilsson, Nucl. Phys., to be published
Table 1: Transitions in $^{199}$Tl

<table>
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<tr>
<th>$E_\gamma$(keV)</th>
<th>$I(\gamma)^a$</th>
<th>$\alpha_K$(90°)</th>
<th>$K/L$(90°)</th>
<th>$A_C^c$</th>
<th>$A_L^c$</th>
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<td>181.2</td>
<td>7</td>
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<td>6.8 ± 1</td>
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(continued)
Table 1 (continued)

a The intensities are corrected for the angular distribution where possible. Otherwise they refer to the intensity measured at 90° to the beam direction.

b These intensities depend on the duty cycle.
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<th>Gamma Ray</th>
<th>Gamma Rays in Coincidence&lt;sup&gt;a&lt;/sup&gt;</th>
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<td>838</td>
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<sup>a</sup>Parentheses indicate uncertain assignments due to poor statistical accuracy or overlapping peaks.
Table 3

State properties

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<tr>
<th>State</th>
<th>Spin</th>
<th>Transition</th>
<th>$A_2(m = \frac{1}{2})$</th>
<th>Multipolarity</th>
<th>$\alpha_k$</th>
<th>$\delta$</th>
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<td>$\frac{11}{2}^-$</td>
<td>369</td>
<td>$-1.2 \pm 0.4$</td>
<td>M1 + E2</td>
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<td>E2</td>
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<td>$-0.80 \pm 0.2$</td>
<td>M1 + E2</td>
<td>0.017 ±0.0003</td>
<td>$-3^{+1}_{-3}$</td>
</tr>
<tr>
<td>1867</td>
<td>$\frac{15}{2}^-$</td>
<td>417</td>
<td>$-0.83 \pm 0.2$</td>
<td>M1 + E2</td>
<td>0.11 ±0.02</td>
<td>$-0.29 \pm 0.14$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>749</td>
<td>$+0.54 \pm 0.26$</td>
<td>E2</td>
<td>0.010 ±0.004</td>
<td></td>
</tr>
<tr>
<td>1985</td>
<td>$\frac{13}{2}^+$</td>
<td>535</td>
<td>$+0.52 \pm 0.17$</td>
<td>E1</td>
<td>0.008 ±0.003</td>
<td>$\pm(0.09^{+0.05}_{-0.09})$</td>
</tr>
<tr>
<td>2080</td>
<td>$\frac{15}{2}^+$</td>
<td>629</td>
<td>$-0.26 \pm 0.09$</td>
<td>E1</td>
<td>0.0045±0.0015</td>
<td>$+0.03 \pm 0.05$</td>
</tr>
</tbody>
</table>
Table 4

Energies for oblate shapes in the Tl nuclei

<table>
<thead>
<tr>
<th>Neutron Number ( N )</th>
<th>( \beta(\text{Hg})^a )</th>
<th>( E_{\text{def}}^b ) (MeV)</th>
<th>Zero Point Energy</th>
<th>( E_{\text{zp}}^c + E_{\text{def}}^d ) (MeV)</th>
<th>( A(\text{Tl}) )</th>
<th>( E_\beta^e(9/2-[505])^c )</th>
<th>( E_{\text{zp}}^f + E_{\text{def}}^g + E_\beta^h ) (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>112</td>
<td>-0.135</td>
<td>-0.93</td>
<td>1.42</td>
<td>0.49</td>
<td>193</td>
<td>-1.50</td>
<td>-1.01</td>
</tr>
<tr>
<td>114</td>
<td>-0.128</td>
<td>-0.77</td>
<td>1.49</td>
<td>0.72</td>
<td>195</td>
<td>-1.41</td>
<td>-0.69</td>
</tr>
<tr>
<td>116</td>
<td>-0.116</td>
<td>-0.61</td>
<td>1.56</td>
<td>0.95</td>
<td>197</td>
<td>-1.28</td>
<td>-0.33</td>
</tr>
<tr>
<td>118</td>
<td>-0.099</td>
<td>-0.38</td>
<td>1.00</td>
<td>0.62</td>
<td>199</td>
<td>-1.10</td>
<td>-0.48</td>
</tr>
<tr>
<td>120</td>
<td>-0.080</td>
<td>-0.15</td>
<td>1.10</td>
<td>0.95</td>
<td>201</td>
<td>-0.88</td>
<td>+0.07</td>
</tr>
<tr>
<td>122</td>
<td>0</td>
<td>0</td>
<td>1.42</td>
<td>1.42</td>
<td>203</td>
<td></td>
<td></td>
</tr>
<tr>
<td>124</td>
<td>0</td>
<td>0</td>
<td>2.52</td>
<td>2.52</td>
<td>205</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\( a \beta(\text{Hg}) \) is the deformation at the oblate minimum in the potential of the Hg nucleus.

\( b \) \( E_{\text{def}} \) is the energy of the oblate potential minimum with respect to the potential energy for \( \beta = 0 \).

\( c \) \( E_\beta^e(9/2-[505]) \) is the difference between the energy of this state at a deformation of \( \beta(\text{Hg}) \) and that at \( \beta = 0 \).

\( d \) The deformation may be stable provided that \( E_{\text{zp}}^f + E_{\text{def}}^g + E_\beta^h \) is < 0.
Figure Captions

Fig. 1. Partial level schemes for the light odd-mass thallium isotopes possessing the $9/2^-$ level. Data are taken from Ref. 1) and 2).

Fig. 2. Gamma-ray spectrum, taken during the beam pulses, produced by 27 MeV helium ions on gold.

Fig. 3. Gamma-ray spectrum, taken between beam pulses, produced by 27 MeV helium ions on gold.

Fig. 4. Conversion-electron spectrum produced by 27 MeV helium ions on gold.

Fig. 5. Yields of various gamma rays, relative to that of the 382 keV gamma ray, as a function of bombarding energy.

Fig. 6. Experimental values for $G_2$ and $G_4$ plotted against the spin J of the decaying state, taken from Ref. 7). A few error bars are shown when the errors are large and only a few data exist for a given J. Errors in the lower values of J are usually small. The continuous lines indicate the average values we take for $G_2$ and $G_4$. The broken lines are placed at what we have taken to be one standard deviation on either side of the average lines.

Fig. 7. Examples of theoretical gamma-ray angular distribution coefficients for various transitions from states completely aligned in the $m = \pm 1/2$ subsate, as functions of $|\delta|(1 + |\delta|)^{-1}$. The dashed line is $A_4$.

Fig. 8. Proposed decay scheme for $^{199}$Tl. Uncertain levels and transitions not definitely placed are shown by broken lines. When more than one spin assignment is given the less likely ones are placed in brackets. Intensities are total transition intensities and include any component between beam pulses.

Fig. 9. Systematics for the $9/2^-$, $11/2^-$, $13/2^-$ and $15/2^-$ states in the odd-thallium nuclei derived from (heavy-ion,xn) experiments.
Fig. 1.
Fig. 2.
Fig. 3.
Fig. 4.
Fig. 5.
Fig. 6.
Fig. 7.
Fig. 8.
Fig. 9.
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